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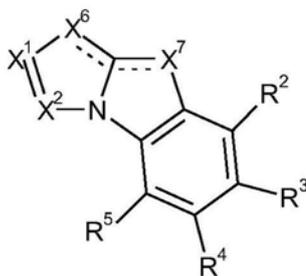
(54) 发明名称

用于电子应用的4H-咪唑并[1,2-a]咪唑

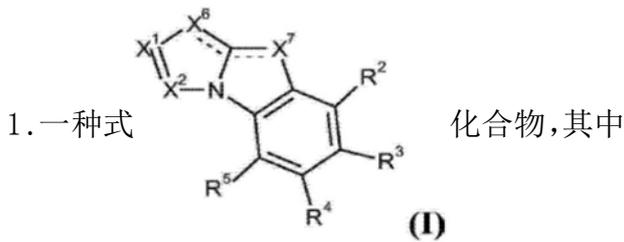
(57) 摘要

本申请涉及用于电子应用的4H-咪唑并[1,2-a]咪唑。本发明涉及式(I)化合物,一种其的生产方法及其在电子器件,尤其是场致发光器件中的用途。当在场致发光器件中的磷光发光体中用作主体材料时,式(I)化合物可以提供具有改进效率、稳定性、可制造性或光谱特性的场致发光

器件。



(I)



X^6 为-N=且 X^7 为-NR¹-, 或

X^7 为=N-且 X^6 为-NR¹-,

R^1 为式-A¹- (A²)_p- (A³)_q- (A⁴)_r-R⁶的基团,

p为0或1, q为0或1, r为0或1,

A¹、A²、A³和A⁴相互独立地为可以任选被G取代的C₆-C₂₄亚芳基, 或可以任选被G取代的C₂-C₃₀亚杂芳基, 其中

基团A¹、A²、A³和A⁴可以被一个或多个基团-(SiR⁷R⁸)-间隔;

R²、R³、R⁴和R⁵相互独立地为H, 可以任选被E取代和/或被D间隔的C₁-C₂₅烷基, 可以任选被G取代的C₆-C₂₄芳基, 或可以任选被G取代的C₂-C₃₀杂芳基;

R⁶为H, 基团-(SiR²⁰R²¹R²²), 可以任选被G取代的C₆-C₂₄芳基, 或可以任选被G取代的C₂-C₃₀杂芳基;

R⁷和R⁸相互独立地为C₁-C₂₅烷基, 或C₆-C₂₄芳基, 其可以任选被G取代; X¹为N或CR⁹;

X²为N或CR¹⁰,

R⁹和R¹⁰相互独立地为H, 可以任选被E取代和/或被D间隔的C₁-C₂₅烷基, 可以任选被G取代的C₆-C₂₄芳基, 或可以任选被G取代的C₂-C₃₀杂芳基, 或

R⁹和R¹⁰一起形成可任选被取代的环,

R²⁰、R²¹和R²²相互独立地为C₁-C₂₅烷基, 或C₆-C₂₄芳基, 其可以任选被G取代;

D为-CO-、-COO-、-S-、-SO-、-SO₂-、-O-、-NR⁶⁵-、-SiR⁷⁰R⁷¹-、-POR⁷²-、-CR⁶³=CR⁶⁴-或-C≡C-,

E为-OR⁶⁹、-SR⁶⁹、-NR⁶⁵R⁶⁶、-COR⁶⁸、-COOR⁶⁷、-CONR⁶⁵R⁶⁶、-CN或卤素,

G为E或C₁-C₁₈烷基, C₆-C₂₄芳基, 任选被F取代的C₆-C₂₄芳基, C₁-C₁₈烷基, 被0间隔的C₁-C₁₈烷基, C₂-C₃₀杂芳基或被F取代的C₂-C₃₀杂芳基, C₁-C₁₈烷基, 被0间隔的C₁-C₁₈烷基;

R⁶³和R⁶⁴相互独立地为C₆-C₁₈芳基, 被C₁-C₁₈烷基、C₁-C₁₈烷氧基取代的C₆-C₁₈芳基, C₁-C₁₈烷基或被-0-间隔的C₁-C₁₈烷基;

R⁶⁵和R⁶⁶相互独立地为C₆-C₁₈芳基, 被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基, C₁-C₁₈烷基或被-0-间隔的C₁-C₁₈烷基; 或者

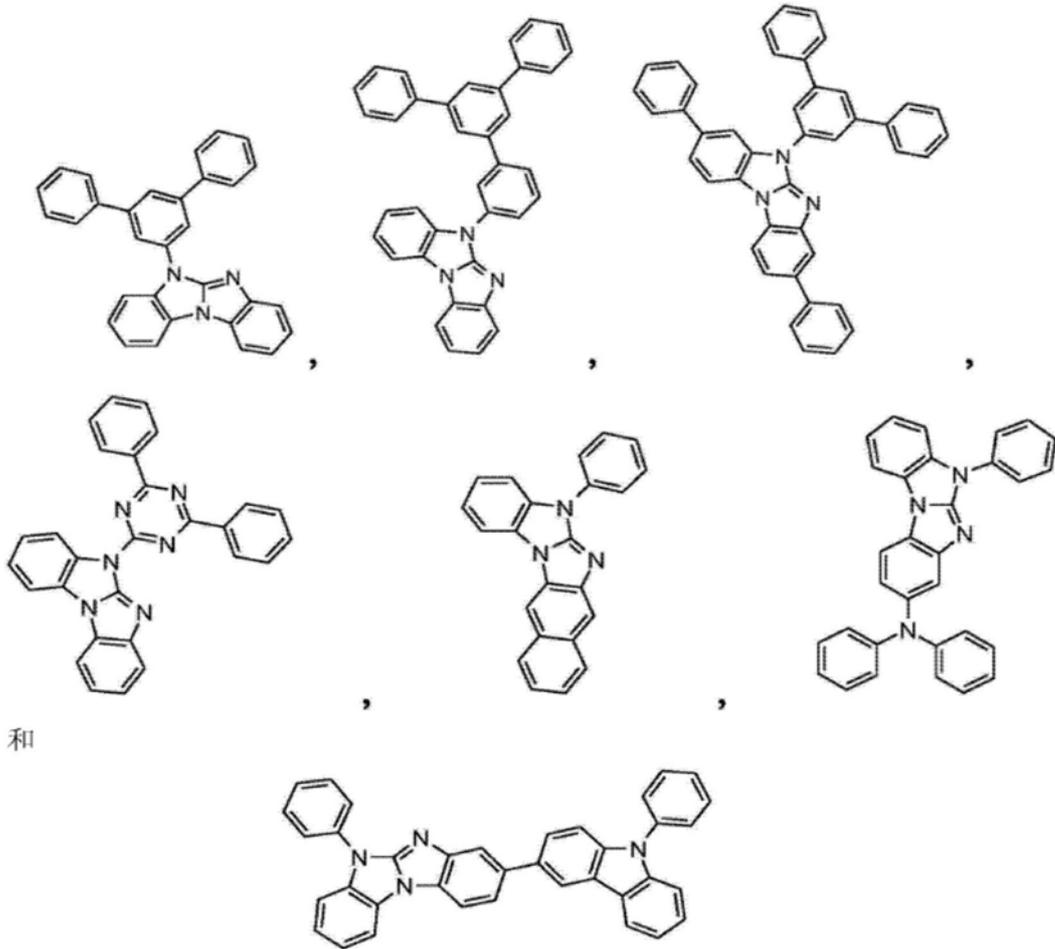
R⁶⁵和R⁶⁶一起形成5或6员环,

R⁶⁷为C₆-C₁₈芳基, 被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基, C₁-C₁₈烷基或被-0-间隔的C₁-C₁₈烷基,

R⁶⁸为H, C₆-C₁₈芳基, 被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基, C₁-C₁₈烷基或被-0-间隔的C₁-C₁₈烷基,

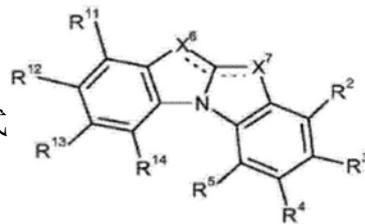
R⁶⁹为C₆-C₁₈芳基, 被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基, C₁-C₁₈烷基或被-0-间隔的C₁-C₁₈烷基,

R⁷⁰和R⁷¹相互独立地为C₁-C₁₈烷基, C₆-C₁₈芳基或被C₁-C₁₈烷基取代的C₆-C₁₈芳基, 以及R⁷²为C₁-C₁₈烷基, C₆-C₁₈芳基或被C₁-C₁₈烷基取代的C₆-C₁₈芳基; 条件是排除如下化合物:



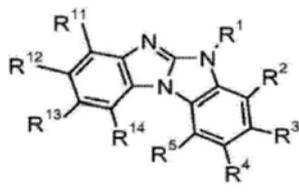
和

2. 根据权利要求1的化合物, 其为式



尤其是

(II),



(II')

的化合物, 其中

X⁶为-N=且X⁷为-NR¹-或

X⁷为=N-且X⁶为-NR¹-,

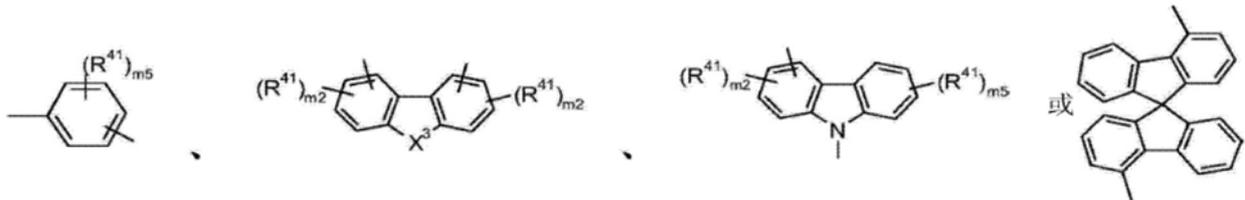
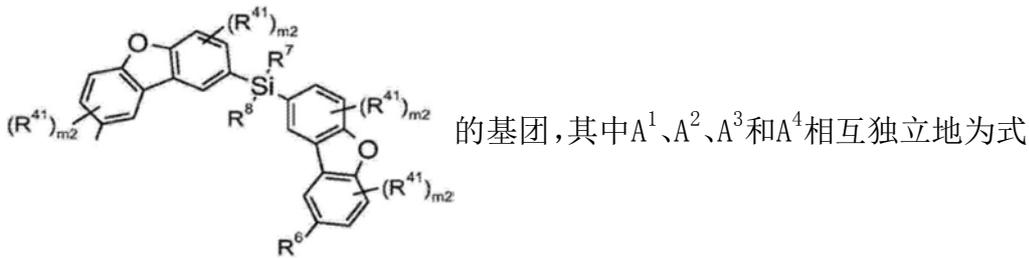
R¹¹、R¹²、R¹³和R¹⁴相互独立地为H, 可任选被E取代和/或被D间隔的C₁-C₂₅烷基, 可以任选被G取代的C₆-C₂₄芳基, 或可以任选被G取代的C₂-C₃₀杂芳基, 和

E、D、G、R¹、R²、R³、R⁴和R⁵如权利要求1所定义。

3. 根据权利要求3的化合物, 其中R²、R³、R⁴、R⁵、R¹¹、R¹²、R¹³和R¹⁴为H和R¹如权利要求1所定

义。

4. 根据权利要求1-3中任一项的化合物,其中R¹为式-A¹-(A²)_p-(A³)_q-(A⁴)_r-R⁶或



的基团,其中

m₅为0或1-4的整数,

m₂为0或整数1-3,

X³为-O-、-S-或-NR¹⁵-,

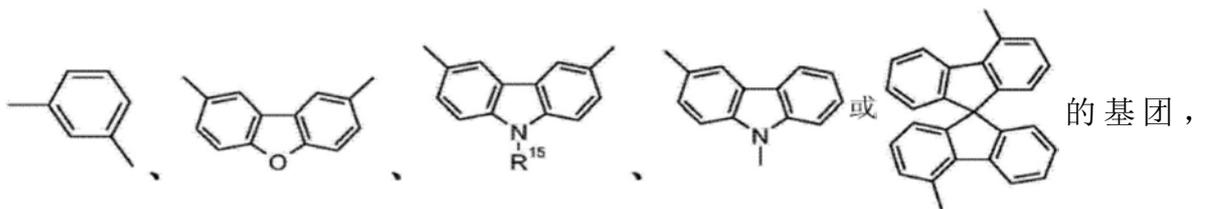
R⁷和R⁸为C₁-C₁₈烷基,

R¹⁵为C₁-C₁₈烷基;或被0-间隔的C₁-C₁₈烷基;C₆-C₁₈芳基;被一个或多个C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基;C₂-C₂₀杂芳基或被一个或多个C₁-C₁₈烷基取代的C₂-C₂₀杂芳基,

R⁴¹在每次出现时可以相同或不同且为F, C₁-C₁₈烷基,被E取代和/或被D间隔的C₁-C₁₈烷基, C₆-C₂₄芳基,被G取代的C₆-C₂₄芳基, C₂-C₂₀杂芳基或被G取代的C₂-C₂₀杂芳基,和

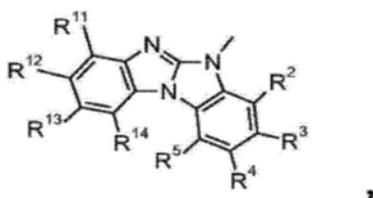
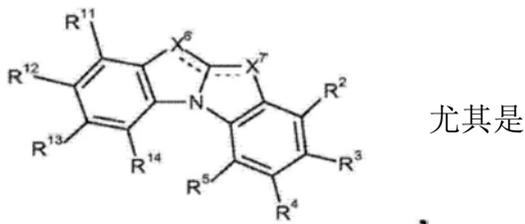
R⁶、p、q、r、E、D和G如权利要求1所定义。

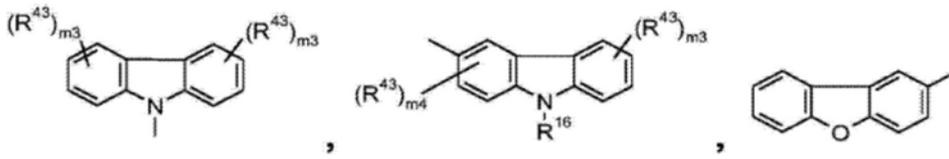
5. 根据权利要求4的化合物,其中A¹、A²、A³和A⁴相互独立地为式



其中R¹⁵为C₆-C₁₈芳基;或被一个或多个C₁-C₁₈烷基取代的C₆-C₁₈芳基。

6. 根据权利要求1-5中任一项的化合物,其中R⁶为式





的基团或基团 - (SiR²⁰R²¹R²²), 其中

X^{6'} 为 -N= 且 X^{7'} 为 -NR¹- 或

X^{7'} 为 =N- 且 X^{6'} 为 -NR¹-,

R²、R³、R⁴、R⁵、R¹¹、R¹²、R¹³ 和 R¹⁴ 相互独立地为 H, 可任选被 E 取代和/或被 D 间隔的 C₁-C₂₅ 烷基; 可以任选被 G 取代的 C₆-C₂₄ 芳基, 或可以任选被 G 取代的 C₂-C₃₀ 杂芳基,

R¹⁶ 为 C₆-C₁₈ 芳基; 或被一个或多个 C₁-C₁₈ 烷基取代的 C₆-C₁₈ 芳基。

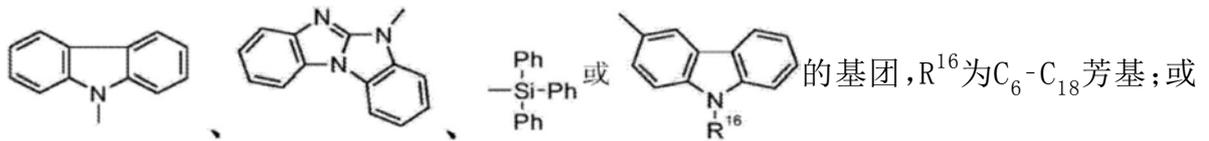
R²⁰、R²¹ 和 R²² 相互独立地为 C₆-C₁₈ 芳基; 或被一个或多个 C₁-C₁₈ 烷基取代的 C₆-C₁₈ 芳基,

R⁴³ 在每次出现时可以相同或不同且为 F, C₁-C₁₈ 烷基, 被 E 取代和/或被 D 间隔的 C₁-C₁₈ 烷基, C₆-C₂₄ 芳基, 被 G 取代的 C₆-C₂₄ 芳基, C₂-C₂₀ 杂芳基或被 G 取代的 C₂-C₂₀ 杂芳基,

m₃ 为 0 或 1-4 的整数, m₄ 为 0 或 1-3 的整数, 和

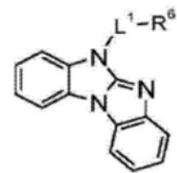
E、D 和 G 如权利要求 1 所定义。

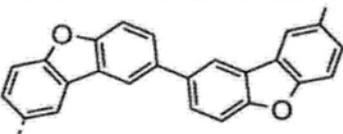
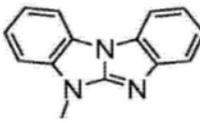
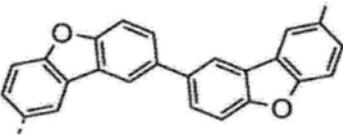
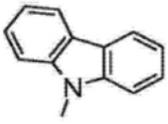
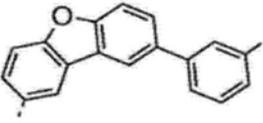
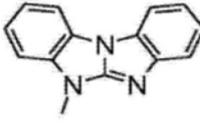
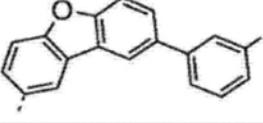
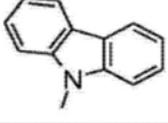
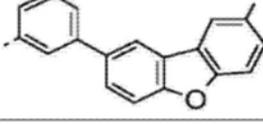
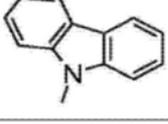
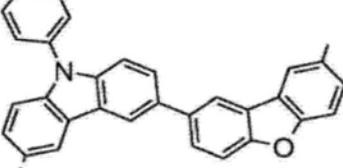
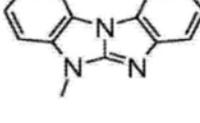
7. 根据权利要求 6 的化合物, 其中 R⁶ 为式

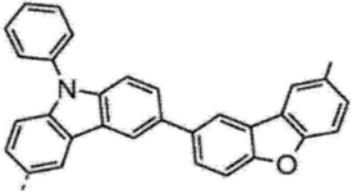
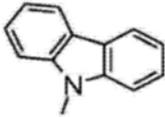
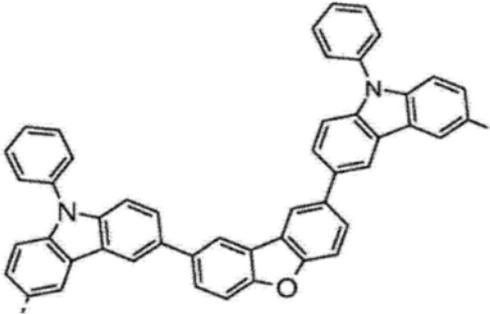
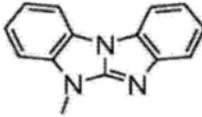
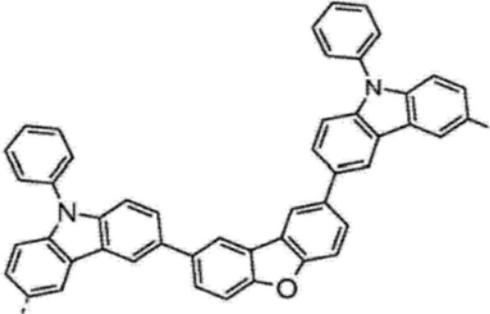
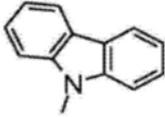
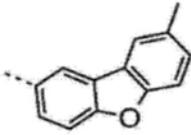
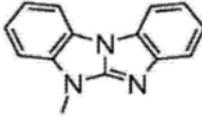
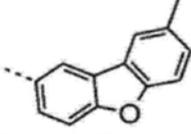
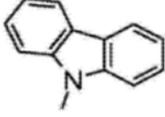
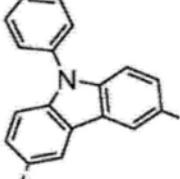
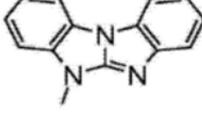


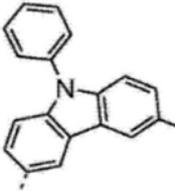
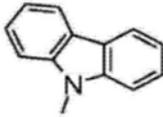
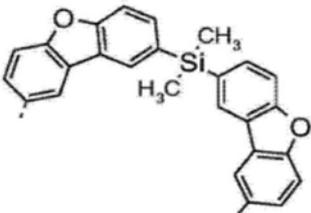
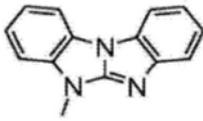
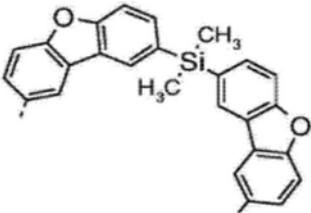
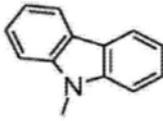
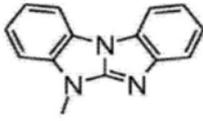
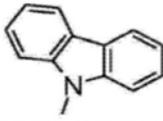
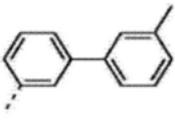
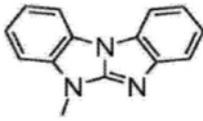
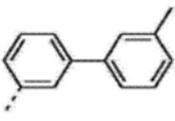
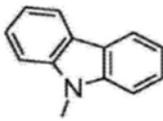
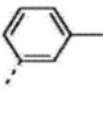
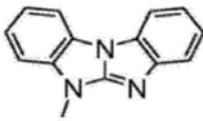
被一个或多个 C₁-C₁₈ 烷基取代的 C₆-C₁₈ 芳基。

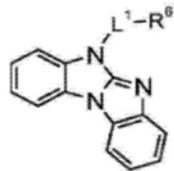
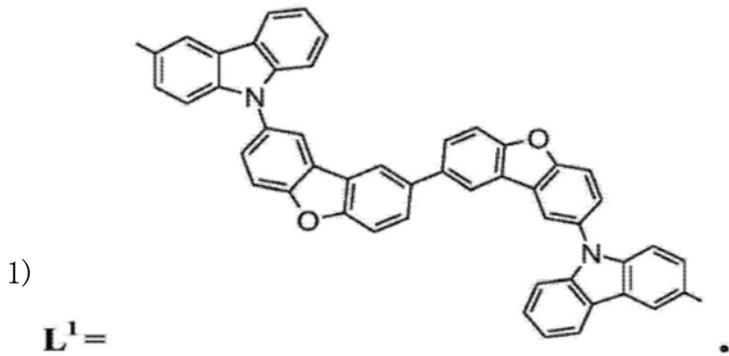
8. 根据权利要求 1 的化合物:



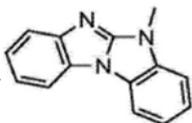
Cpd.	L ¹²⁾	R ⁶
A-1		
A-2		
A-3		
A-4		
A-5		
A-6		

<p>A-7</p>		
<p>A-8</p>		
<p>A-9</p>		
<p>A-10</p>		
<p>A-11</p>		
<p>A-12</p>		

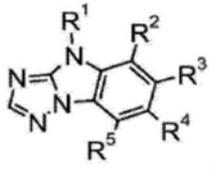
A-13		
A-14		
A-15		
A-16	D)	
A-17	D)	
A-18		
A-19		
A-20		



Cpd.	$L^{1,2)}$	R^6
B-1		
B-2		
B-3		
B-4		
B-5		

2) 虚线表示与式  的基团的键。

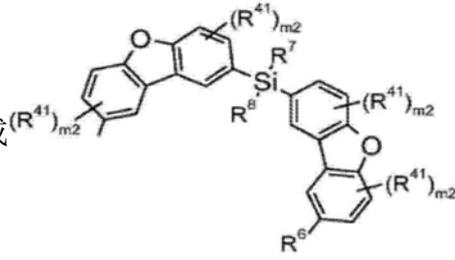


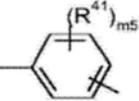


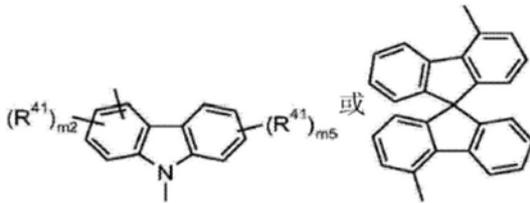
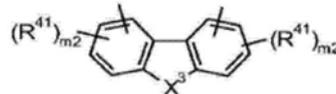
(Ib)

的化合物,其中

R¹为式-A¹- (A²)_p- (A³)_q- (A⁴)_r-R⁶的基团或



A¹、A²、A³和A⁴相互独立为式  ,



的基团,其中

m₅为0或1-4的整数,

m₂为0或整数1-3,

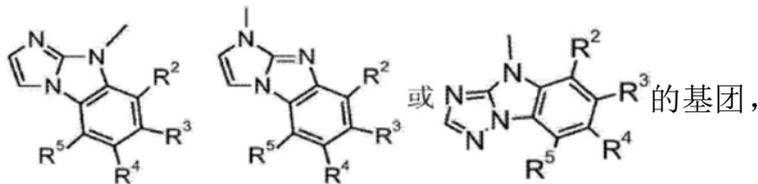
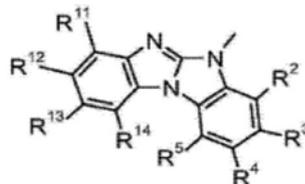
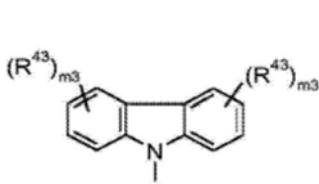
X³为-O-、-S-或-NR¹⁵-,

R⁷和R⁸为C₁-C₁₈烷基,

R¹⁵为C₁-C₁₈烷基;或被0-间隔的C₁-C₁₈烷基;C₆-C₁₈芳基;被一个或多个C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基;C₂-C₂₀杂芳基或被一个或多个C₁-C₁₈烷基取代的C₂-C₂₀杂芳基,

R⁴¹在每次出现时可以相同或不同且为F, C₁-C₁₈烷基, 被E取代和/或被D间隔的C₁-C₁₈烷基, C₆-C₂₄芳基, 被G取代的C₆-C₂₄芳基, C₂-C₂₀杂芳基或被G取代的C₂-C₂₀杂芳基,

R⁶为式

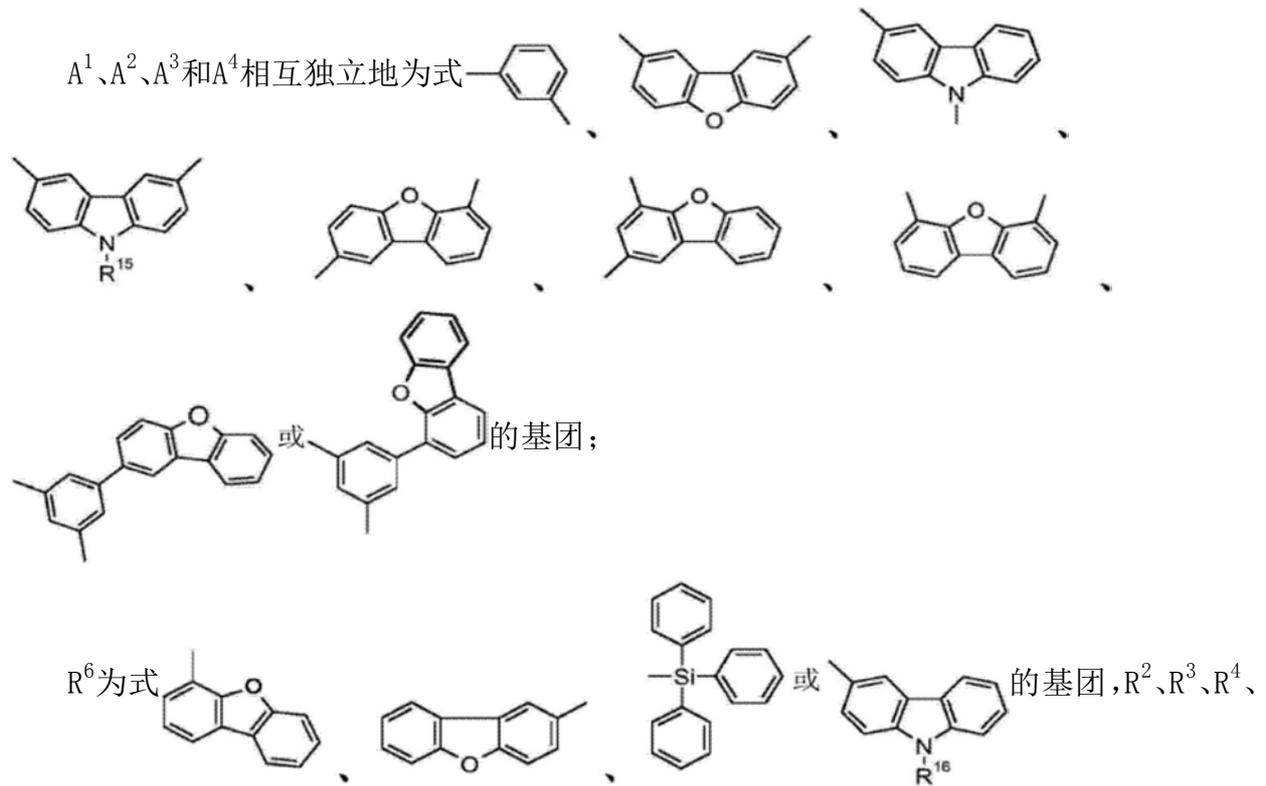


的基团,

R⁴³在每次出现时可以相同或不同且为F, C₁-C₁₈烷基, 被E取代和/或被D间隔的C₁-C₁₈烷基, C₆-C₂₄芳基, 被G取代的C₆-C₂₄芳基, C₂-C₂₀杂芳基, 或被G取代的C₂-C₂₀杂芳基,

m₃为0或1-4的整数;或

R¹为式-A¹- (A²)_p- (A³)_q- (A⁴)_r-R⁶的基团,其中



R^5 、p、q、r、E、D和G如权利要求1所定义，和 R^{11} 、 R^{12} 、 R^{13} 和 R^{14} 如权利要求2所定义。

10. 一种电子器件，包含根据权利要求1-9中任一项的化合物。

11. 根据权利要求10的电子器件，其为场致发光器件。

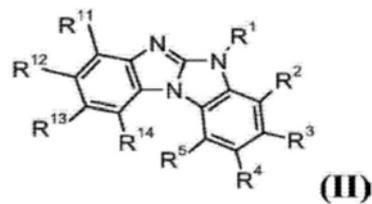
12. 一种空穴传输层或发光层，包含根据权利要求1-9中任一项的化合物。

13. 根据权利要求12的发光层，包含作为主体材料的根据权利要求1-9中任一项的化合物与磷光发光体组合。

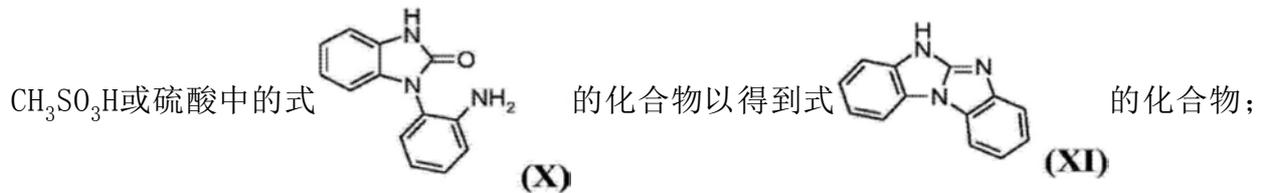
14. 一种装置，选自固定可视显示单元，例如计算机、电视的可视显示单元，打印机、厨房用具和广告板、照明、信息板中的可视显示单元，和移动可视显示单元，例如移动电话、台式PC、便携式电脑、数码相机、MP3播放器、车辆以及公共汽车和火车上的目的地显示中的可视显示单元；照明单元；键盘；衣物；家具；壁纸，包含根据权利要求10或11的有机电子器件或根据权利要求12的空穴传输层或发光层。

15. 根据权利要求1-9中任一项的式I化合物在电摄影感光体、光电转换器、有机太阳能电池（有机光伏器件）、开关元件、有机发光场效应晶体管（OLEFET）、图像传感器、染料激光器和场致发光器件中的用途。

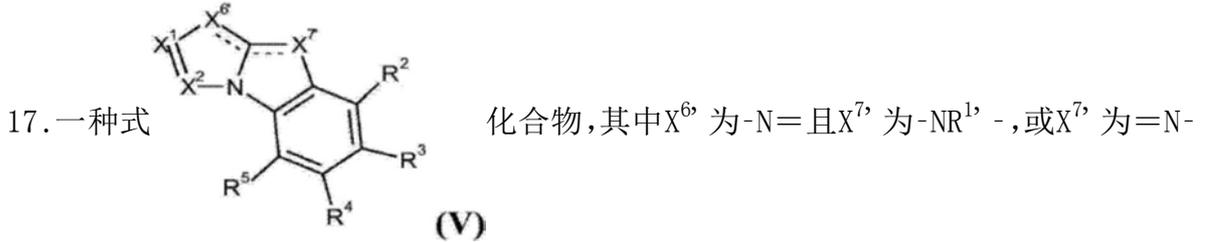
16. 一种制备其中 R^2 、 R^3 、 R^4 、 R^5 、 R^{11} 、 R^{12} 、 R^{13} 和 R^{14} 为H和 R^1 如权利要求1所定义的式



的化合物的方法，包括(a)加热在 H_3PO_4 、多磷酸、 CH_3SO_3H/P_2O_5 、



和 (b) 使式 XI 化合物反应而得到式 II 化合物。

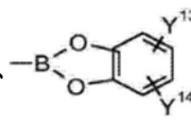
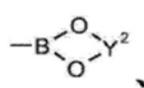


且 X⁶ 为 -NR^{1b} -,

R^{1b} 为式 -A¹ - (A²)_p - (A³)_q - (A⁴)_r - (R^{6a})_t 的基团, t 为 1 或 2;

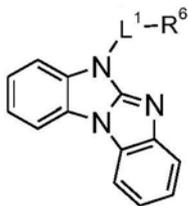
p, q, r, A¹, A², A³, A⁴, X¹, X², R², R³, R⁴ 和 R⁵ 如权利要求 1 所定义, 和

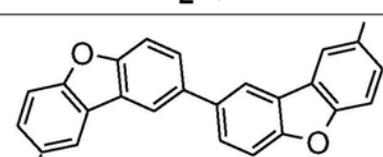
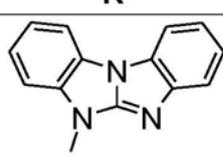
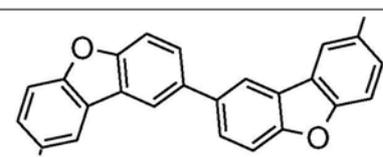
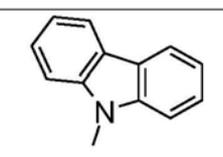
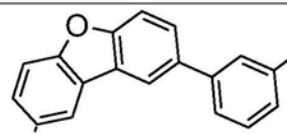
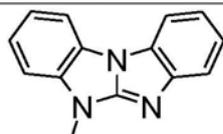
R^{6a} 为卤素; ZnX¹²; -SnR²⁰⁷R²⁰⁸R²⁰⁹, 其中 R²⁰⁷, R²⁰⁸ 和 R²⁰⁹ 相同或不同且为 H 或 C₁-C₆ 烷基, 其中两个基团任选形成公用环且这些基团任选支化或未支化; 和 X¹² 为卤原子、-OS(O)₂CF₃、-

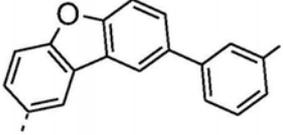
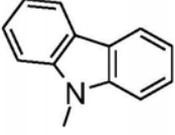
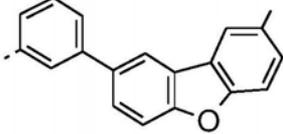
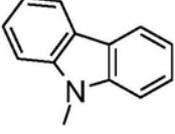
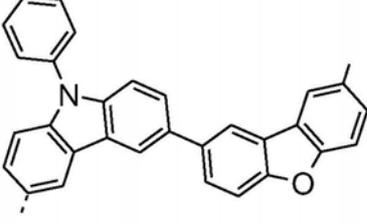
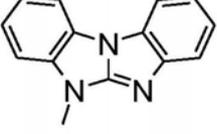
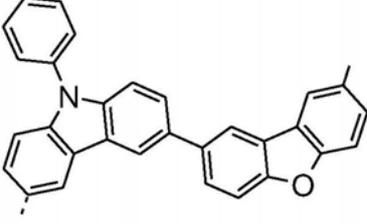
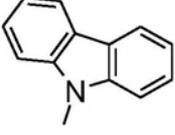
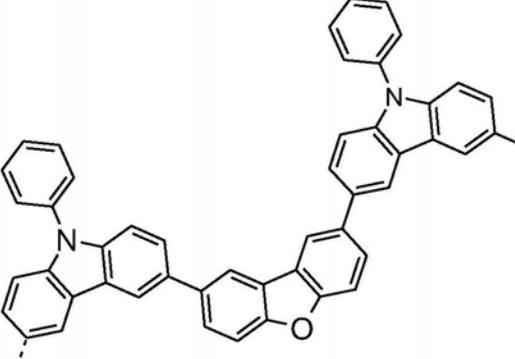
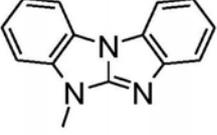
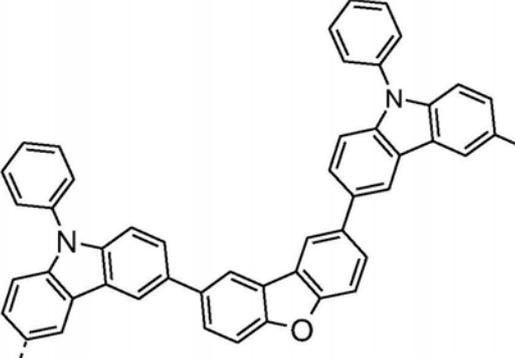
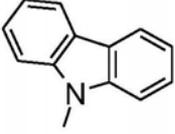
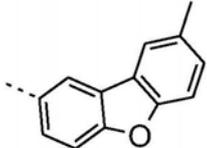
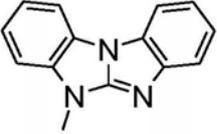
OS(O)₂-芳基、-OS(O)₂CH₃、-B(OH)₂、-B(OY¹)₂、、 -BF₄Na 或 -BF₄K, 其

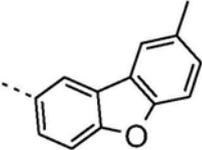
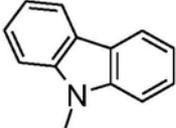
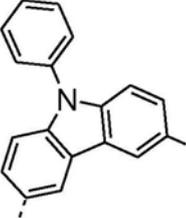
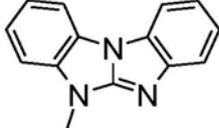
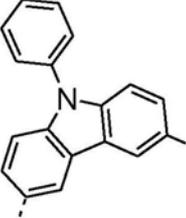
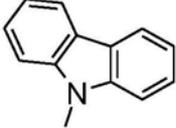
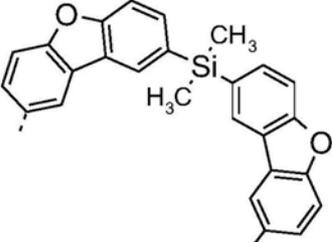
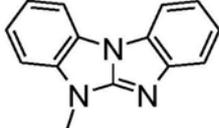
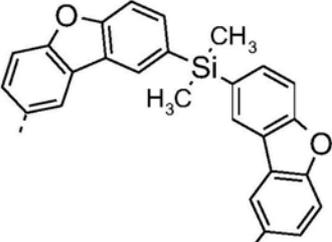
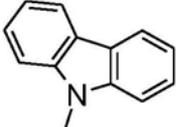
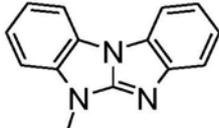
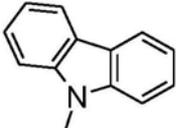
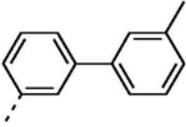
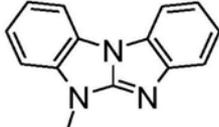
中 Y¹ 在每次出现时独立地为 C₁-C₁₀ 烷基。Y² 在每次出现时独立地为 C₂-C₁₀ 亚烷基, 和 Y¹³ 和 Y¹⁴ 相互独立地为氢或 C₁-C₁₀ 烷基。

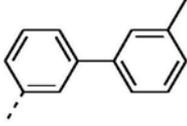
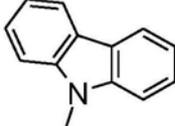
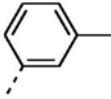
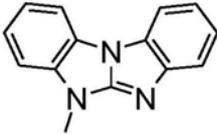
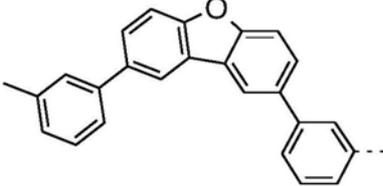
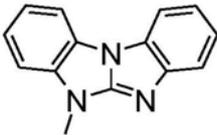
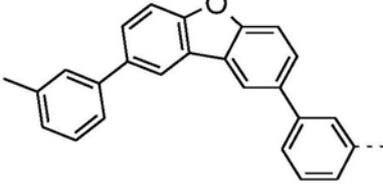
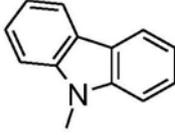
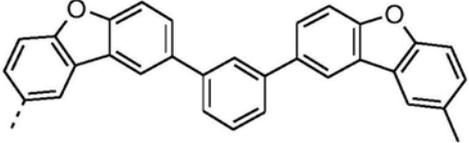
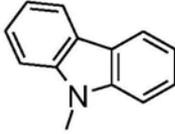
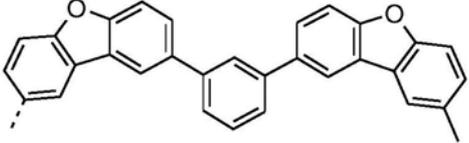
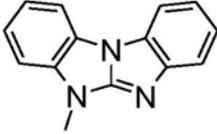
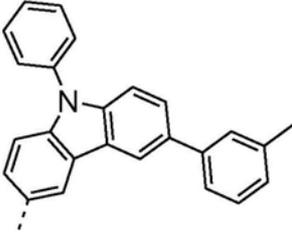
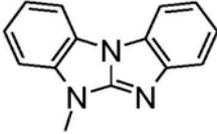
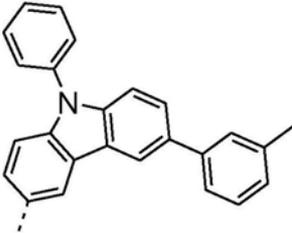
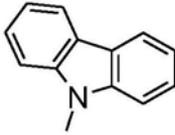
18. 一种化合物, 其选自由以下组成的群组:

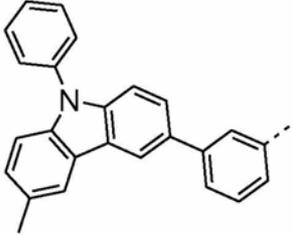
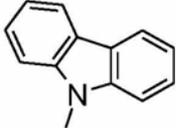
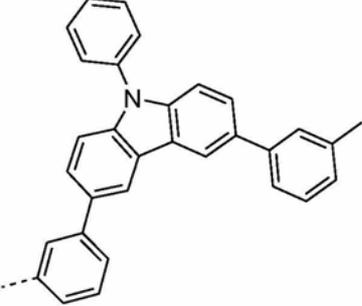
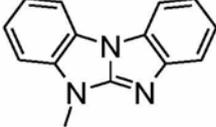
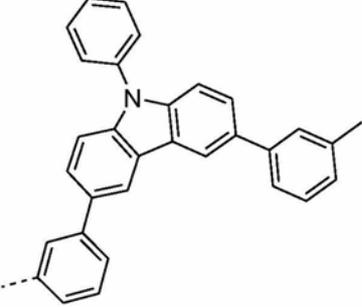
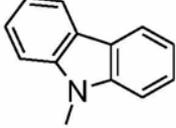
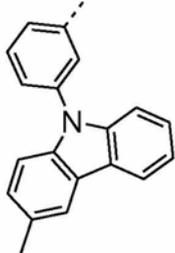
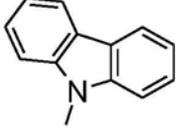
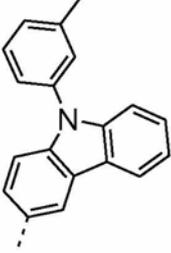
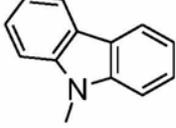
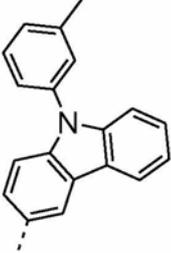
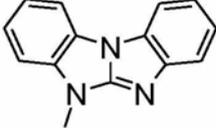


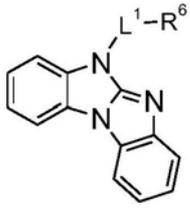
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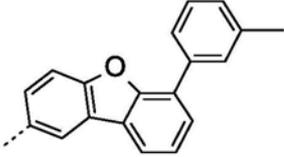
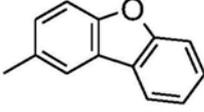
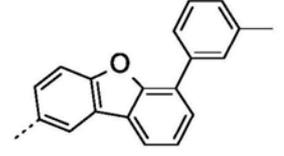
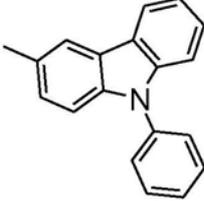
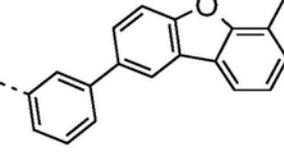
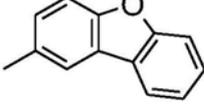
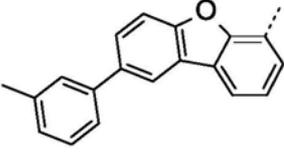
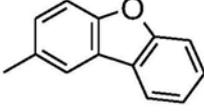
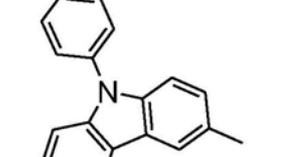
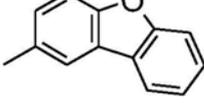
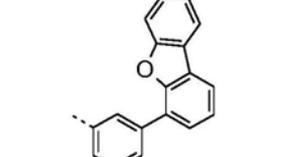
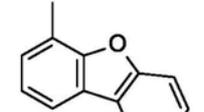
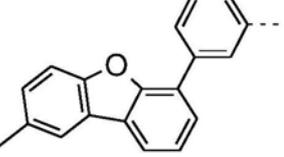
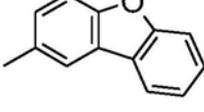
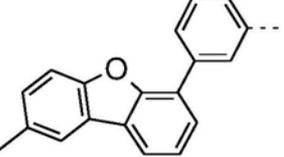
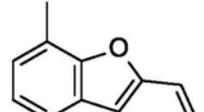
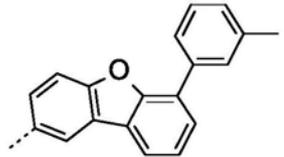
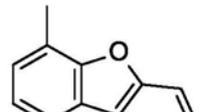
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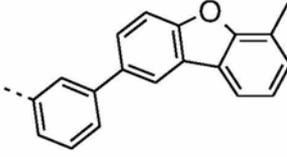
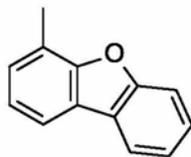
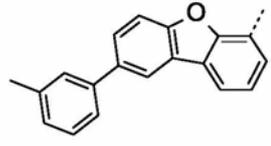
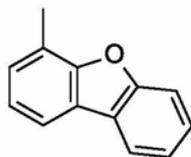
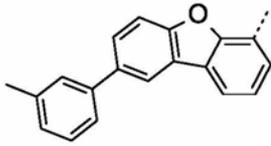
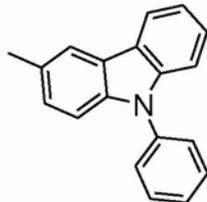
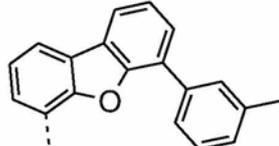
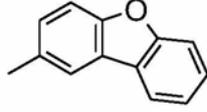
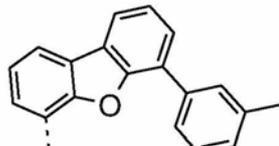
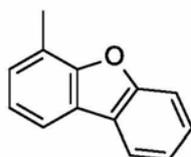
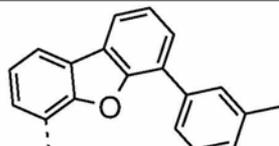
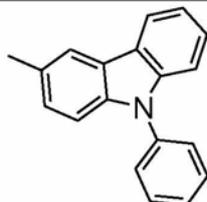
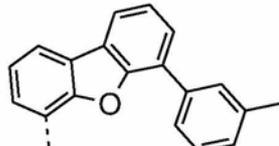
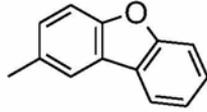
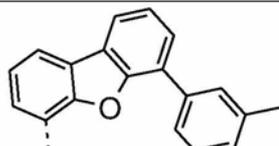
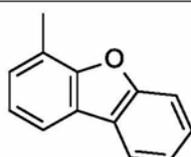
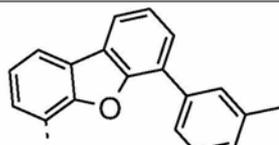
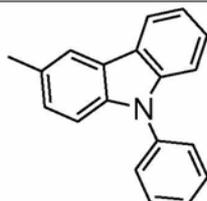
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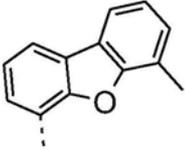
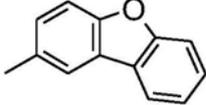
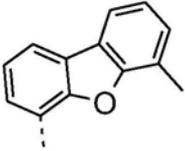
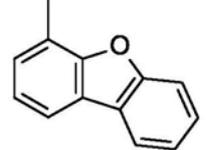
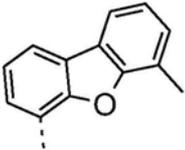
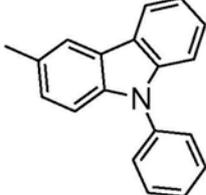
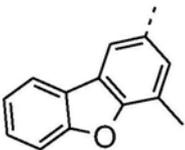
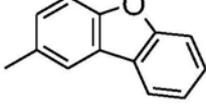
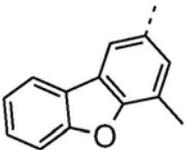
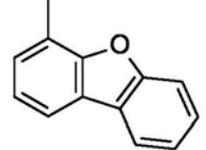
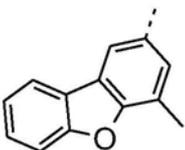
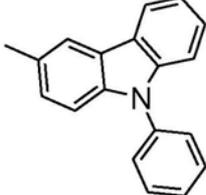
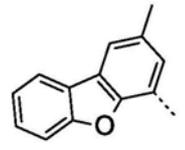
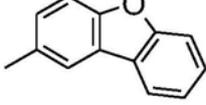
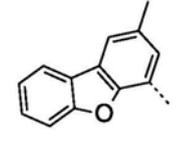
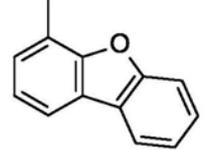
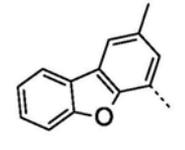
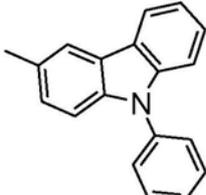
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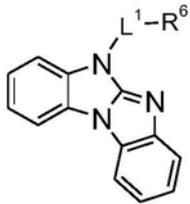
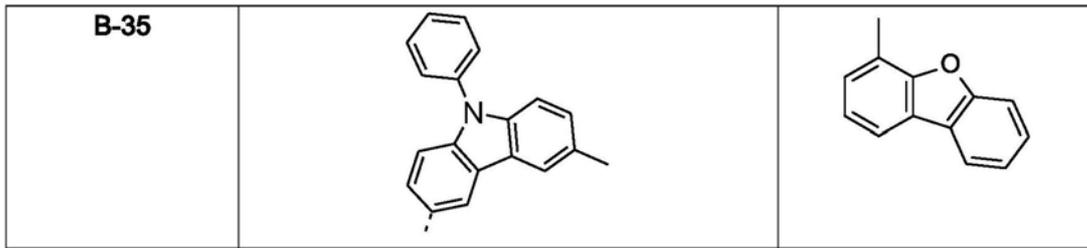


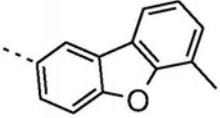
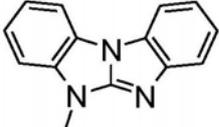
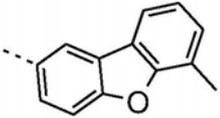
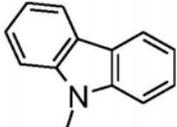
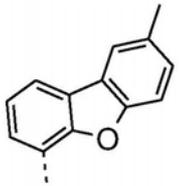
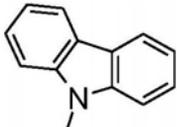
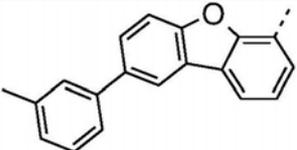
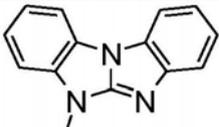
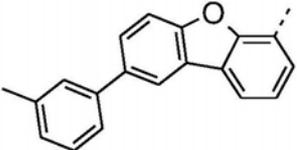
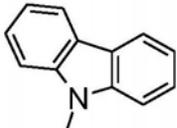
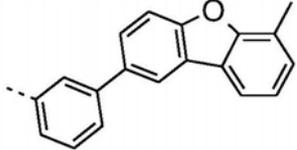
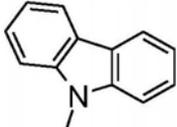
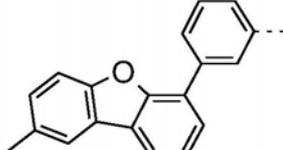
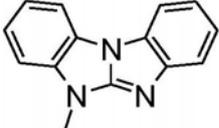
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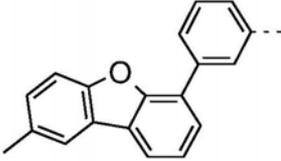
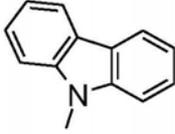
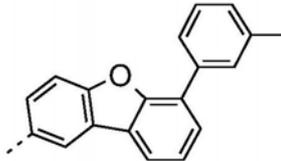
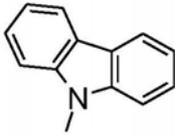
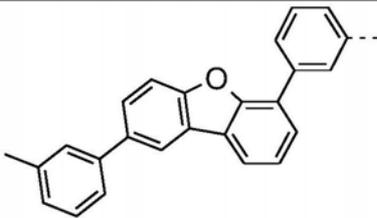
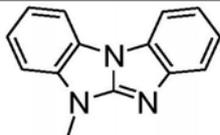
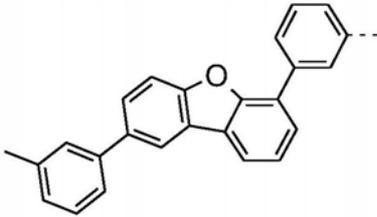
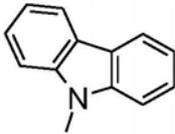
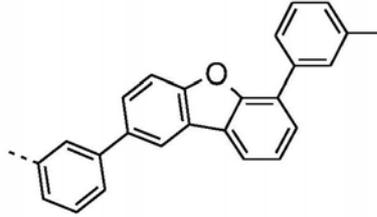
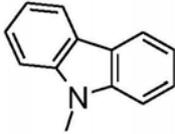
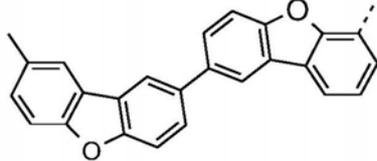
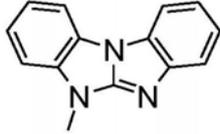
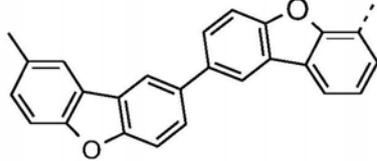
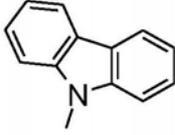
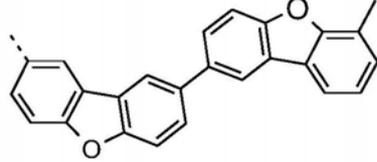
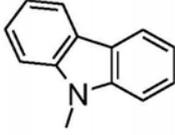
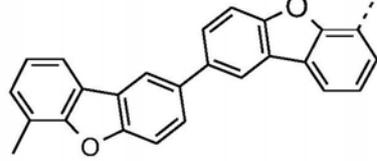
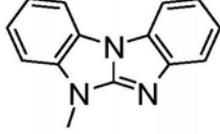
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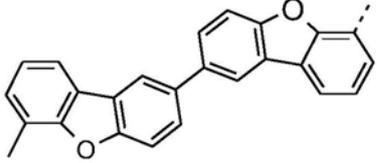
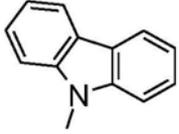
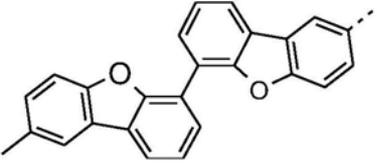
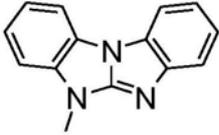
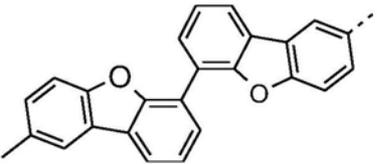
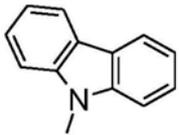
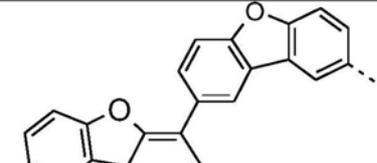
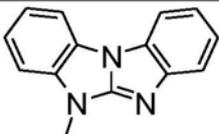
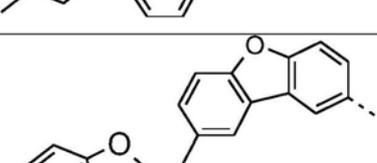
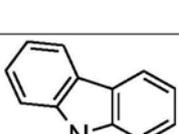
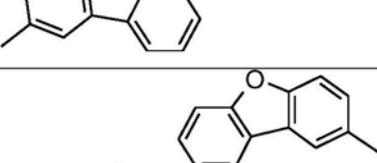
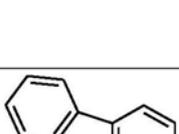
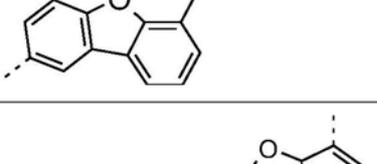
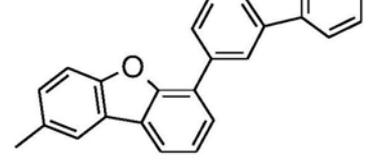
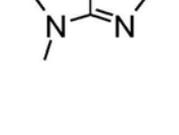
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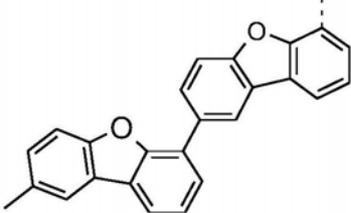
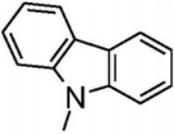
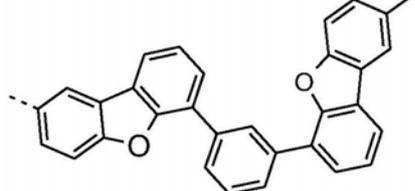
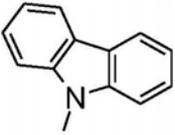
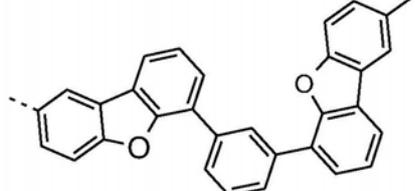
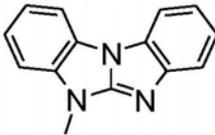
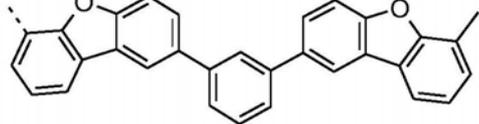
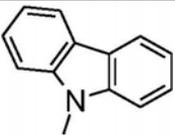
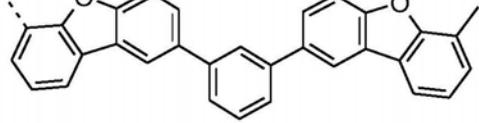
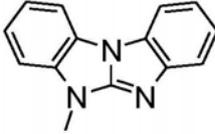
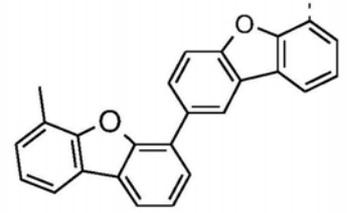
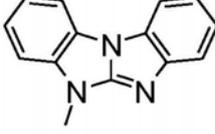
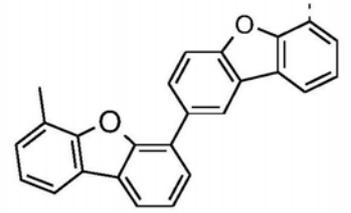
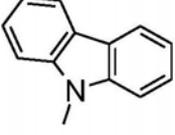
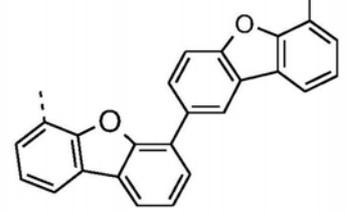
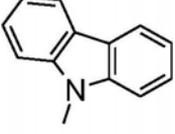
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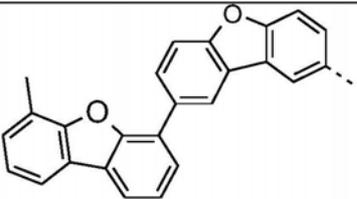
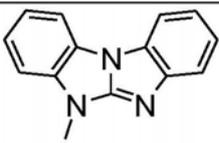
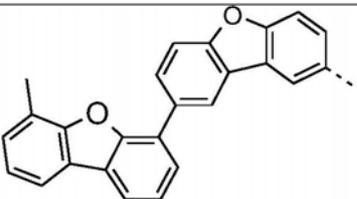
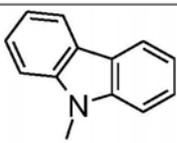
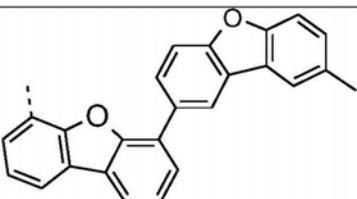
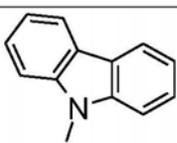
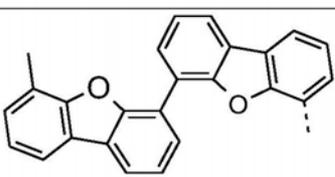
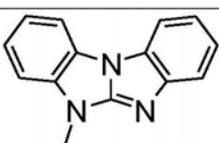
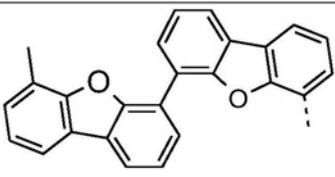
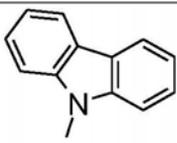
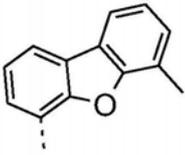
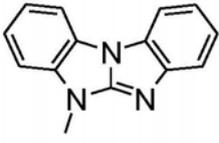
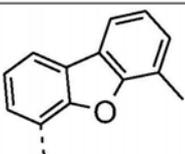
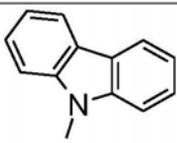
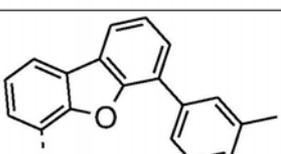
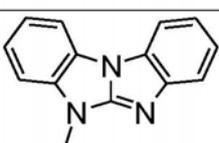
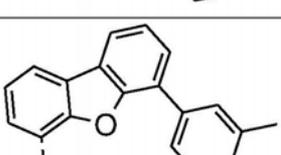
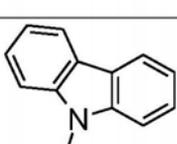


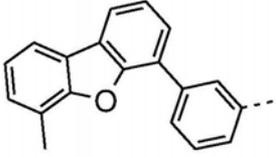
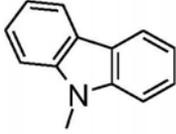
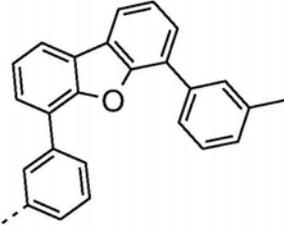
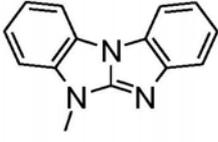
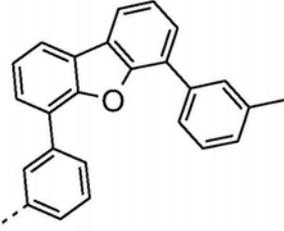
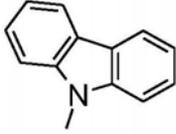
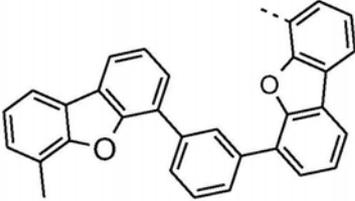
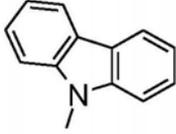
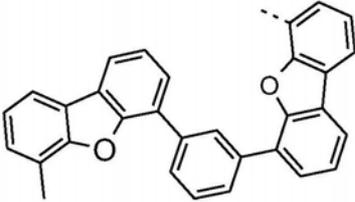
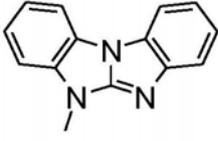
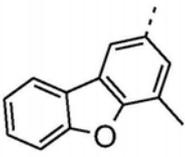
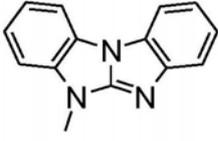
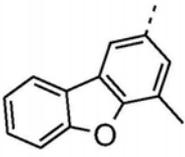
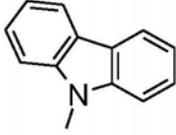
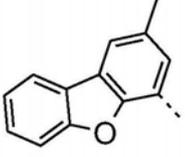
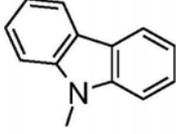
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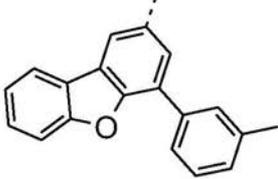
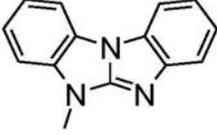
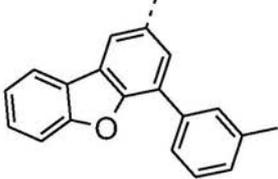
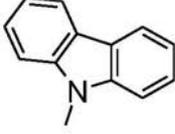
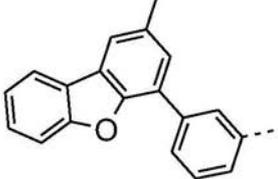
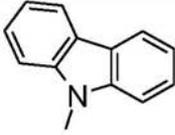
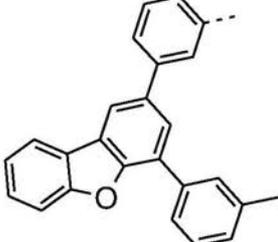
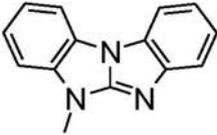
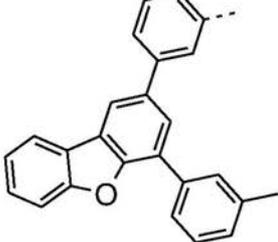
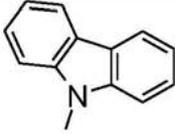
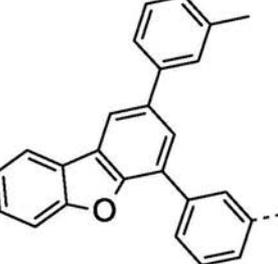
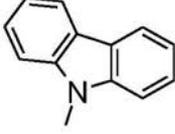
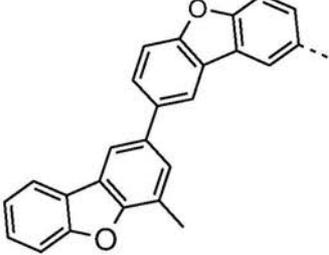
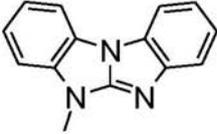
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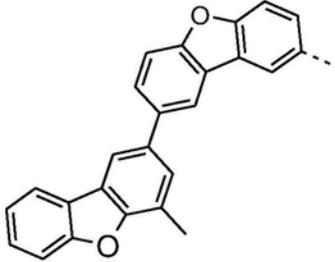
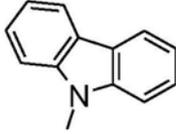
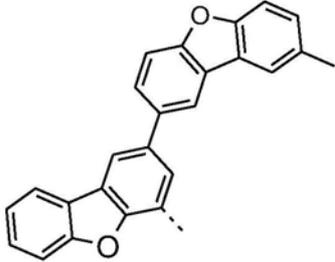
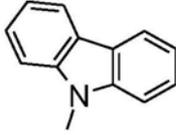
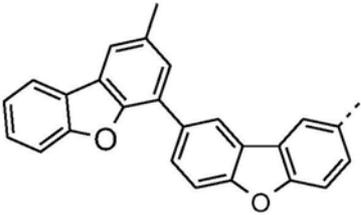
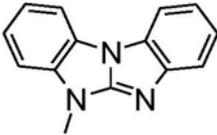
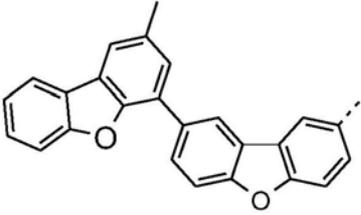
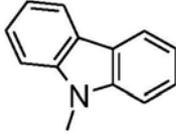
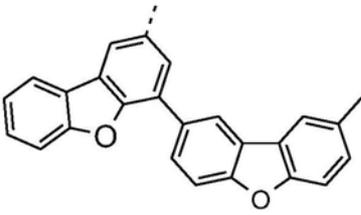
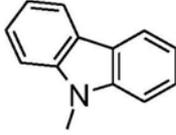
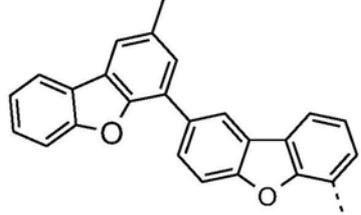
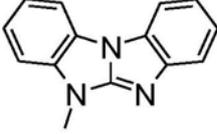
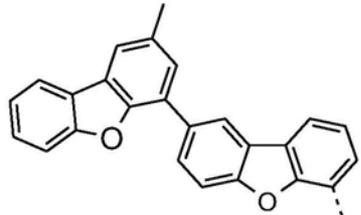
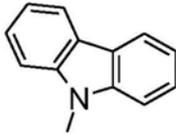
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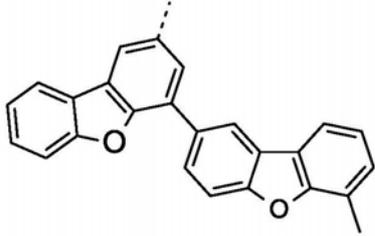
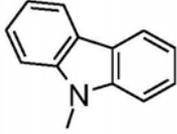
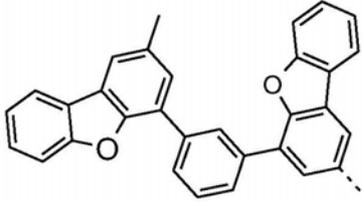
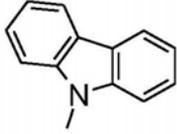
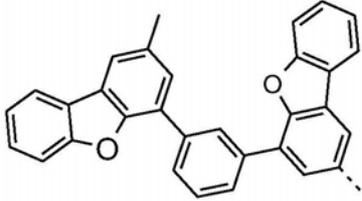
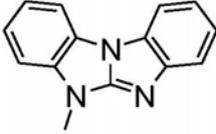
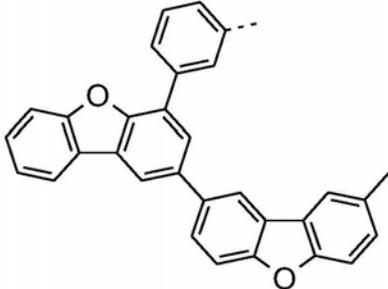
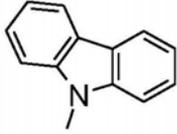
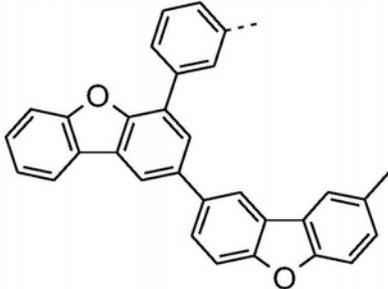
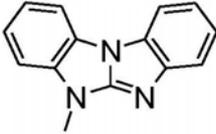
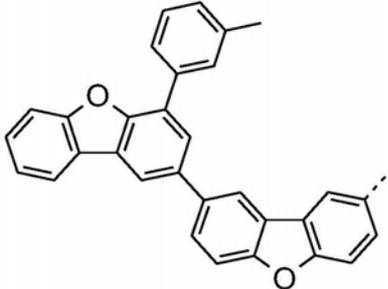
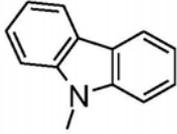
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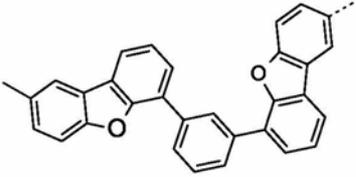
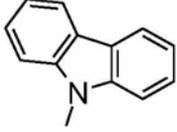
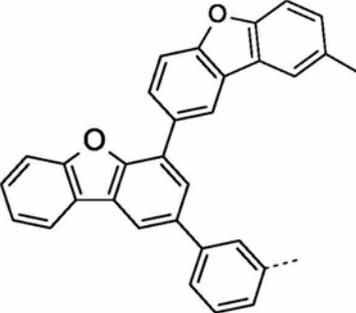
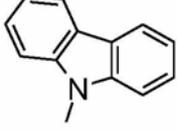
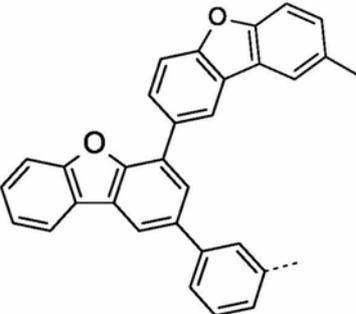
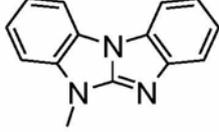
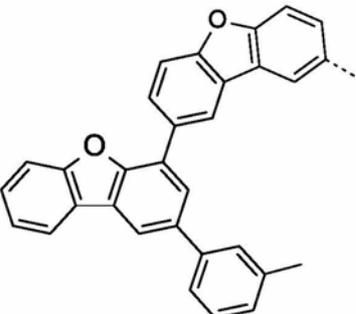
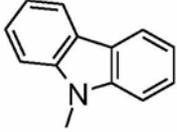
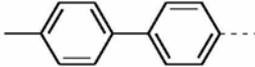
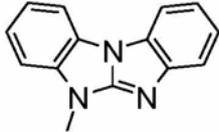
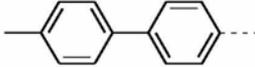
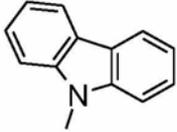
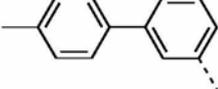
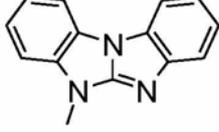
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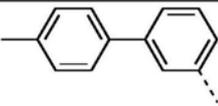
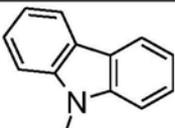
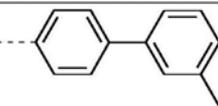
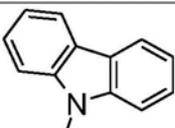
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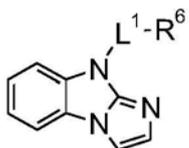
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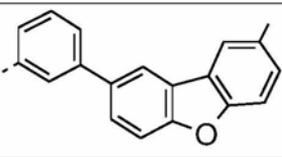
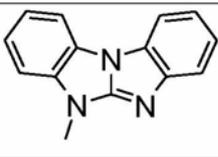
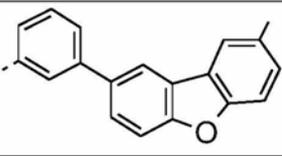
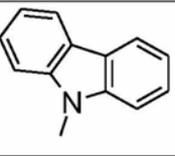
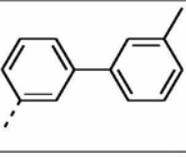
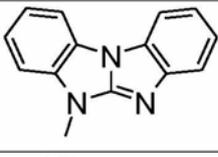
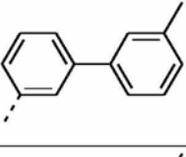
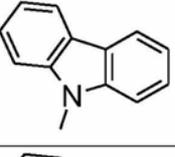
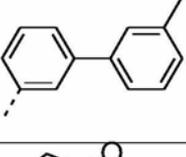
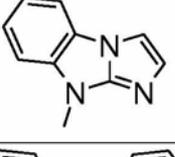
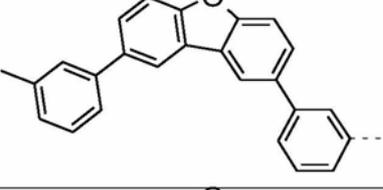
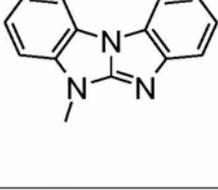
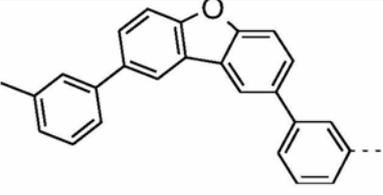
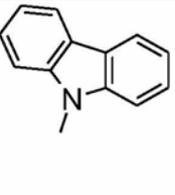
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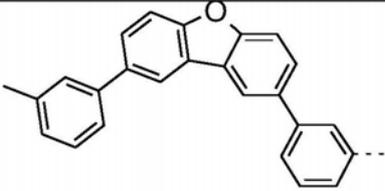
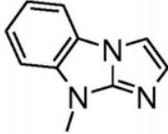
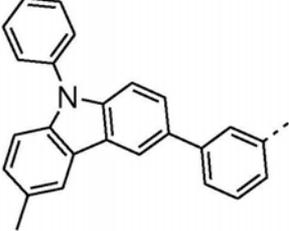
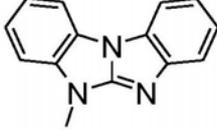
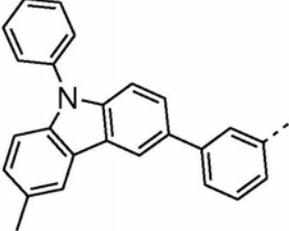
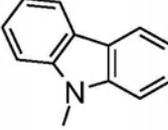
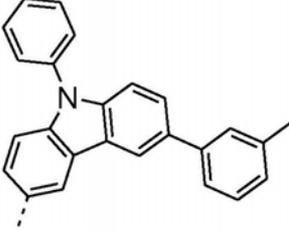
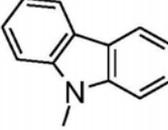
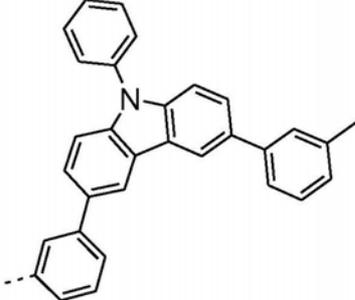
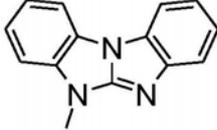
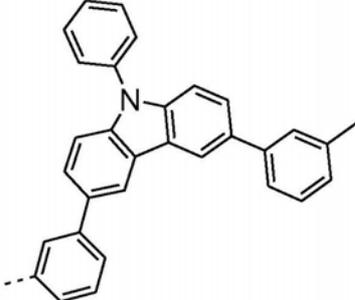
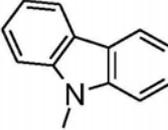
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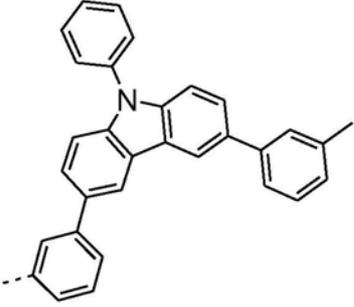
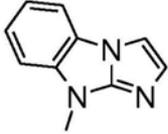
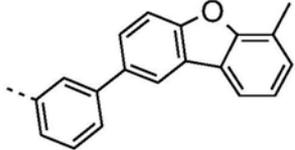
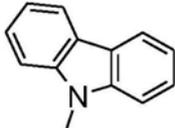
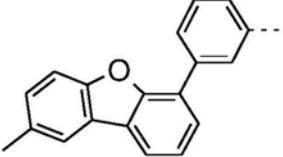
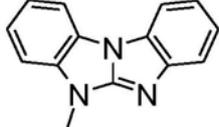
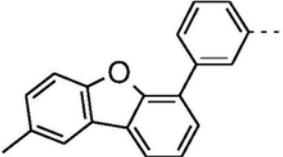
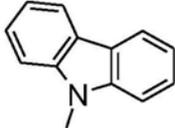
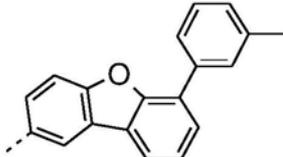
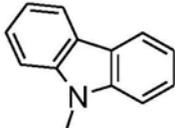
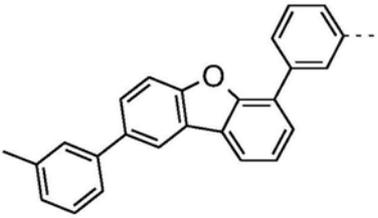
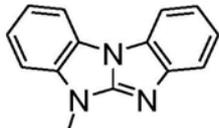
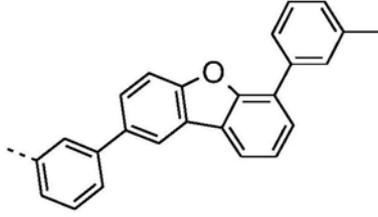
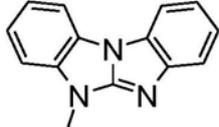
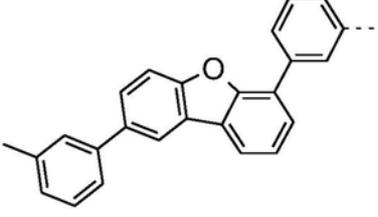
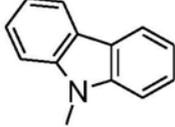
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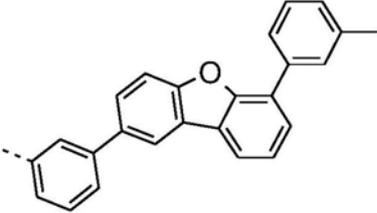
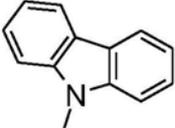
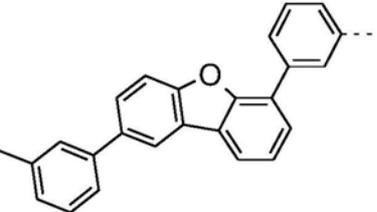
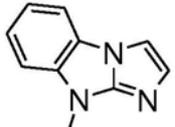
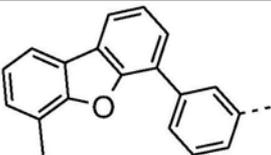
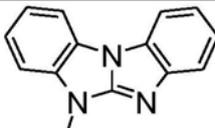
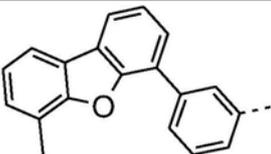
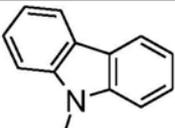
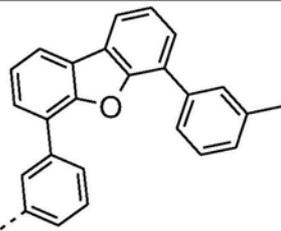
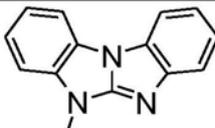
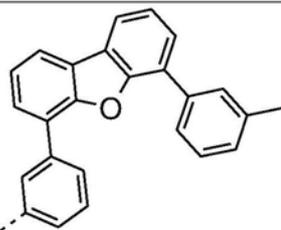
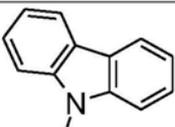
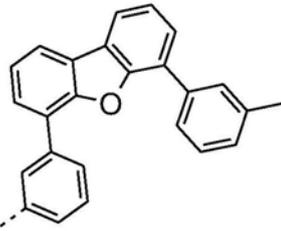
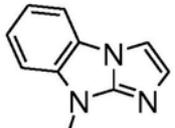
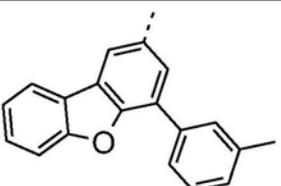
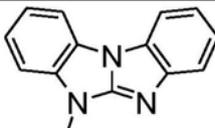
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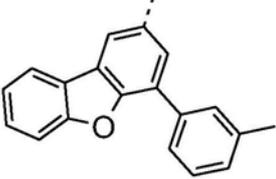
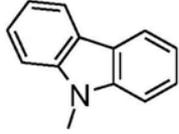
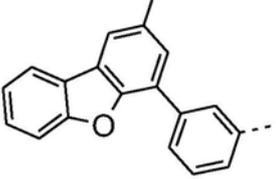
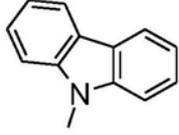
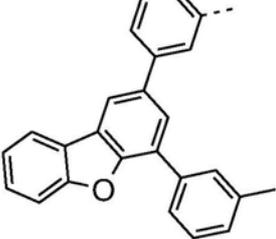
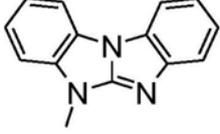
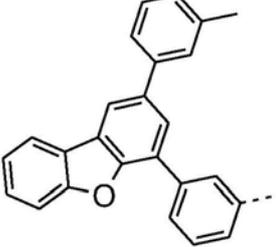
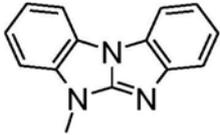
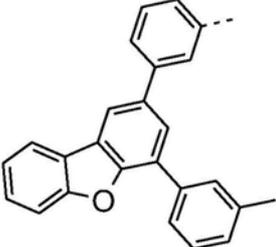
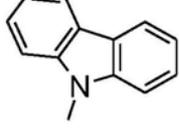
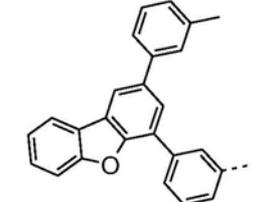
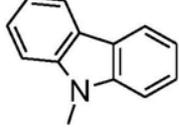
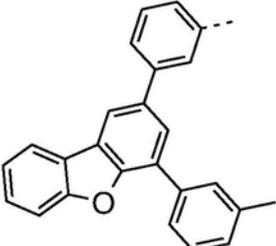
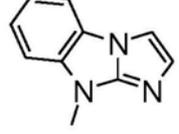


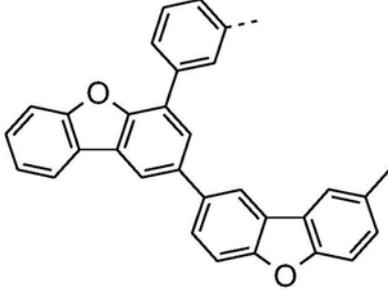
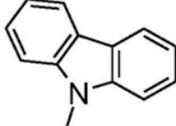
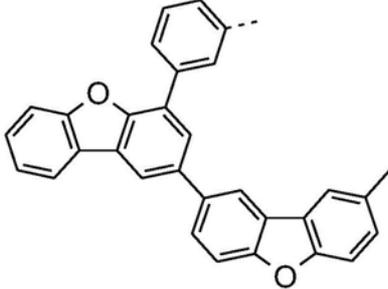
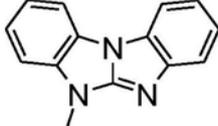
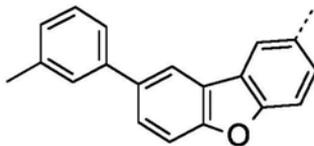
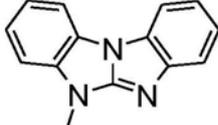
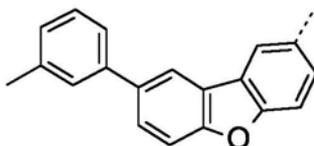
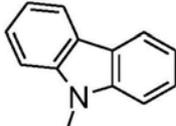
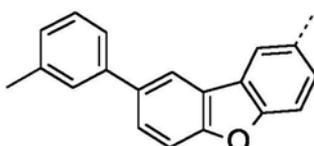
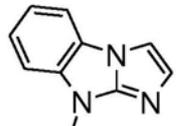
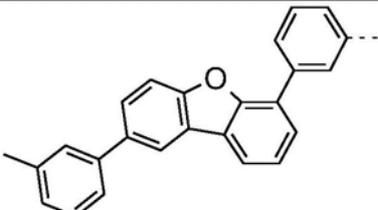
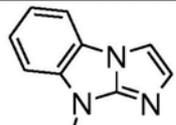
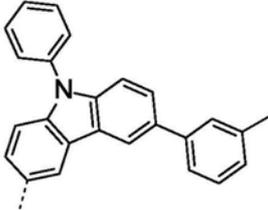
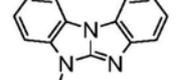
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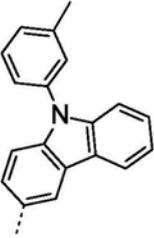
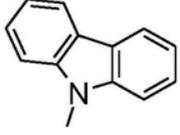
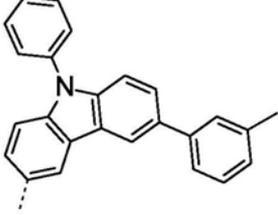
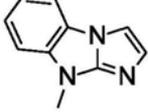
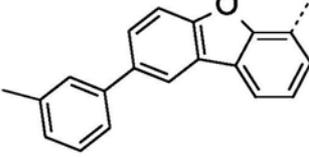
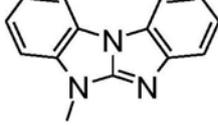
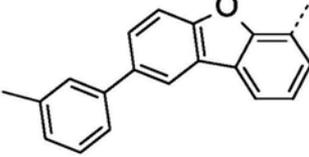
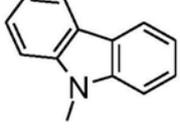
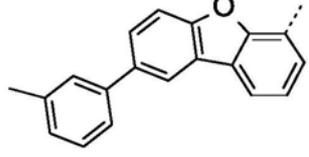
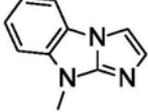
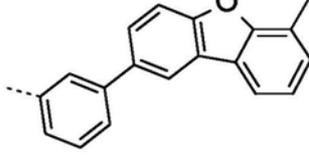
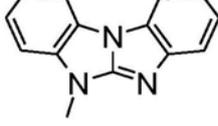
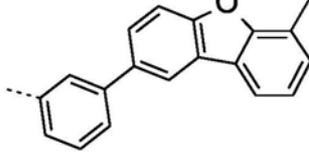
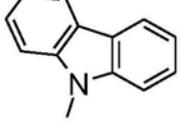
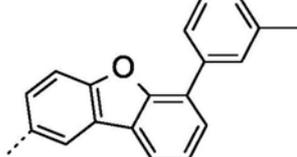
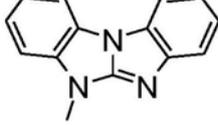
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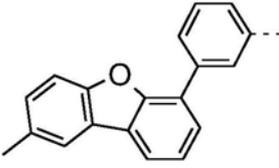
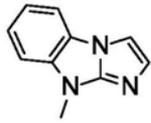
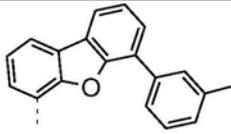
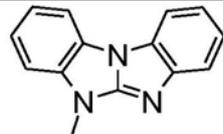
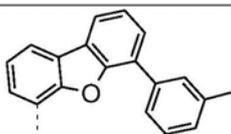
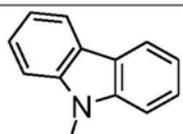
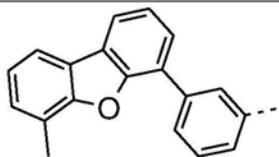
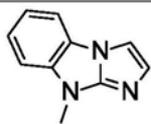
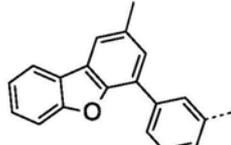
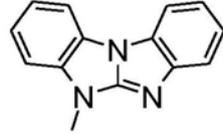
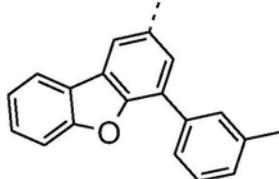
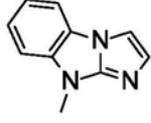
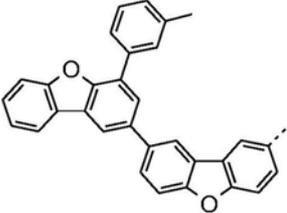
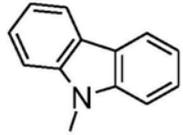
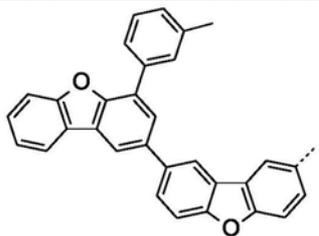
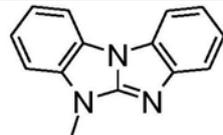
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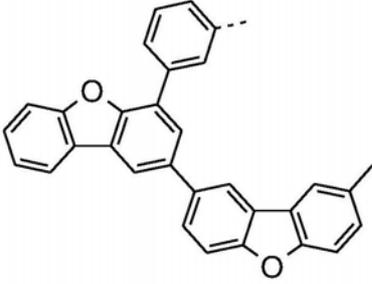
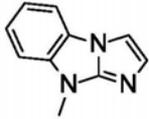
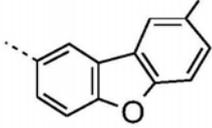
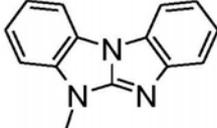
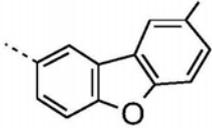
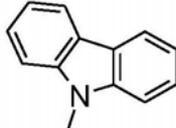
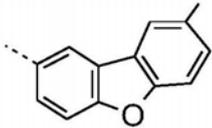
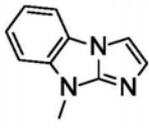
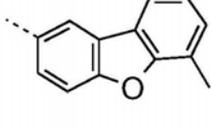
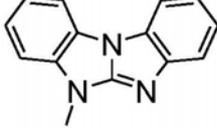
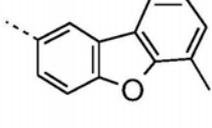
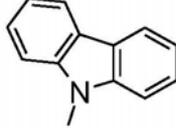
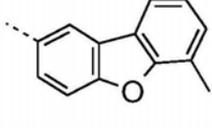
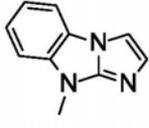
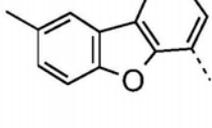
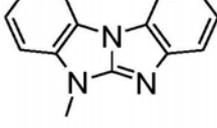
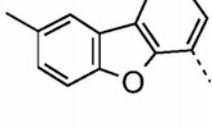
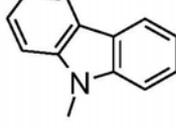
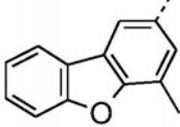
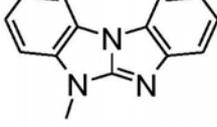
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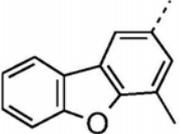
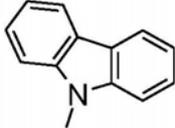
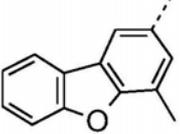
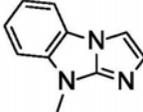
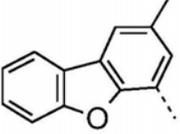
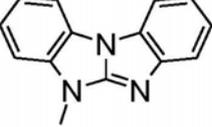
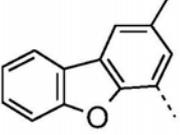
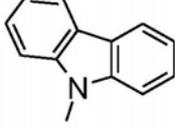
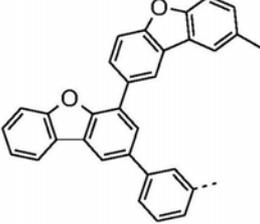
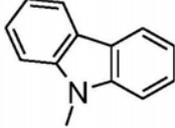
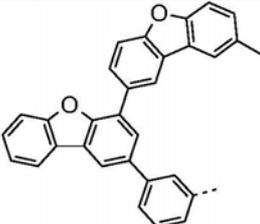
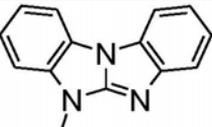
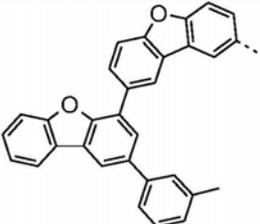
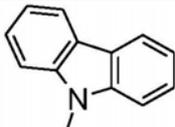
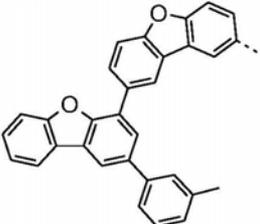
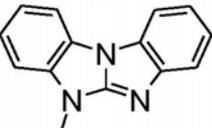
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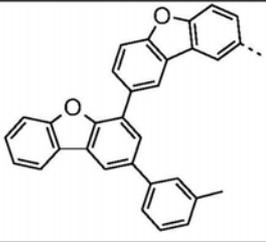
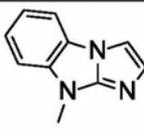
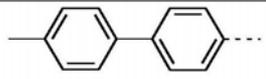
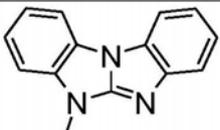
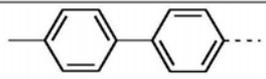
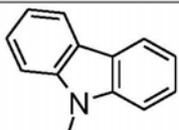
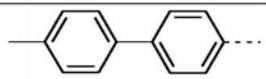
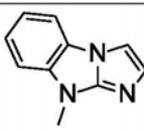
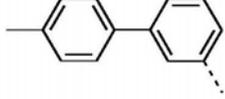
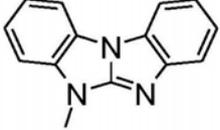
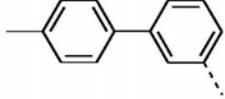
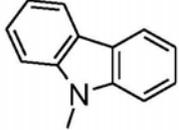
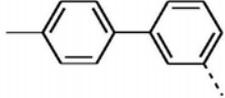
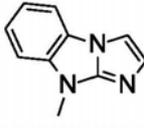
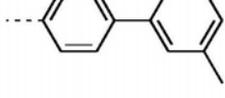
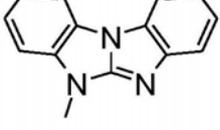
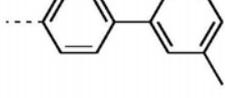
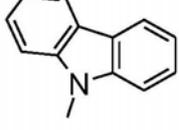
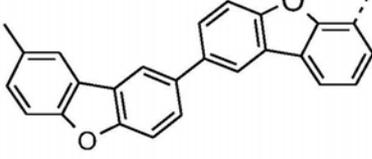
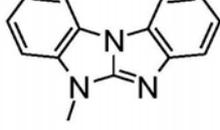
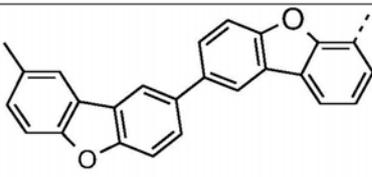
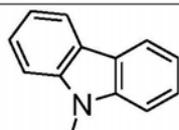
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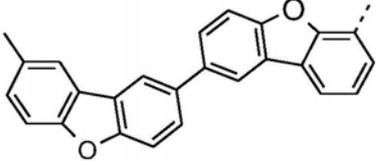
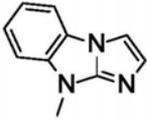
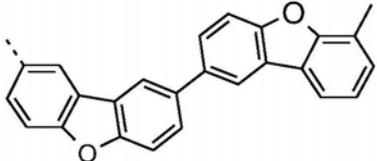
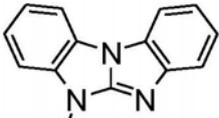
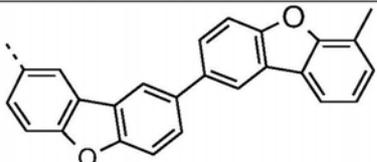
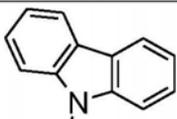
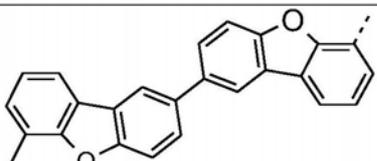
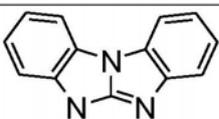
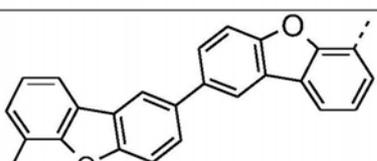
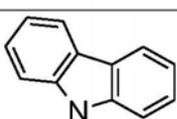
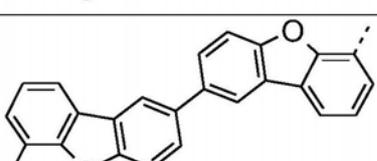
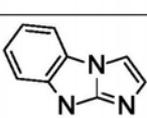
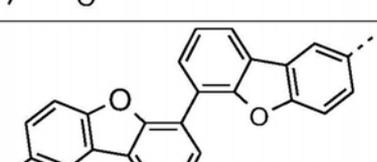
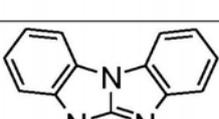
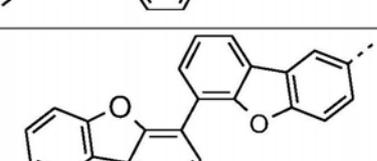
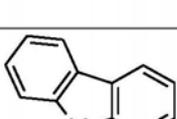
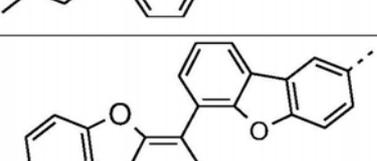
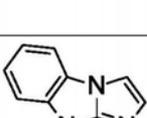
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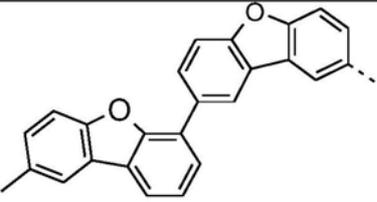
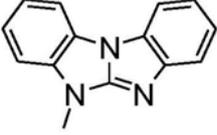
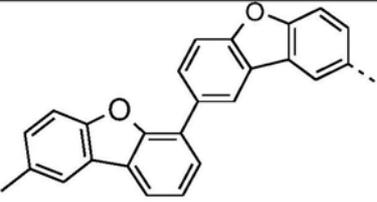
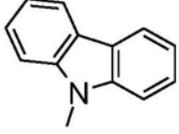
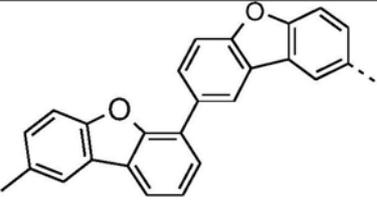
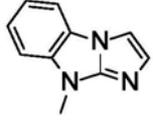
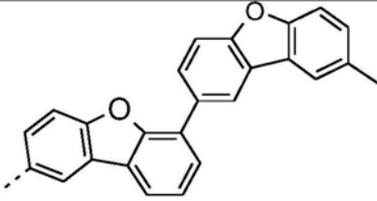
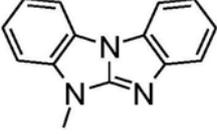
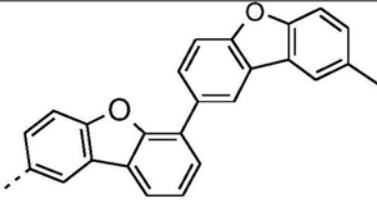
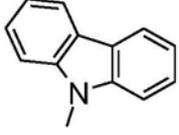
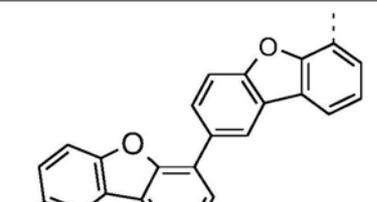
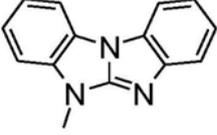
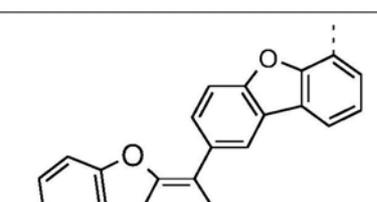
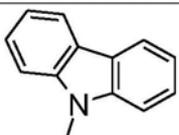
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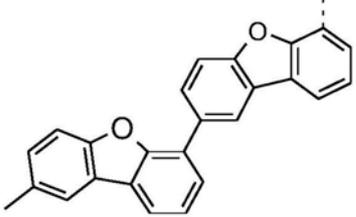
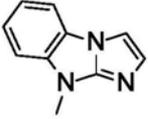
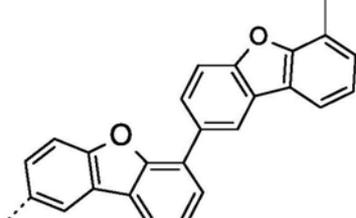
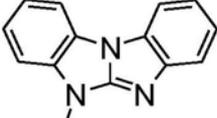
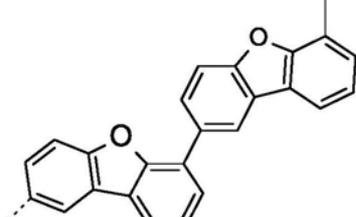
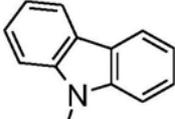
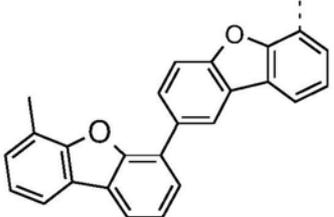
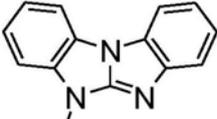
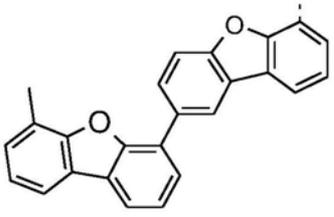
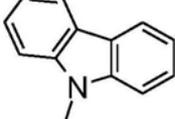
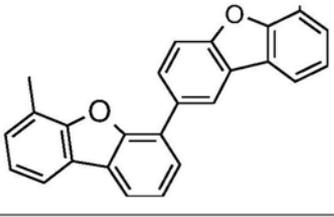
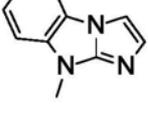
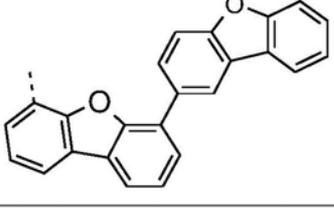
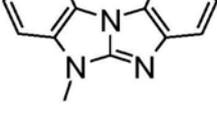
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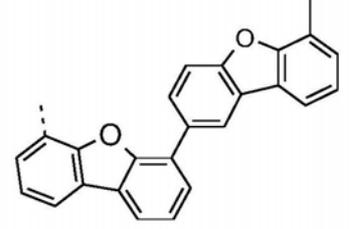
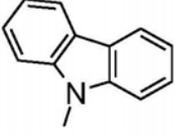
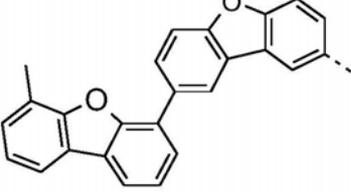
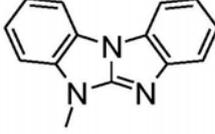
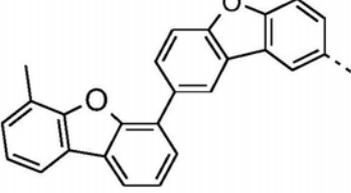
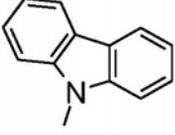
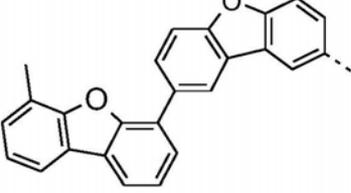
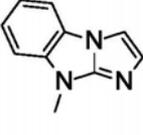
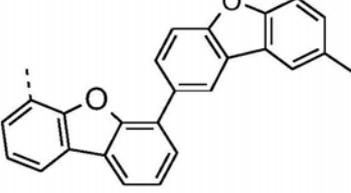
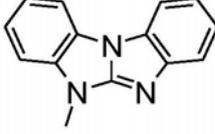
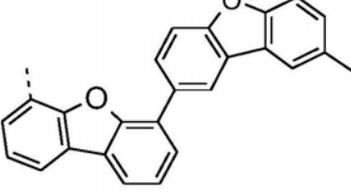
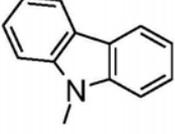
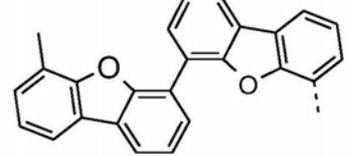
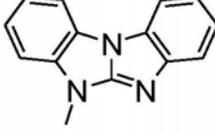
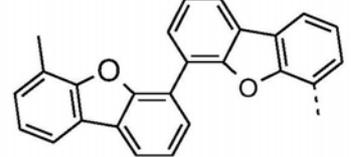
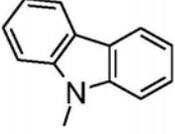
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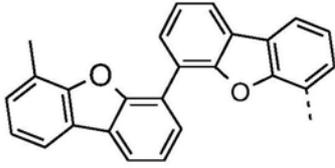
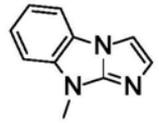
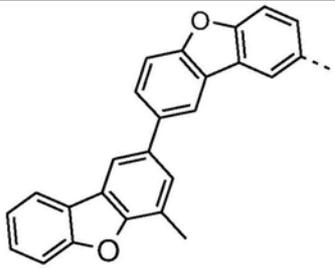
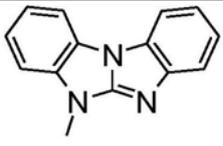
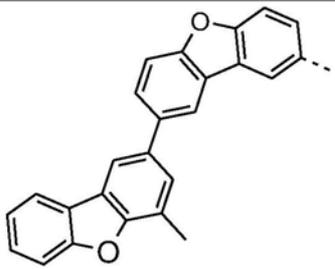
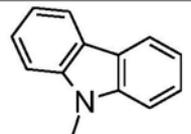
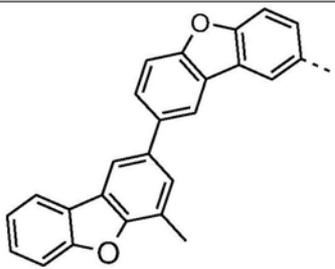
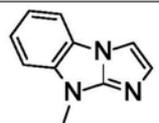
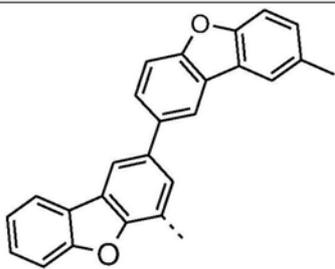
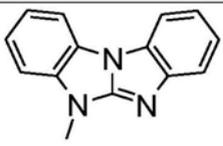
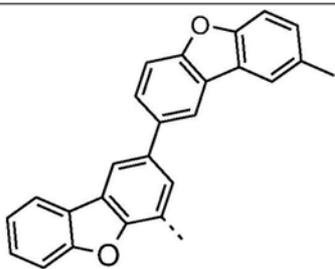
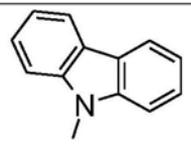
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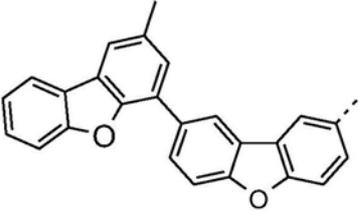
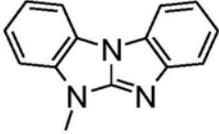
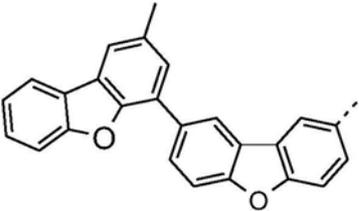
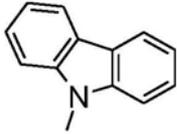
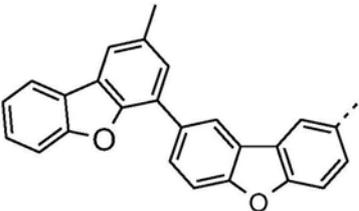
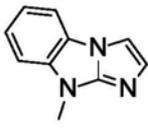
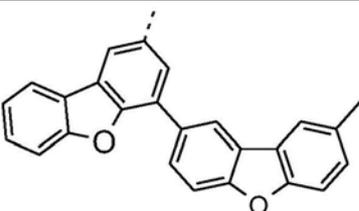
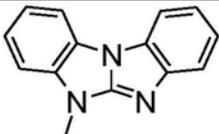
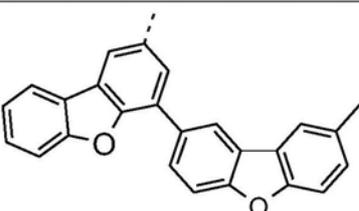
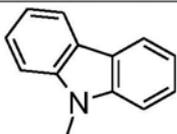
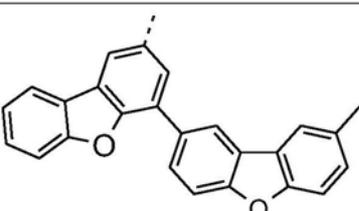
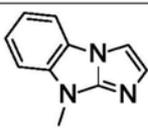
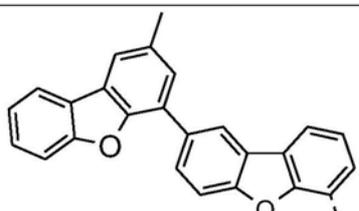
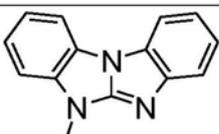
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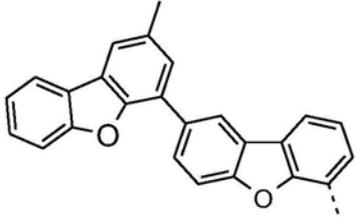
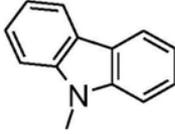
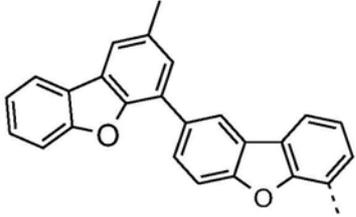
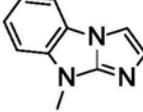
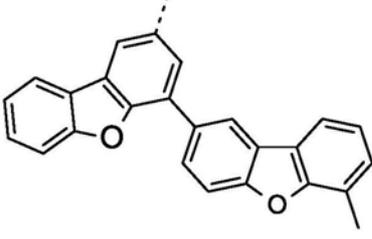
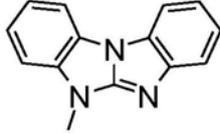
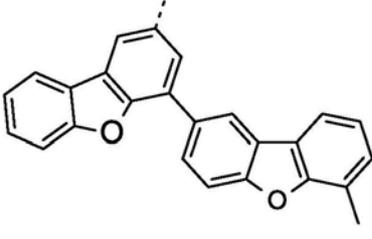
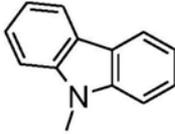
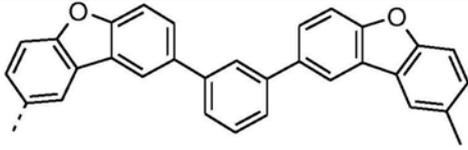
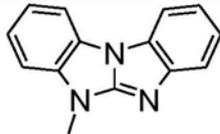
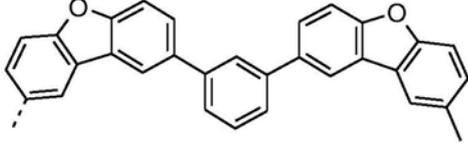
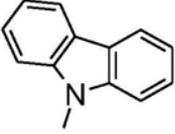
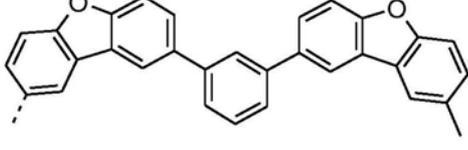
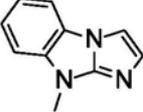
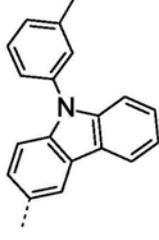
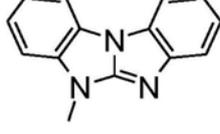
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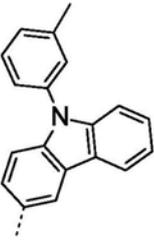
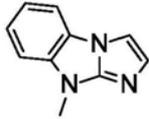
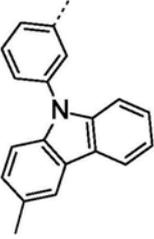
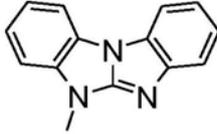
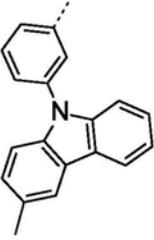
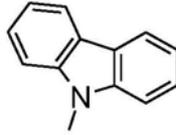
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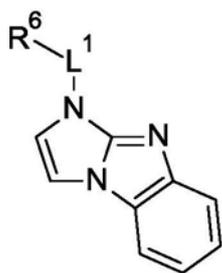
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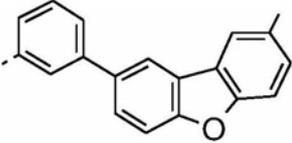
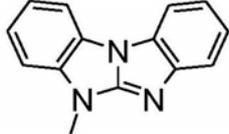
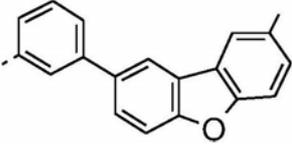
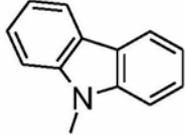
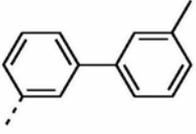
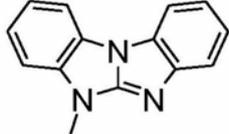
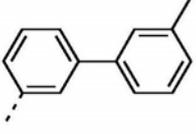
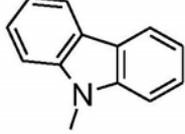
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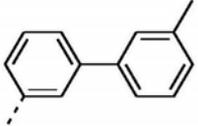
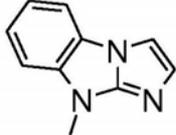
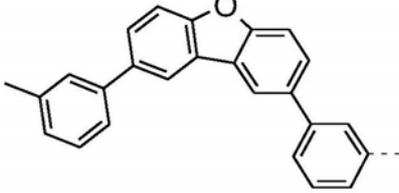
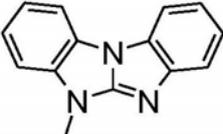
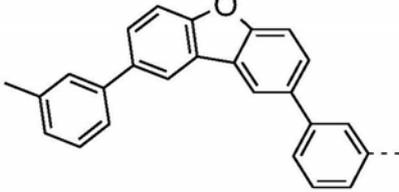
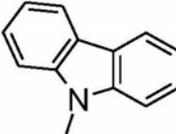
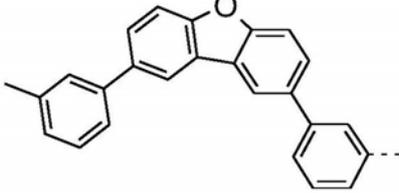
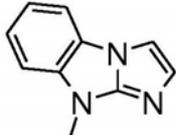
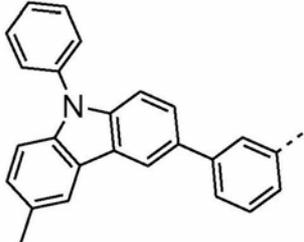
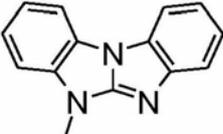
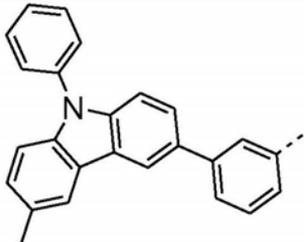
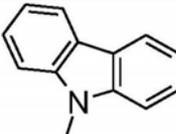
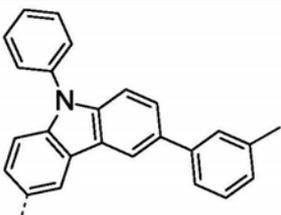
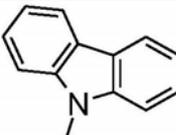
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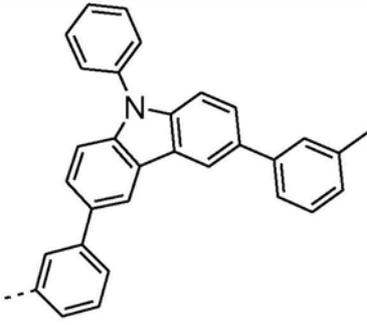
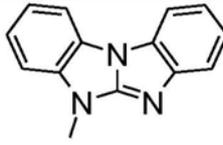
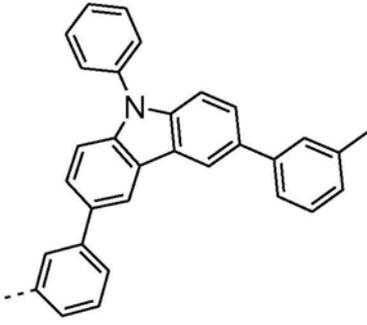
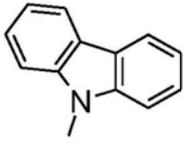
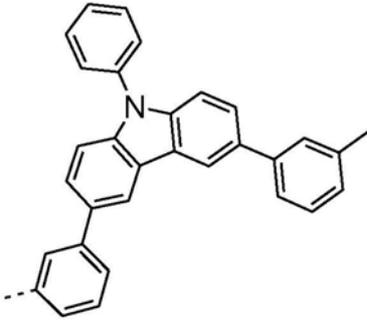
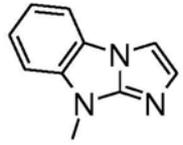
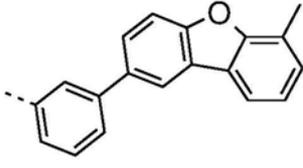
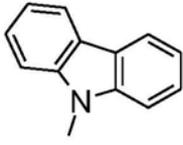
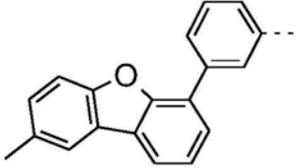
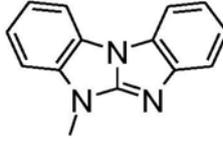
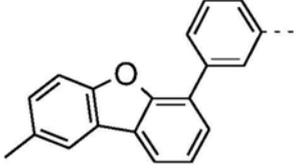
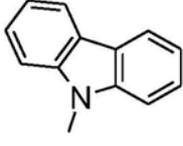
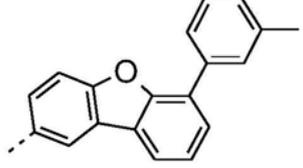
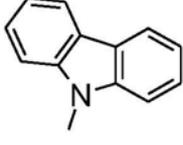
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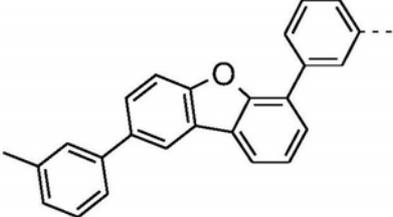
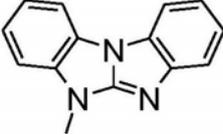
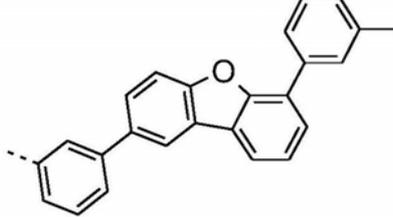
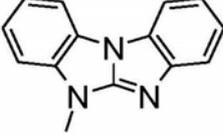
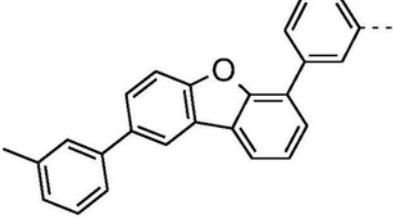
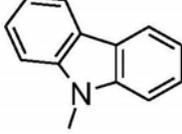
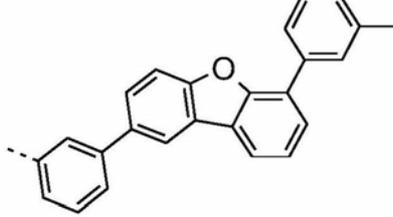
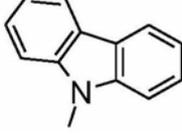
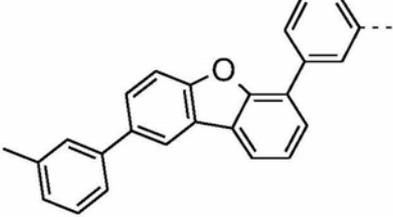
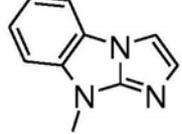
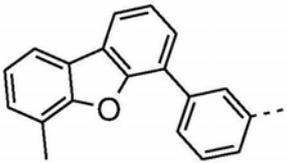
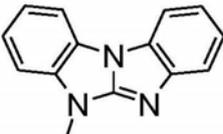
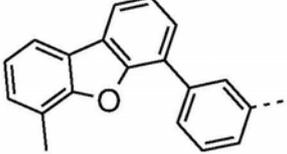
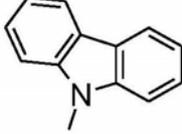
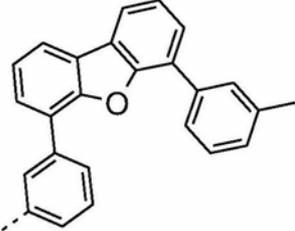
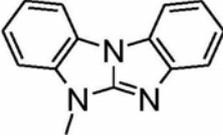
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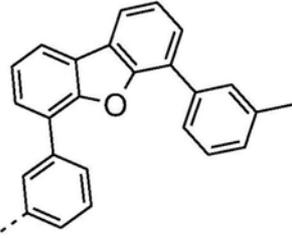
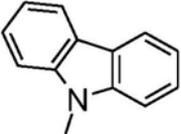
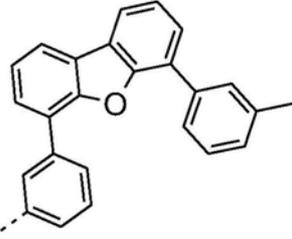
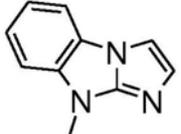
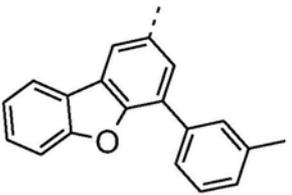
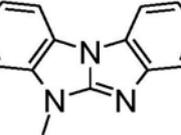
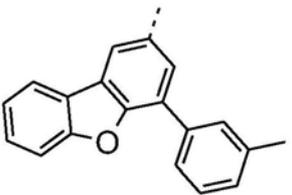
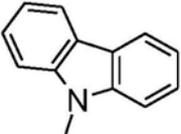
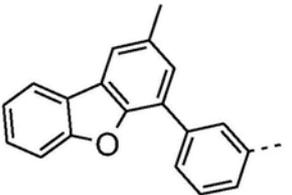
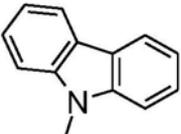
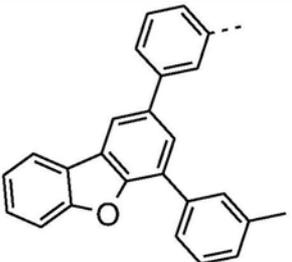
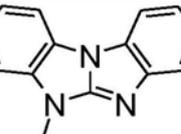
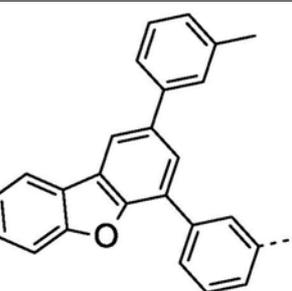
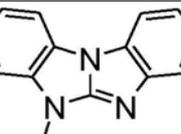


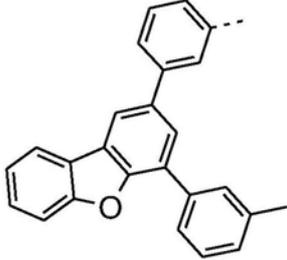
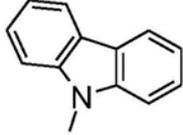
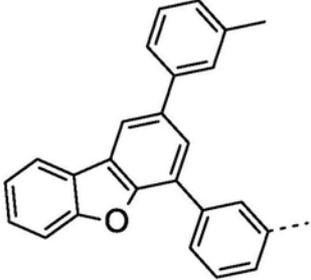
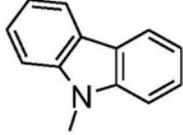
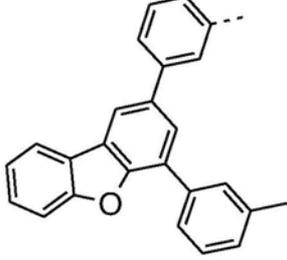
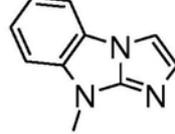
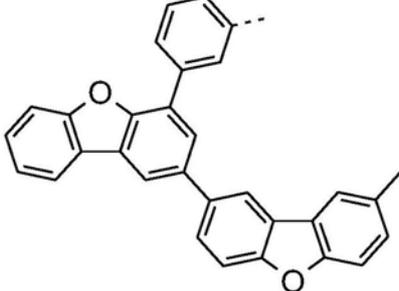
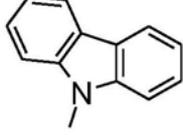
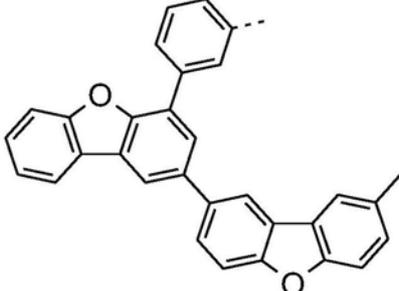
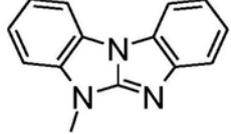
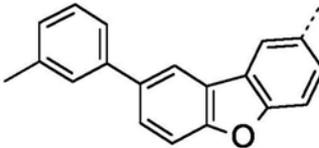
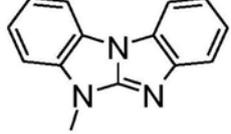
CpE.	$L^{1(3)}$	R^6
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<p>E-2</p>		
<p>E-3</p>		
<p>E-4</p>		

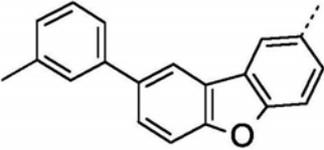
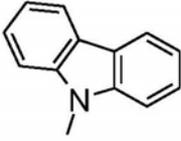
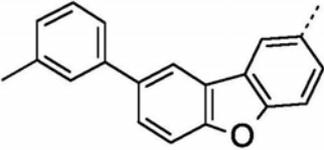
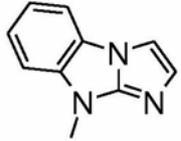
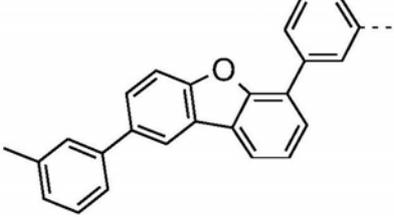
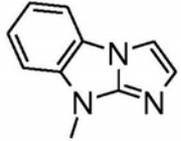
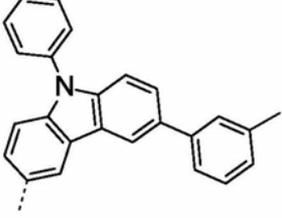
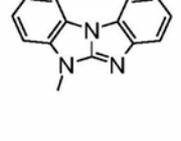
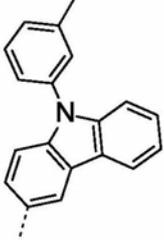
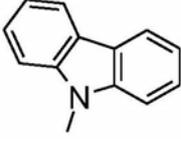
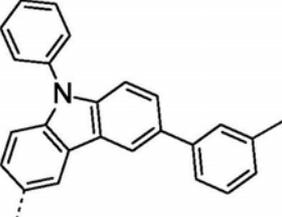
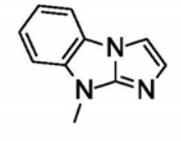
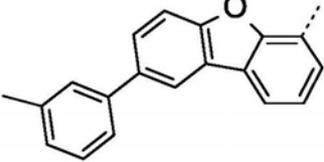
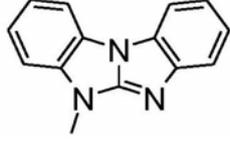
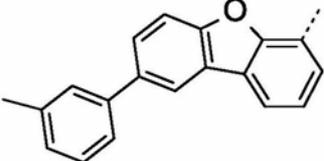
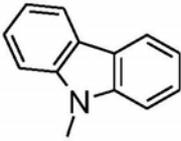
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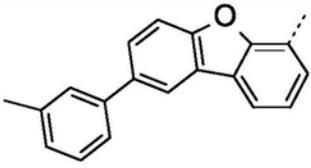
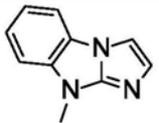
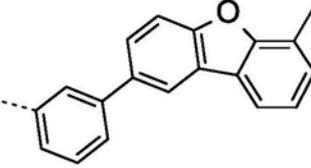
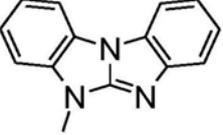
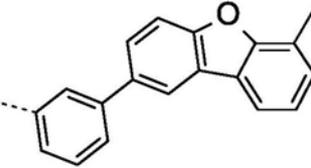
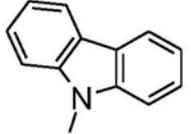
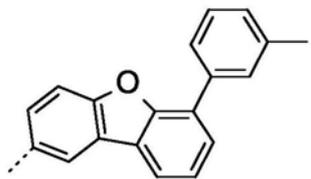
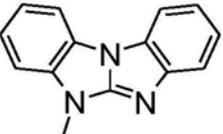
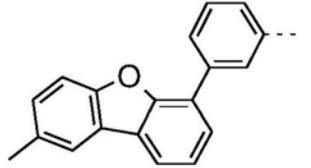
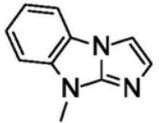
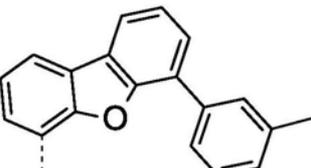
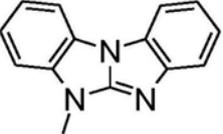
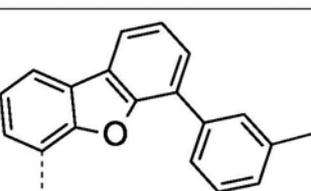
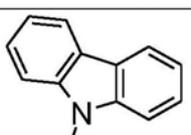
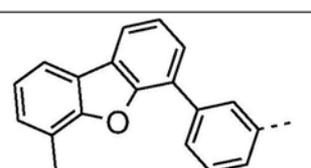
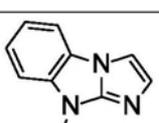
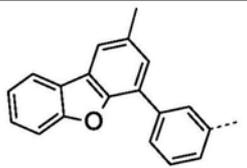
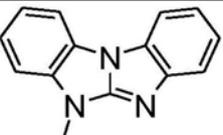
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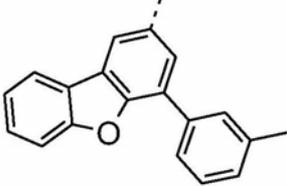
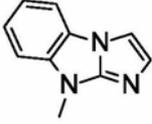
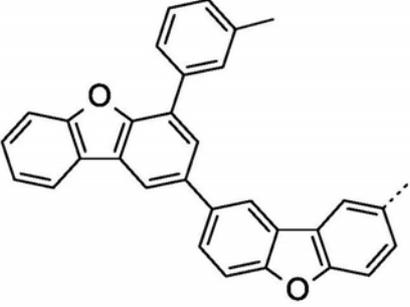
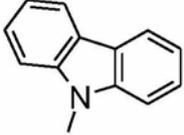
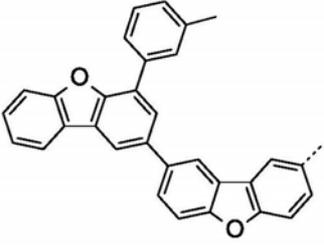
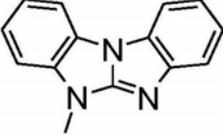
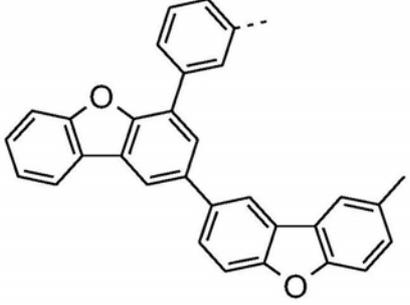
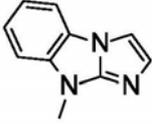
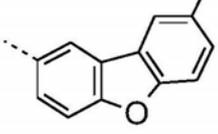
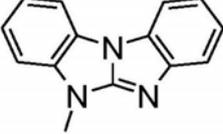
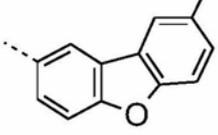
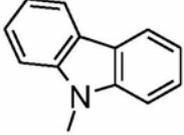
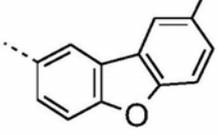
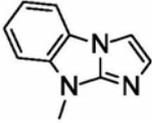
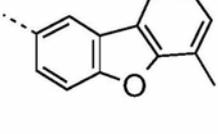
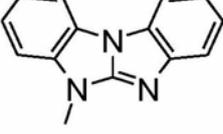
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<p>E-20</p>		
<p>E-21</p>		
<p>E-22</p>		
<p>E-23</p>		
<p>E-24</p>		
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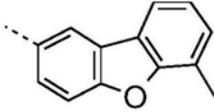
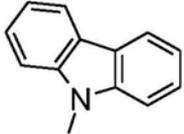
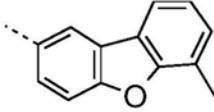
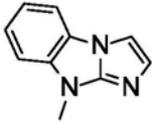
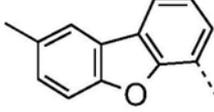
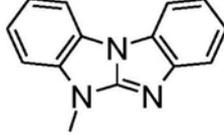
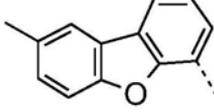
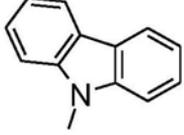
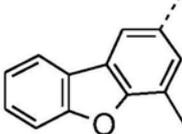
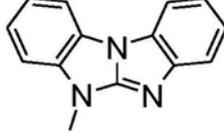
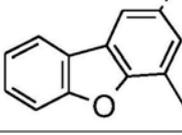
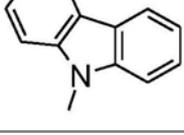
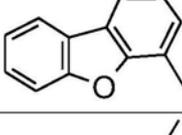
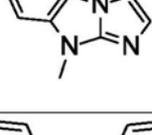
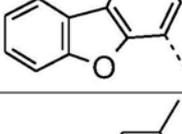
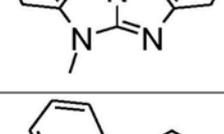
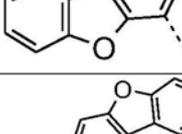
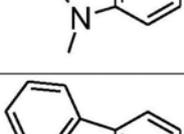
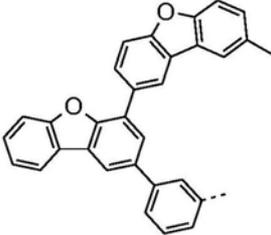
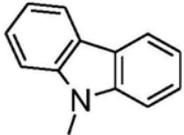
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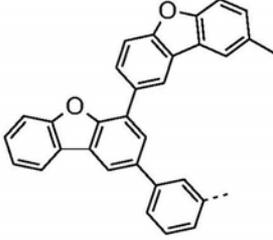
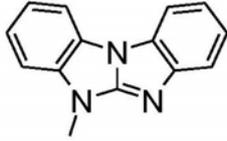
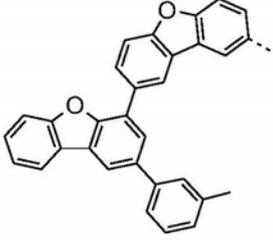
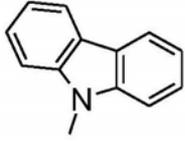
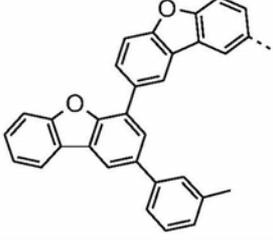
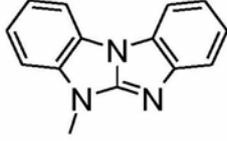
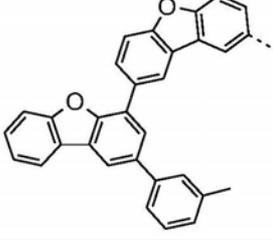
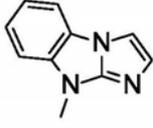
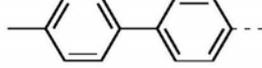
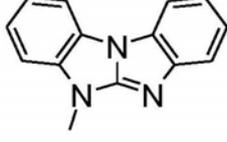
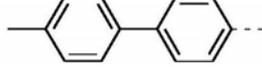
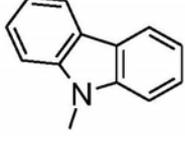
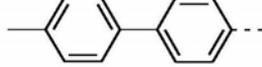
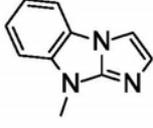
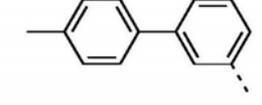
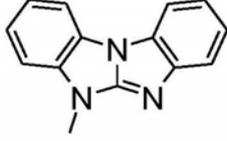
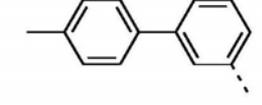
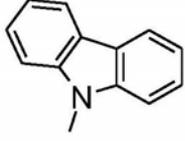
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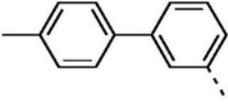
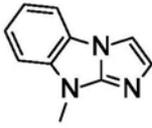
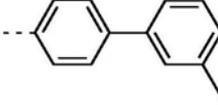
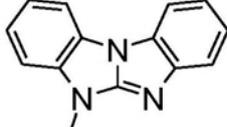
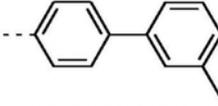
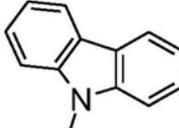
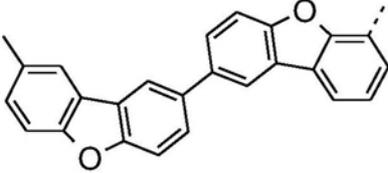
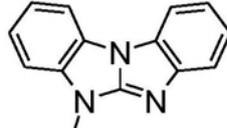
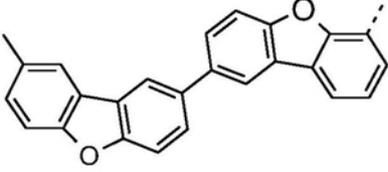
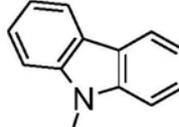
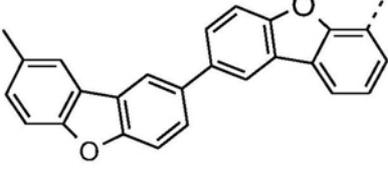
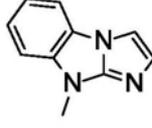
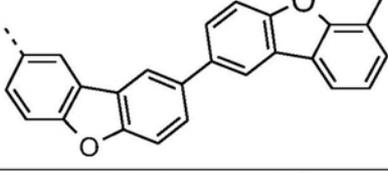
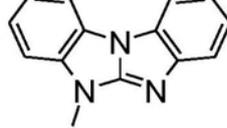
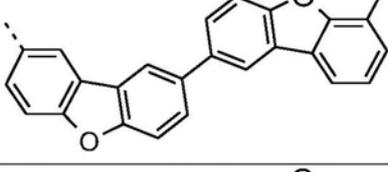
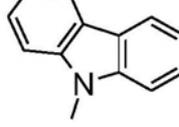
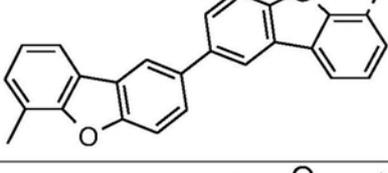
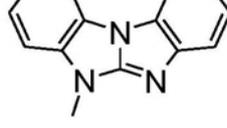
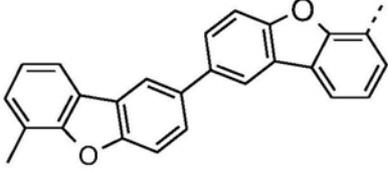
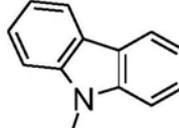
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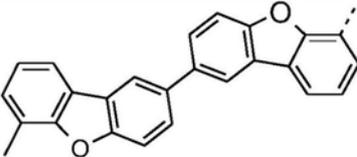
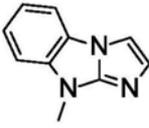
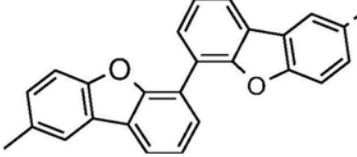
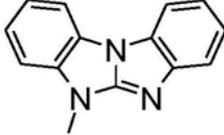
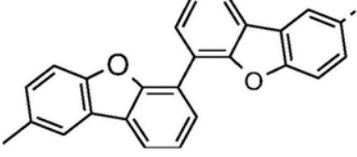
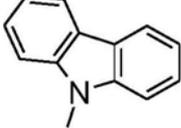
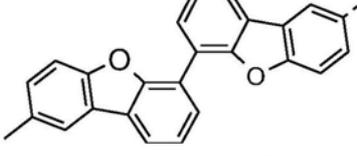
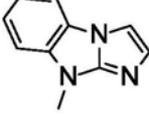
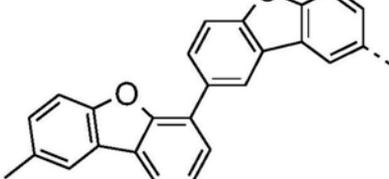
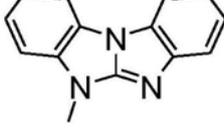
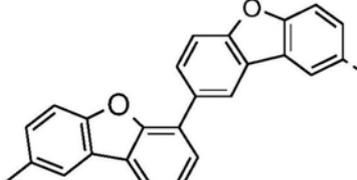
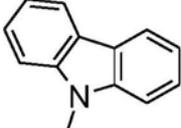
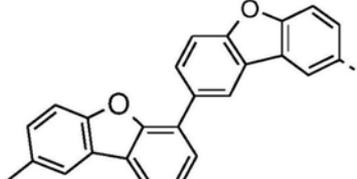
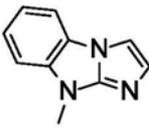
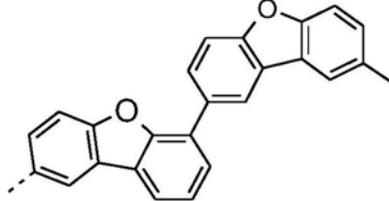
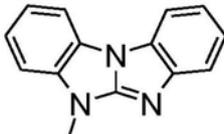
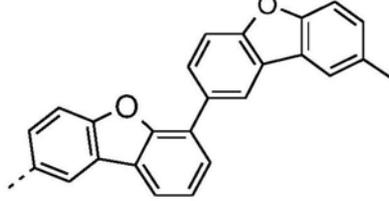
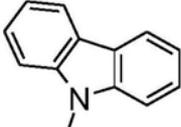
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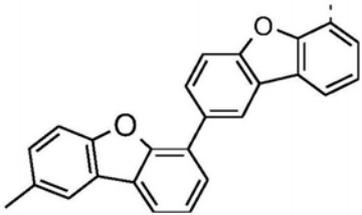
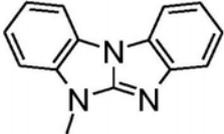
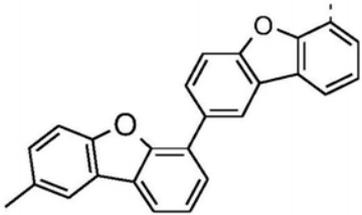
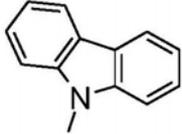
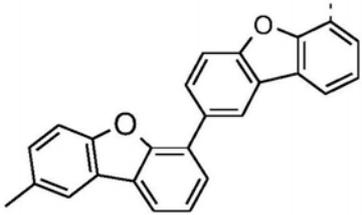
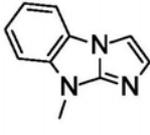
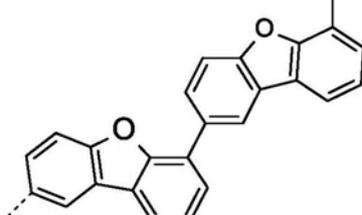
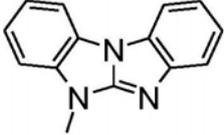
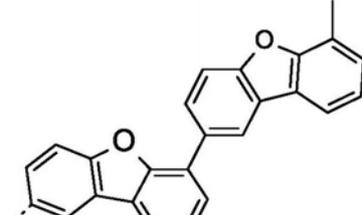
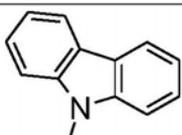
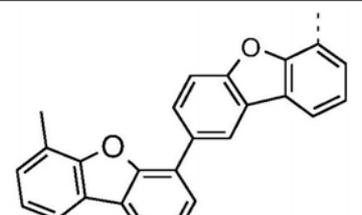
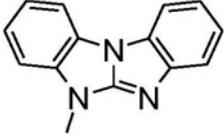
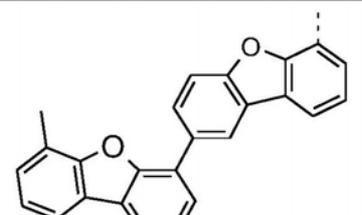
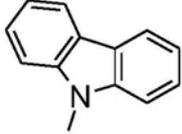
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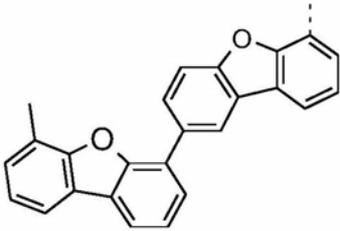
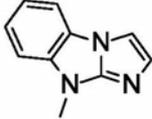
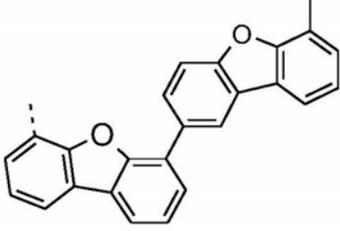
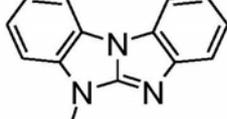
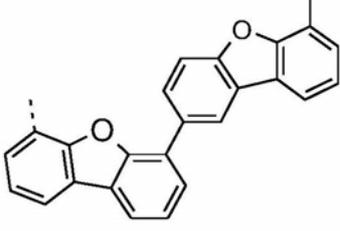
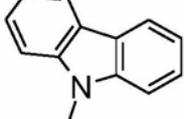
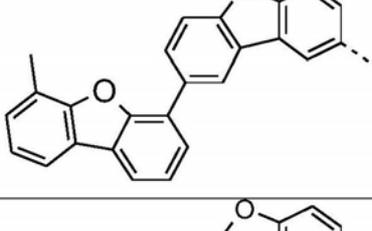
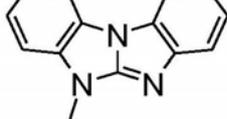
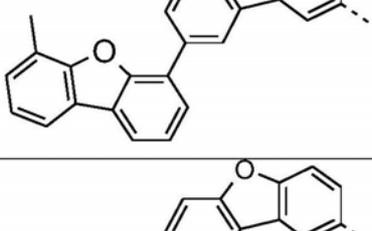
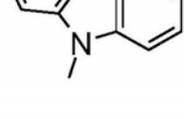
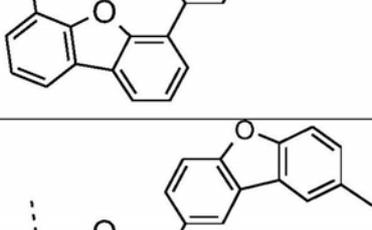
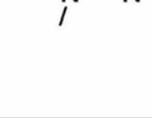
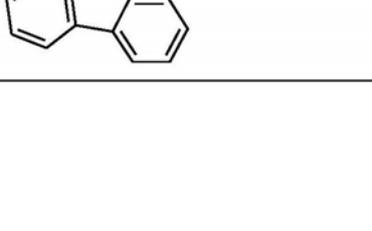
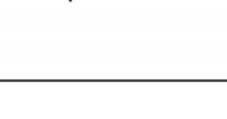
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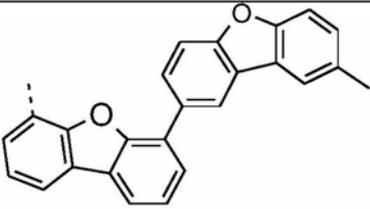
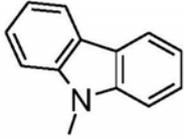
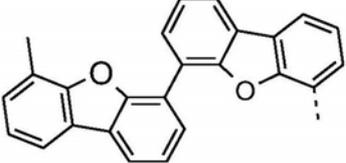
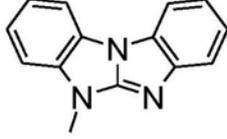
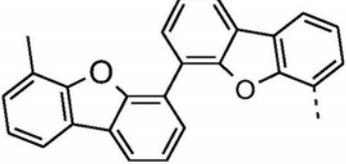
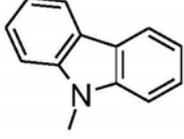
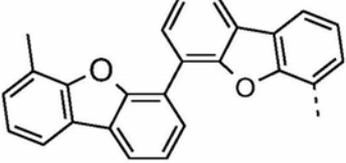
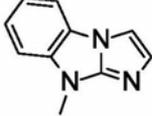
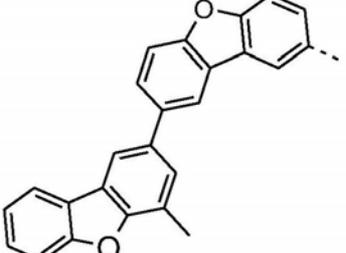
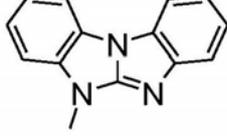
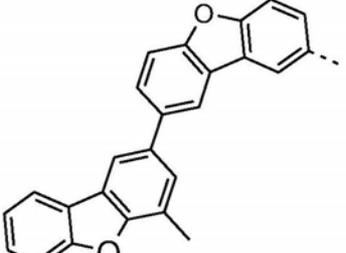
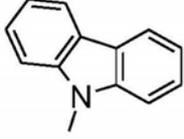
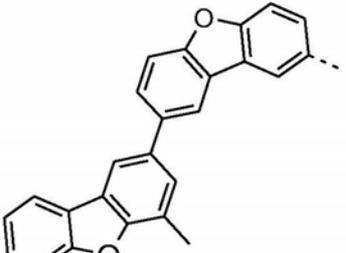
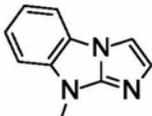
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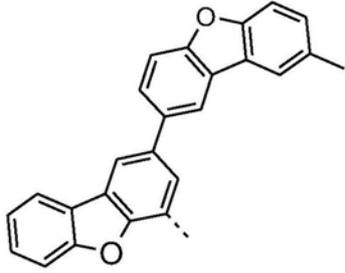
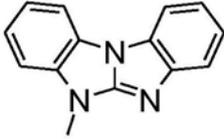
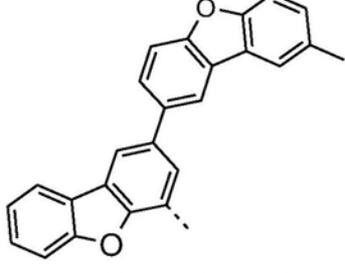
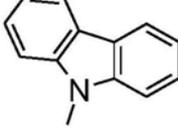
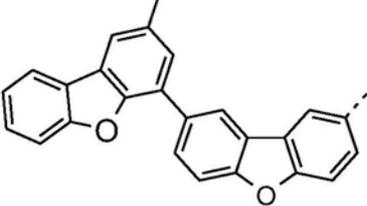
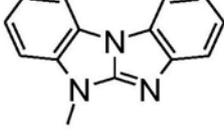
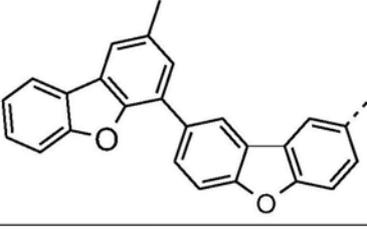
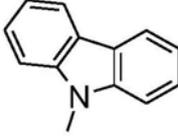
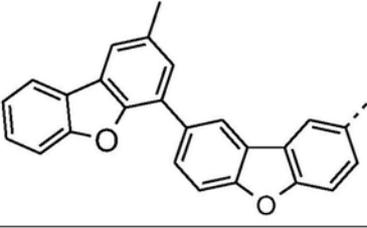
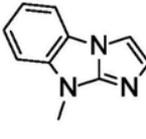
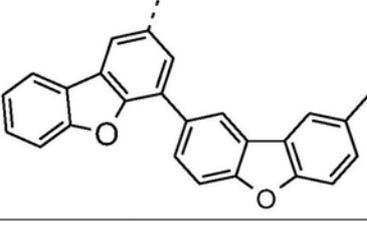
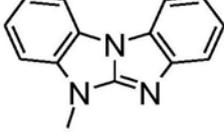
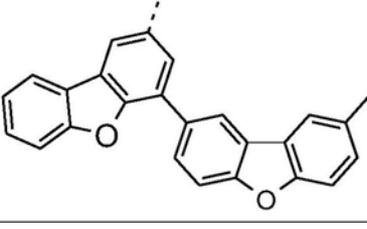
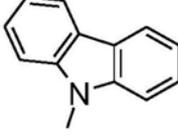
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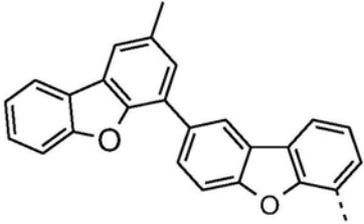
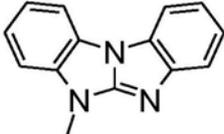
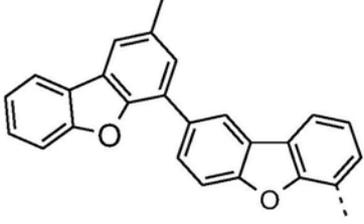
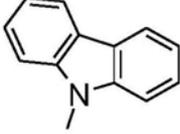
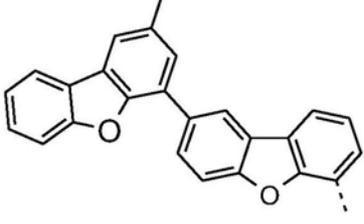
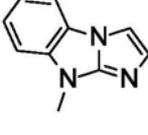
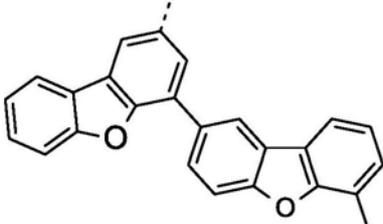
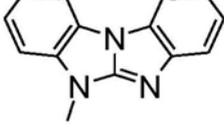
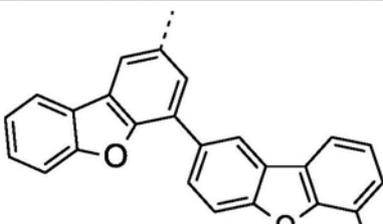
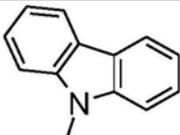
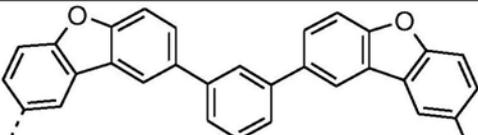
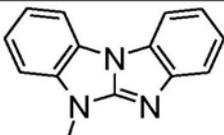
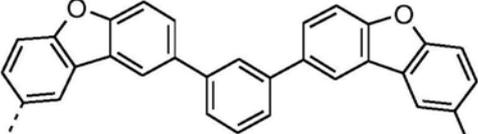
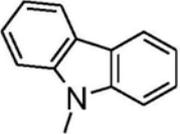
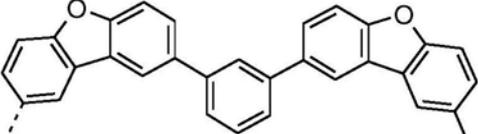
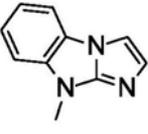
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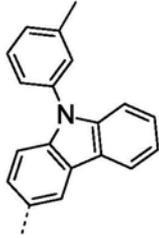
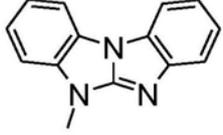
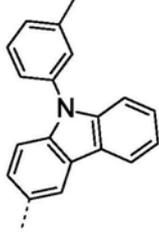
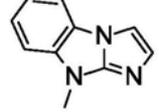
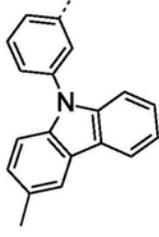
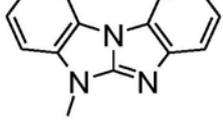
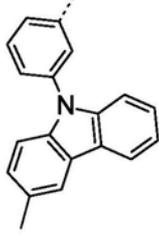
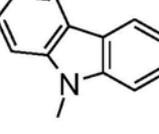
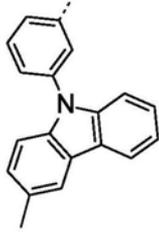
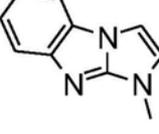
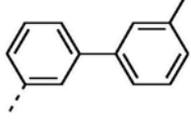
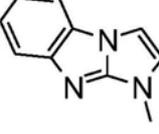
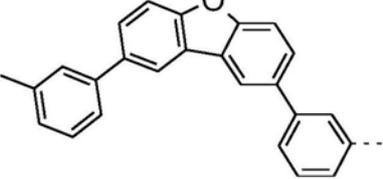
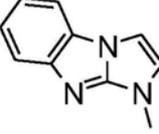
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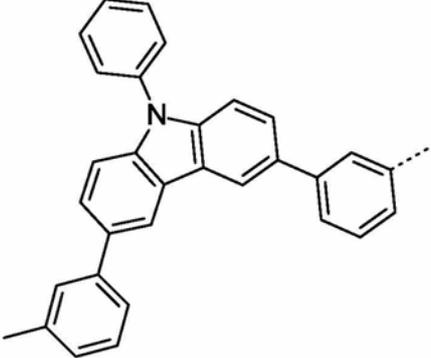
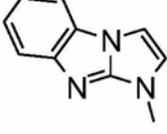
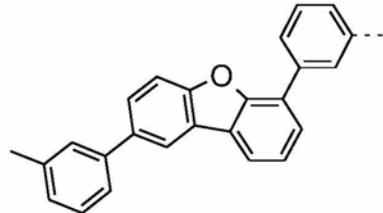
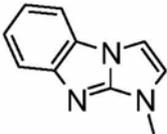
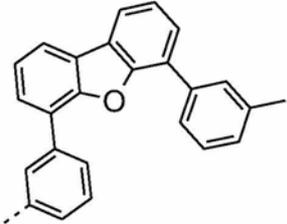
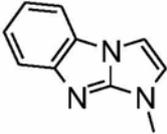
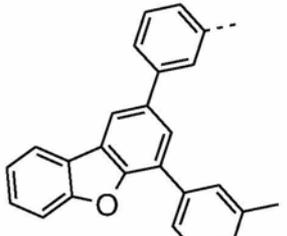
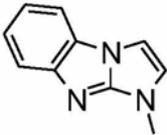
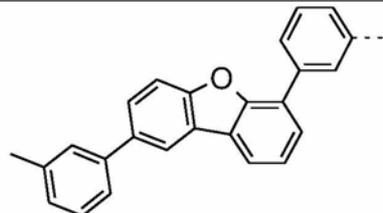
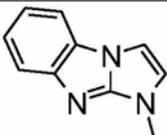
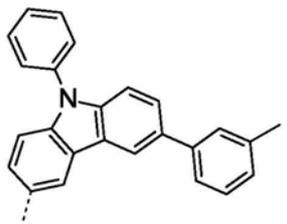
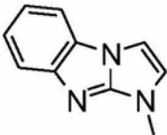
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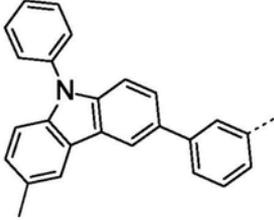
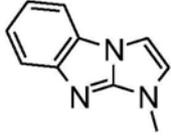
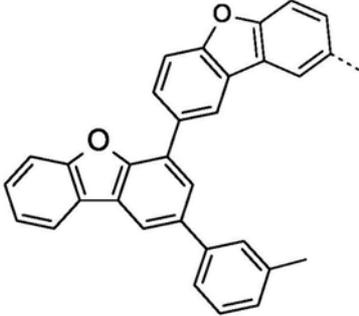
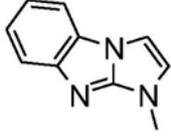
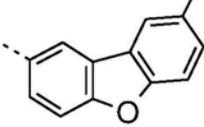
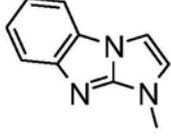
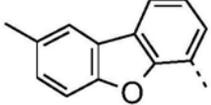
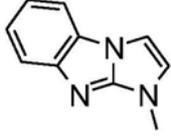
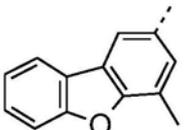
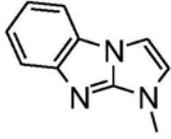
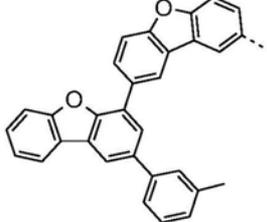
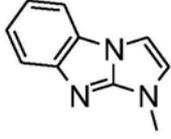
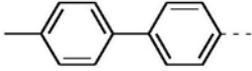
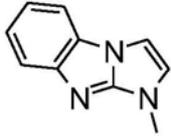
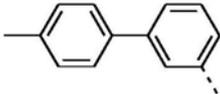
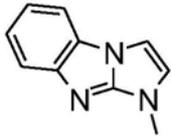
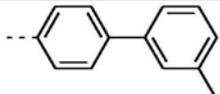
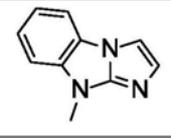
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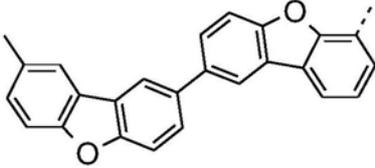
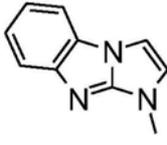
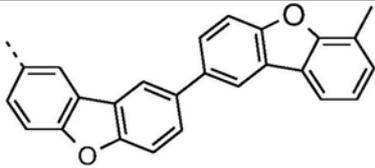
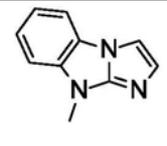
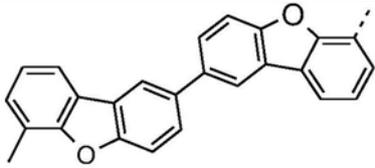
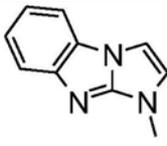
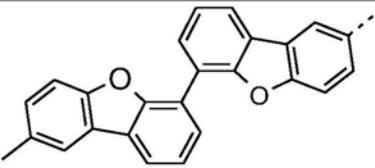
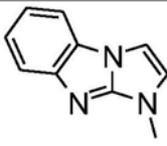
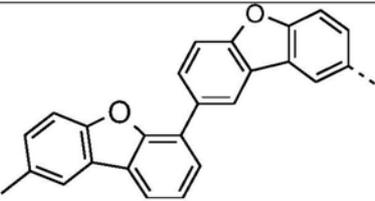
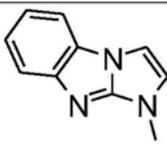
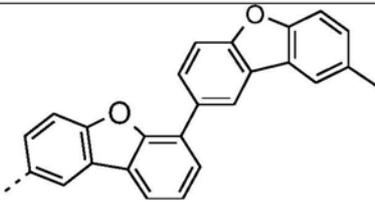
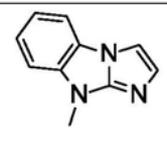
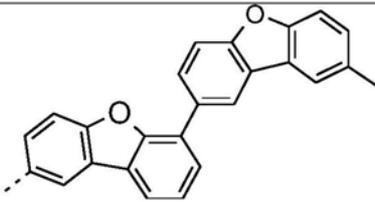
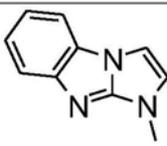
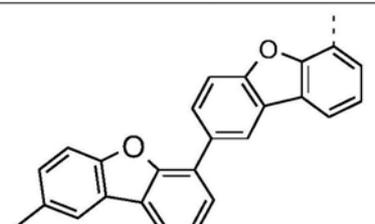
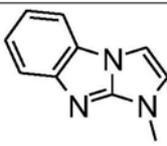
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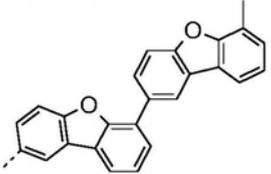
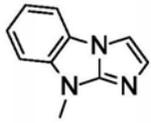
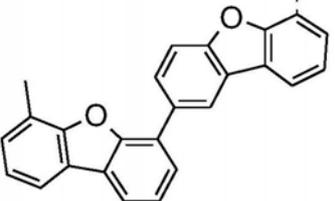
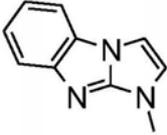
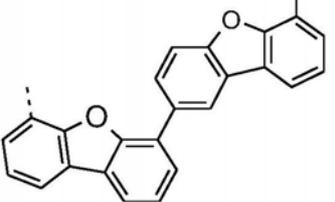
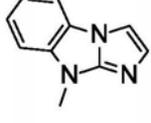
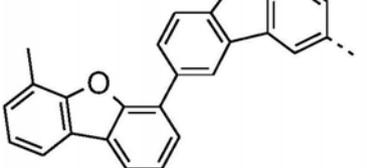
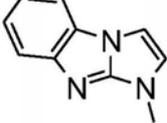
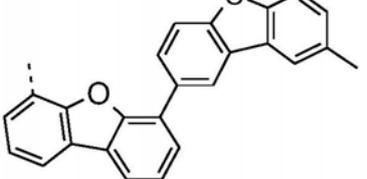
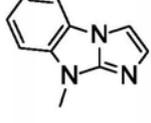
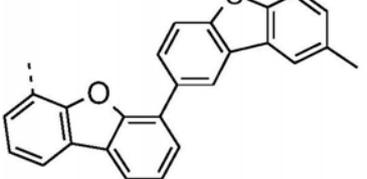
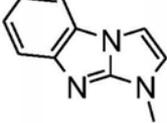
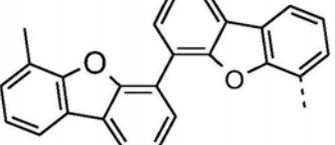
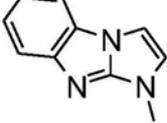
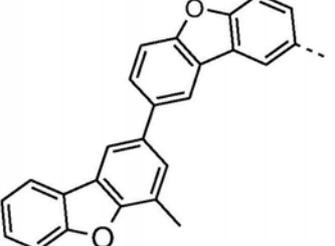
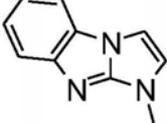
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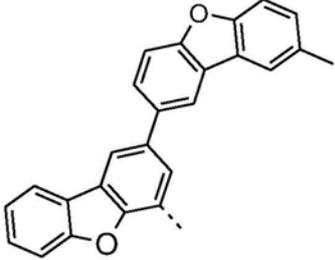
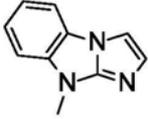
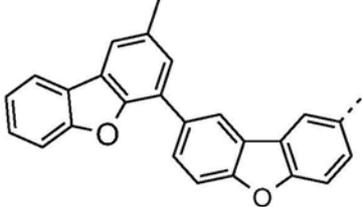
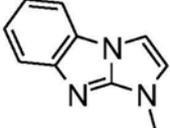
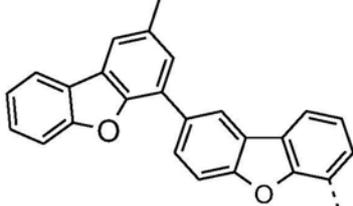
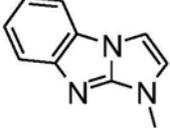
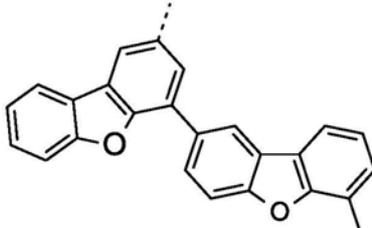
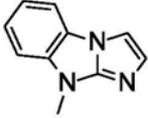
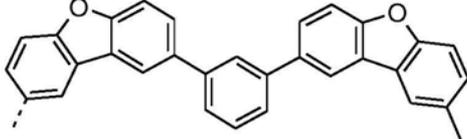
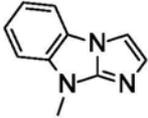
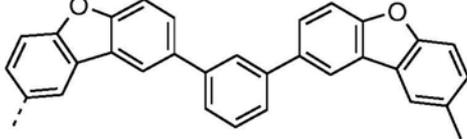
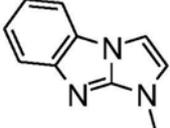
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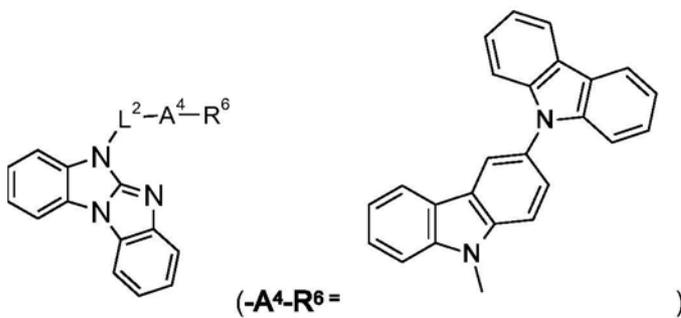
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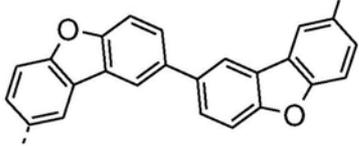
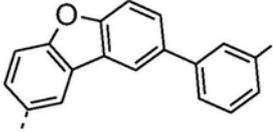
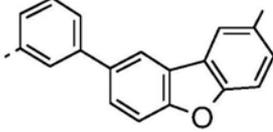
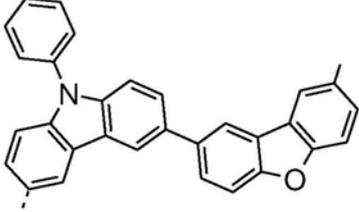
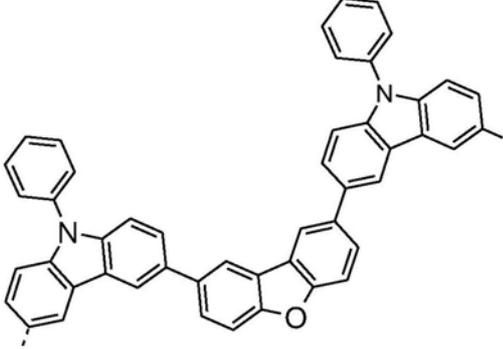
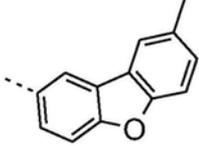
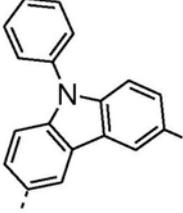
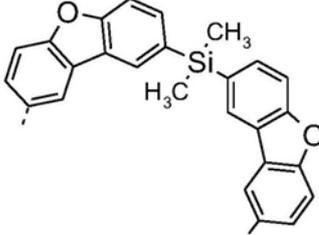
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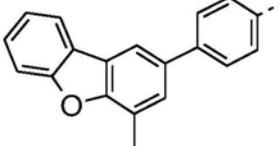
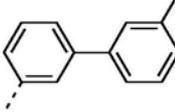
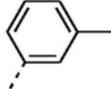
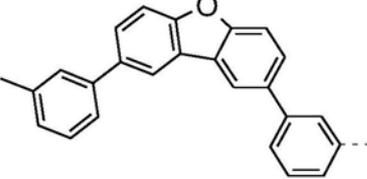
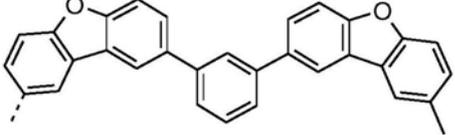
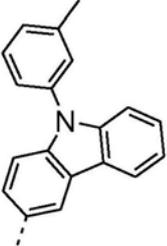
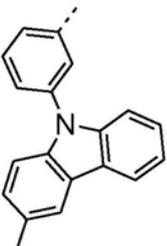
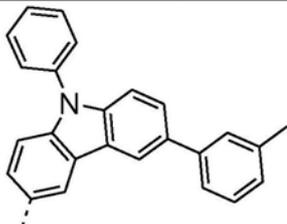
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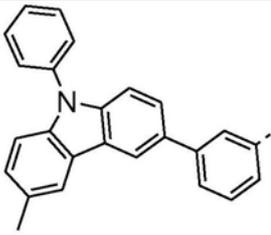
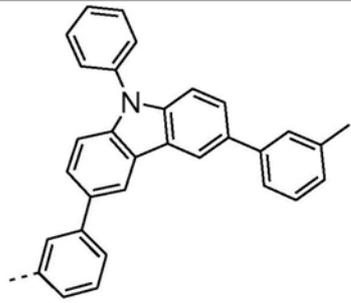
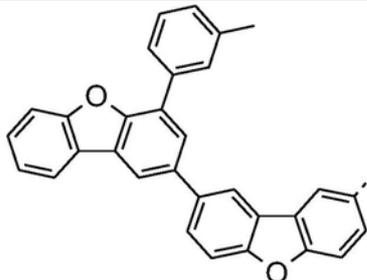
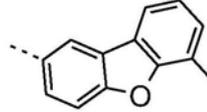
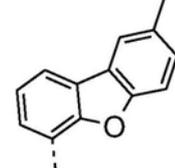
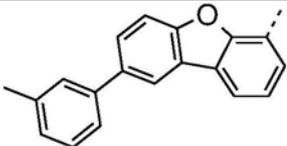
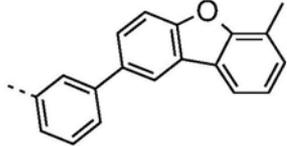
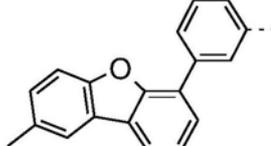
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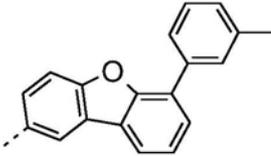
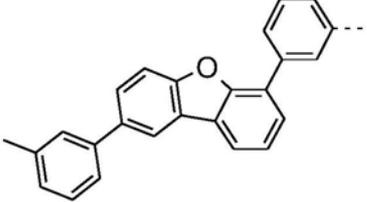
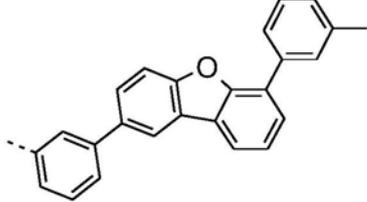
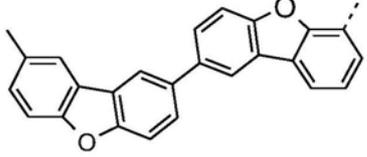
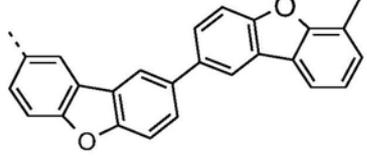
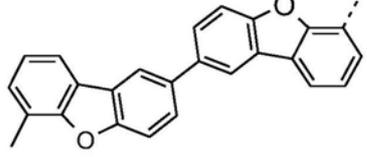
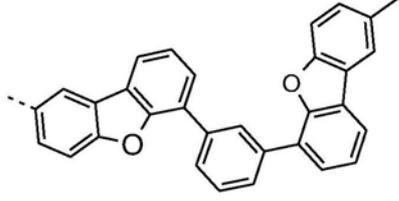
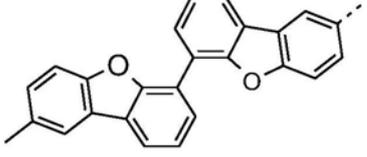


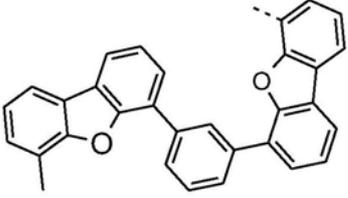
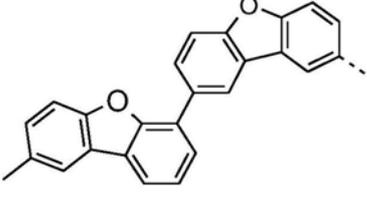
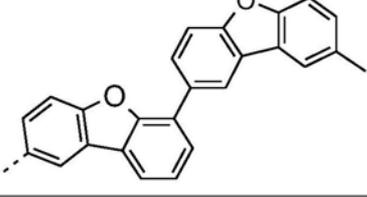
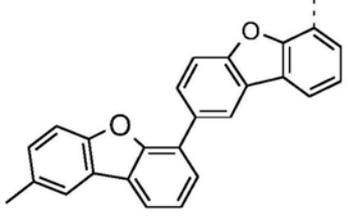
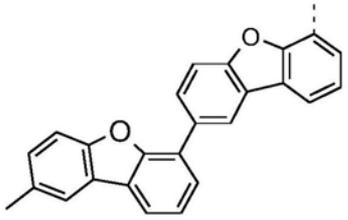
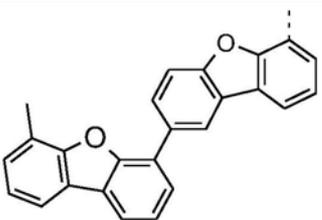
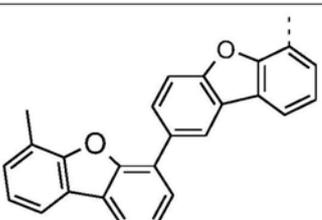
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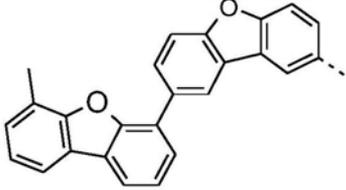
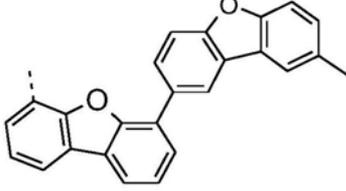
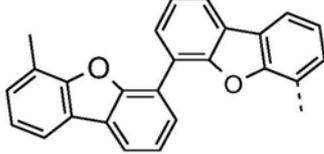
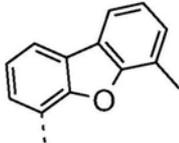
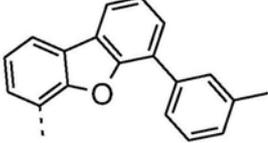
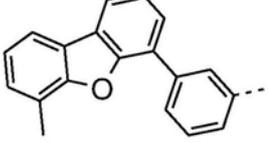
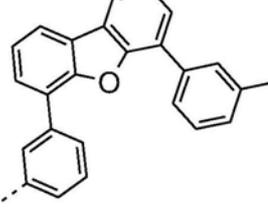
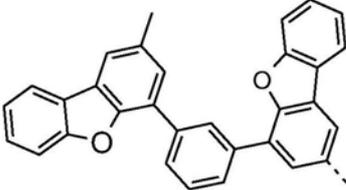
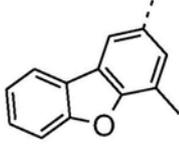
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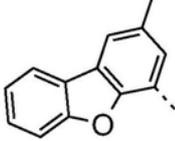
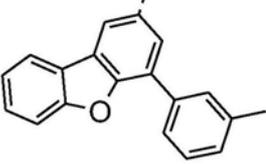
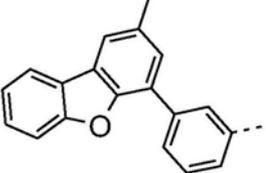
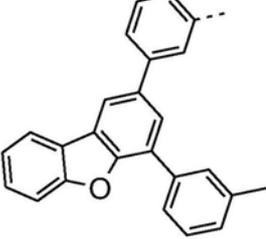
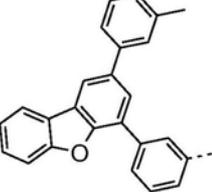
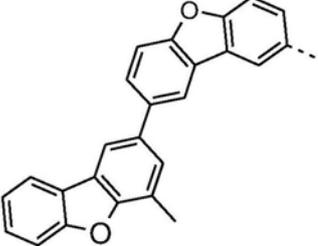
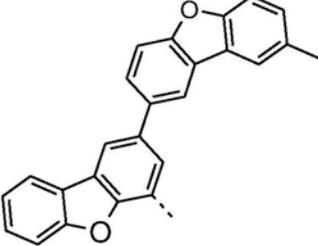
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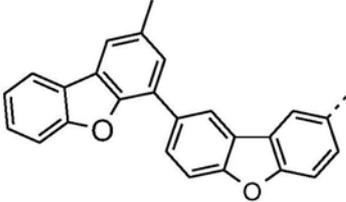
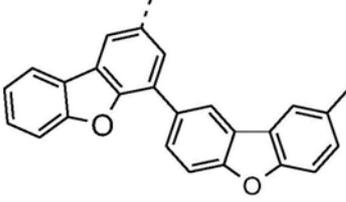
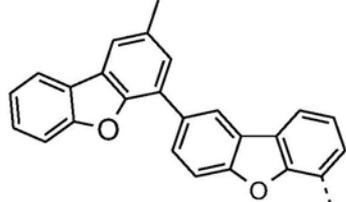
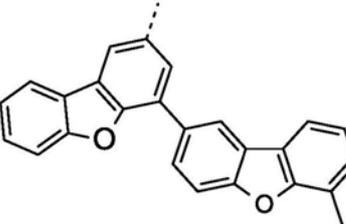
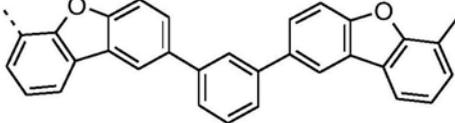
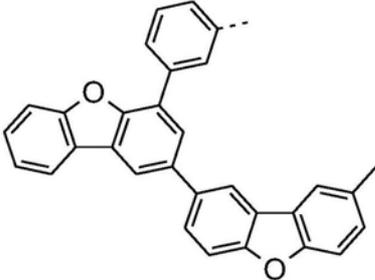
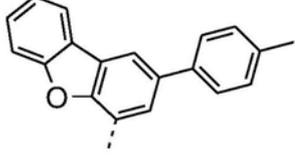
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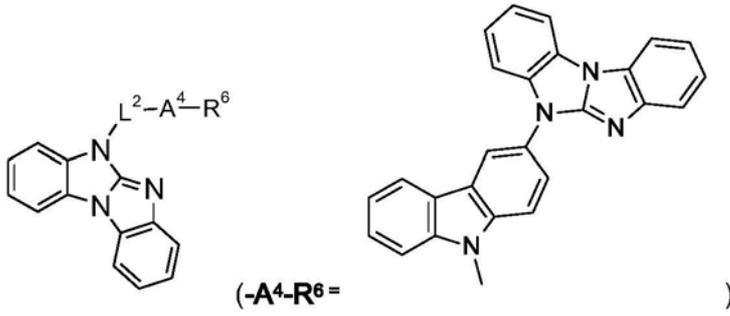
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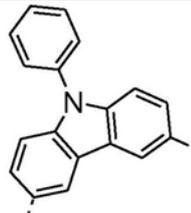
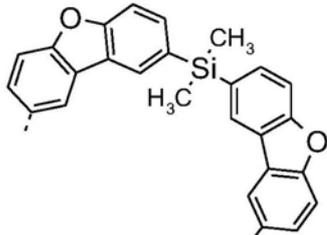
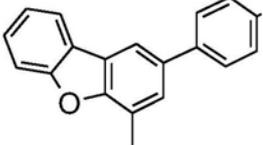
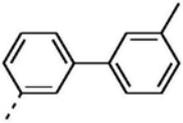
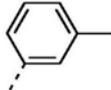
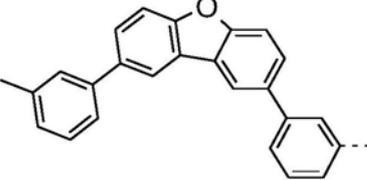
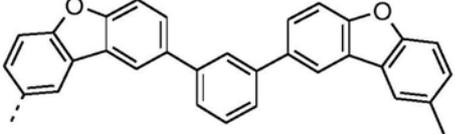
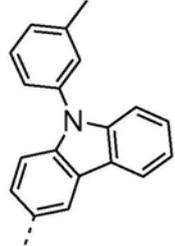
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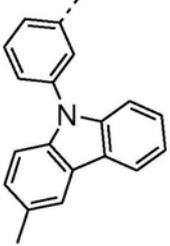
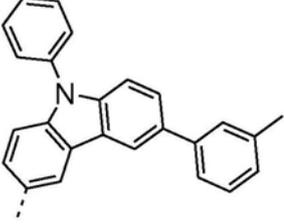
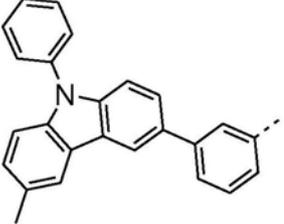
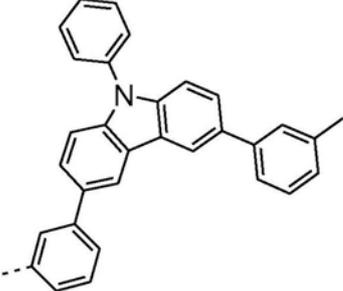
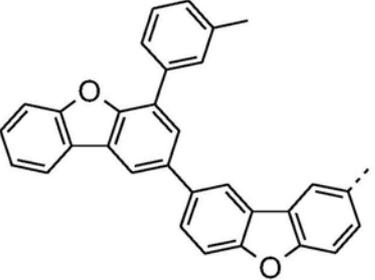
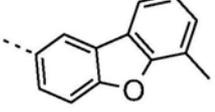
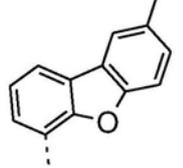
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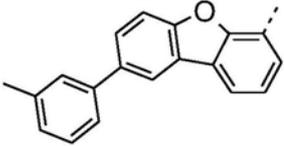
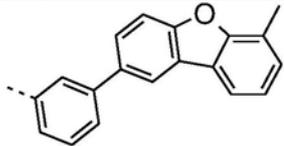
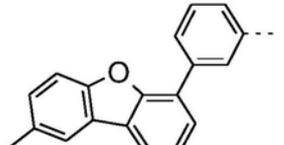
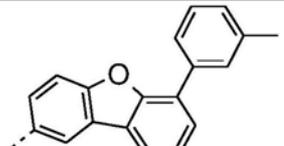
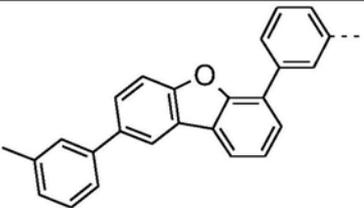
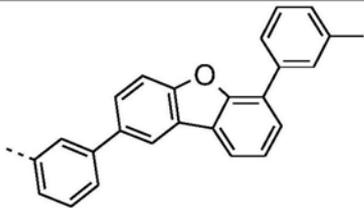
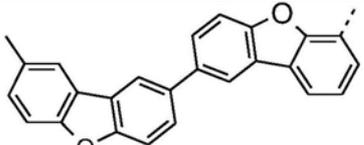
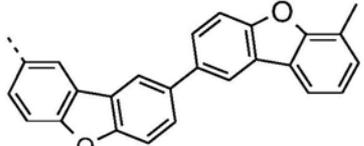
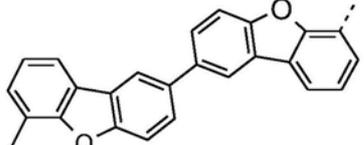
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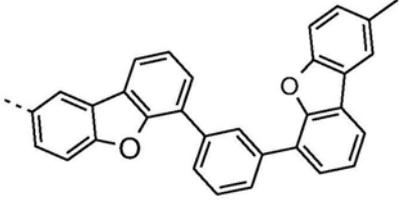
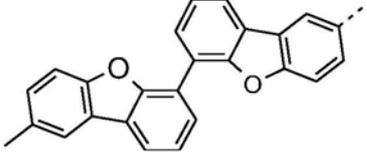
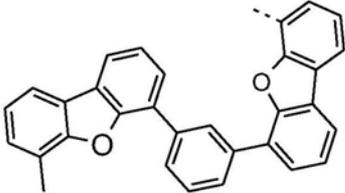
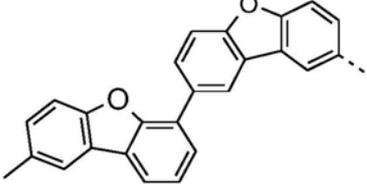
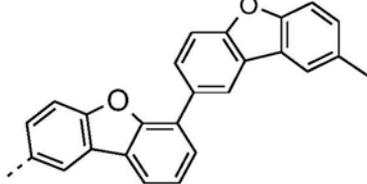
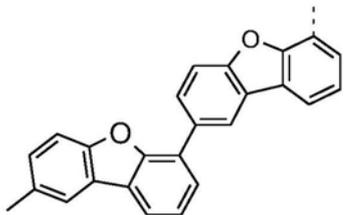
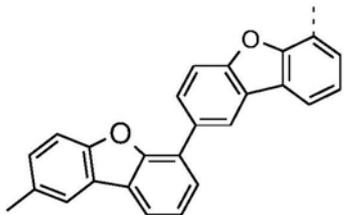
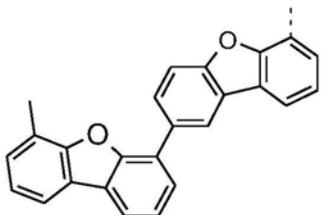


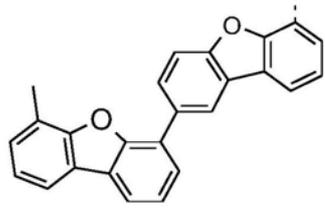
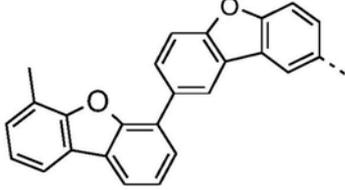
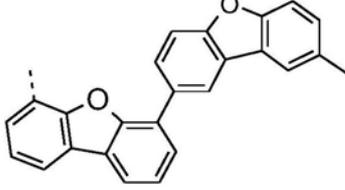
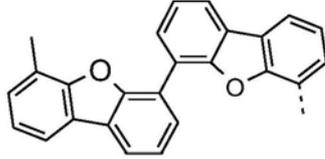
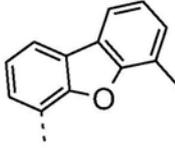
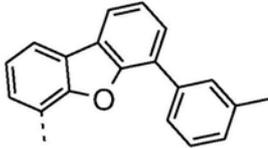
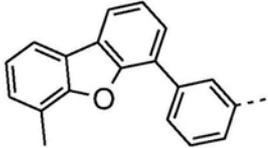
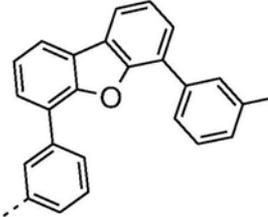
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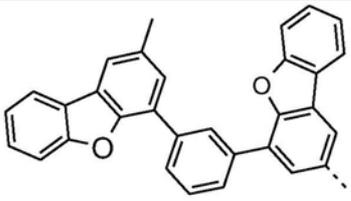
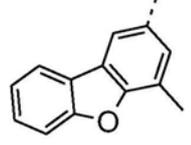
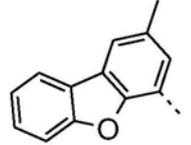
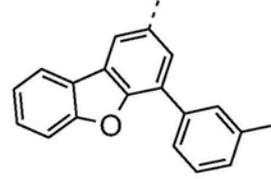
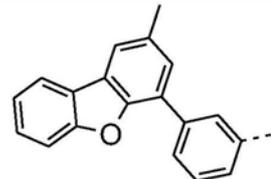
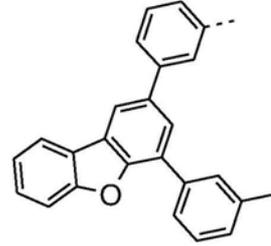
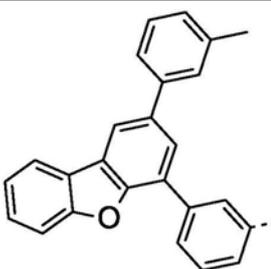
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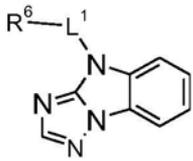
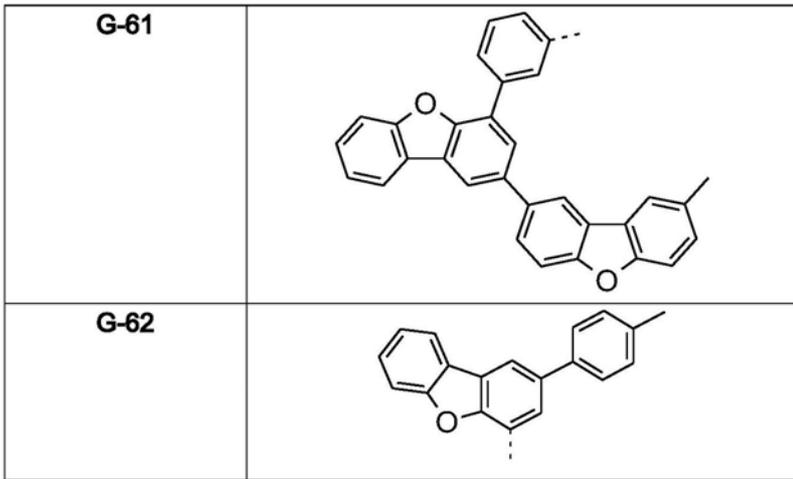
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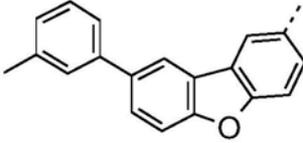
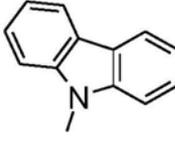
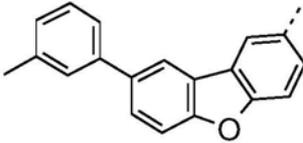
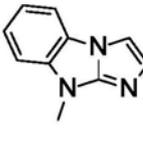
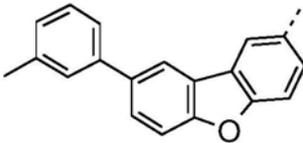
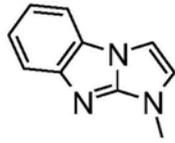
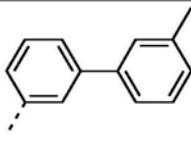
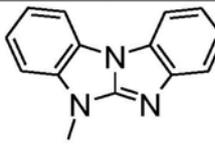
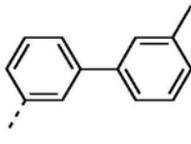
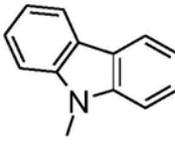
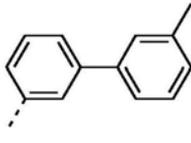
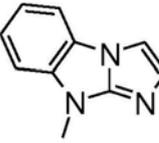
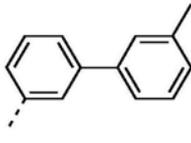
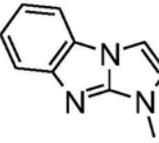
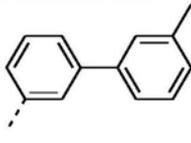
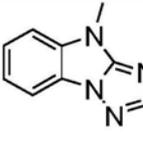
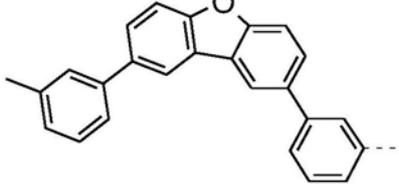
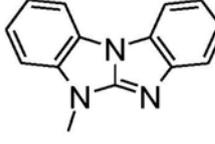
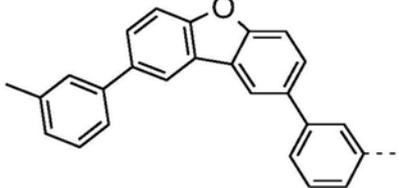
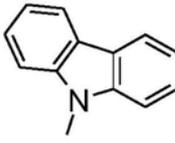
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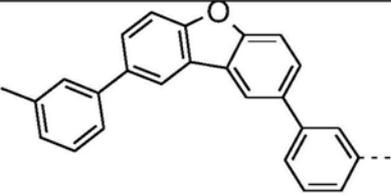
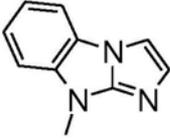
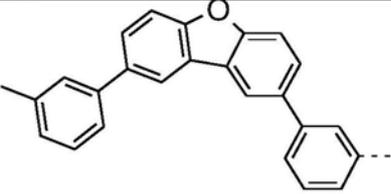
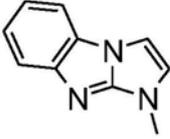
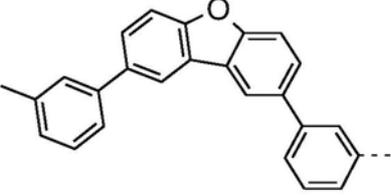
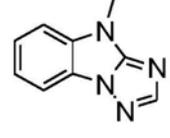
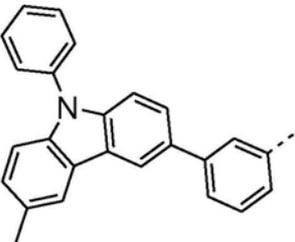
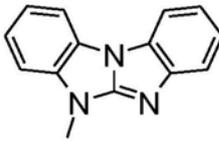
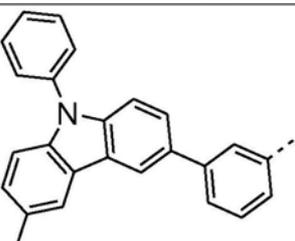
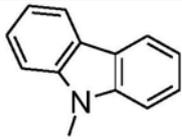
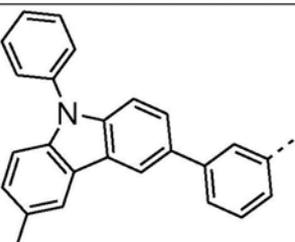
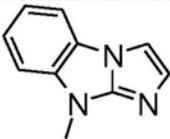
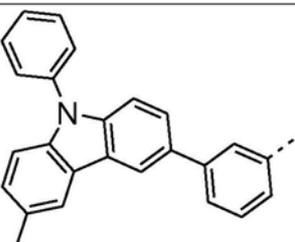
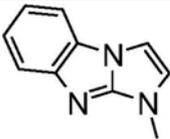
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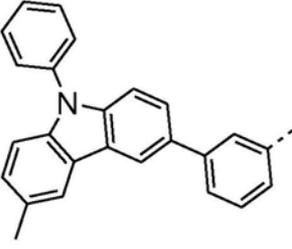
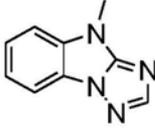
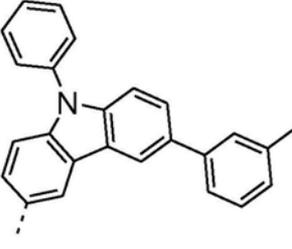
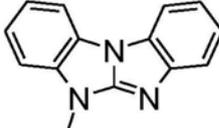
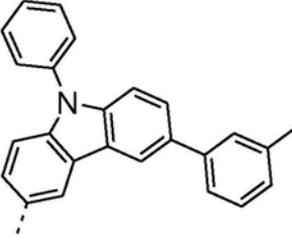
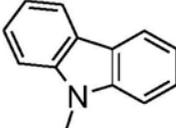
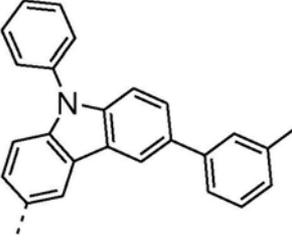
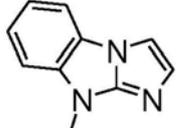
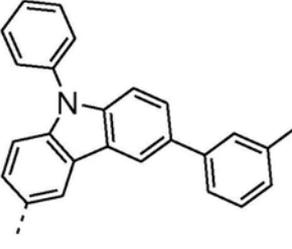
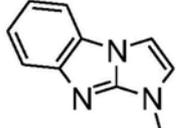
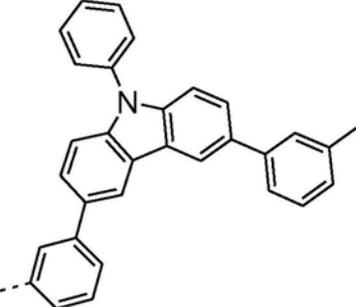
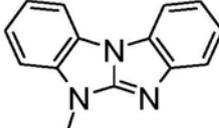
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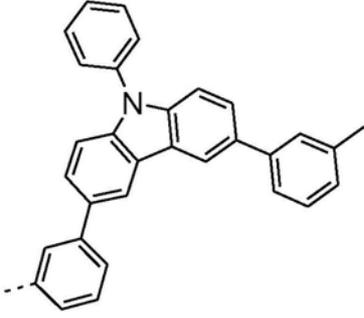
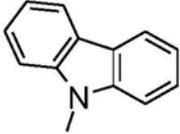
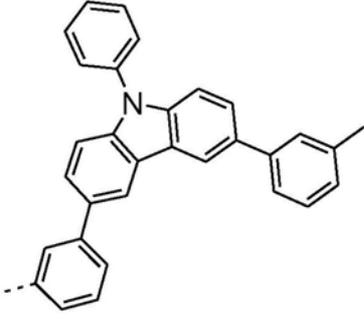
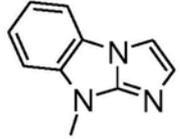
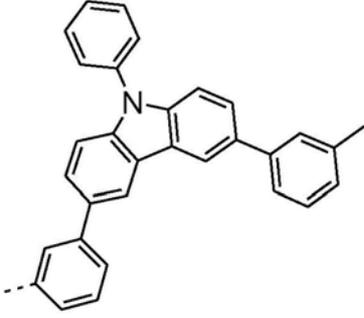
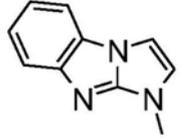
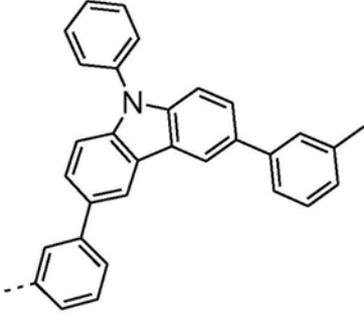
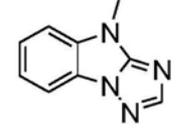
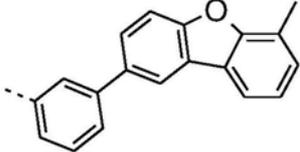
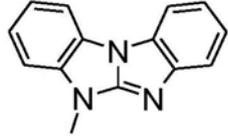
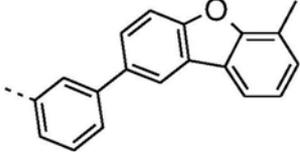
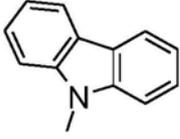


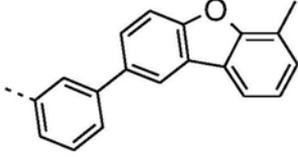
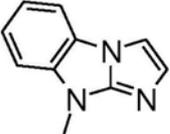
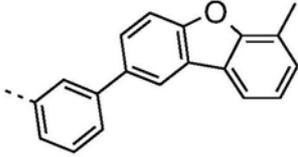
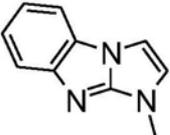
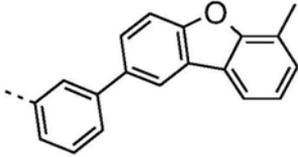
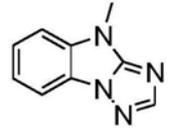
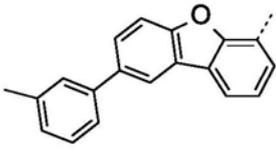
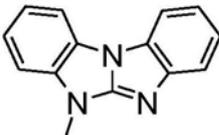
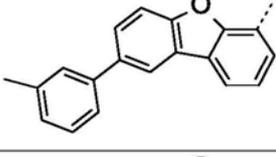
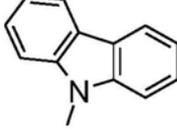
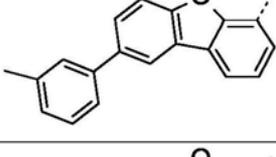
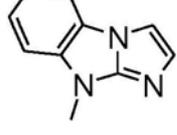
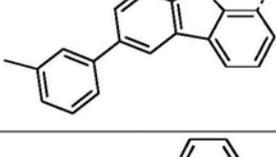
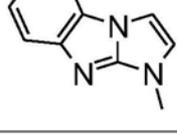
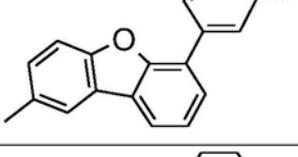
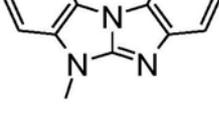
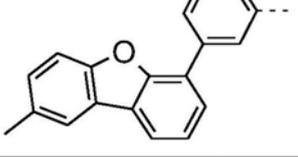
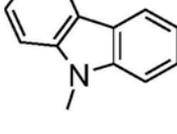
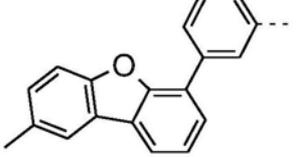
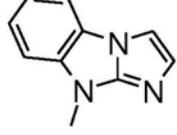
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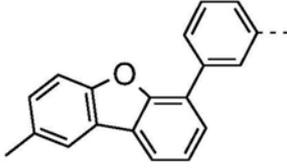
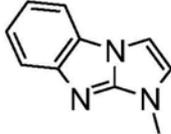
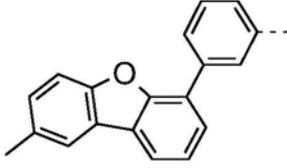
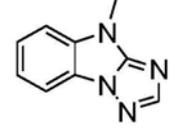
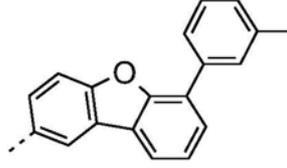
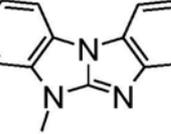
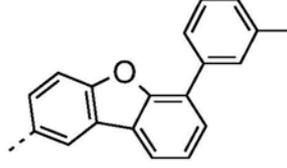
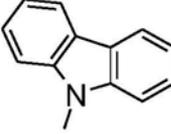
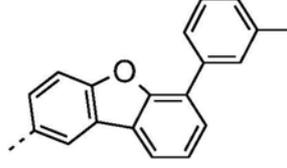
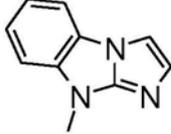
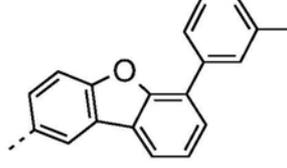
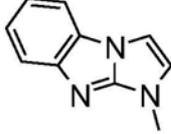
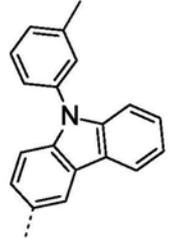
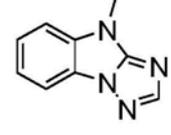
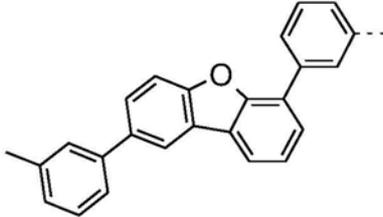
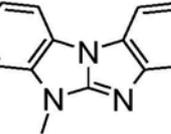
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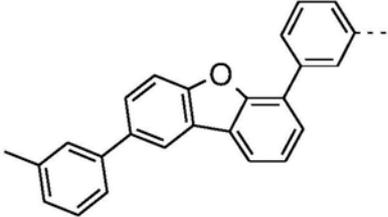
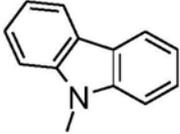
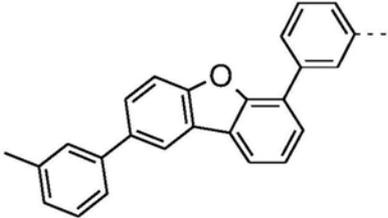
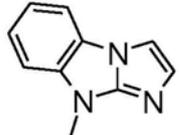
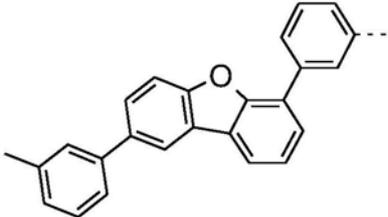
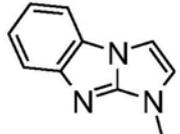
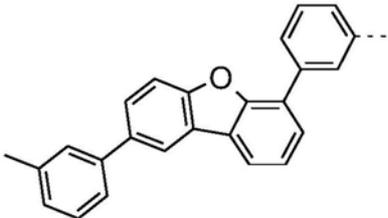
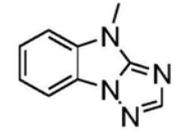
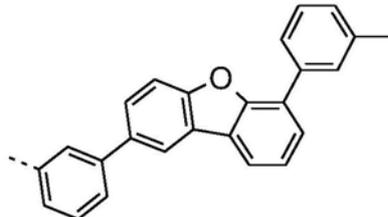
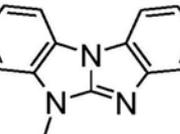
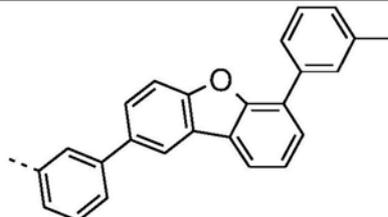
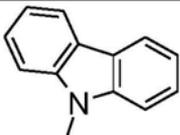
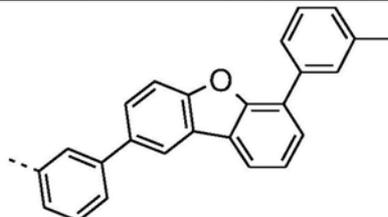
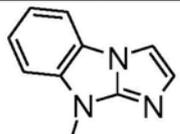
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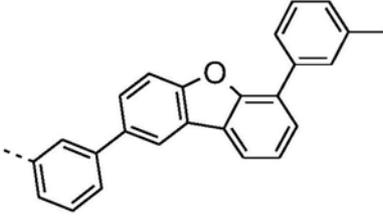
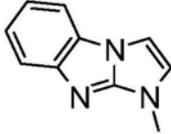
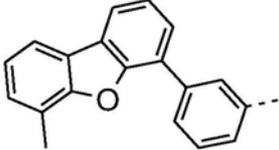
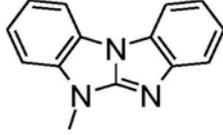
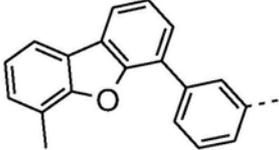
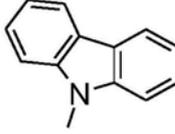
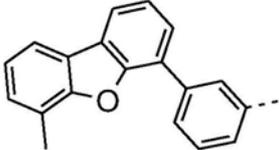
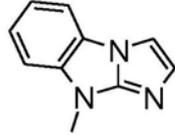
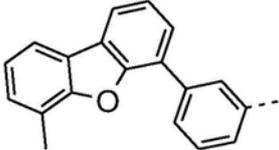
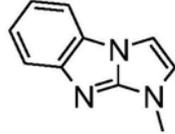
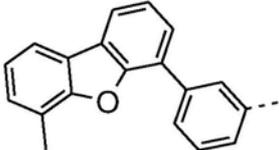
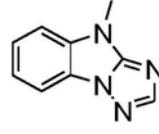
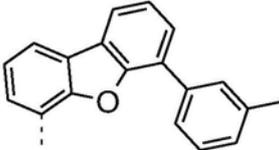
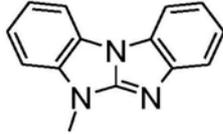
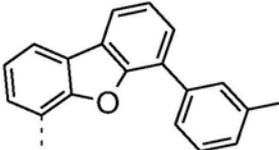
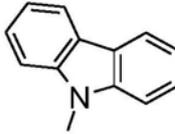
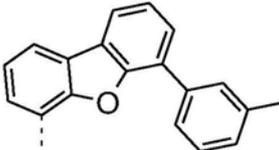
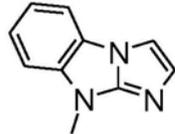
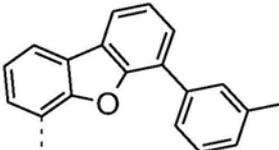
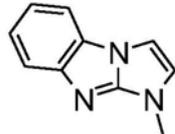
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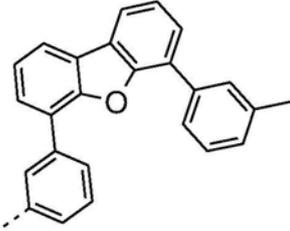
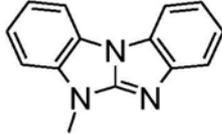
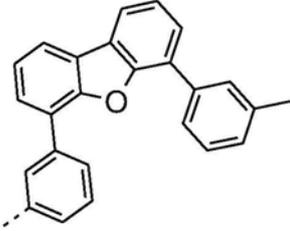
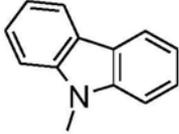
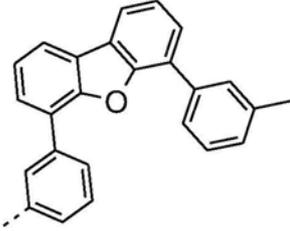
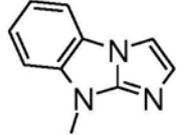
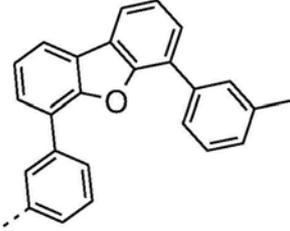
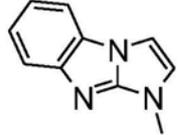
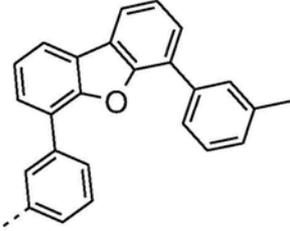
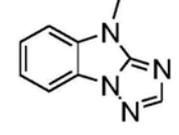
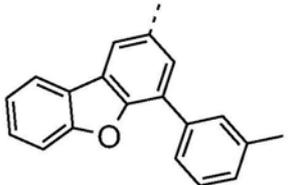
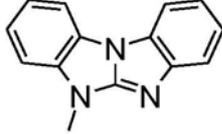
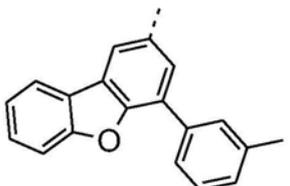
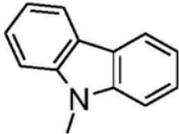
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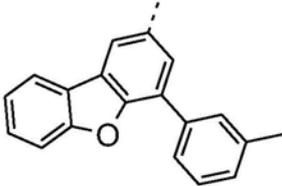
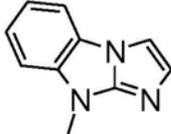
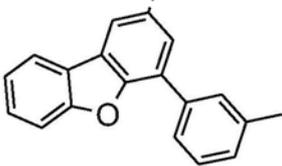
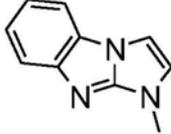
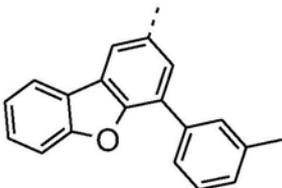
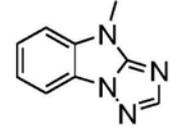
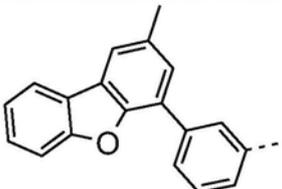
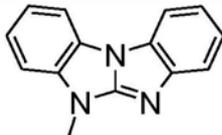
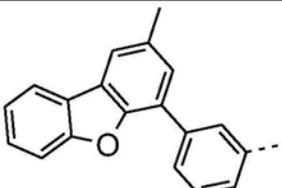
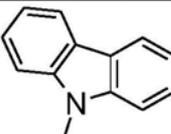
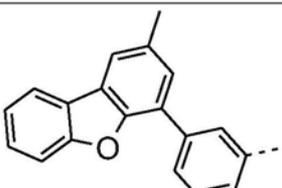
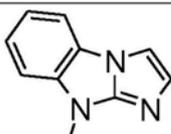
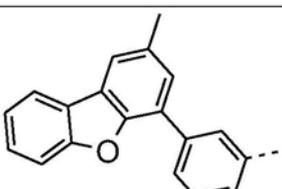
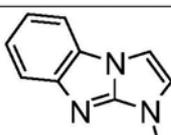
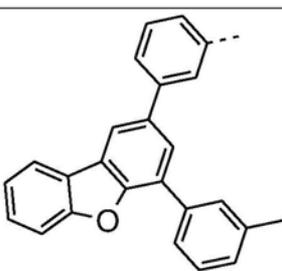
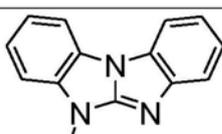
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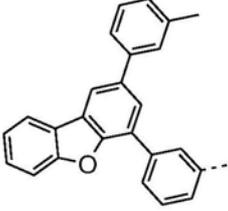
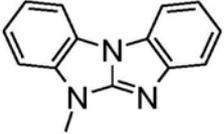
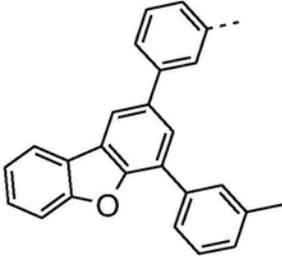
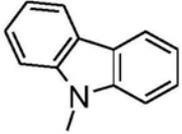
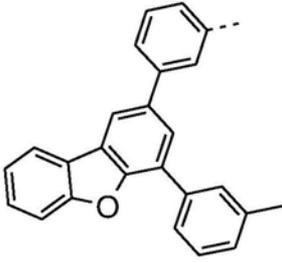
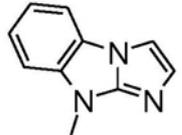
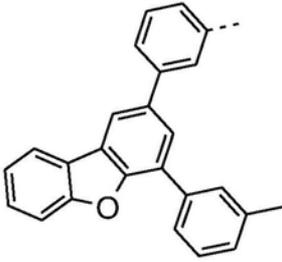
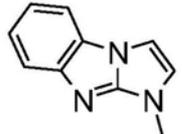
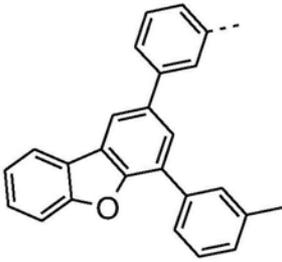
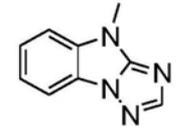
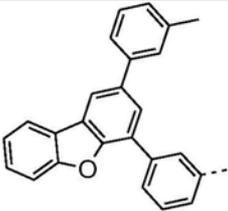
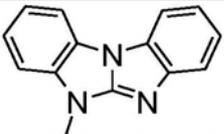
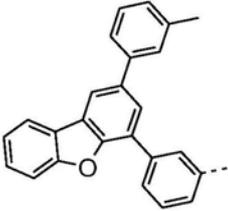
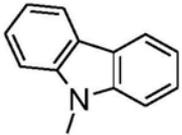
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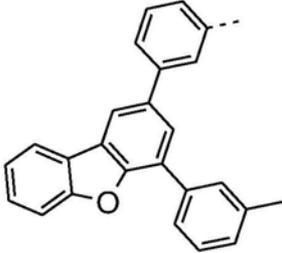
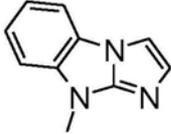
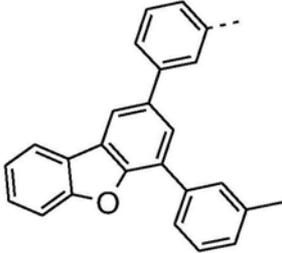
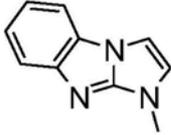
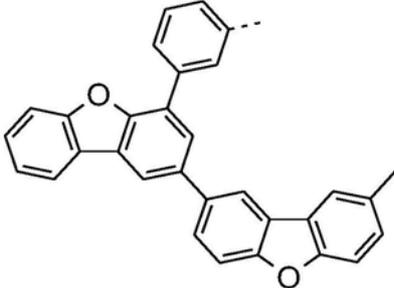
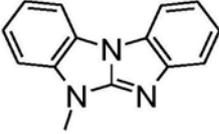
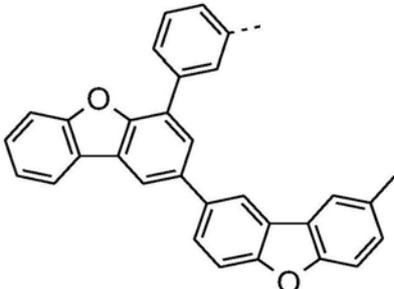
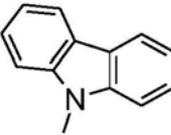
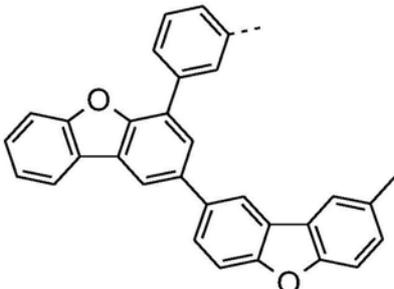
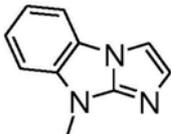
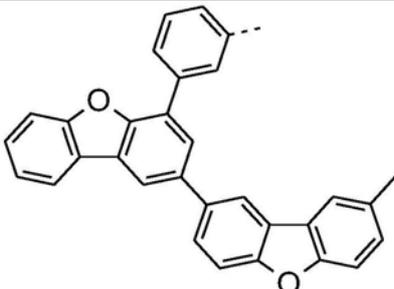
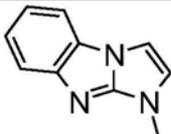
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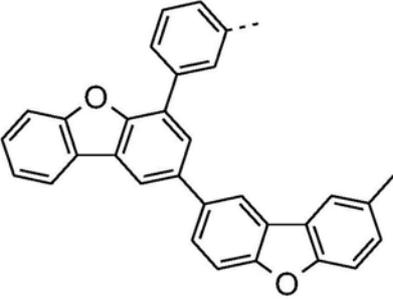
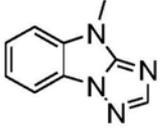
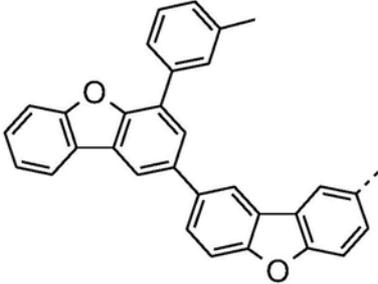
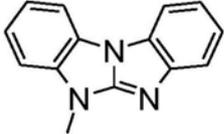
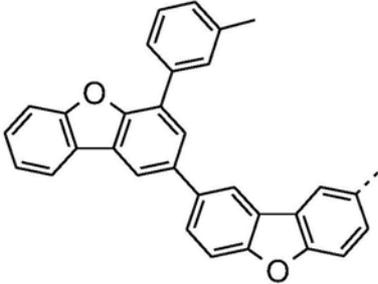
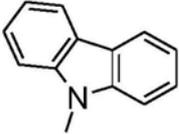
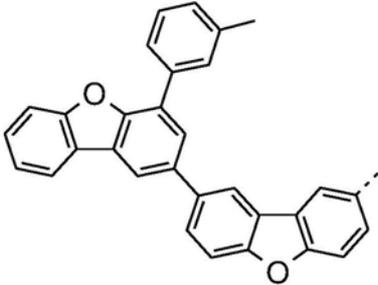
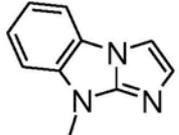
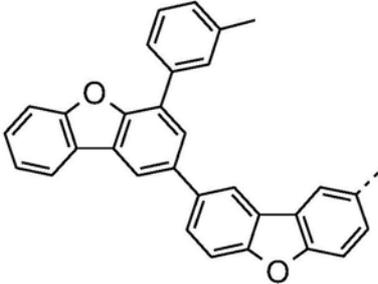
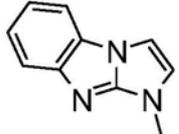
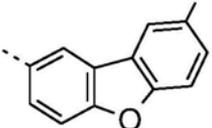
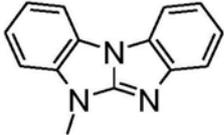
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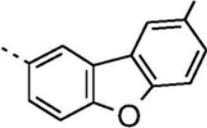
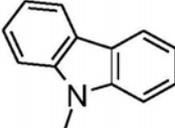
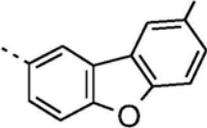
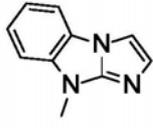
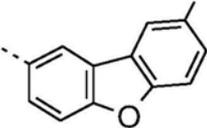
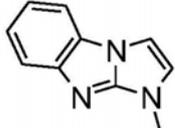
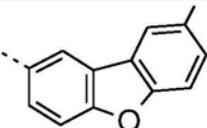
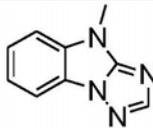
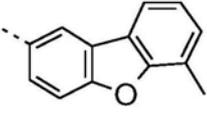
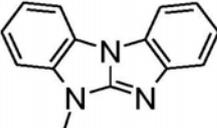
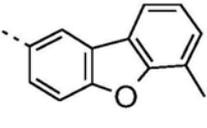
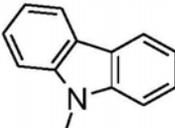
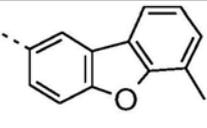
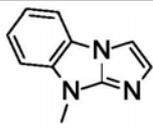
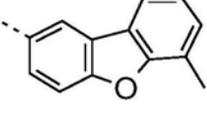
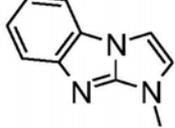
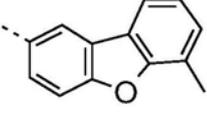
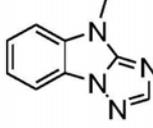
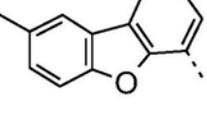
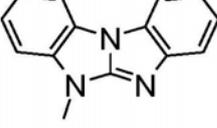
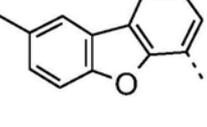
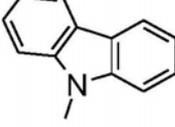
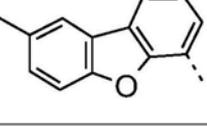
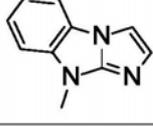
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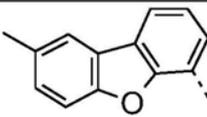
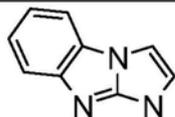
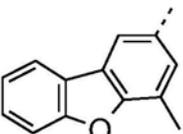
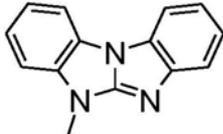
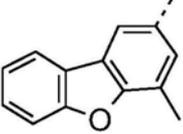
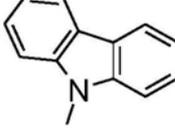
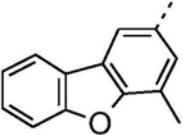
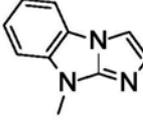
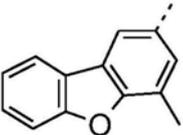
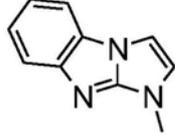
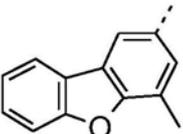
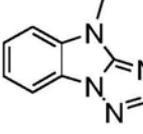
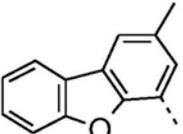
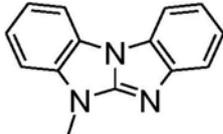
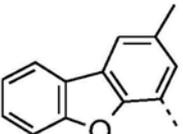
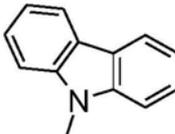
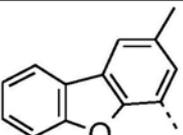
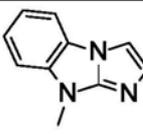
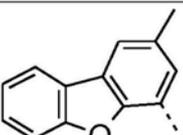
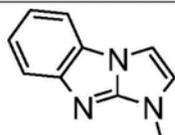
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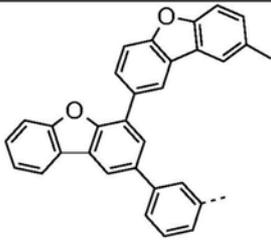
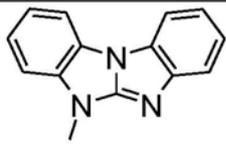
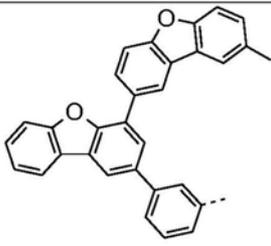
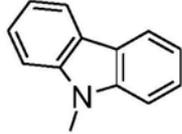
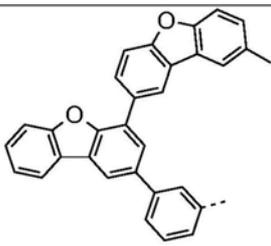
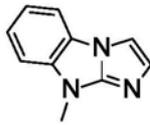
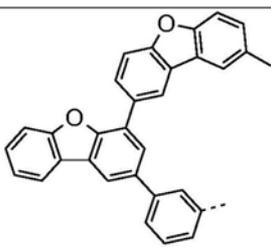
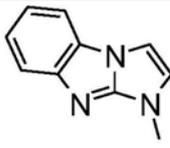
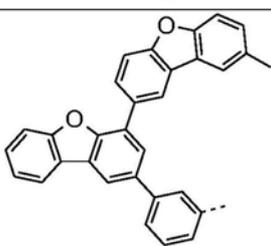
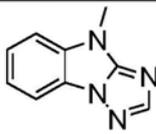
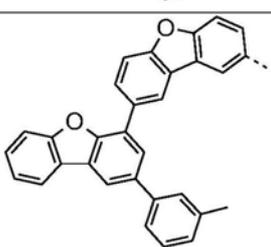
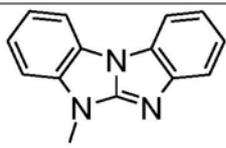
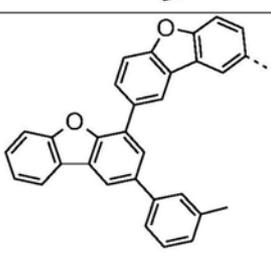
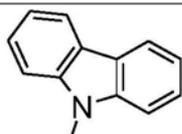
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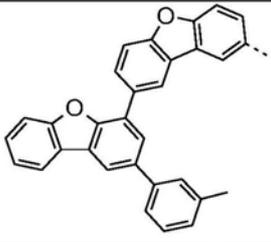
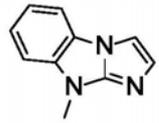
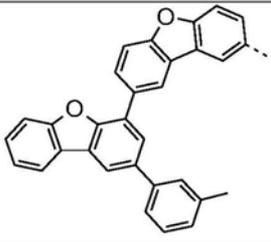
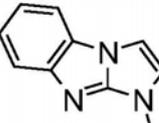
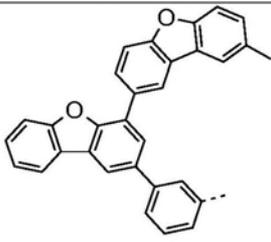
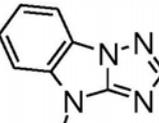
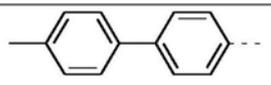
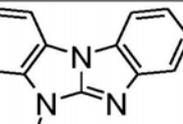
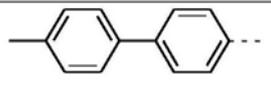
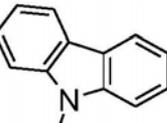
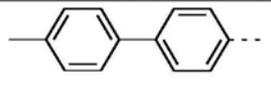
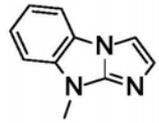
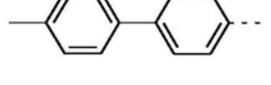
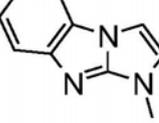
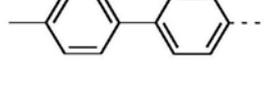
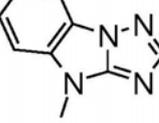
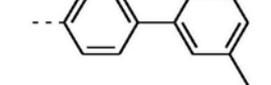
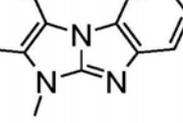
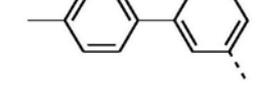
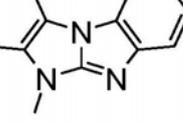
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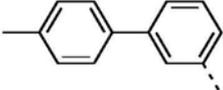
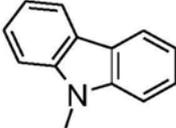
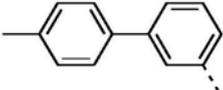
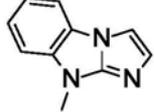
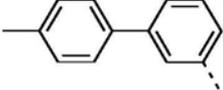
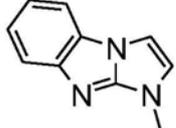
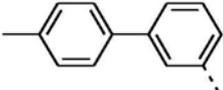
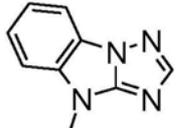
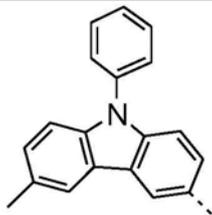
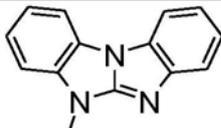
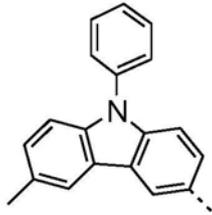
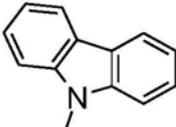
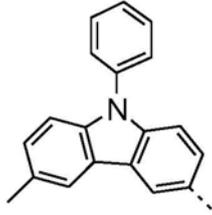
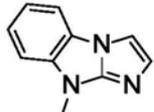
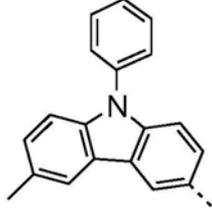
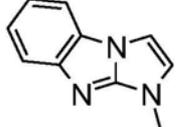
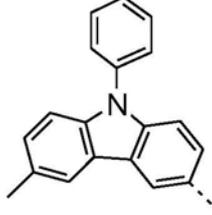
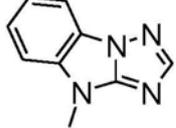
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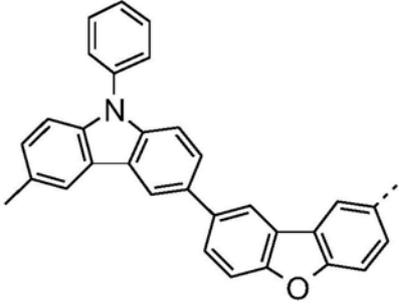
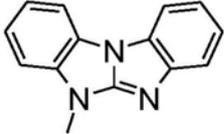
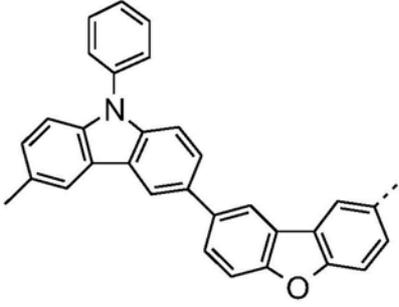
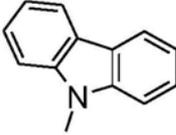
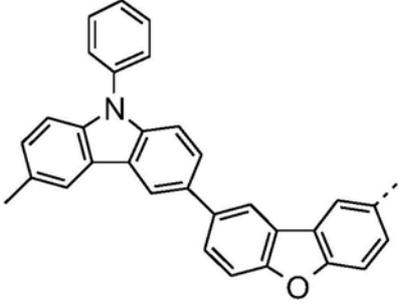
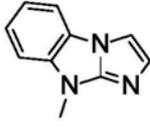
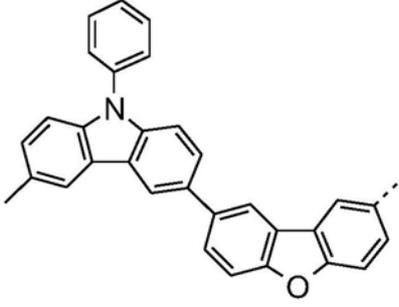
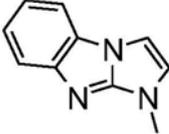
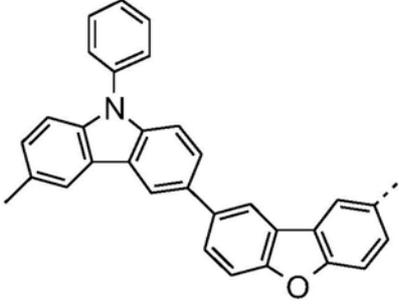
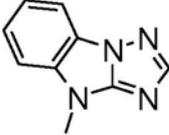
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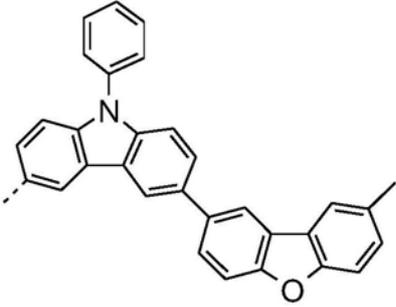
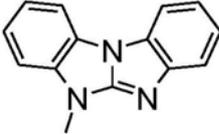
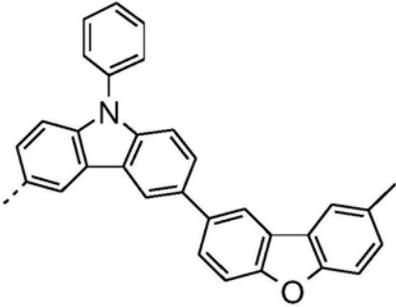
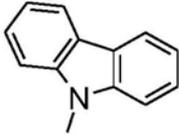
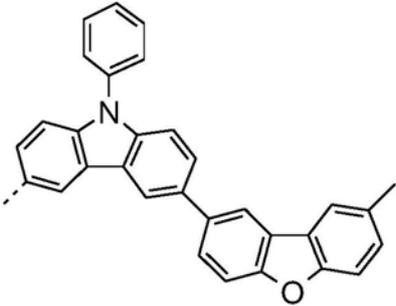
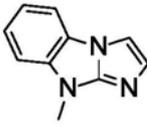
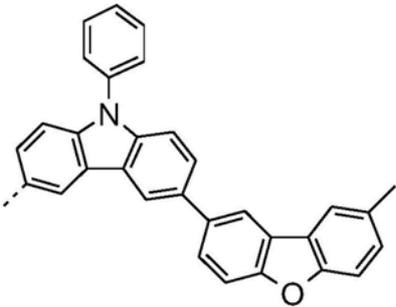
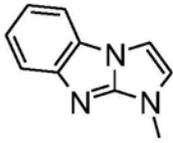
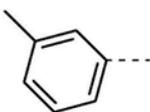
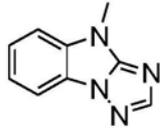
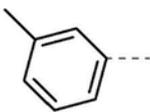
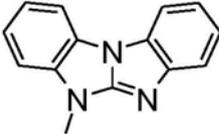
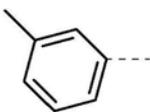
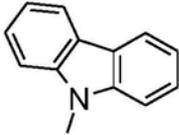
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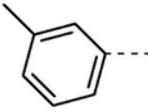
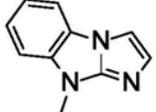
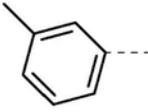
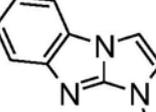
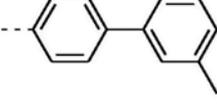
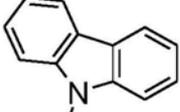
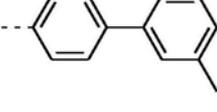
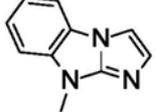
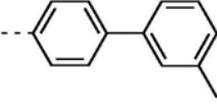
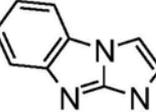
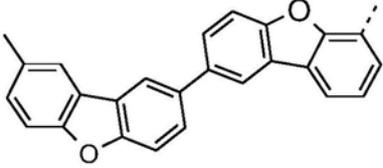
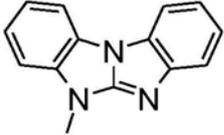
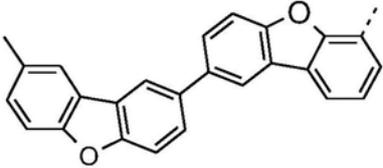
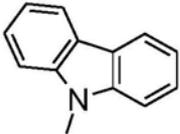
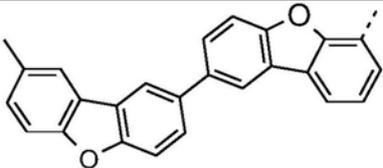
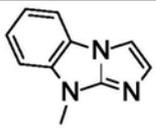
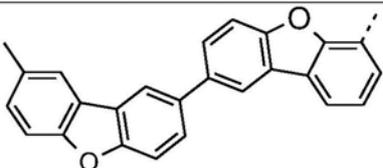
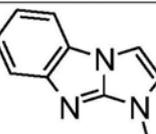
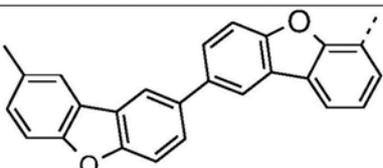
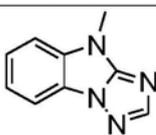
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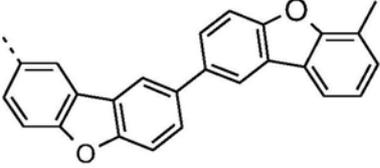
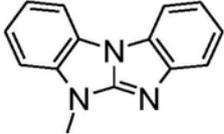
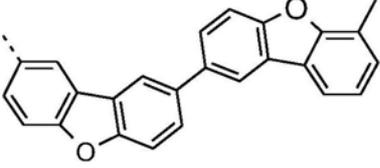
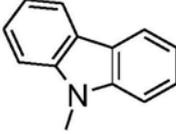
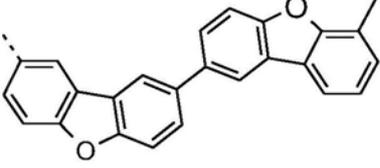
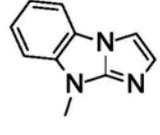
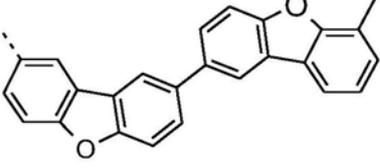
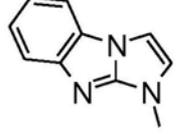
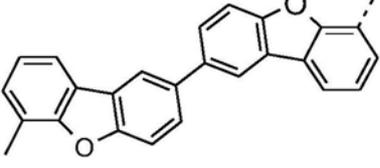
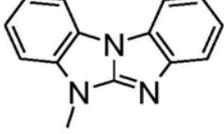
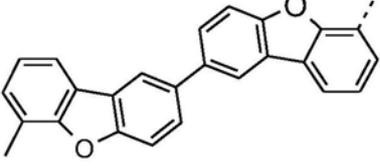
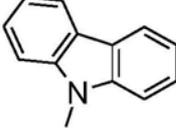
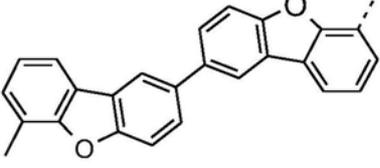
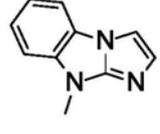
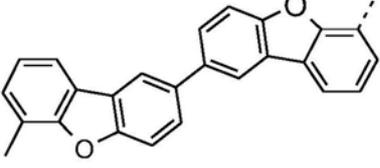
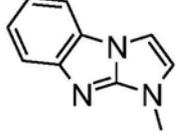
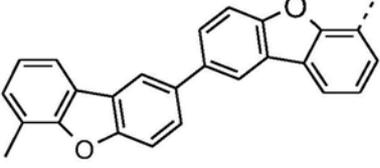
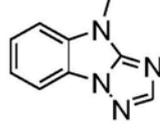
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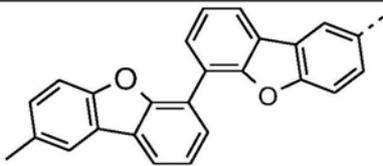
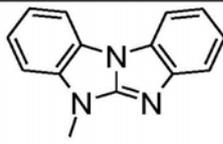
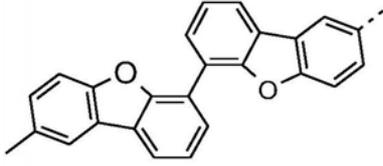
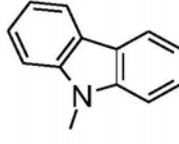
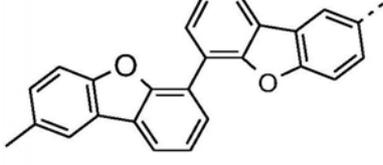
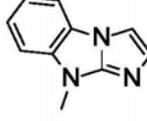
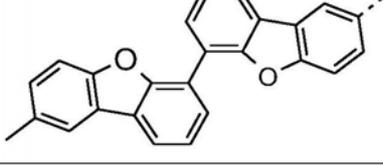
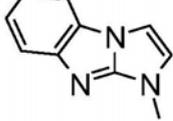
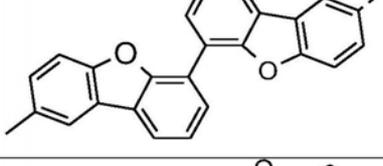
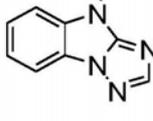
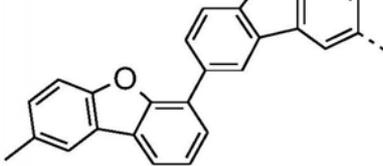
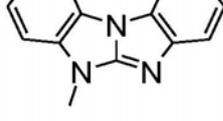
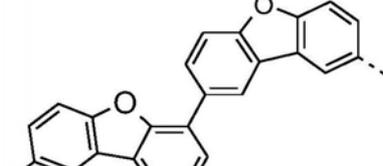
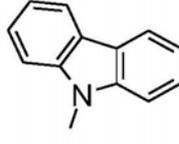
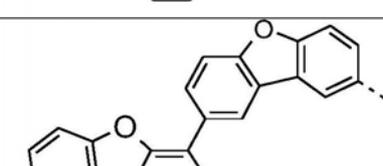
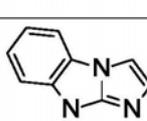
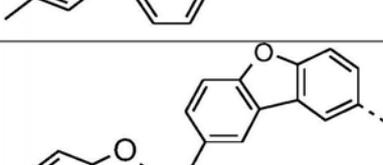
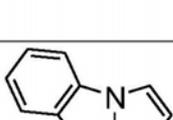
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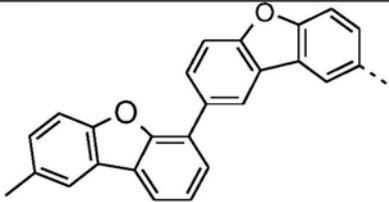
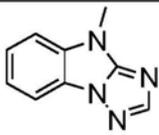
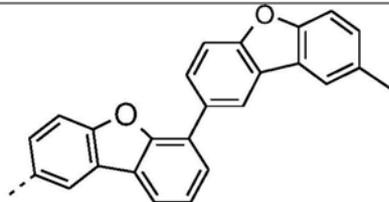
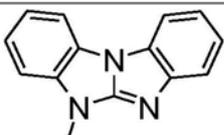
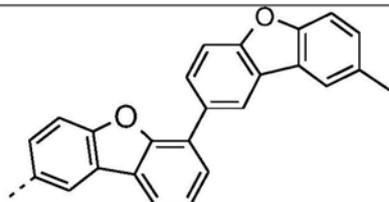
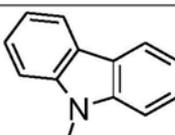
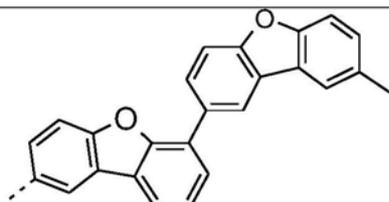
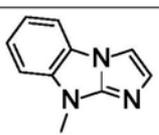
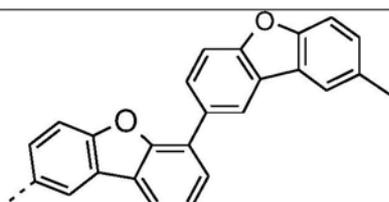
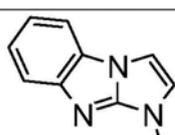
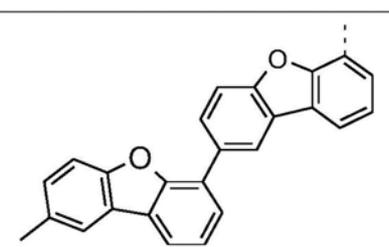
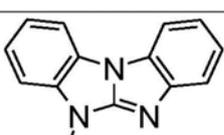
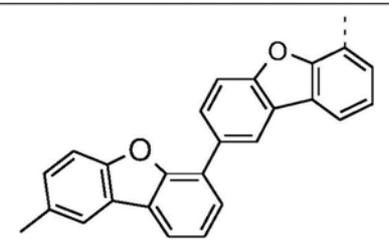
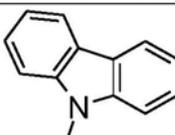
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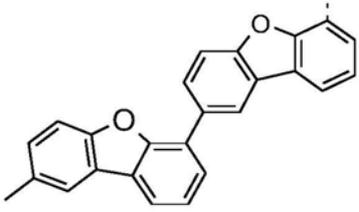
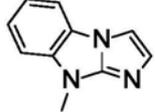
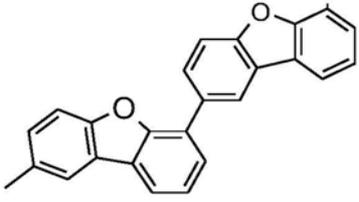
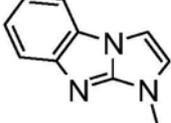
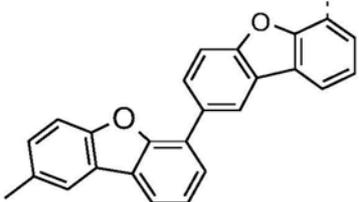
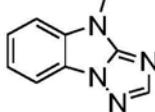
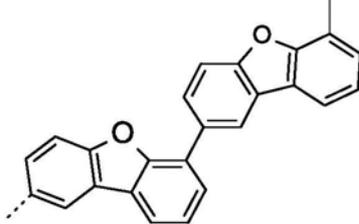
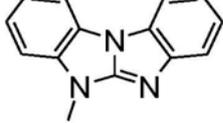
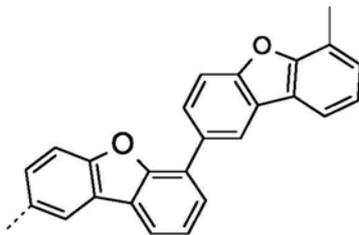
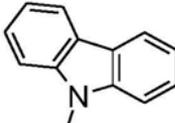
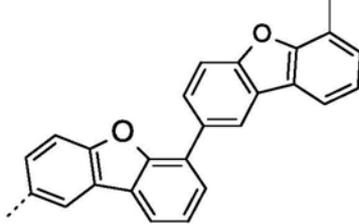
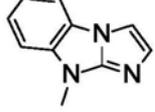
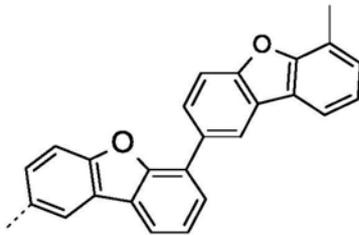
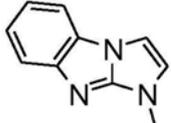
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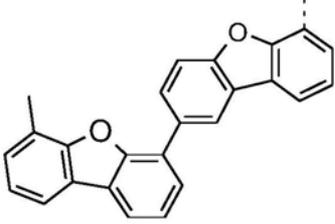
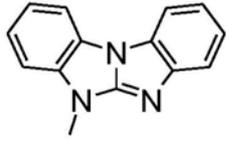
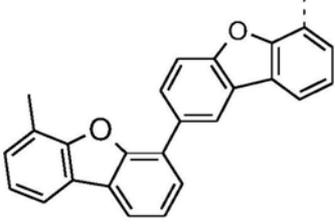
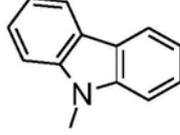
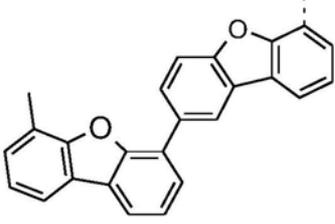
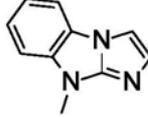
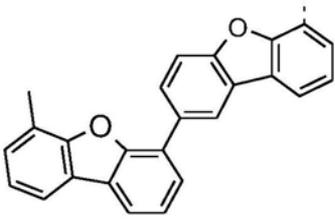
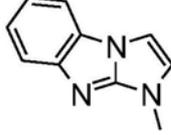
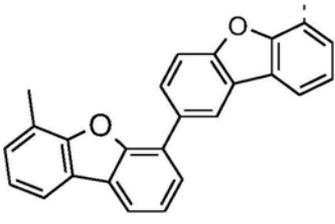
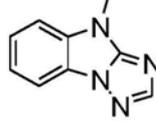
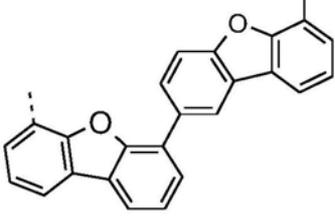
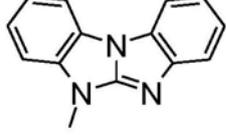
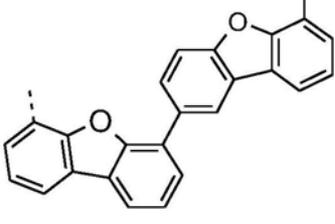
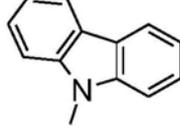
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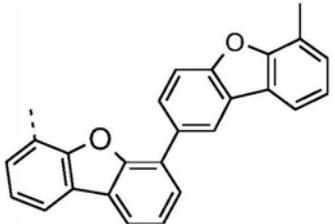
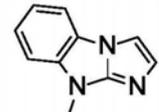
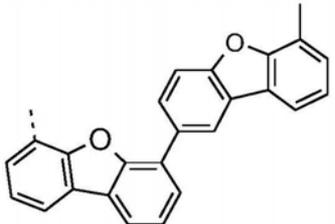
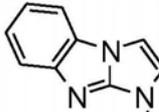
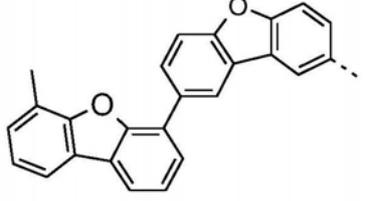
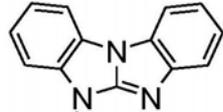
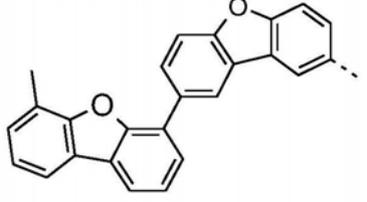
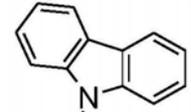
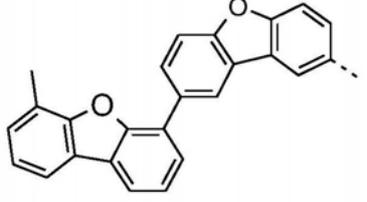
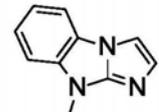
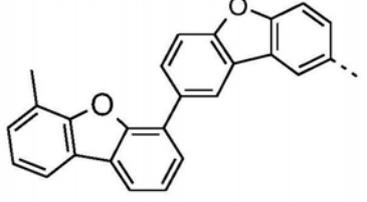
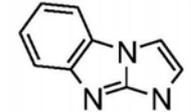
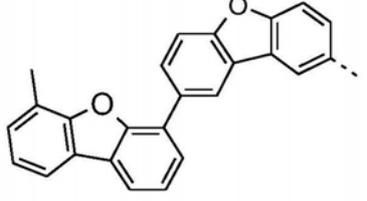
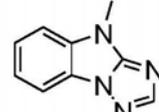
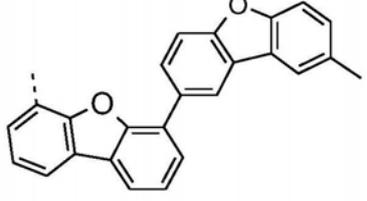
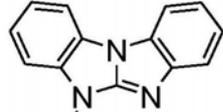
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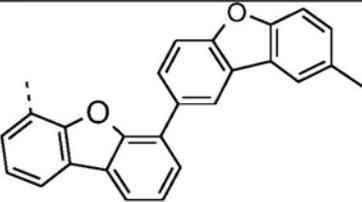
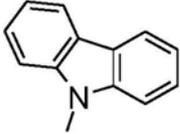
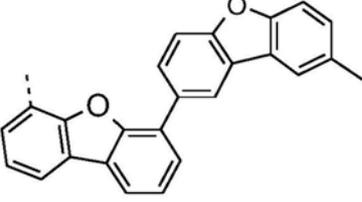
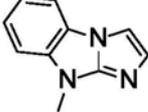
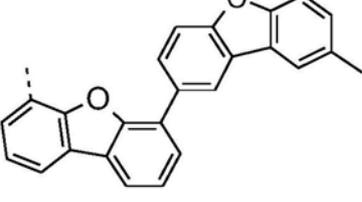
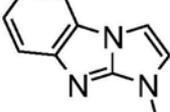
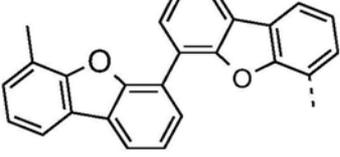
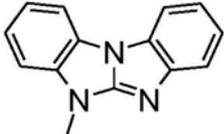
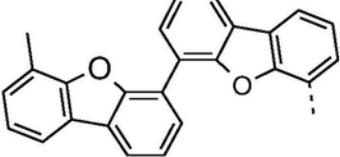
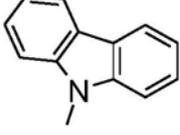
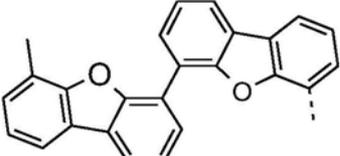
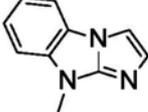
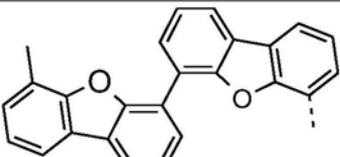
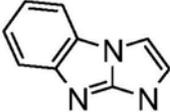
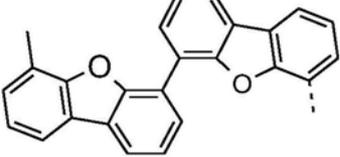
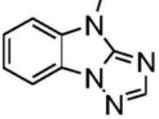
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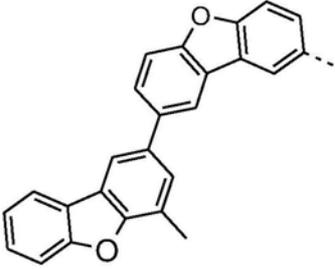
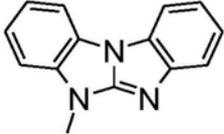
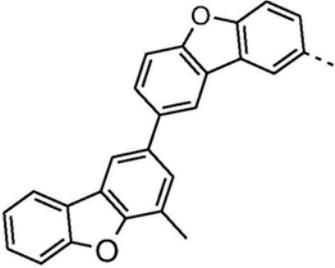
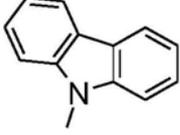
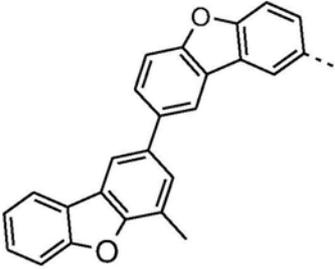
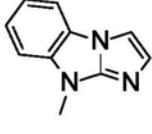
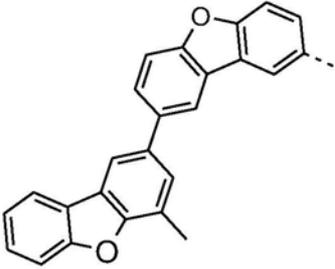
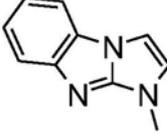
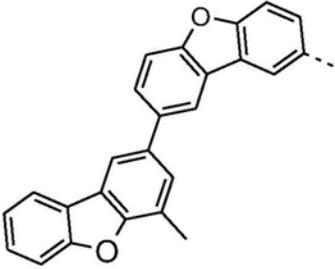
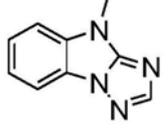
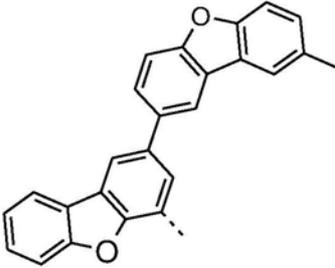
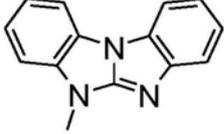
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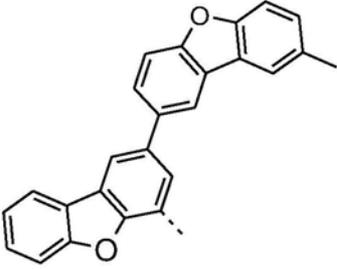
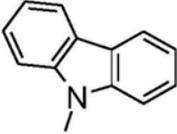
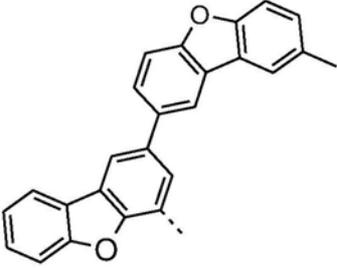
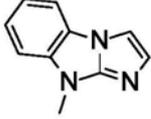
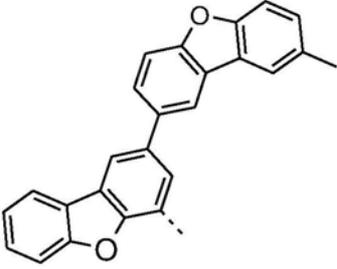
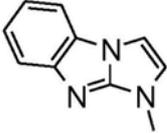
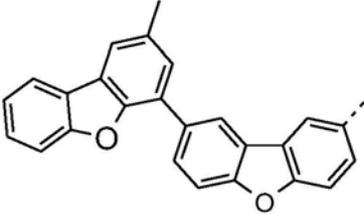
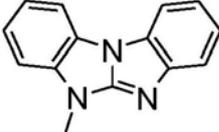
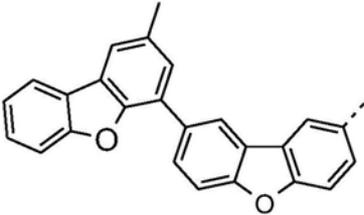
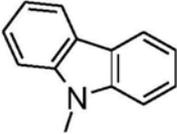
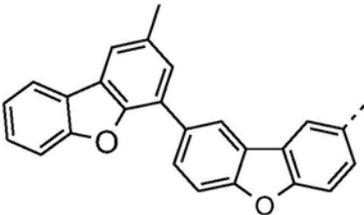
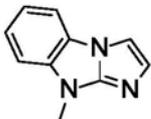
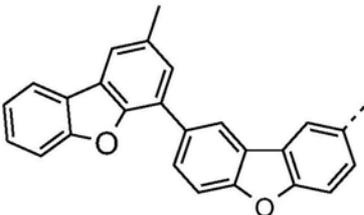
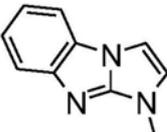
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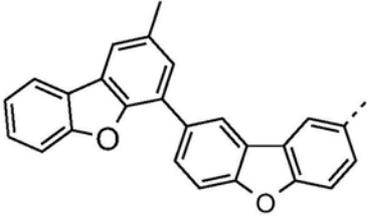
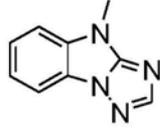
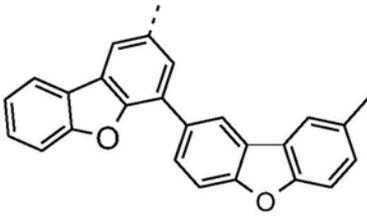
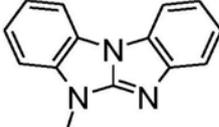
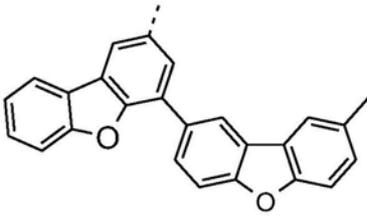
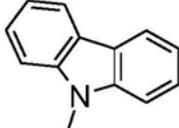
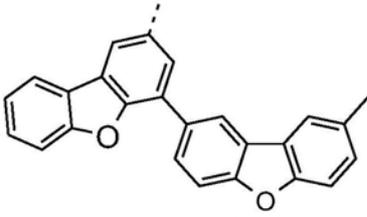
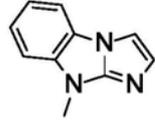
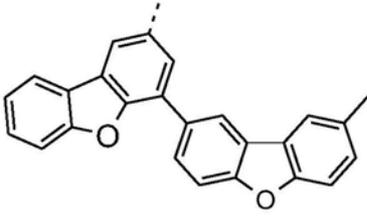
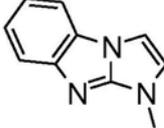
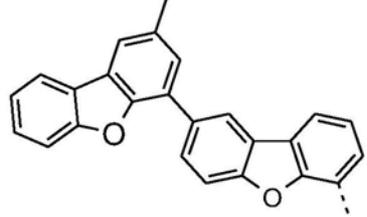
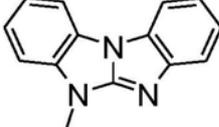
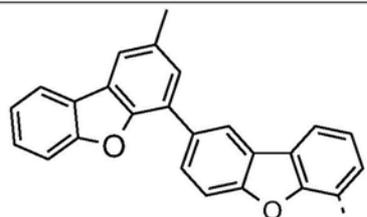
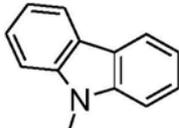
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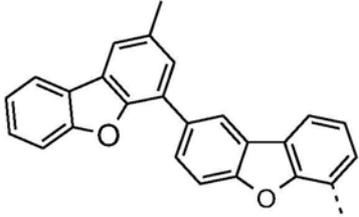
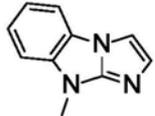
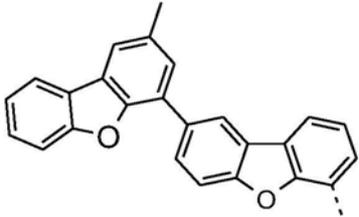
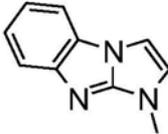
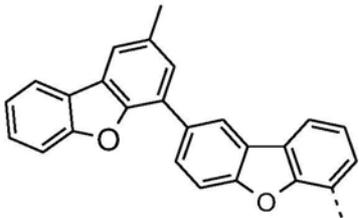
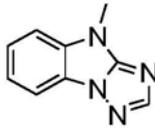
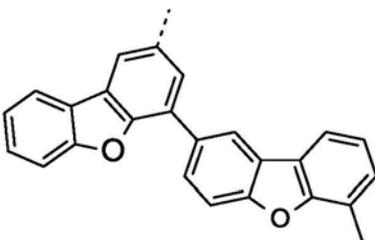
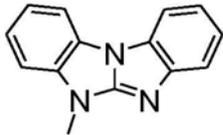
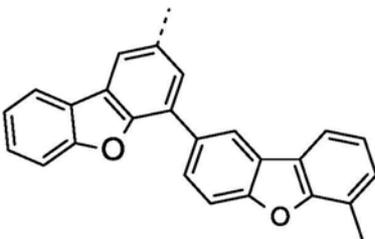
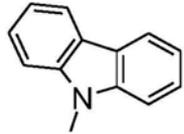
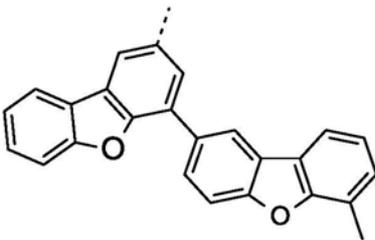
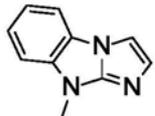
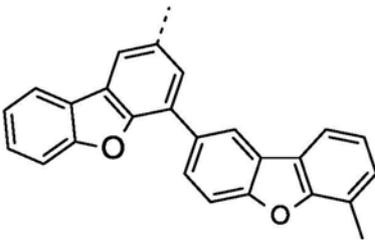
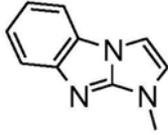
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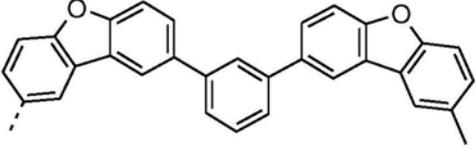
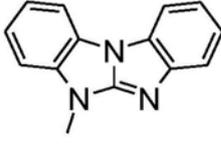
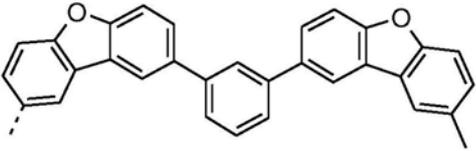
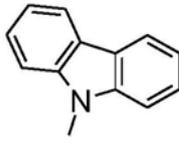
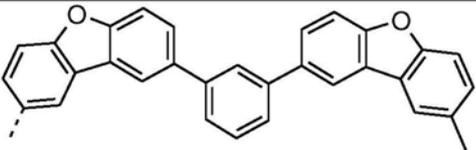
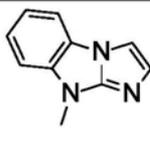
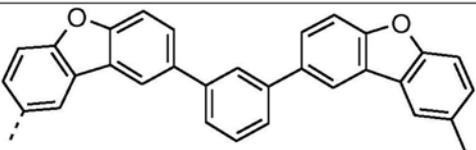
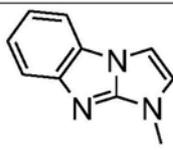
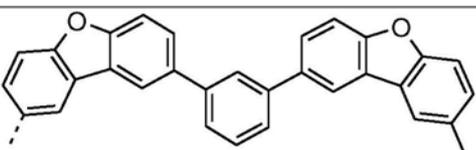
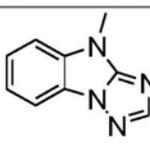
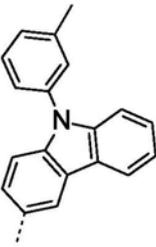
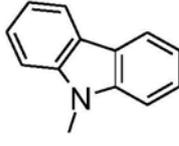
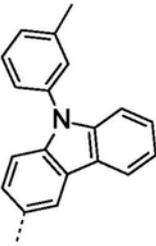
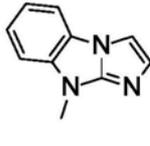
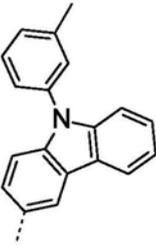
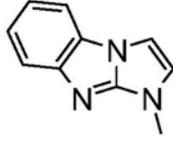
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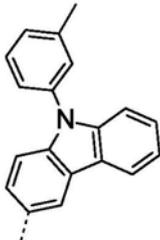
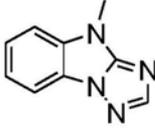
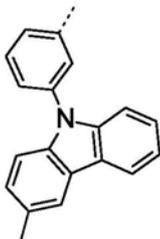
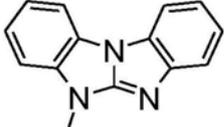
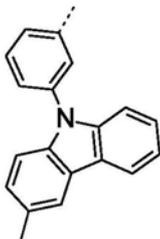
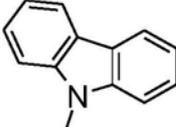
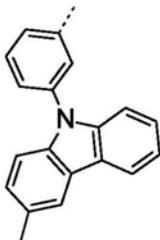
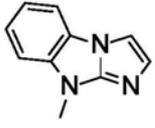
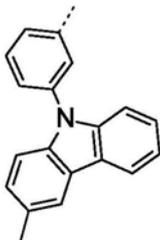
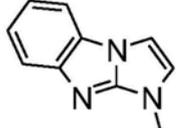
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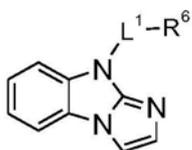
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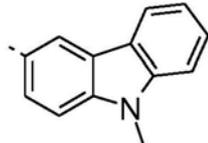
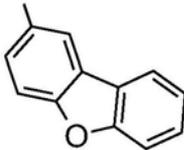
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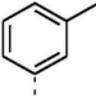
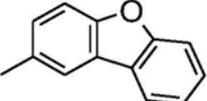
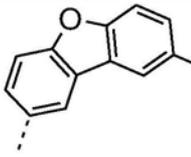
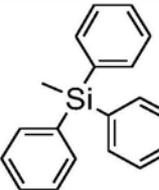
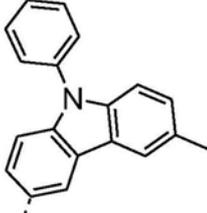
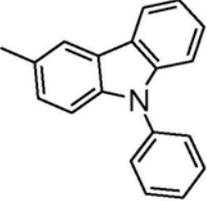
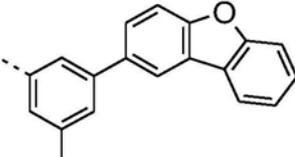
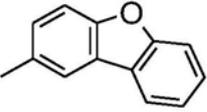
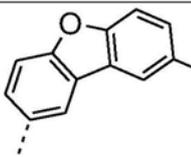
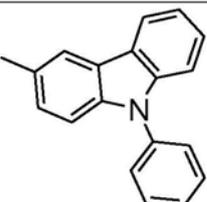
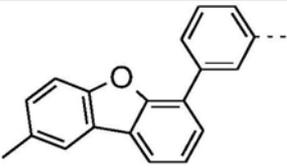
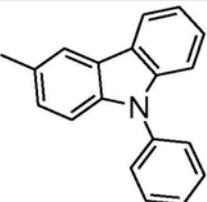
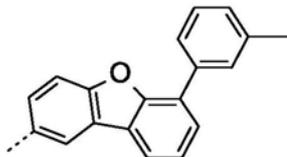
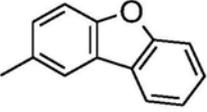
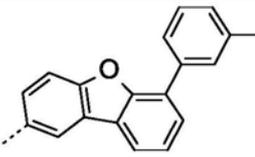
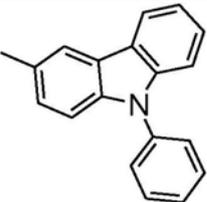
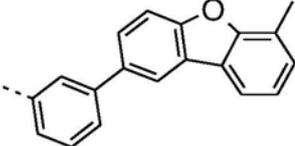
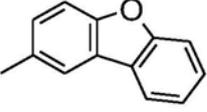
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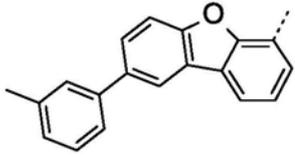
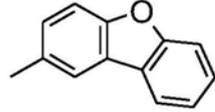
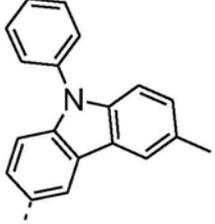
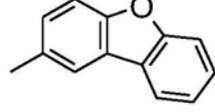
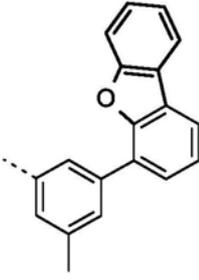
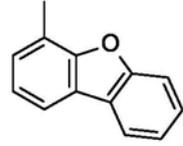
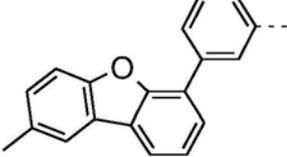
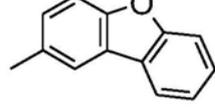
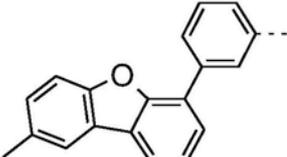
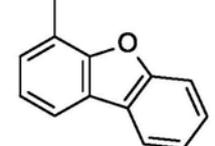
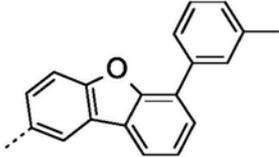
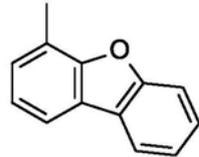
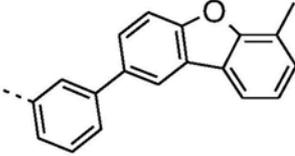
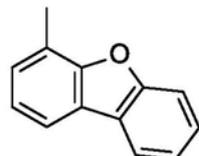
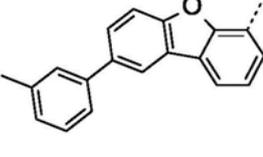
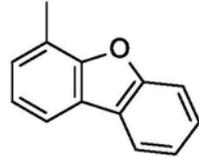
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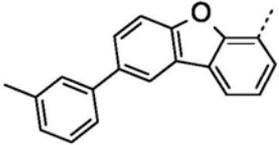
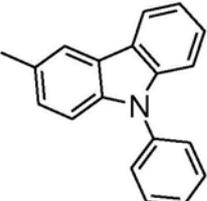
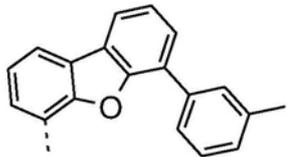
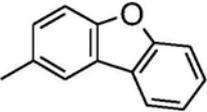
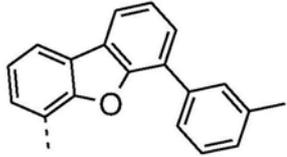
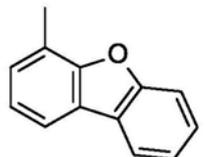
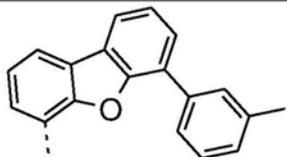
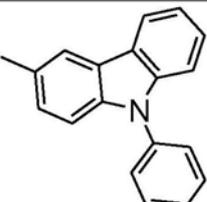
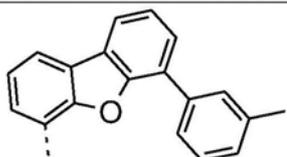
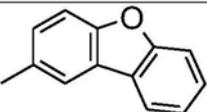
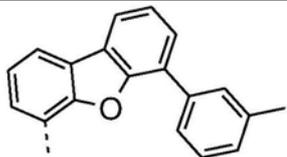
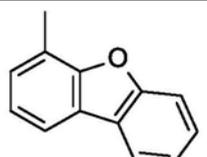
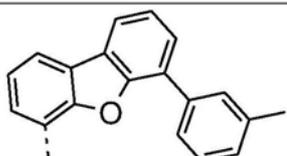
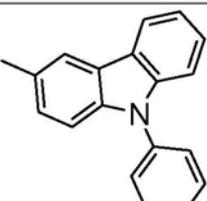
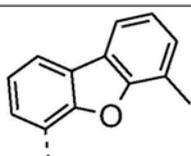
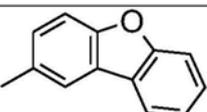
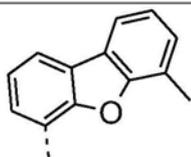
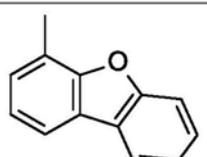
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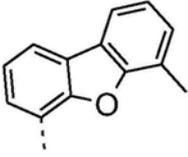
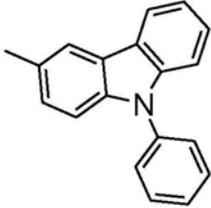
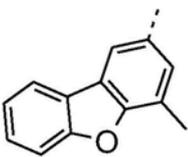
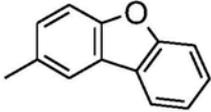
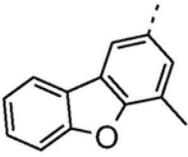
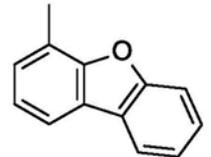
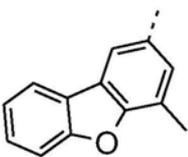
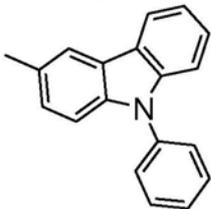
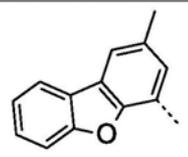
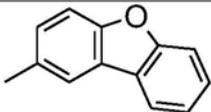
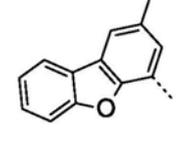
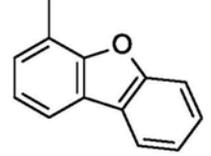
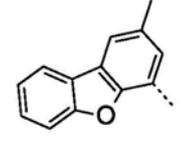
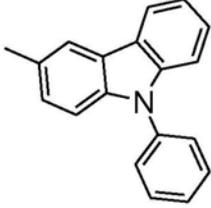
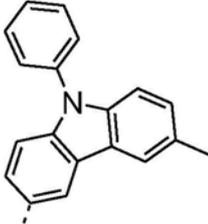
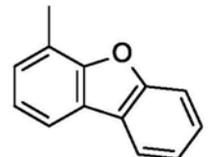


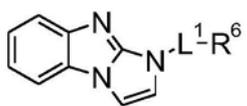
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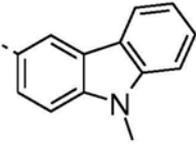
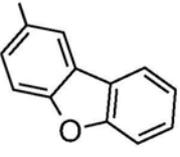
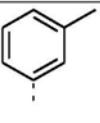
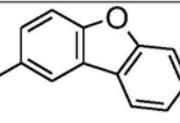
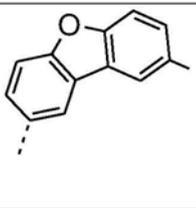
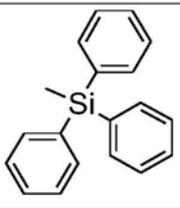
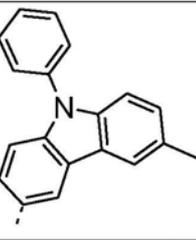
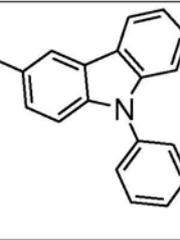
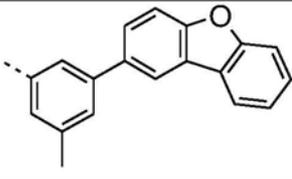
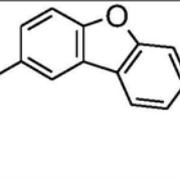
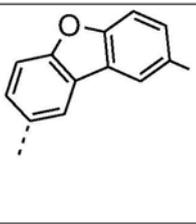
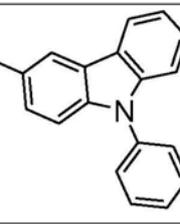
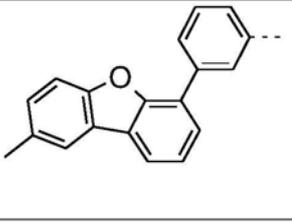
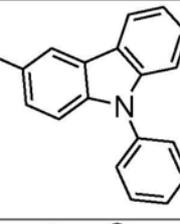
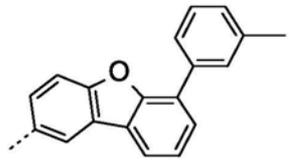
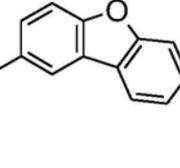
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<p>J-4</p>		
<p>J-5</p>		
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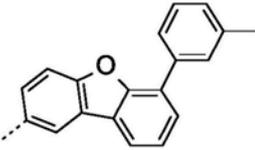
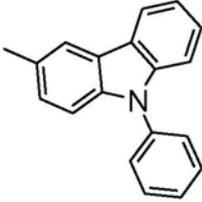
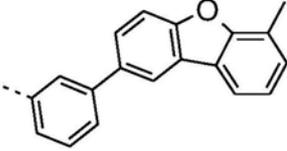
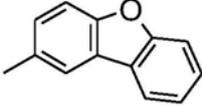
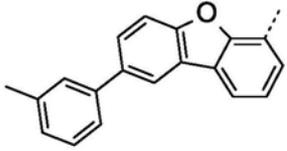
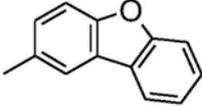
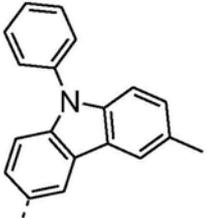
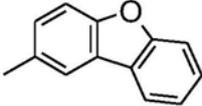
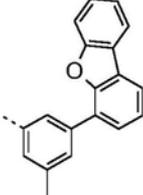
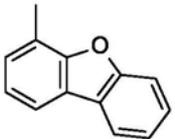
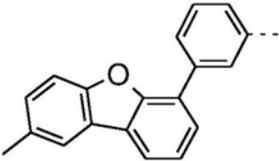
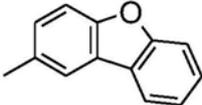
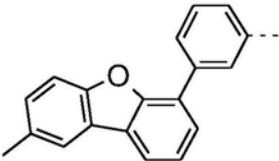
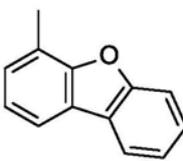
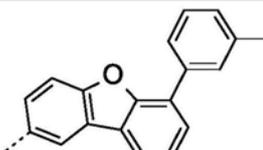
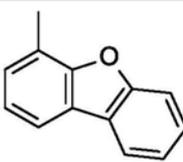
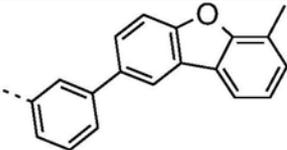
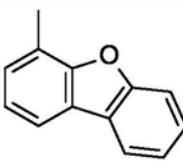
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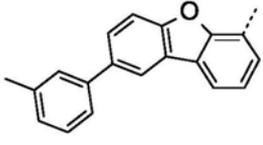
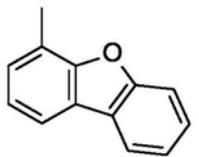
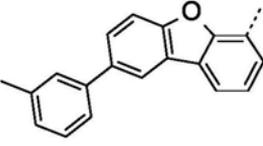
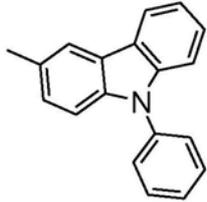
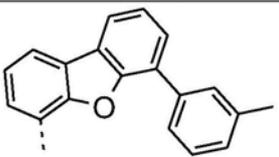
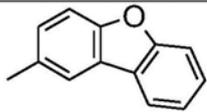
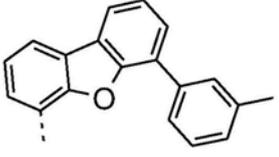
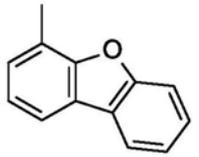
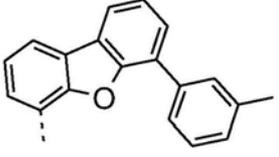
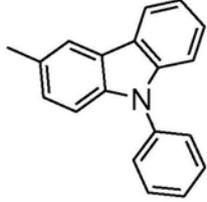
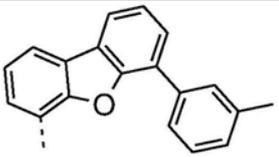
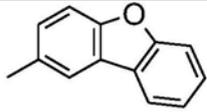
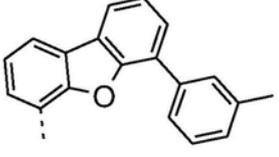
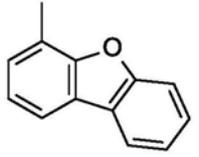
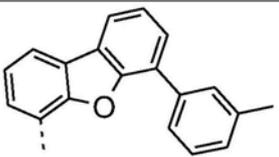
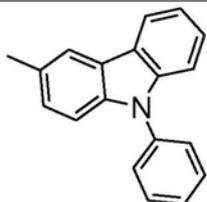
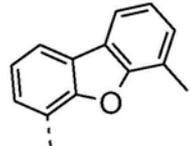
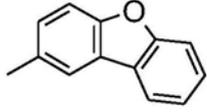
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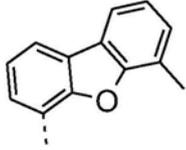
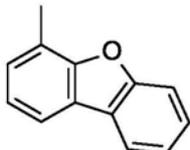
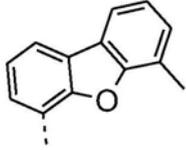
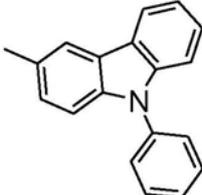
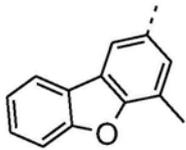
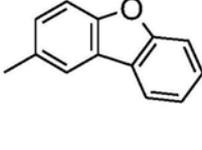
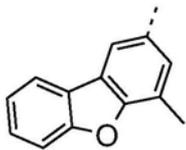
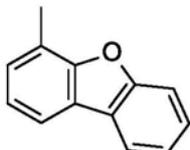
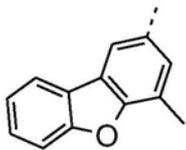
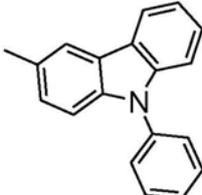
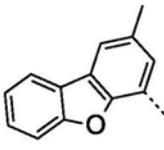
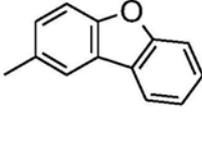
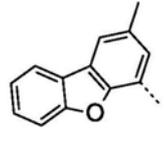
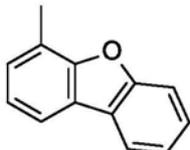
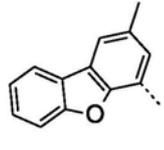
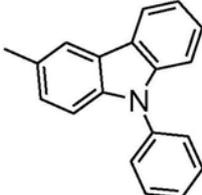
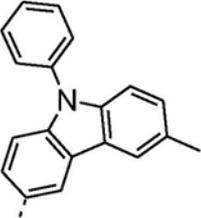
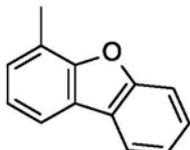
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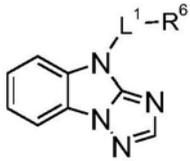


Cpd.	L ¹³⁾	R ⁶
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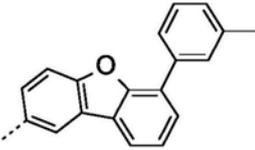
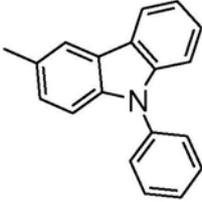
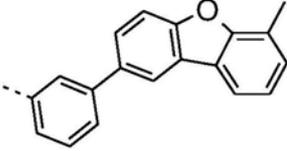
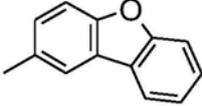
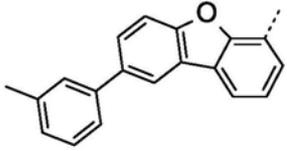
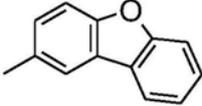
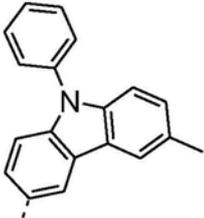
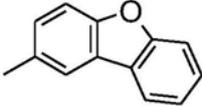
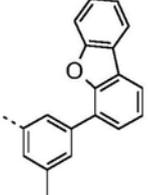
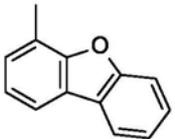
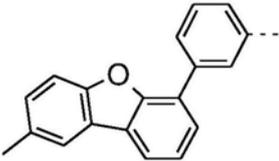
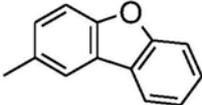
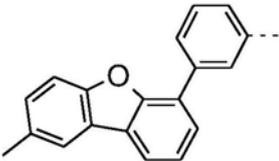
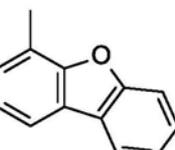
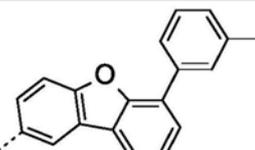
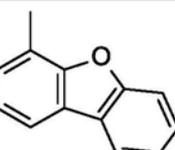
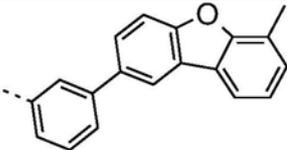
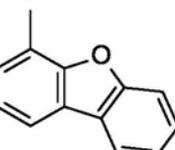
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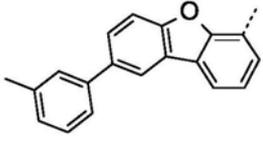
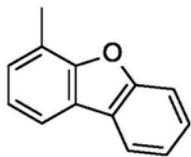
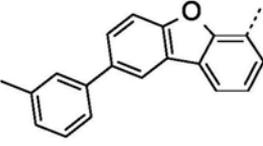
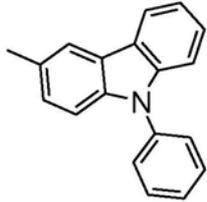
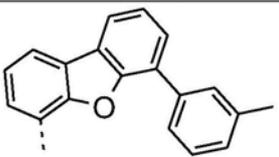
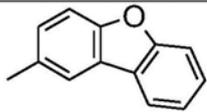
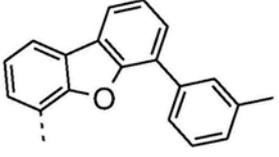
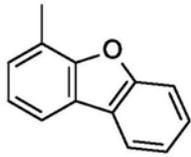
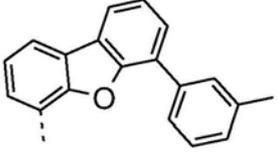
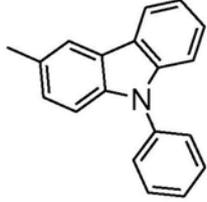
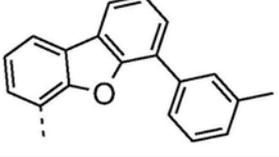
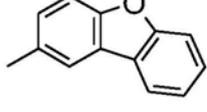
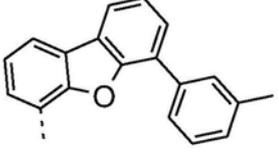
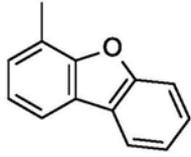
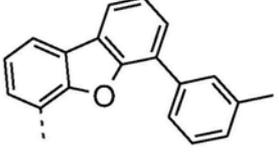
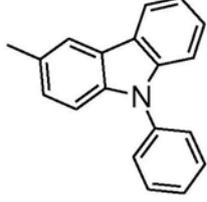
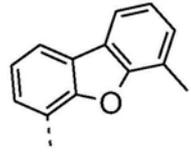
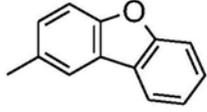
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<p>K-26</p>		

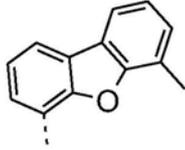
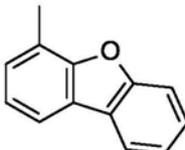
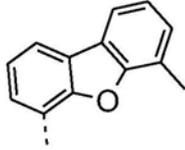
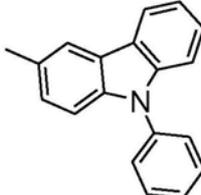
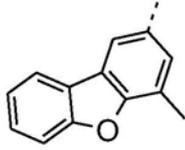
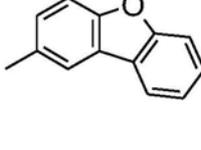
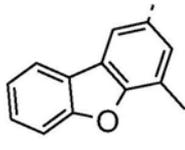
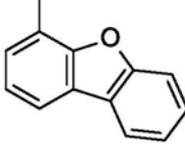
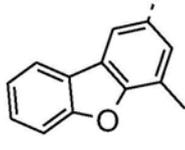
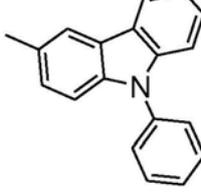
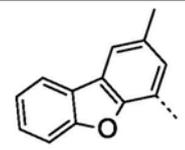
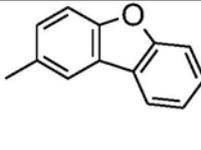
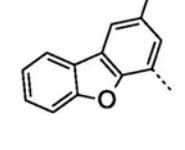
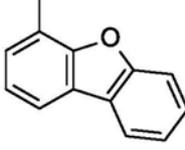
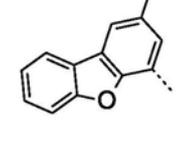
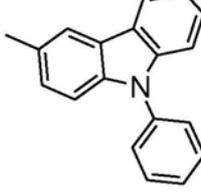
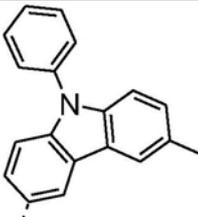
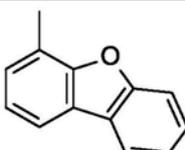
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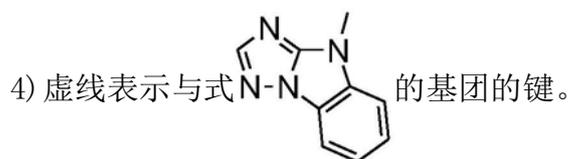
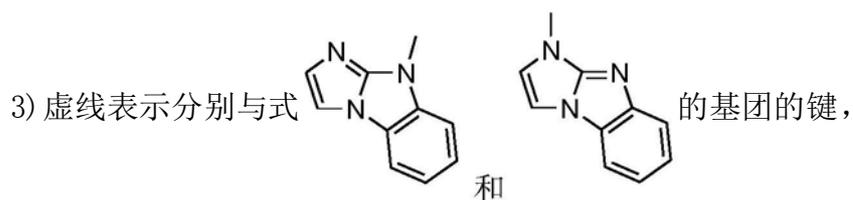
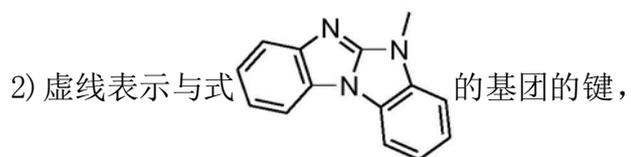
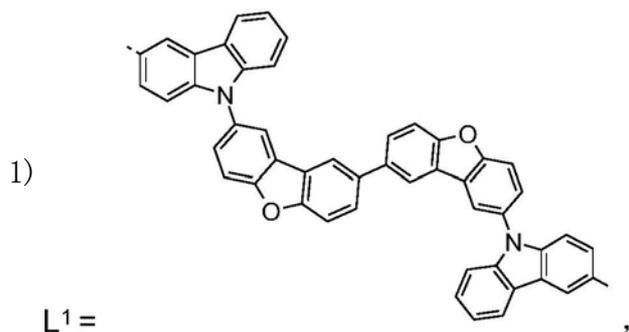


Cpd.	L ¹⁴⁾	R ⁶
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<p>L-9</p>		
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19. 一种有机发光二极管OLED,其包括:

阳极;

阴极;和

设置在所述阳极和所述阴极之间的发光层,所述发光层包含至少一种根据权利要求1-18中任一项所述的化合物。

用于电子应用的4H-咪唑并[1,2-a]咪唑

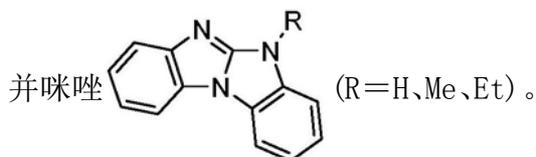
[0001] 本申请是申请号为201610552418.2、申请日为2012年3月22日、发明名称为“用于电子应用的4H-咪唑并[1,2-a]咪唑”的专利申请的分案申请,其中申请号为201610552418.2的专利申请为申请号为201280022301.3、申请日为2012年3月22日、发明名称为“用于电子应用的4H-咪唑并[1,2-a]咪唑”的专利申请的分案申请。

技术领域

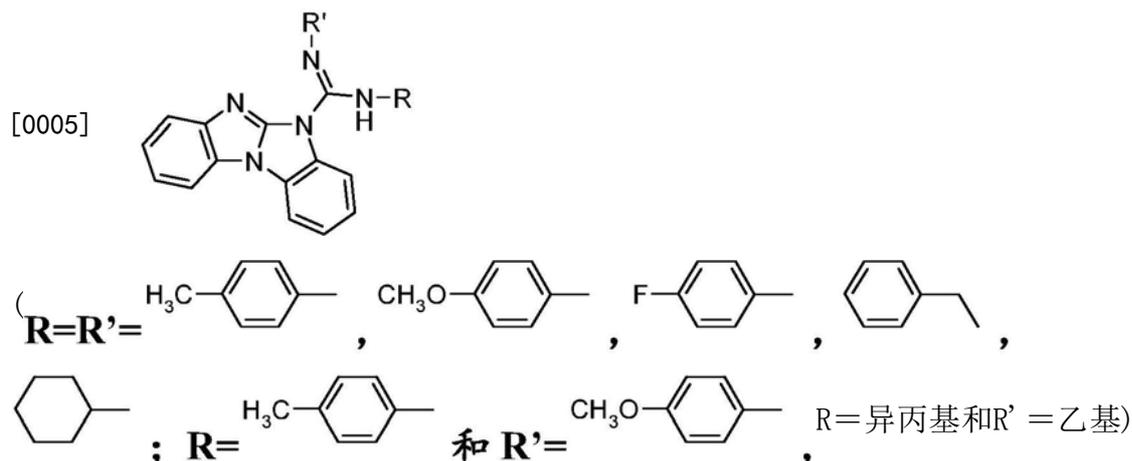
[0002] 本发明涉及式I化合物,一种其的生产方法及其在电子器件,尤其是场致发光器件中的用途。当在场致发光器件中用作空穴传输材料时,式I化合物可以提供具有改进效率、稳定性、可制造性或光谱特性的场致发光器件。

背景技术

[0003] Khan,Misbahul Ain;Ribeiro,Vera Lucia Teixeira,Pakistan Journal of Scientific and Industrial Research 43 (2000) 168-170描述了通过亚磷酸三烷基酯诱导脱氧和和1-(邻硝基苯基)-和1-(邻叠氮基苯基)苯并咪唑的热解合成苯并咪唑并[1,2-a]苯

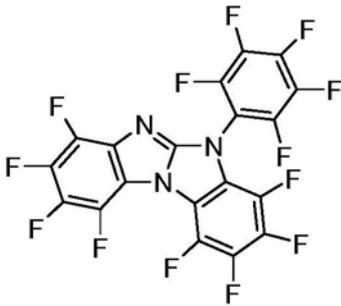


[0004] Pedro Molina等.Tetrahedron (1994) 10029-10036报道了由二(2-氨基苯基)胺与2当量异氰酸酯衍生的二(亚氨基正磷)的氮杂Wittig类型反应直接提供苯并咪唑并[1,2,a]苯并咪唑衍生物。



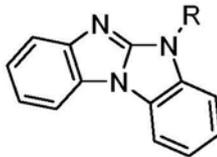
[0006] Kolesnikova,I.V.;Zhurnal Organicheskoi Khimii 25(1989) 1689-95描述了1,2,3,4,7,8,9,10-八氟-5-(2,3,4,5,6-五氟苯基-5H-苯并咪唑并[1,2-a]苯并咪唑)的合成。

[0007]



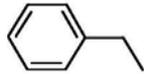
[0008] Achour,Reddouane;Zniber,Rachid,Bulletin des Societes Chimiques Belges

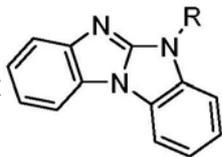
96 (1987) 787-92描述了苯并咪唑并苯并咪唑 (R=H, -CH(CH₃)₂) 的合成,其



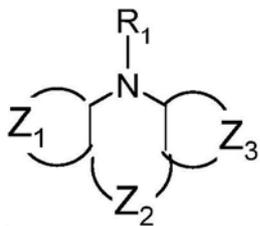
由苯并咪唑啉酮衍生物制备。

[0009] Hubert,Andre J.;Reimplinger,Hans,Chemische Berichte 103 (1970) 2828-35描

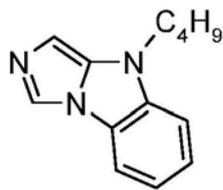
述了苯并咪唑并苯并咪唑 (R=H, CH₃, ) 的合成。



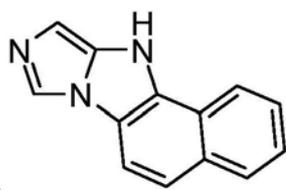
[0010] JP2001160488描述了一种场致发光元件,其在相对的阳极和阴极之间具有发光层,其具有单层或多层有机化合物膜,其中至少一层有机化合物膜含有至少一种由式



所示的化合物。明确公开如下化合物:

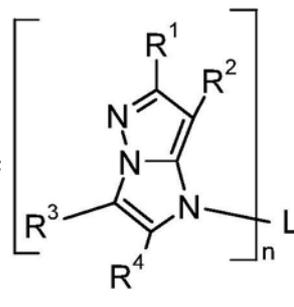


和



[0011] US20100244006涉及一种有机场致发光器件,该器件包括:阴极;阳极;和在阴极和阳极之间的至少一个有机层。所述至少一个有机层包括含有至少一种发光材料的发光层。

在所述至少一个有机层中含有由下式所表示的化合物:

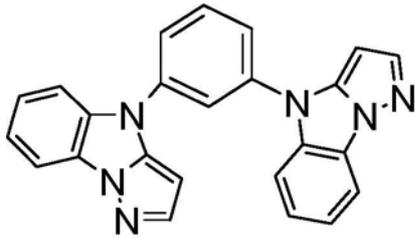


其中n为

(I)。

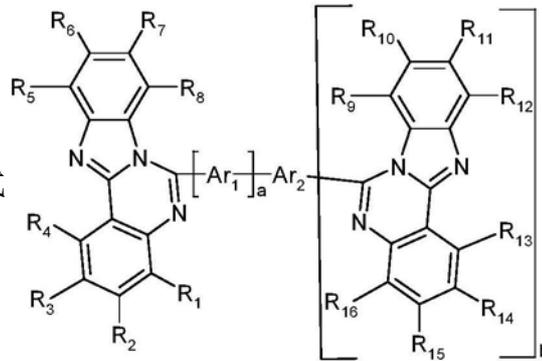
2或更大的整数,L为n价连接基团且R¹、R²、R³和R⁴各自独立地为氢原子或取代基。

[0012] US 20100244006中所述的化合物优选在发光层中用作主体。



代表了US20100244006中所公开的化合物的一个实例。

[0013] KR1020110008784涉及式

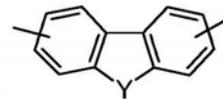
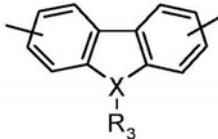
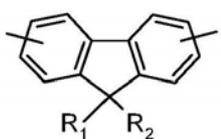
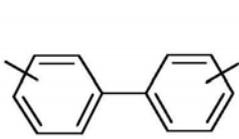


的新型有机发光

化合物和包含其的有机场致发光器件。

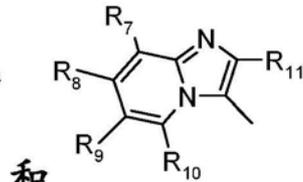
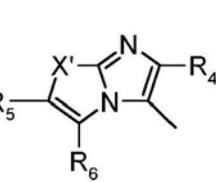
[0014] US2005079387涉及一种含有式Ar₁-Ar₂-Ar₃化合物的咪唑环(发蓝光主体化合物)

和使用其的有机场致发光(EL)显示器件。Ar₂选自



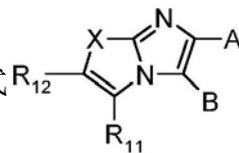
Ar₁和Ar₃

各自独立地选自



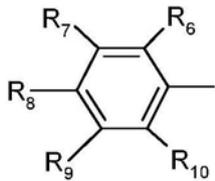
其中X'为O或S。

[0015] US2005074632涉及一种含有式

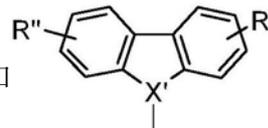


场致发光(EL)显示器件。含咪唑环化合物尤其可以单独或与掺杂剂组合在有机膜如场致发

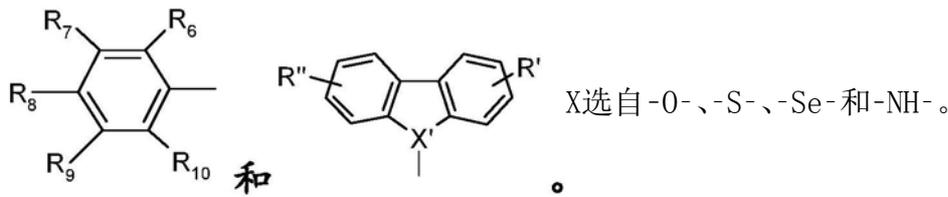
光层中用作材料。A选自



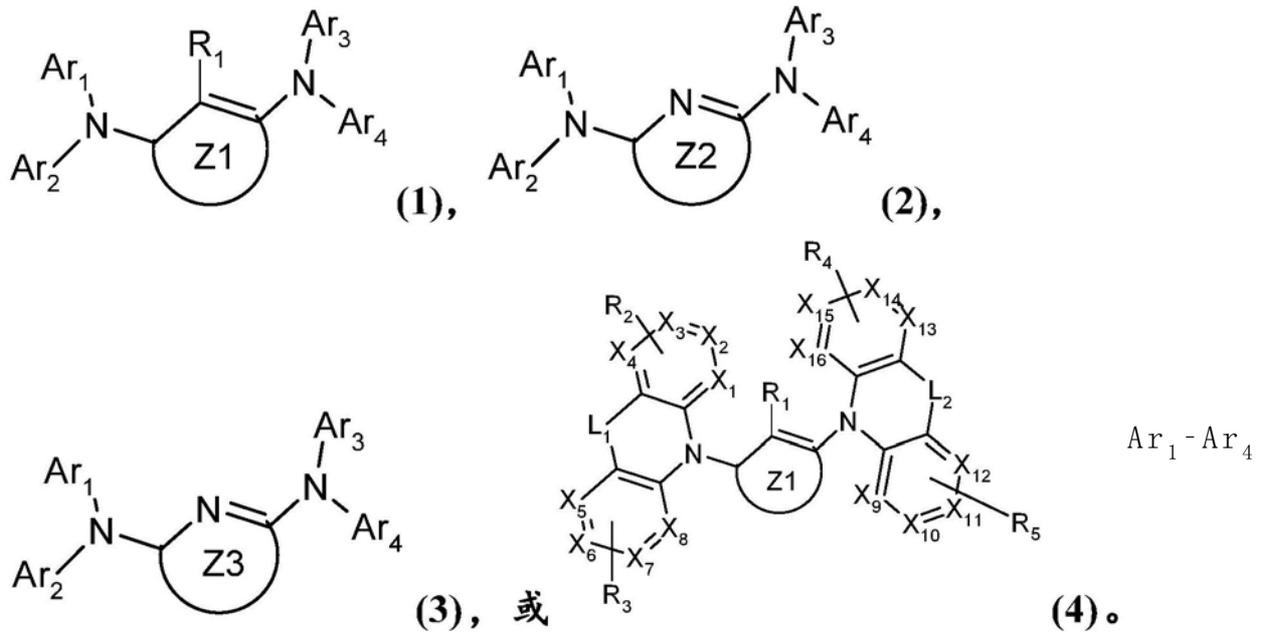
-N(R₁₃,R₁₄)和



B选自

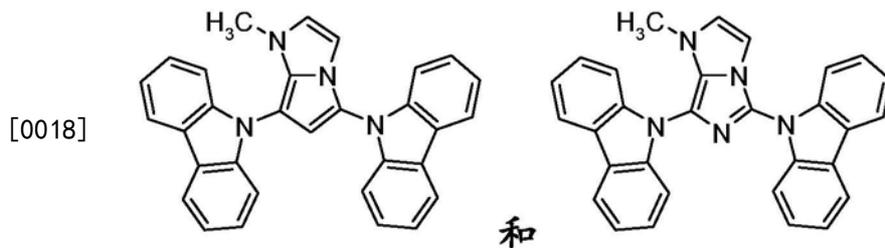


[0016] JP2007180147涉及一种有机场致发光元件,其夹在阳极和阴极之间且含有至少一个发光层,其含有由通式1、2、3或4所表示的化合物:

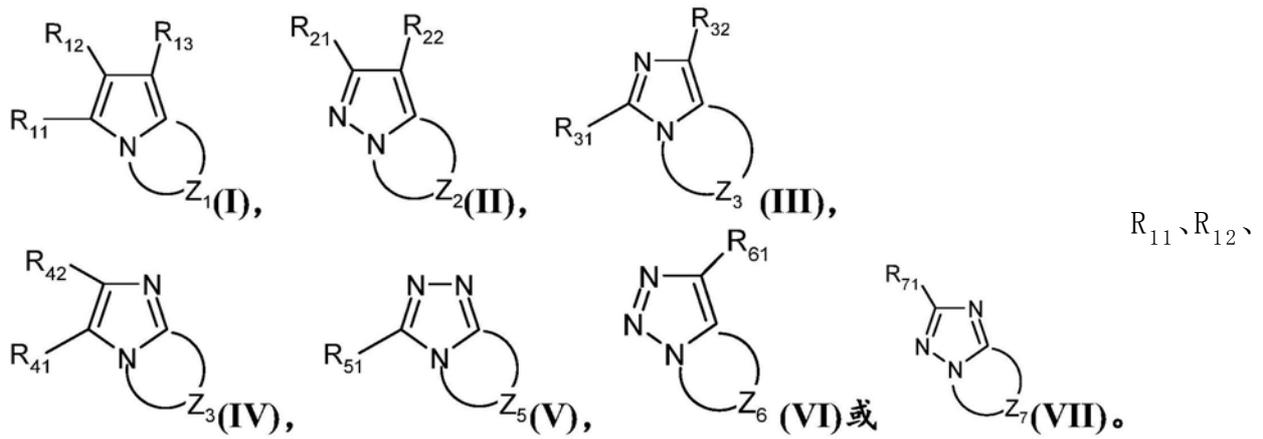


=芳族基团或芳族杂环基团; $R_1 - R_5 = H$ 或取代基; $Z_1 =$ 形成5或6员杂环所需残基; $L_1, L_2 =$ 键或偶联基团;和 $X_1 - X_{16} =$ 碳或氮。在 Ar_1 和 Ar_2 以及 Ar_3 和 Ar_4 的一个部分中可以形成新环。

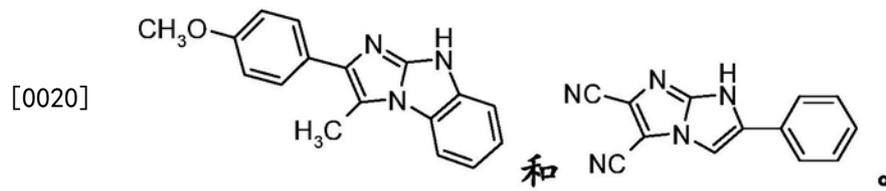
[0017] 明确公开了如下化合物:



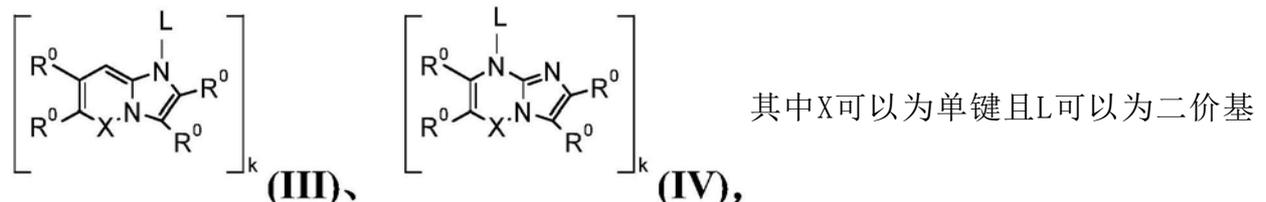
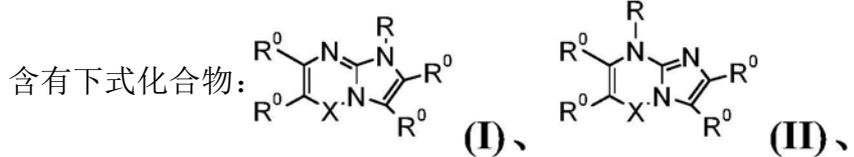
[0019] US6551723涉及一种有机场致发光元件,其在一对电极之间包含一个发光层或含有发光层的多个有机化合物薄层,其中有机场致发光元件中的至少一个层包含至少一种由式(I)至(VII)所表示的杂环化合物:



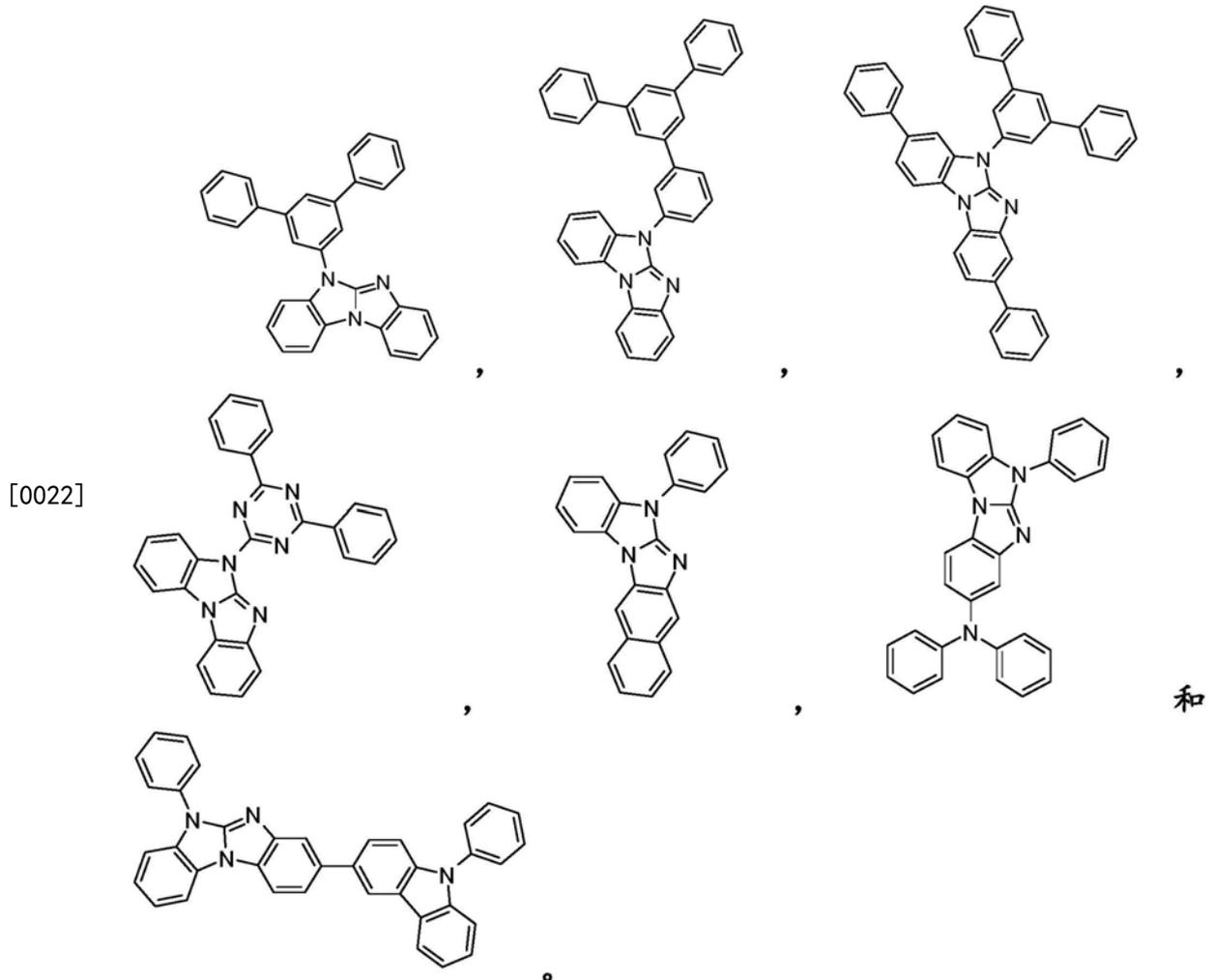
R_{13} 、 R_{21} 、 R_{22} 、 R_{31} 、 R_{32} 、 R_{41} 、 R_{42} 、 R_{51} 、 R_{61} 和 R_{71} 各自独立地为氢原子或取代基； Z_1 、 Z_2 、 Z_3 、 Z_4 、 Z_5 、 Z_6 和 Z_7 为形成5或6员环所必需的原子的基团。尤其将由式 (I) 至 (VII) 所表示的化合物加入发光层和/或电子注入/传输层。明确公开了如下化合物：



[0021] W02011160757涉及一种电子器件,其包含阳极、阴极和至少一个有机层,该有机层



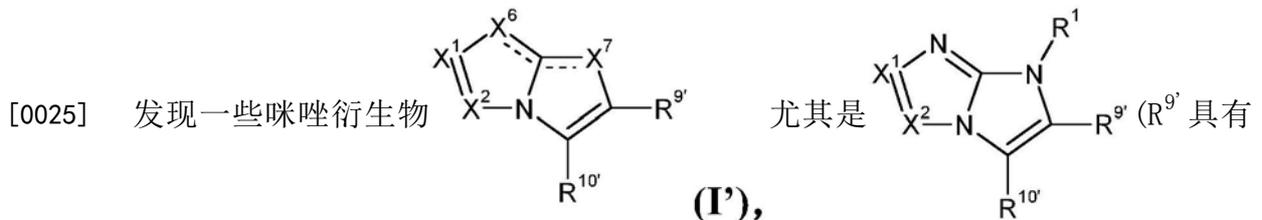
团。明确公开了如下4H-咪唑并[1,2-a]咪唑化合物：



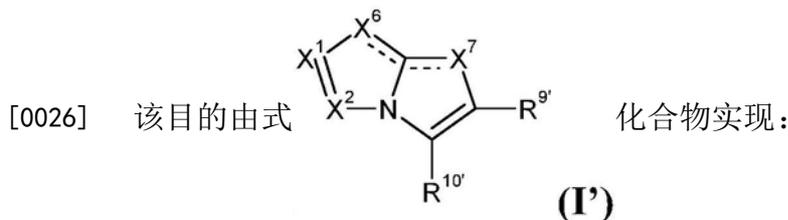
[0023] 尽管存在这些进展,仍然需要包含新型空穴传输材料以提供具有改进效率、稳定性、可制造性和/或光谱特性的场致发光器件的有机发光器件。

发明内容

[0024] 因此,本发明的目的是相对于上述现有技术提供适合用于OLED和有机电子中的其他应用的其他材料。更特别地,应可以提供用于OLED的空穴传输材料、电子/激子阻断剂材料和基体材料。该材料应尤其适合用于包含至少一个磷光发光体,尤其是至少一个绿色发光体或至少一个蓝色发光体的OLED。此外,该材料应适合用于提供OLED,其确保OLED具有良好效率、良好操作寿命和对热应力的高稳定性以及低使用电压和操作电压。



R^9 的含义, R^{10} 具有 R^{10} 的含义) 适合用于有机场致发光器件中。尤其是一些咪唑衍生物为具有良好效率和耐久性的磷光发光体的合适空穴传输材料或主体材料。



[0027] 其中

[0028] X^6 为-N=且 X^7 为-NR¹-,或

[0029] X^7 为=N-且 X^6 为-NR¹-,

[0030] R¹为式-A¹-(A²)_p-(A³)_q-(A⁴)_r-R⁶的基团,

[0031] p为0或1,q为0或1,r为0或1,

[0032] A¹、A²、A³和A⁴相互独立地为可以任选被G取代的C₆-C₂₄亚芳基,或可以任选被G取代的C₂-C₃₀亚杂芳基;其中

[0033] 基团A¹、A²、A³和A⁴可以被一个或多个基团-(SiR⁷R⁸)-间隔;

[0034] R⁶为H,基团-(SiR²⁰R²¹R²²),可以任选被G取代的C₆-C₂₄芳基,或可以任选被G取代的C₂-C₃₀杂芳基;

[0035] R⁷和R⁸相互独立地为C₁-C₂₅烷基,或C₆-C₂₄芳基,其可以任选被G取代;

[0036] X¹为N或CR⁹;

[0037] X²为N或CR¹⁰,

[0038] R⁹、R¹⁰、R^{9'}和R^{10'}相互独立地为H,可以任选被E取代和/或被D间隔的C₁-C₂₅烷基,可以任选被G取代的C₆-C₂₄芳基,或可以任选被G取代的C₂-C₃₀杂芳基;或R⁹和R¹⁰和/或R^{9'}和R^{10'}一起形成可任选被取代的环,

[0039] R²⁰、R²¹和R²²相互独立地为C₁-C₂₅烷基,或C₆-C₂₄芳基,其可以任选被G取代;D为-CO-、-COO-、-S-、-SO-、-SO₂-、-O-、-NR⁶⁵-、-SiR⁷⁰R⁷¹-、-POR⁷²-、-CR⁶³=CR⁶⁴-或-C≡C-

[0040] E为-OR⁶⁹、-SR⁶⁹、-NR⁶⁵R⁶⁶、-COR⁶⁸、-COOR⁶⁷、-CONR⁶⁵R⁶⁶、-CN或卤素,

[0041] G为E或C₁-C₁₈烷基,C₆-C₂₄芳基,任选被F取代的C₆-C₂₄芳基,C₁-C₁₈烷基或任选被O间隔的C₁-C₁₈烷基,C₂-C₃₀杂芳基或任选被F取代的C₂-C₃₀杂芳基,C₁-C₁₈烷基或任选被O间隔的C₁-C₁₈烷基;

[0042] R⁶³和R⁶⁴相互独立地为C₆-C₁₈芳基,被C₁-C₁₈烷基、C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基;

[0043] R⁶⁵和R⁶⁶相互独立地为C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基;或者

[0044] R⁶⁵和R⁶⁶一起形成5或6员环,

[0045] R⁶⁷为C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基,

[0046] R⁶⁸为H,C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基,

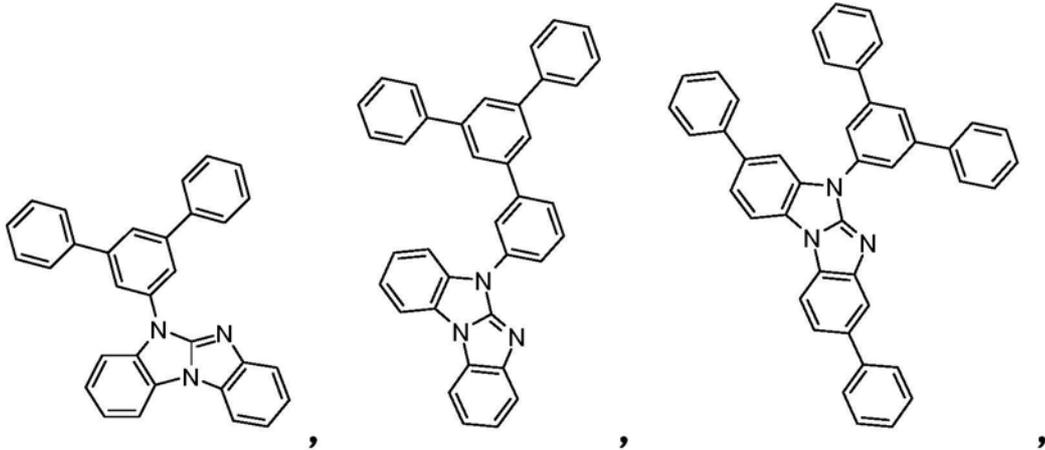
[0047] R⁶⁹为C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基,

[0048] R⁷⁰和R⁷¹相互独立地为C₁-C₁₈烷基,C₆-C₁₈芳基或被C₁-C₁₈烷基取代的C₆-C₁₈芳基,以

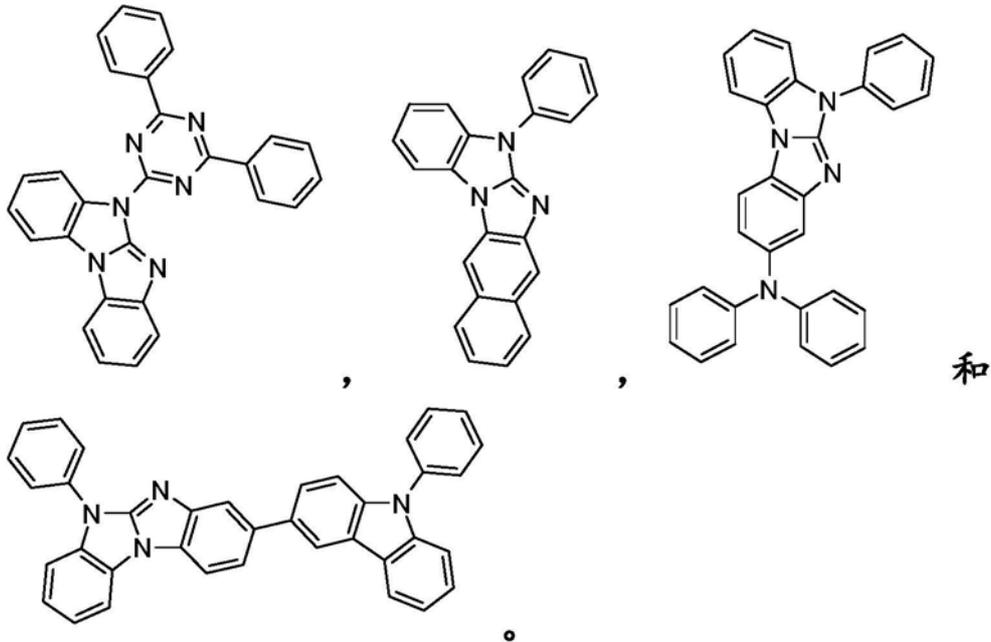
及

[0049] R^{72} 为 C_1-C_{18} 烷基, C_6-C_{18} 芳基或被 C_1-C_{18} 烷基取代的 C_6-C_{18} 芳基;

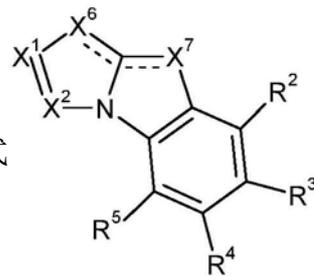
[0050] 条件是排除如下化合物:



[0051]



[0052] 尤其是所述目的由式



化合物,非常特别是

(I)



[0053] X^6 为 $-N=$ 且 X^7 为 $-NR^1-$,或

- [0054] X^7 为=N-且 X^6 为-NR¹-,其中
- [0055] R¹为式-A¹-(A²)_p-(A³)_q-(A⁴)_r-R⁶的基团,
- [0056] p为0或1,q为0或1,r为0或1,
- [0057] A¹、A²、A³和A⁴相互独立地为可以任选被G取代的C₆-C₂₄亚芳基,或可以任选被G取代的C₂-C₃₀亚杂芳基;其中
- [0058] 基团A¹、A²、A³和A⁴可以被一个或多个基团-(SiR⁷R⁸)-间隔;
- [0059] R²、R³、R⁴和R⁵相互独立地为H,可任选被E取代和/或被D间隔的C₁-C₂₅烷基;可以任选被G取代的C₆-C₂₄芳基,或可以任选被G取代的C₂-C₃₀杂芳基;
- [0060] R⁶为H,基团-(SiR²⁰R²¹R²²),可以任选被G取代的C₆-C₂₄芳基,或可以任选被G取代的C₂-C₃₀杂芳基;
- [0061] R⁷和R⁸相互独立地为C₁-C₂₅烷基,或C₆-C₂₄芳基,其可以任选被G取代;
- [0062] X¹为N或CR⁹;
- [0063] X²为N或CR¹⁰,
- [0064] R⁹和R¹⁰相互独立地为H,可以任选被E取代和/或被D间隔的C₁-C₂₅烷基,可以任选被G取代的C₆-C₂₄芳基,或可以任选被G取代的C₂-C₃₀杂芳基;或
- [0065] R⁹和R¹⁰一起形成可任选被取代的环,
- [0066] R²⁰、R²¹和R²²相互独立地为C₁-C₂₅烷基,或C₆-C₂₄芳基,其可以任选被G取代;D为-CO-、-COO-、-S-、-SO-、-SO₂-、-O-、-NR⁶⁵-、-SiR⁷⁰R⁷¹-、-POR⁷²-、-CR⁶³=CR⁶⁴-或-C≡C-,
- [0067] E为-OR⁶⁹、-SR⁶⁹、-NR⁶⁵R⁶⁶、-COR⁶⁸、-COOR⁶⁷、-CONR⁶⁵R⁶⁶、-CN或卤素,
- [0068] G为E或C₁-C₁₈烷基,C₆-C₂₄芳基,任选被C₁-C₁₈烷基取代的C₆-C₂₄芳基;
- [0069] R⁶³和R⁶⁴相互独立地为C₆-C₁₈芳基,被C₁-C₁₈烷基、C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基;
- [0070] R⁶⁵和R⁶⁶相互独立地为C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基;或者
- [0071] R⁶⁵和R⁶⁶一起形成5或6员环,
- [0072] R⁶⁷为C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基,
- [0073] R⁶⁸为H,C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基,
- [0074] R⁶⁹为C₆-C₁₈芳基,被C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基,C₁-C₁₈烷基或被-O-间隔的C₁-C₁₈烷基,
- [0075] R⁷⁰和R⁷¹相互独立地为C₁-C₁₈烷基,C₆-C₁₈芳基或被C₁-C₁₈烷基取代的C₆-C₁₈芳基,以及
- [0076] R⁷²为C₁-C₁₈烷基,C₆-C₁₈芳基或被C₁-C₁₈烷基取代的C₆-C₁₈芳基。
- [0077] 本发明化合物可以用于电摄影感光体,光电转换器,有机太阳能电池(有机光伏器件),开关元件,如有机晶体管,例如有机FET和有机TFT,有机发光场效应晶体管(OLEFET),图像传感器,染料激光器和场致发光器件,例如有机发光二极管(OLED)。
- [0078] 因此,本发明的另一主题涉及一种包含本发明化合物的电子器件。该电子器件优选为场致发光器件。

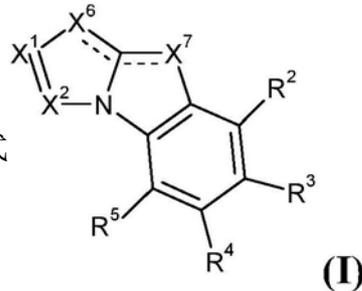
[0079] 式I化合物原则上可以用于EL器件的任何层,但是优选用作主体、空穴传输和电子阻断材料。特别地,式I化合物在发蓝光的磷光发光体中用作主体材料。

[0080] 因此,本发明的另一主题涉及一种包含本发明式I化合物的空穴传输层。

[0081] 本发明的另一主题涉及一种包含本发明式I化合物的发光层。在所述实施方案中,式I化合物优选作为主体材料与磷光发光体组合使用。

[0082] 本发明的另一主题涉及一种包含本发明式I化合物的电子阻断层。

[0083] 式I化合物尤其是式



的化合物,其中

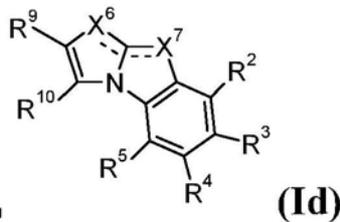
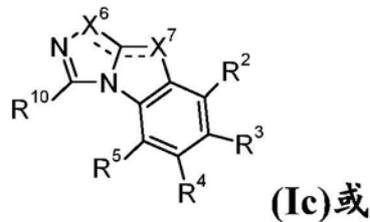
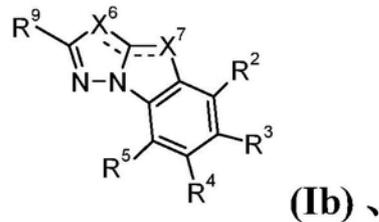
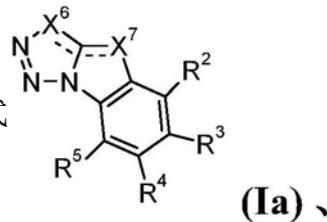
[0084] X^6 为 $-N=$ 且 X^7 为 $-NR^1-$,或

[0085] X^7 为 $=N-$ 且 X^6 为 $-NR^1-$,

[0086] R^2 、 R^3 、 R^4 和 R^5 相互独立地为H,可以任选被E取代和/或被D间隔的 C_1-C_{25} 烷基,可以任选被G取代的 C_6-C_{24} 芳基,或可以任选被G取代的 C_2-C_{30} 杂芳基,

[0087] X^1 、 X^2 、 R^1 、E、D和G如上所定义。

[0088] 式I化合物可以为式



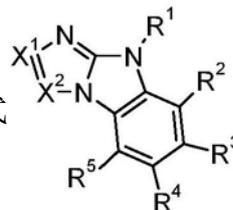
的化合物, X^6 为 $-N=$ 且 X^7 为 $-NR^1-$ 或 X^7 为

$=N-$ 且 X^6 为 $-NR^1-$ 。尤其优选如下式Id化合物,其中 R^9 和 R^{10} 一起形成环



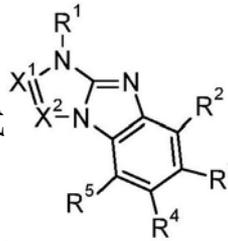
R^{11} 、 R^{12} 、 R^{13} 和 R^{14} 如下所定义。 R^{11} 、 R^{12} 、 R^{13} 和 R^{14} 优选为H。 R^2 、 R^3 、 R^4 和 R^5 优选为H。

[0089] 在一个优选实施方案中,式(I)化合物为式



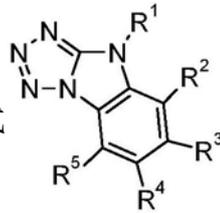
的化合物。在另一优选

实施方案中,式(I)化合物为式

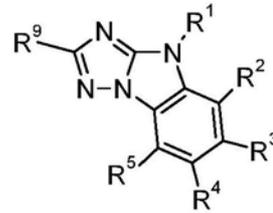


的化合物。

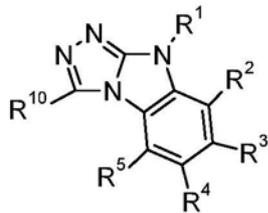
[0090] 优选地,式I化合物为式



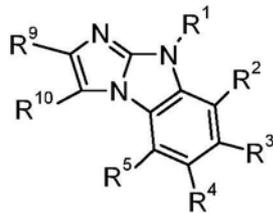
(Ia')、



(Ib')、



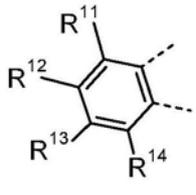
(Ic')或



(Id')

的化合物。优选式Ib'和Id'的化合

物。更优选式Id'化合物。甚至更优选如下式Id'化合物,其中R⁹和R¹⁰一起形成环

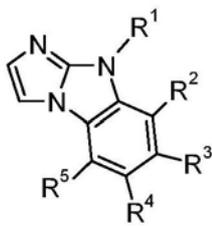


其中R¹¹、R¹²、R¹³和R¹⁴如下所定义。R¹¹、R¹²、R¹³和R¹⁴优选为H。

[0091] R²、R³、R⁴和R⁵优选为H。

[0092] 不呈轴向对称的式I化合物,例如其中R⁹和R¹⁰为H的式Id'化合物可以以两种异构体形式存在:

[0093]



(Id')和

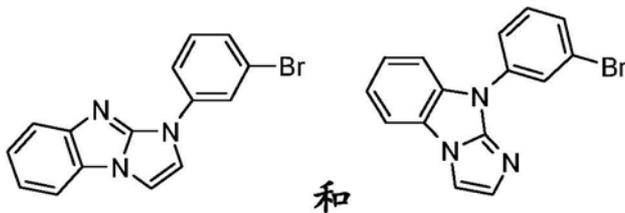


(Id'')。

参考实施例2,其描述了如下化

合物的混合物的合成:

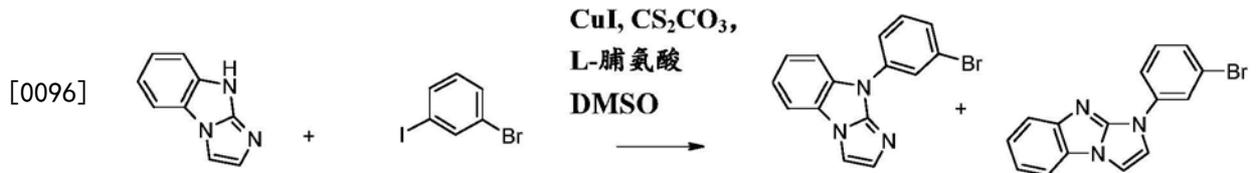
[0094]



和

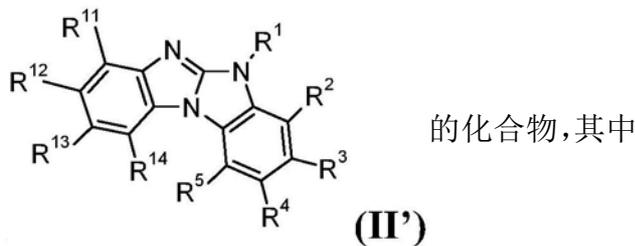
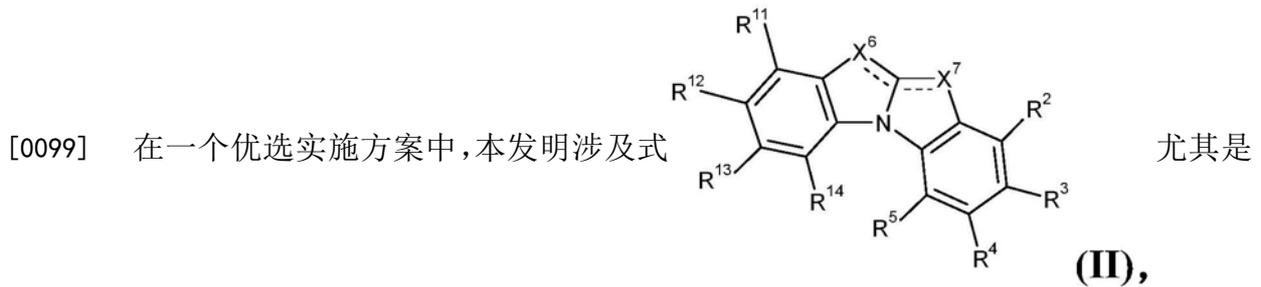
。

[0095] 4H-咪唑并[1,2-a]苯并咪唑如ARKIVOC 2002 (v) 48-61中所述制备。4H-咪唑并[1,2-a]苯并咪唑与1-溴-3-碘苯的Ullmann反应得到比值af约为1:1的两种异构体。



[0097] R^{20} 、 R^{21} 和 R^{22} 优选为 C_1 - C_{18} 烷基,例如甲基、乙基、正丙基、异丙基、正丁基、异丁基、仲丁基、叔丁基、2-甲基丁基、正戊基、异戊基、正己基、2-乙基己基或正庚基, C_6 - C_{14} 芳基,例如苯基、萘基或联苯基,或被G取代的 C_6 - C_{14} 芳基,例如 $-C_6H_4OCH_3$ 、 $-C_6H_4OCH_2CH_3$ 、 $-C_6H_3(OCH_3)_2$ 或 $-C_6H_3(OCH_2CH_3)_2$ 、 $-C_6H_4CH_3$ 、 $-C_6H_3(CH_3)_2$ 、 $-C_6H_2(CH_3)_3$ 或 $-C_6H_4tBu$ 。

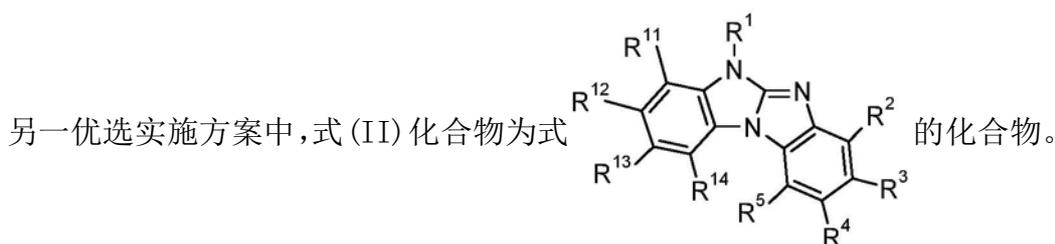
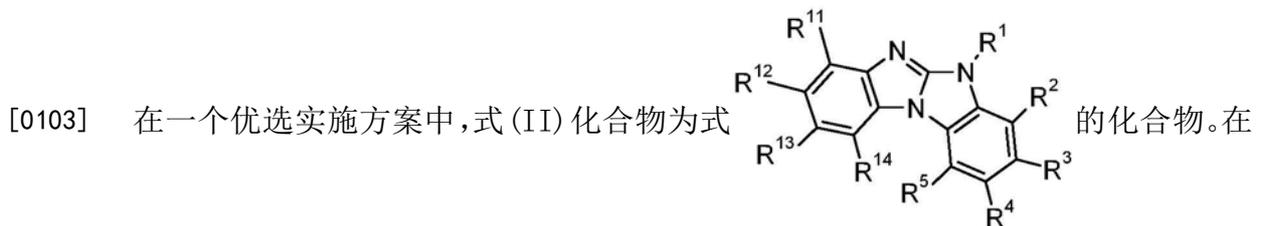
[0098] R^9 和 R^{10} 优选为H或 C_6 - C_{14} 芳基,例如苯基、萘基或联苯基,其可以任选被一个或多个 C_1 - C_8 烷基取代;或 C_2 - C_{30} 杂芳基,例如二苯并咪唑基,其可以任选被一个或多个 C_1 - C_8 烷基取代。



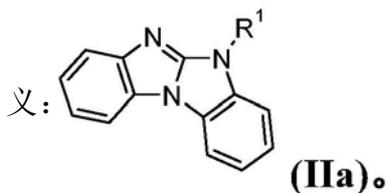
[0100] X^6 为 $-N=$ 且 X^7 为 $-NR^1-$ 或

[0101] X^7 为 $=N-$ 且 X^6 为 $-NR^1-$,

[0102] R^{11} 、 R^{12} 、 R^{13} 和 R^{14} 相互独立地为H,可任选被E取代和/或被D间隔的 C_1 - C_{25} 烷基,可以任选被G取代的 C_6 - C_{24} 芳基,或可以任选被G取代的 C_2 - C_{30} 杂芳基,和E、D、G、 R^1 、 R^2 、 R^3 、 R^4 和 R^5 如上所定义。



[0104] 甚至更优选如下式II化合物,其中 R^2 、 R^3 、 R^4 、 R^5 、 R^{11} 、 R^{12} 、 R^{13} 和 R^{14} 为H和 R^1 如上所定



[0105] R^6 可以为可以任选被G取代的 C_6-C_{24} 芳基,或可以任选被G取代的 C_2-C_{30} 杂芳基。

[0106] 可以任选被G取代的 C_6-C_{24} 芳基 R^6 通常为苯基、4-甲基苯基、4-甲氧基苯基,萘基,尤其是1-萘基或2-萘基,联苯基,三联苯基,苊基,2-或9-苈基,菲基或蒽基,其可为未取代或取代的。

[0107] 可以任选被G取代的 C_2-C_{30} 杂芳基 R^6 表示具有5-7个环原子的环或稠合环体系,其中氮、氧或硫为可能的杂原子,并且通常为具有5-30个原子且具有至少6个共轭 π -电子的杂环基,例如噻吩基、苯并噻吩基、二苯并噻吩基、噻蒽基、呋喃基、糠基、2H-吡喃基、苯并呋喃基、异苯并呋喃基、二苯并呋喃基、苯氧基噻吩基、吡咯基、咪唑基、吡唑基、吡啶基、联吡啶基、三嗪基、嘧啶基、吡嗪基、哒嗪基、中氮茛基、异氮茛基、吡啶基、吡唑基、嘌呤基、喹啉基、喹啶基、异喹啶基、2,3-二氮杂萘基、1,5-二氮杂萘基、喹啉基、喹啶基、肉喹啶基、蝶啶基、咪唑基、苯并三唑基、苯并噁唑基、菲啶基、吡啶基、嘧啶基、菲咯啶基、吩嗪基、异噻唑基、吩噻唑基、异噁唑基、呋咱基、4-咪唑并[1,2-a]苯并咪唑基、5-苯并咪唑并[1,2-a]苯并咪唑基、咪唑基或吩噁唑基,其可为未取代或取代的。此外, C_2-C_{30} 杂芳基 R^6 包括4H-[1,2,4]三唑并[1,5-a]苯并咪唑基,其可为未取代或取代的。

[0108] C_6-C_{24} 芳基和 C_2-C_{30} 杂芳基可以被G取代且优选被一个或多个 C_1-C_8 烷基取代。

[0109] 优选的 C_2-C_{30} 杂芳基为吡啶基、三嗪基、嘧啶基、4-咪唑并[1,2-a]苯并咪唑基、5-苯并咪唑并[1,2-a]苯并咪唑基、咪唑基、二苯并呋喃基,其可为未取代或被取代的,尤其是被 C_6-C_{10} 芳基或被 C_1-C_4 烷基取代的 C_6-C_{10} 芳基或 C_2-C_5 杂芳基取代。4H-[1,2,4]三唑并[1,5-a]苯并咪唑基也代表了一个优选的 C_2-C_{30} 杂芳基 R^6 。

[0110] A^1 、 A^2 、 A^3 和 A^4 相互独立地为可以任选被G取代的 C_6-C_{24} 亚芳基,或可以任选被G取代的 C_2-C_{30} 亚杂芳基。可以任选被G取代的 C_6-C_{24} 亚芳基 A^1 、 A^2 、 A^3 和 A^4 通常为亚苯基、4-甲基亚苯基、4-甲氧基亚苯基,亚萘基,尤其是1-亚萘基或2-亚萘基,亚联苯基,三亚联苯基,亚苊基,2-或9-亚苈基,亚菲基或亚蒽基,其可为未取代或取代的。

[0111] 可以任选被G取代的 C_2-C_{30} 亚杂芳基 A^1 、 A^2 、 A^3 和 A^4 表示具有5-7个环原子的环或稠合环体系,其中氮、氧或硫为可能的杂原子,并且通常为具有5-30个原子且具有至少6个共轭电子的杂环基,例如亚噻吩基、亚苯并噻吩基、亚二苯并噻吩基、亚噻蒽基、亚呋喃基、亚糠基、2H-亚吡喃基、亚苯并呋喃基、亚异苯并呋喃基、亚二苯并呋喃基、亚苯氧基噻吩基、亚吡咯基、亚咪唑基、亚吡唑基、亚吡啶基、亚联吡啶基、亚三嗪基、亚嘧啶基、亚吡嗪基、亚哒嗪基、亚中氮茛基、亚异氮茛基、亚吡啶基、亚吡唑基、亚嘌呤基、亚喹啉基、亚喹啶基、亚肉喹啶基、亚蝶啶基、亚咪唑基、亚苯并三唑基、亚苯并噁唑基、亚菲啶基、亚吡啶基、亚嘧啶基、亚菲咯啶基、亚吩嗪基、亚异噻唑基、亚吩噻唑基、亚异噁唑基、亚呋咱基、亚咪唑基或亚吩噁唑基,其可为未取代或取代的。

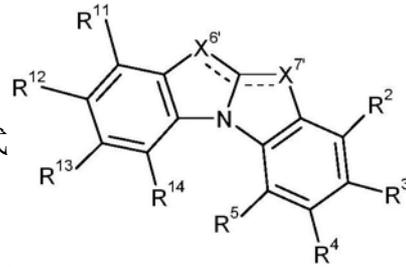
[0112] 优选的C₆-C₂₄亚芳基为1,3-亚苯基、3,3'-亚联苯基、3,3'-亚间三联苯基、2-或9-亚茚基,亚菲基,其可为未取代或取代的。

[0113] 优选的C₂-C₃₀亚杂芳基为亚吡啶基、亚三嗪基、亚嘧啶基、亚咪唑基、亚二苯并咪唑基,其可为未取代或被取代的,尤其是被C₆-C₁₀芳基或被C₁-C₄烷基取代的C₆-C₁₀芳基或C₂-C₅杂芳基取代。

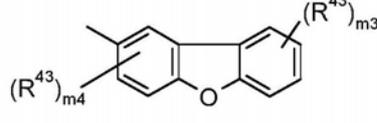
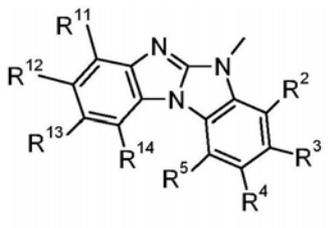
[0114] 更优选C₂-C₃₀亚杂芳基为亚咪唑基和亚二苯并咪唑基,其可以任选被C₆-C₁₀芳基取代,C₆-C₁₀芳基可任选被一个或多个C₁-C₄烷基取代。

[0115] C₆-C₂₄亚芳基和C₂-C₃₀亚杂芳基可以被G取代且优选被一个或多个C₁-C₈烷基取代。

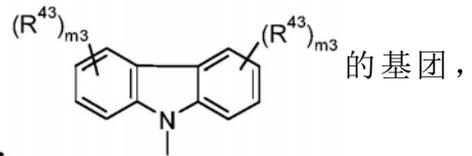
[0116] 作为术语“C₂-C₃₀杂芳基”包括例如式



尤其是

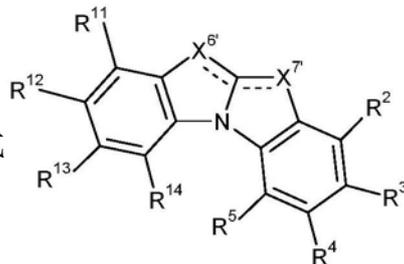


和

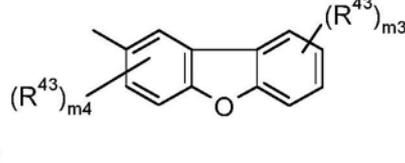
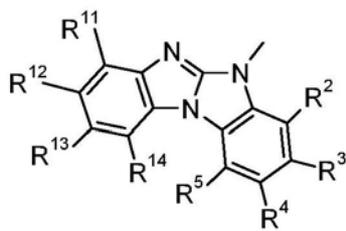


的基团,

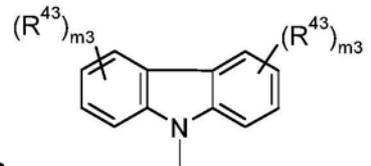
基团A¹、A²、A³和A⁴可以例如被一个或多个式



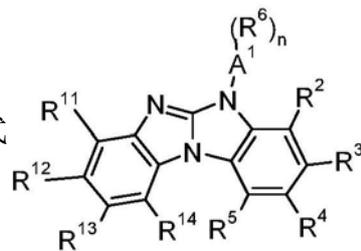
尤其是



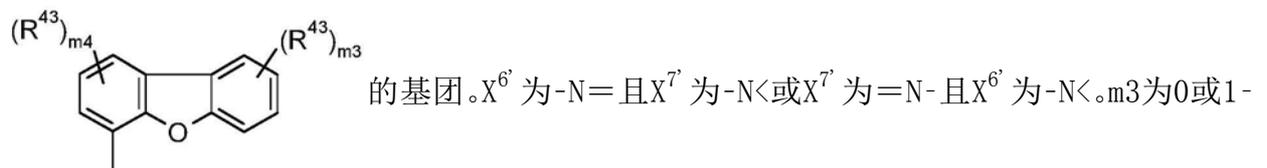
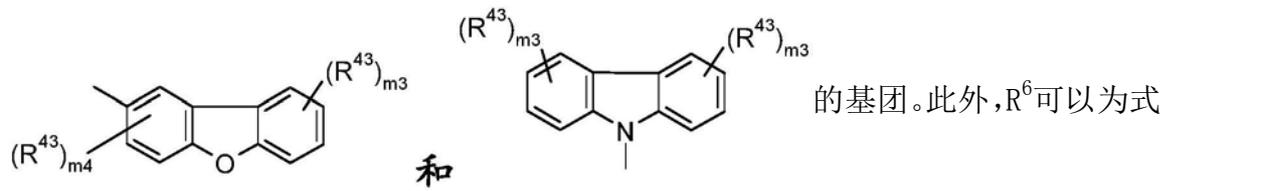
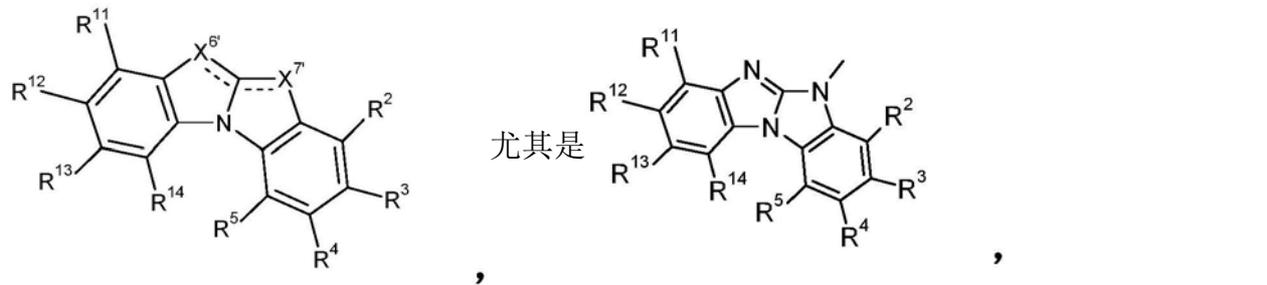
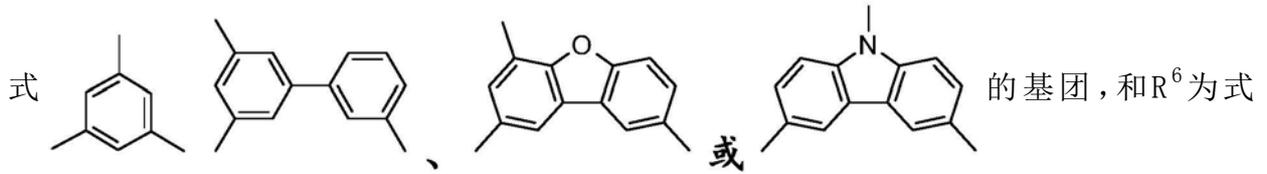
和



的基团取代,例如得到式

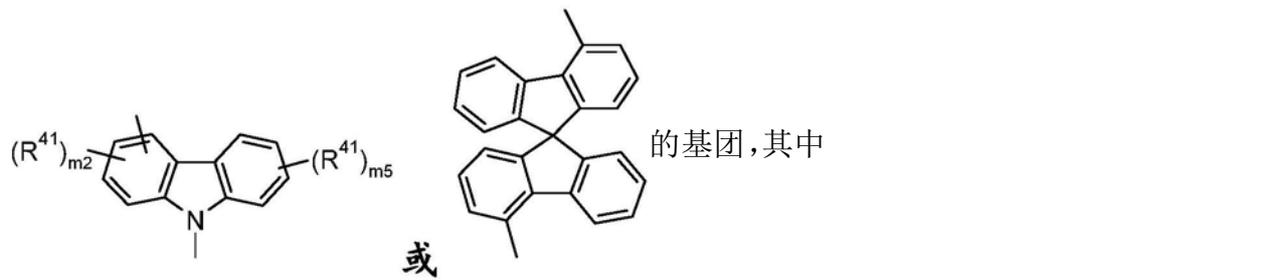
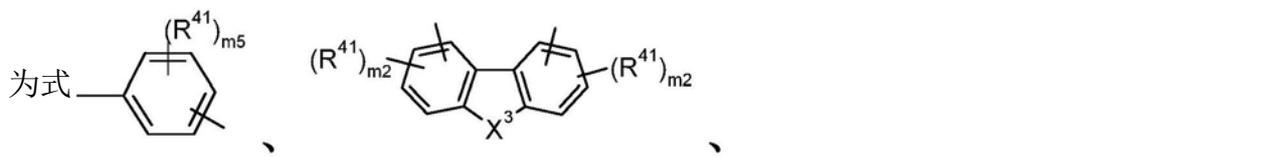
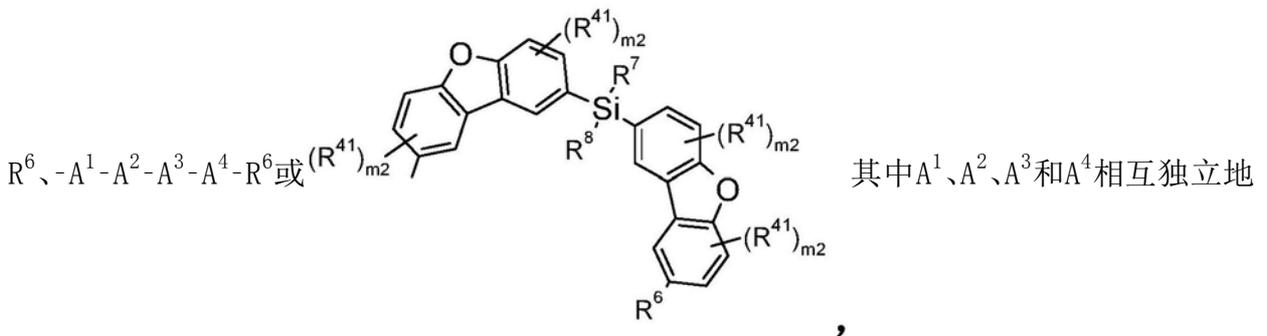


的化合物[n为2或3,尤其是2],其中A¹为



4的整数。m₄为0或整数1-3。

[0117] R¹优选为式-A¹-(A²)_p-(A³)_q-(A⁴)_r-R⁶的基团, 例如-A¹-R⁶、-A¹-A²-R⁶、-A¹-A²-A³-



[0118] m₅为0或1-4的整数,

[0119] m₂为0或整数1-3,

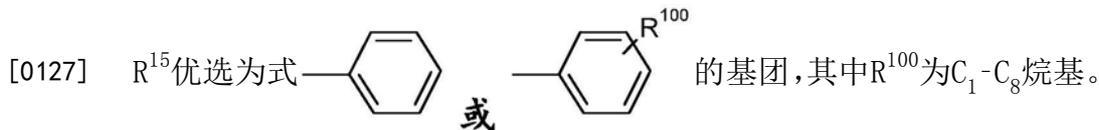
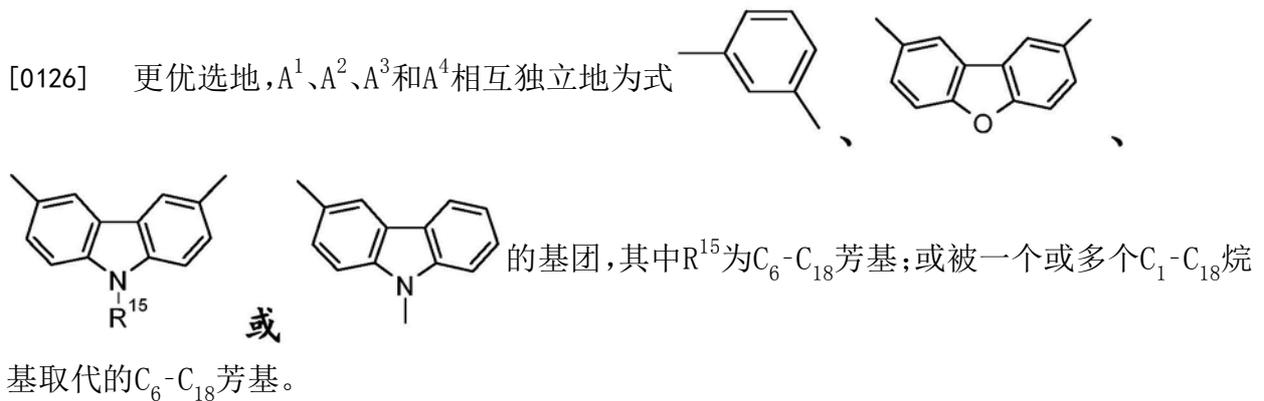
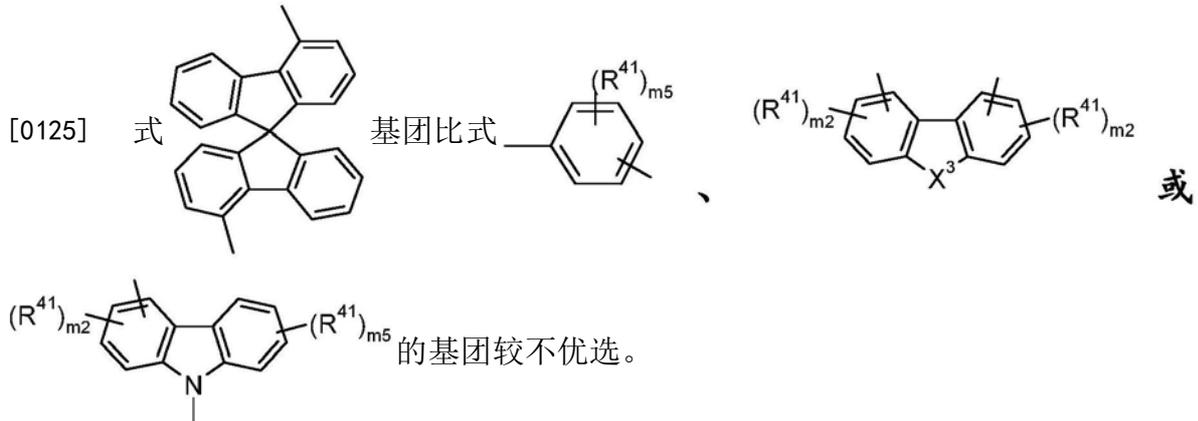
[0120] X³为-O-、-S-或-NR¹⁵-,

[0121] R⁷和R⁸为C₁-C₁₈烷基,

[0122] R¹⁵为C₁-C₁₈烷基;或被-O-间隔的C₁-C₁₈烷基;C₆-C₁₈芳基;被一个或多个C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基;C₂-C₂₀杂芳基或被一个或多个C₁-C₁₈烷基取代的C₂-C₂₀杂芳基,

[0123] R⁴¹在每次出现时可以相同或不同且为F,C₁-C₁₈烷基,被E取代和/或被D间隔的C₁-C₁₈烷基,C₆-C₂₄芳基,被G取代的C₆-C₂₄芳基,C₂-C₂₀杂芳基或被G取代的C₂-C₂₀杂芳基,和

[0124] R⁶、p、q、r、E、D和G如上文或下文所定义。

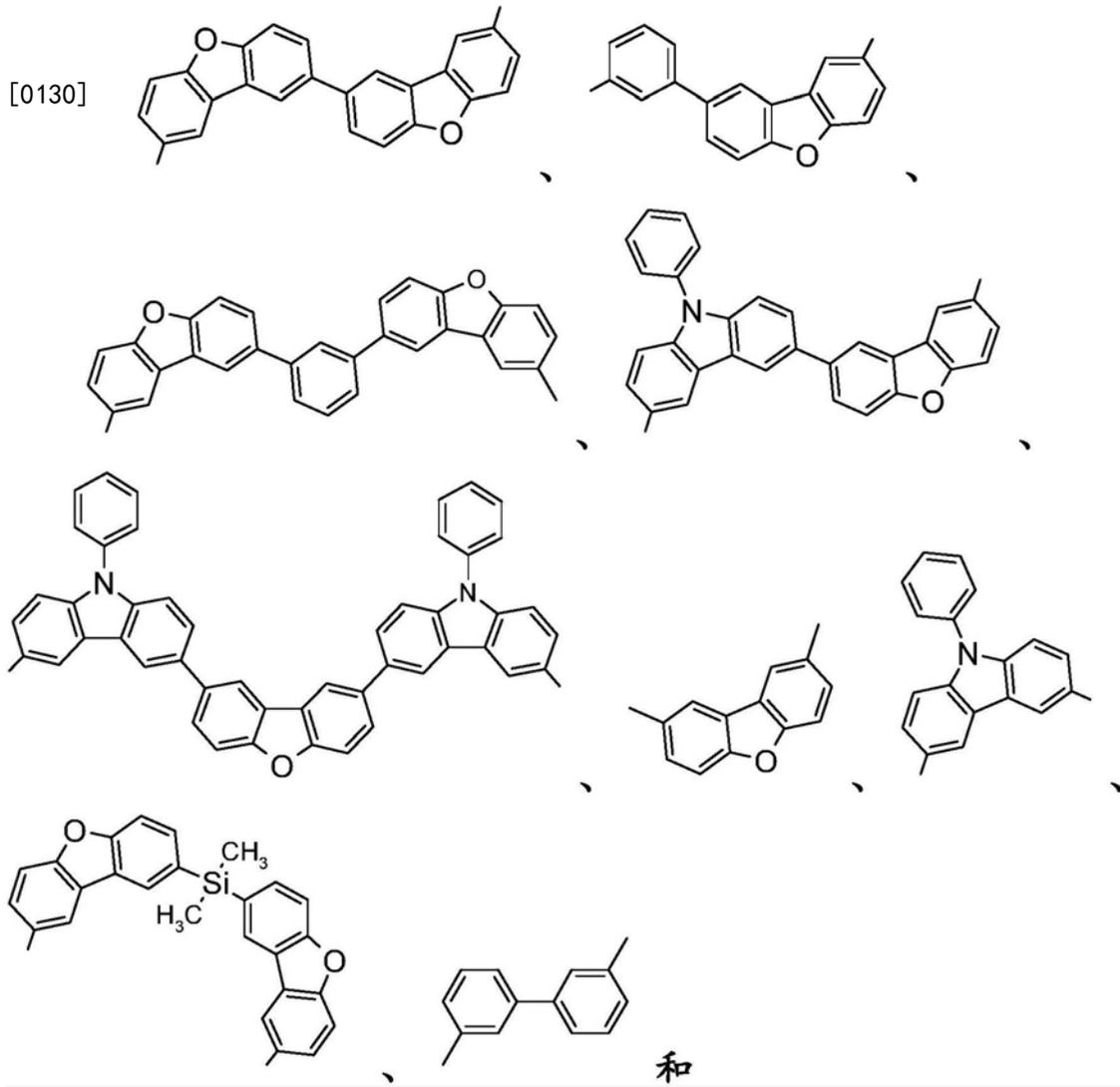


[0128] 优选基团-A¹-(A²)_p-(A³)_q-(A⁴)_r-,其中A¹、A²、A³和A⁴中至少一个为式



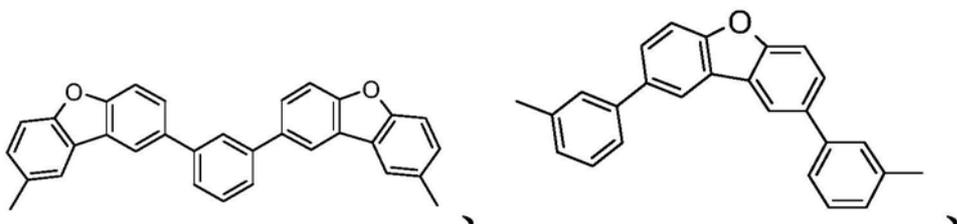
[0129] 优选的基团-A¹-(A²)_p-(A³)_q-(A⁴)_r-的实例如下所示:

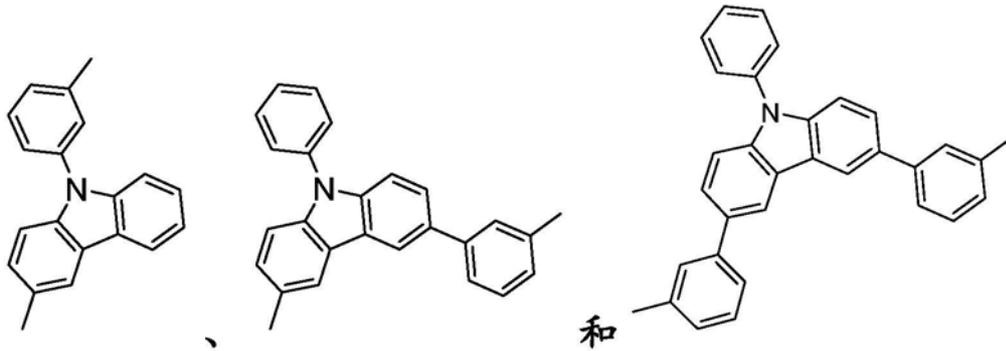
[0130]



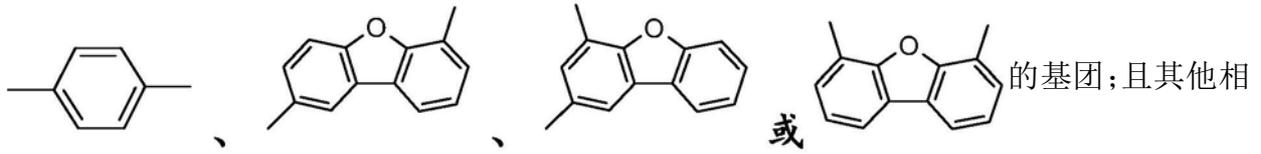
式 $-A^1-$ - $(A^2)_p-$ - $(A^3)_q-$ - $(A^4)_r-$ 的基团的

额外实例为式

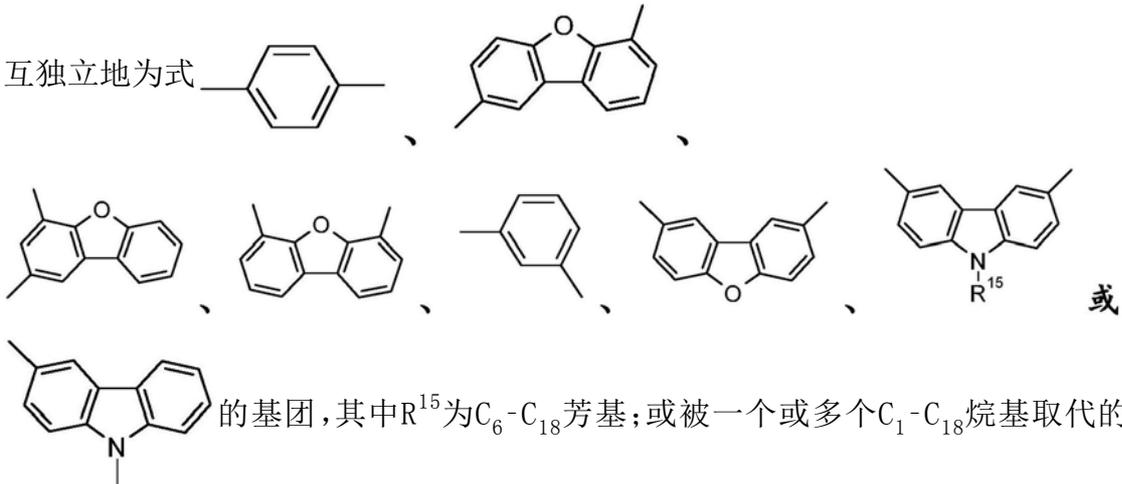




[0131] 在另一优选实施方案中, R¹为式 -A¹ - (A²)_p - (A³)_q - (A⁴)_r - R⁶ 的基团, 例如 -A¹ - R⁶、-A¹ - A² - R⁶、-A¹ - A² - A³ - R⁶ 或 -A¹ - A² - A³ - A⁴ - R⁶, 其中基团 A¹、A²、A³ 和 A⁴ 中至少一个为式

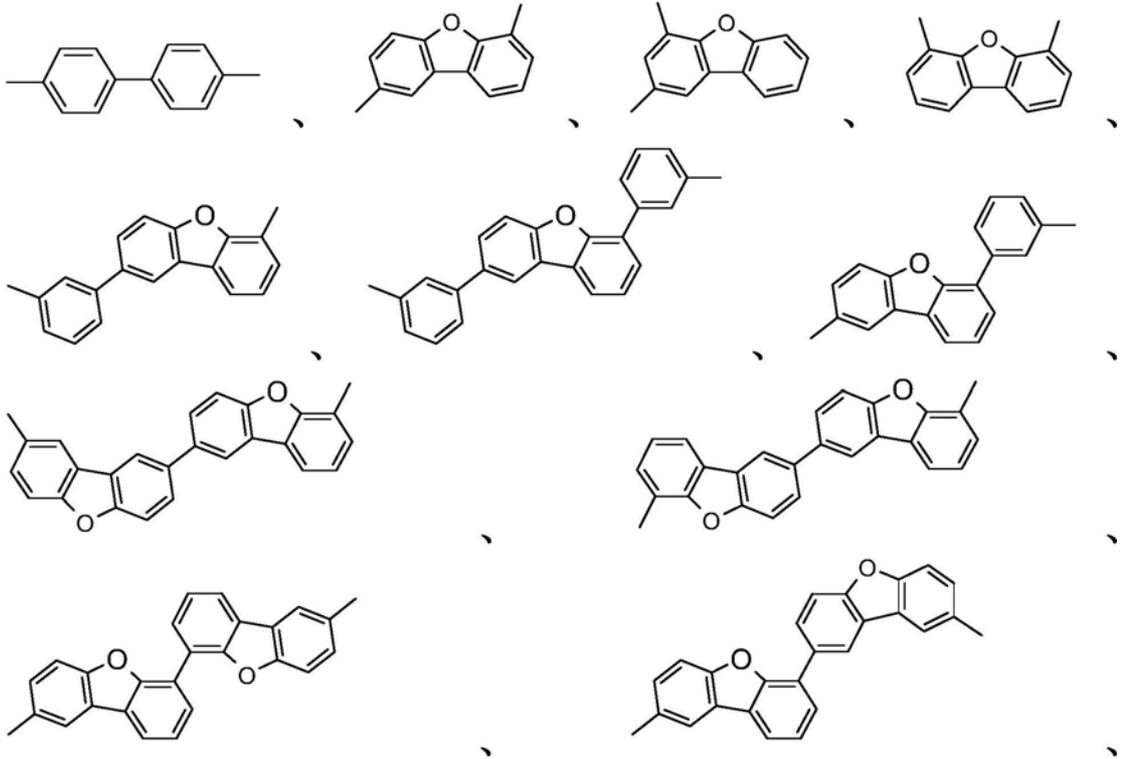


互独立地为式

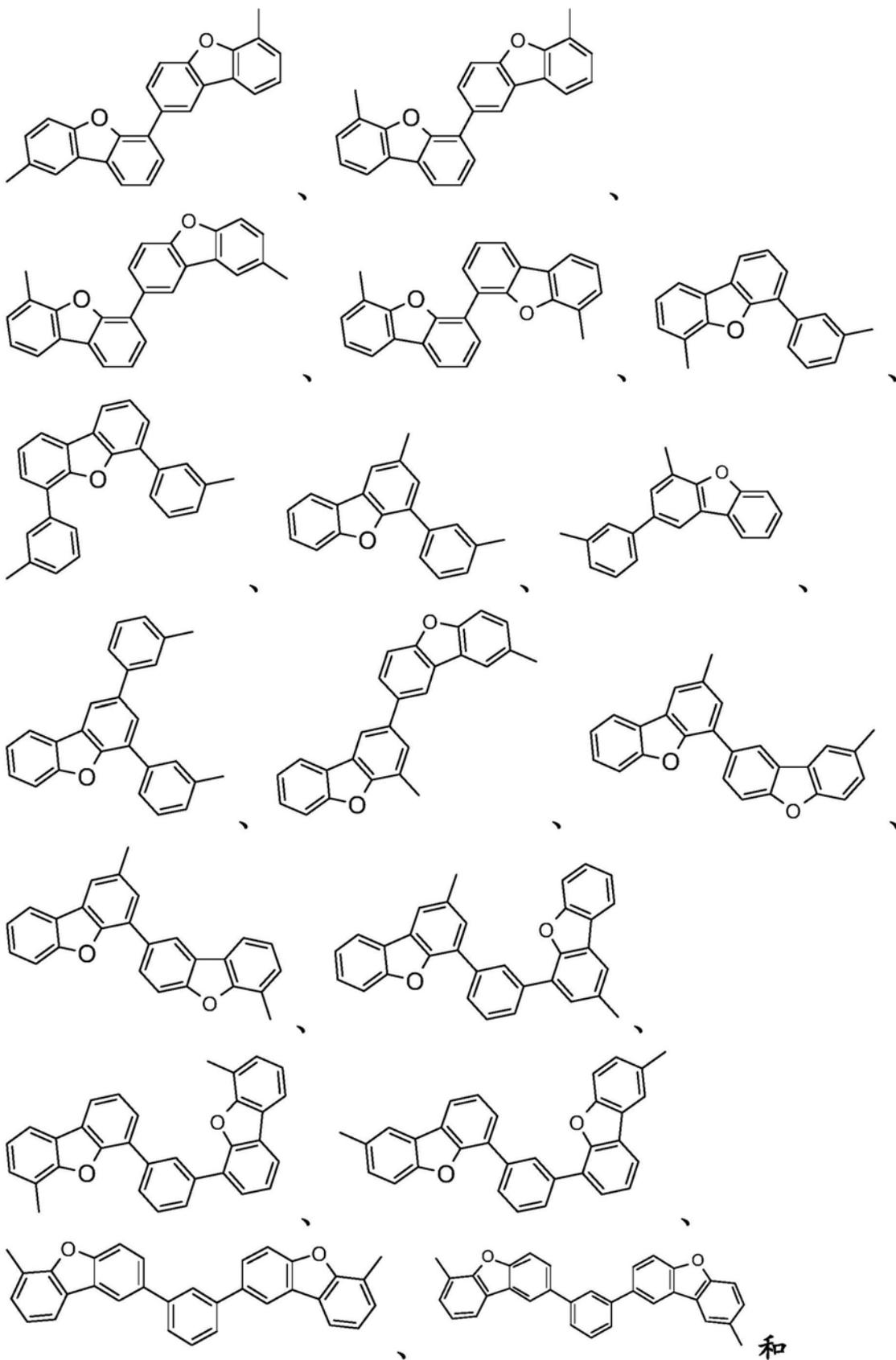


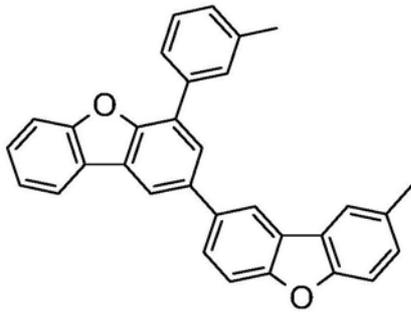
基。优选的基团 -A¹ - (A²)_p - (A³)_q - (A⁴)_r - 的实例如下所示:

[0132]

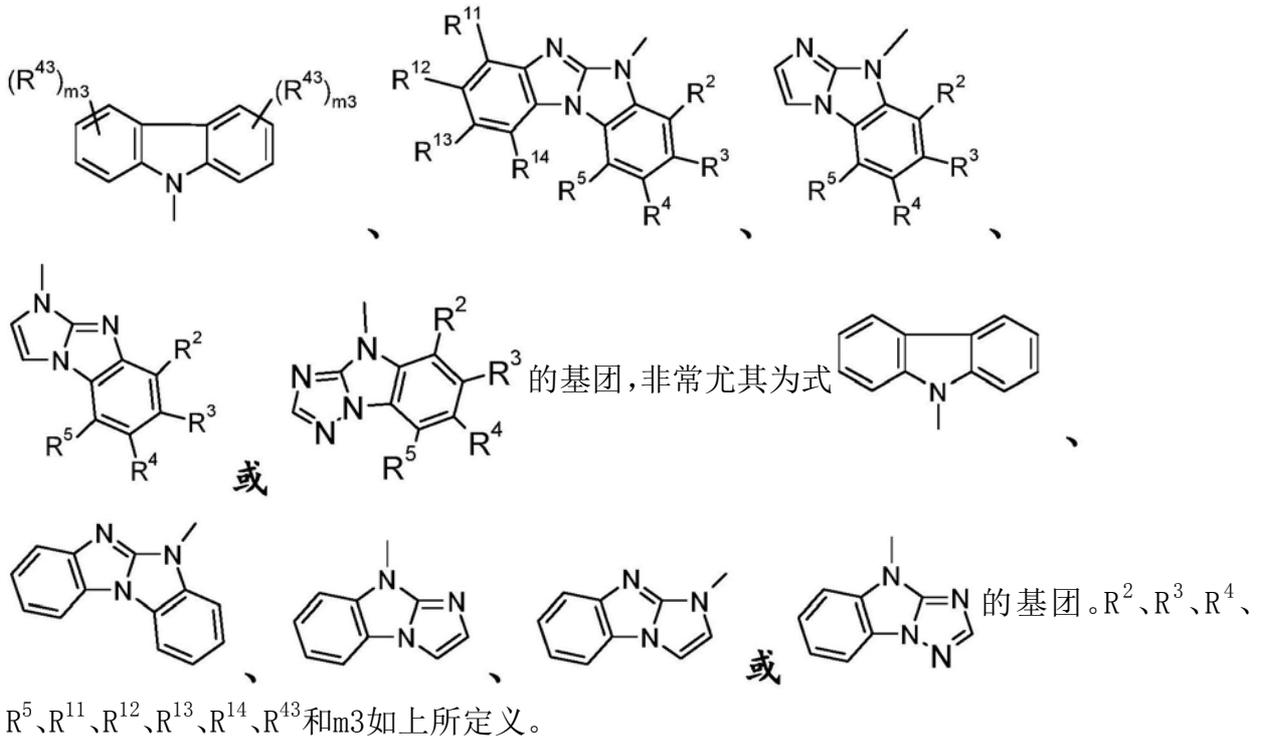


[0133]

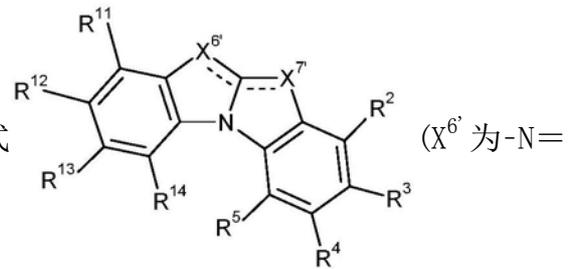




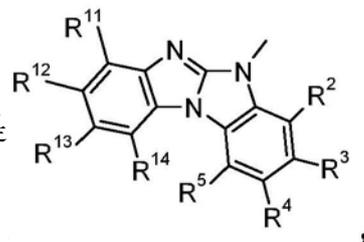
[0134] R⁶尤其为式

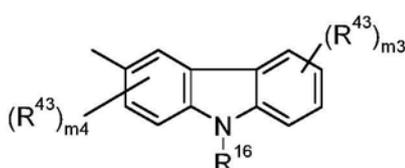
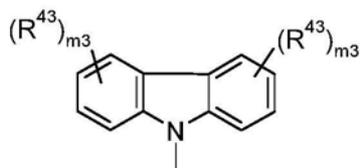


[0135] 在本发明的一个优选实施方案中,R⁶为式



且X⁷ 为-N<或X⁷ 为=N-且X⁶ 为-N<),尤其是





的基团或基团-

(SiR²⁰R²¹R²²),

[0136] 其中R²、R³、R⁴、R⁵、R¹¹、R¹²、R¹³和R¹⁴相互独立地为H,可任选被E取代和/或被D间隔的C₁-C₂₅烷基;可以任选被G取代的C₆-C₂₄芳基,或可以任选被G取代的C₂-C₃₀杂芳基,

[0137] R¹⁶为C₆-C₁₈芳基;或被一个或多个C₁-C₁₈烷基取代的C₆-C₁₈芳基。R²⁰、R²¹和R²²相互独立地为C₆-C₁₈芳基;或被一个或多个C₁-C₁₈烷基取代的C₆-C₁₈芳基,

[0138] R⁴³在每次出现时可以相同或不同且为F, C₁-C₁₈烷基,被E取代和/或被D间隔的C₁-C₁₈烷基, C₆-C₂₄芳基,被G取代的C₆-C₂₄芳基, C₂-C₂₀杂芳基或被G取代的C₂-C₂₀杂芳基,

[0139] m3为0或1-4的整数, m4为0或1-3的整数, 和

[0140] E、D和G如上文或下文所定义。

[0141] D优选为-CO-、-COO-、-S-、-SO-、-SO₂-、-O-、-NR⁶⁵-, 其中R⁶⁵为C₁-C₁₈烷基, 例如甲基、乙基、正丙基、异丙基、正丁基、异丁基或仲丁基, 或C₆-C₁₄芳基, 例如苯基、甲苯基、萘基或联苯基。

[0142] E优选为-OR⁶⁹、-SR⁶⁹、-NR⁶⁵R⁶⁵、-COR⁶⁸、-COOR⁶⁷、-CONR⁶⁵R⁶⁵; 或-CN; 其中R⁶⁵、R⁶⁷、R⁶⁸和R⁶⁹相互独立地为C₁-C₁₈烷基, 例如甲基、乙基、正丙基、异丙基、正丁基、异丁基、仲丁基、己基、辛基或2-乙基己基, 或C₆-C₁₄芳基, 例如苯基、甲苯基、萘基或联苯基。

[0143] G具有与E相同的优选情形或为C₁-C₁₈烷基, 例如甲基、乙基、正丙基、异丙基、正丁基、异丁基、仲丁基、己基、辛基或2-乙基己基或为C₁-C₁₈全氟烷基如-CF₃。

[0144] R²、R³、R⁴、R⁵、R¹¹、R¹²、R¹³和R¹⁴优选为H。

[0145] R⁴³优选为H, 或C₆-C₁₄芳基, 例如苯基、甲苯基、萘基或联苯基, 其可以任选被取代。

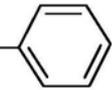
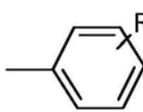
[0146] m3优选为0或1, 最优选0。m4优选为0或1, 最优选0。

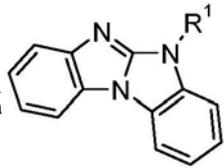
[0147] M2优选为0或1, 最优选0。m5优选为0或1, 最优选0。

[0148] R²⁰、R²¹和R²²优选为式 或 的基团, 其中R¹⁰⁰为C₁-C₈烷基。

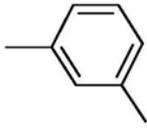
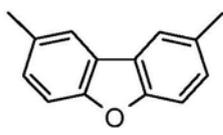
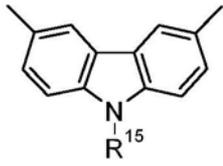
[0149] 更优选地, R⁶为式 、、 或

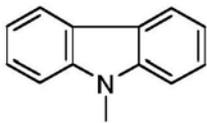
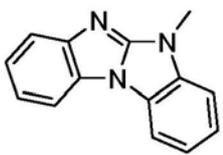
的基团, 其中R¹⁶为C₆-C₁₈芳基; 或被一个或多个C₁-C₁₈烷基取代的C₆-C₁₈芳基。

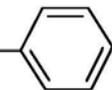
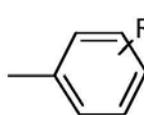
[0150] R¹⁶优选为式  或  的基团,其中R¹⁰⁰为C₁-C₈烷基。

[0151] 在一个优选实施方案中,本发明涉及式  的化合物,其中R¹为 **(II')**

式-A¹- (A²)_p- (A³)_q- (A⁴)_r-R⁶的基团,A¹、A²、A³和A⁴相互独立地为式

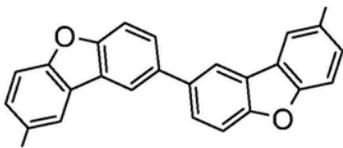
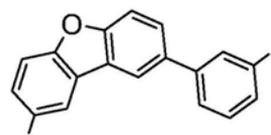
、 或  的基团,

[0152] R⁶为式  或  的基团,和

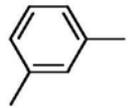
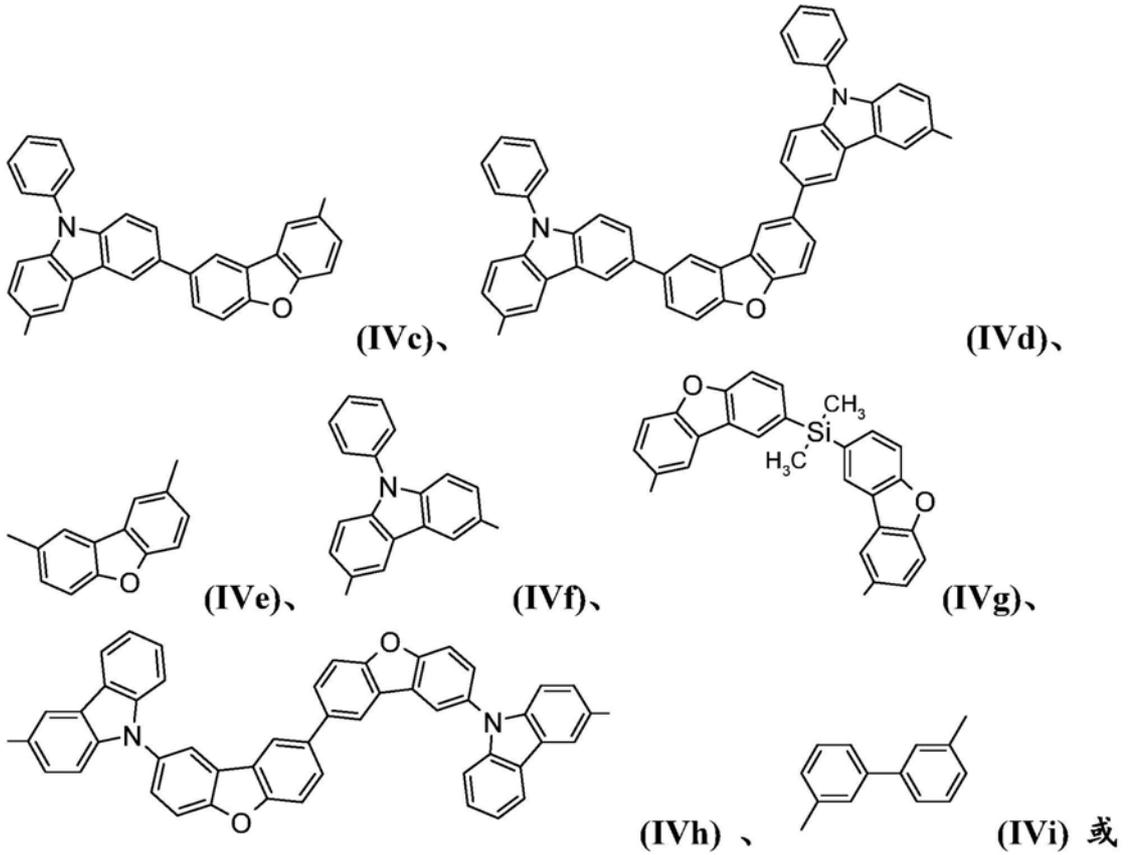
[0153] R¹⁵为式  或  的基团,其中R¹⁰⁰为C₁-C₈烷基,且p、q和r如上

所定义。

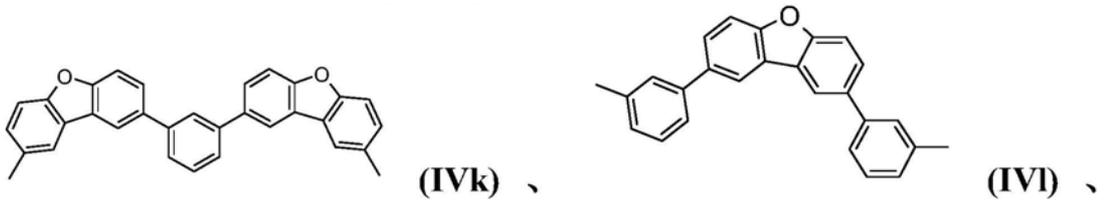
[0154] 在所述实施方案中,式-A¹- (A²)_p- (A³)_q- (A⁴)_r-的基团尤其为下式的基团:

[0155]  **(IVa)**、 **(IVb)**、

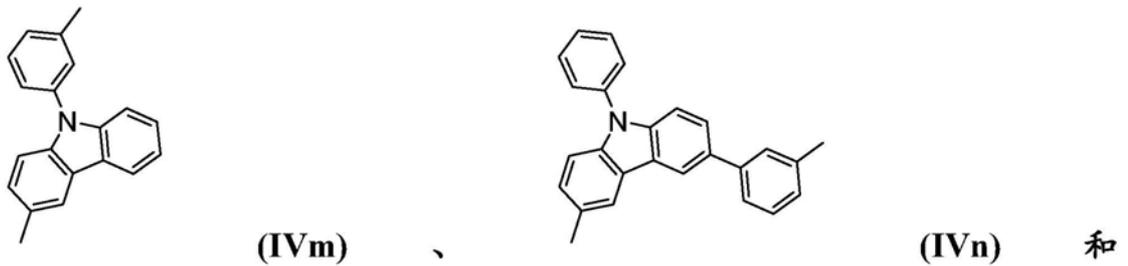
[0156]



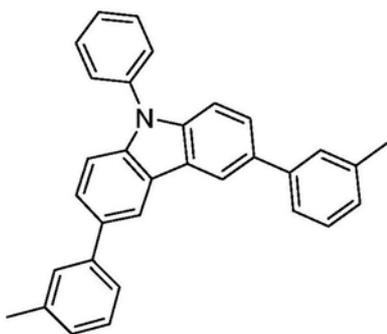
式 $-A^1-(A^2)_p-(A^3)_q-(A^4)_r-$ 的基团的额外实例为下式的基团：



[0157]



[0158]

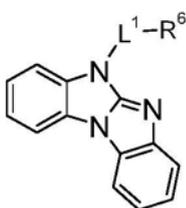


(IVo)。

[0159] 目前最优式 $-A^1-$ 、 $(A^2)_p-$ 、 $(A^3)_q-$ 、 $(A^4)_r-$ 的基团为式 (IVa)、(IVb)、(IVe)、(IVl)、(IVk)、(IVs)、(IVv) 和 (VIj) 的基团。

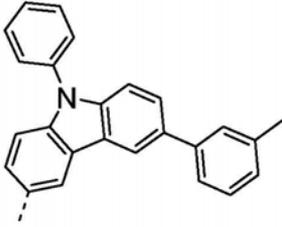
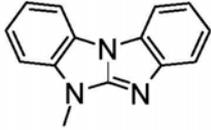
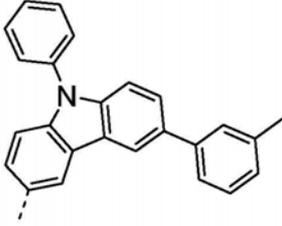
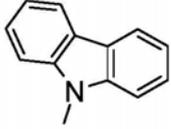
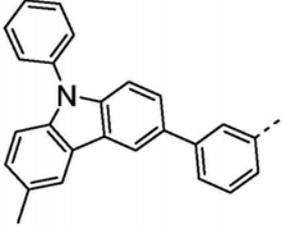
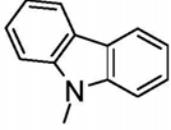
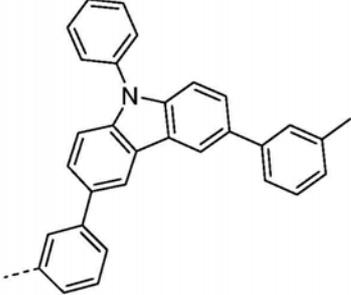
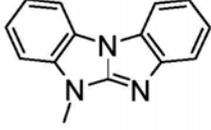
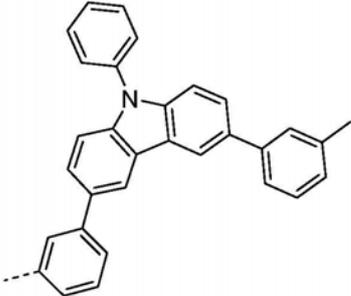
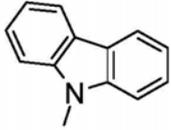
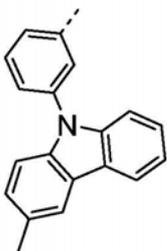
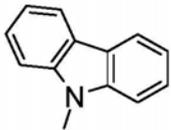
[0160] 优选的化合物的实例为化合物 A-1 至 A-20, 尤其是显示在权利要求 8 中的 A-1 至 A-19, 和显示在下表中的化合物 A-21 至 A-32:

[0161]

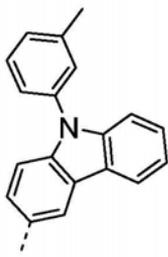
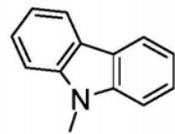
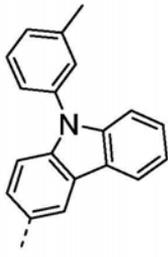
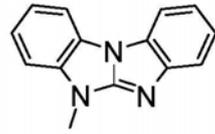


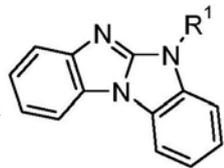
[0162]

Cpd.	L ¹²⁾	R ⁶⁾
A-21		
A-22		
A-23		
A-24		

A-25		
A-26		
A-27		
A-28		
A-29		
A-30		

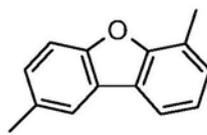
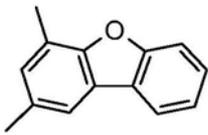
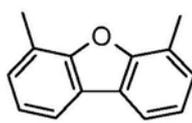
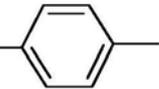
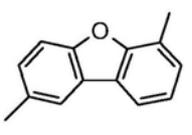
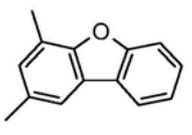
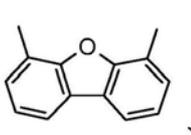
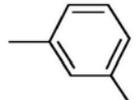
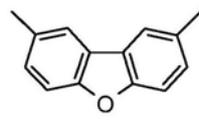
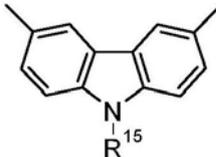
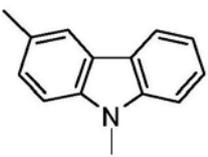
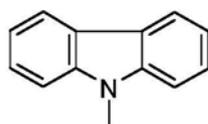
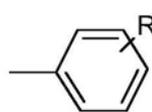
[0163]

<p>[0164]</p>	<p>A-31</p> 	
<p>[0164]</p>	<p>A-32</p> 	

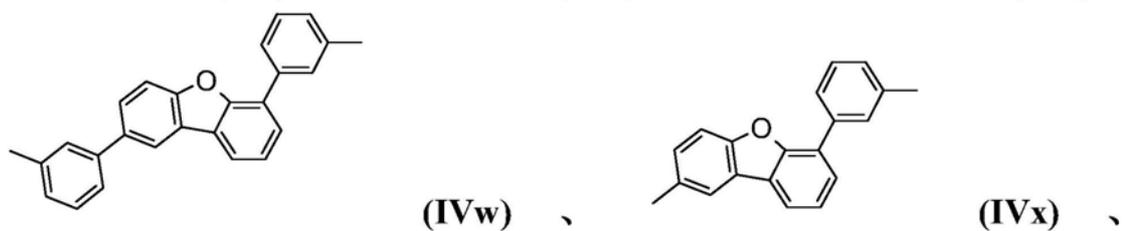
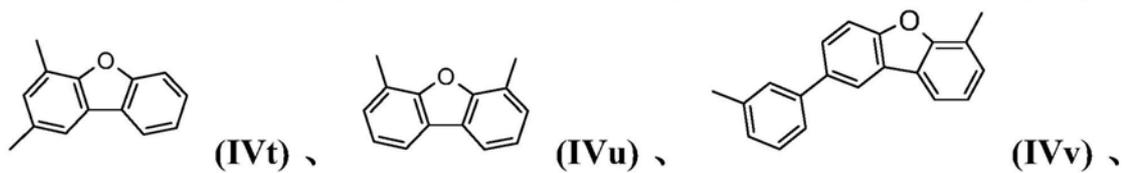
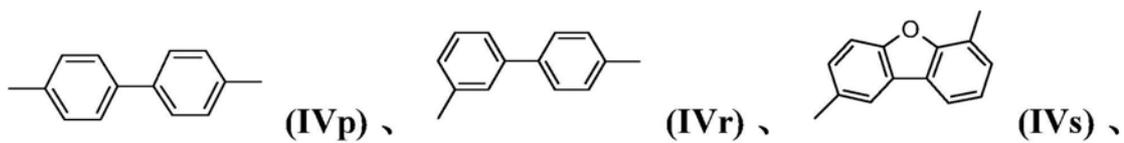
[0165] 在另一优选实施方案中,本发明涉及式  的化合物,其中R¹为

(II')

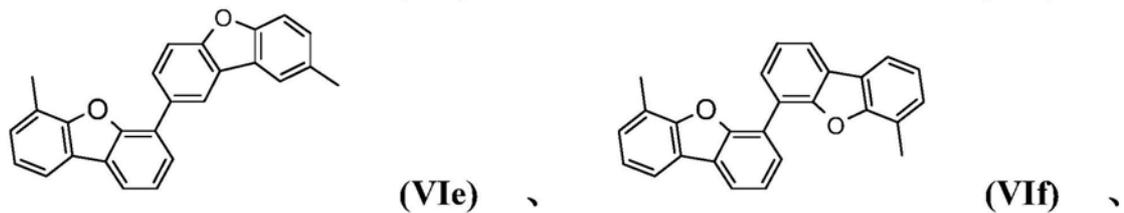
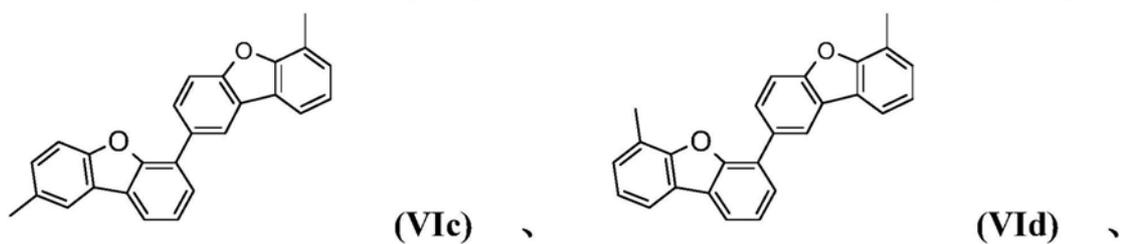
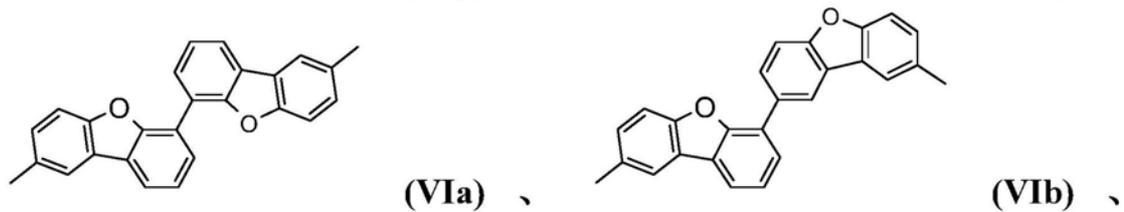
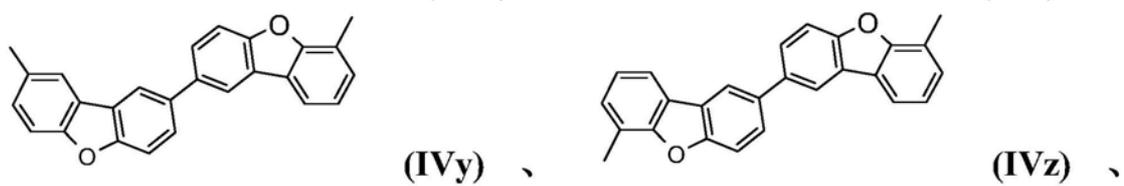
式-A¹-(A²)_p-(A³)_q-(A⁴)_r-R⁶的基团,

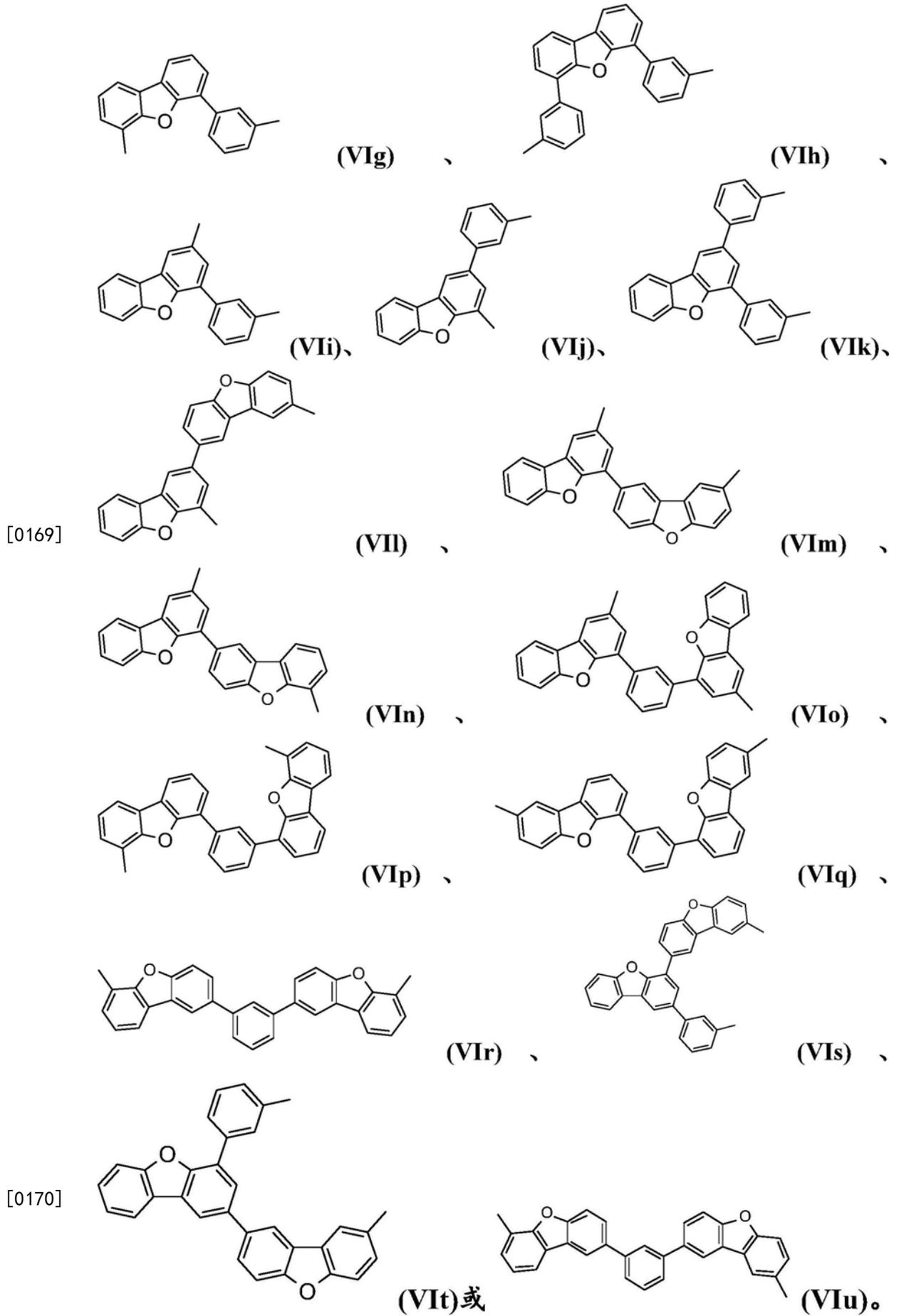
[0166] 其中基团A¹、A²、A³和A⁴中至少一个为式 、、、 的基团;且其他相互独立地为式 、、、、、、、 的基团,R⁶为式  或  的基团,和R¹⁵为式  或  的基团,其中R¹⁰⁰为C₁-C₈烷基,且p、q和r如上所定义。

[0167] 在所述实施方案中,式-A¹-(A²)_p-(A³)_q-(A⁴)_r-的基团尤其为下式的基团:

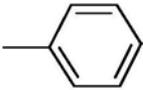


[0168]



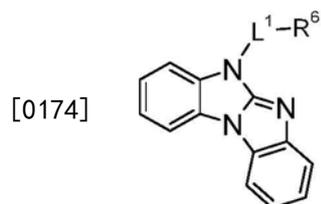


[0171] 目前最优选式 $-A^1-$ 、 $(A^2)_p-$ 、 $(A^3)_q-$ 、 $(A^4)_r-$ 的基团为式 (IVa)、(IVb)、(IVe)、(IVl)、(IVk)、(IVs)、(IVv) 和 (VI j) 的基团。

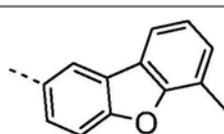
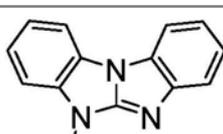
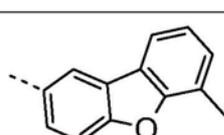
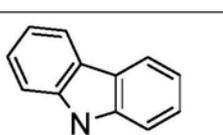
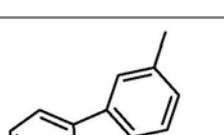
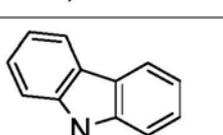
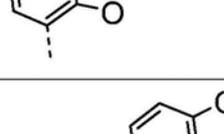
[0172] 在将式 I 化合物在蓝色或绿色磷光发光体中用作主体材料或用作电子/激子阻断材料的情况下, 较不优选如下基团 A^1- 、 $(A^2)_p-$ 、 $(A^3)_q-$ 、 $(A^4)_r-$, 其含有式  的基团,

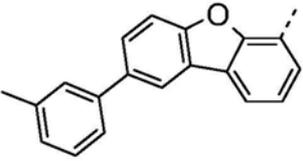
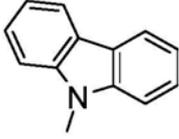
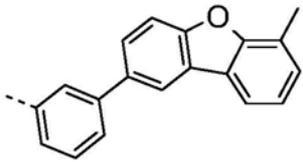
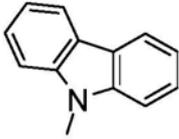
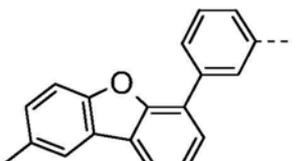
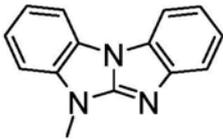
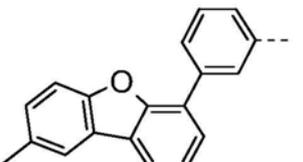
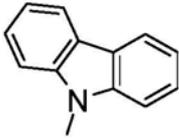
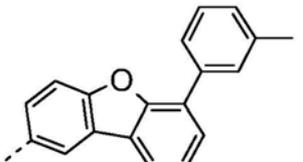
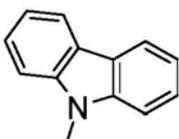
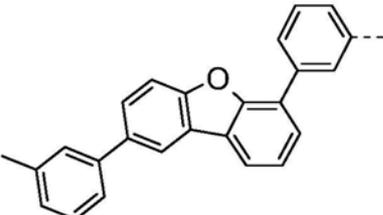
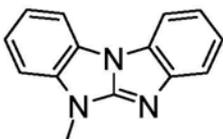
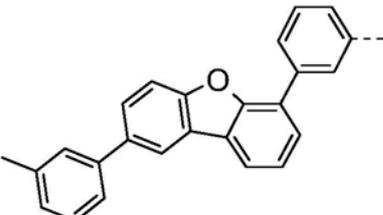
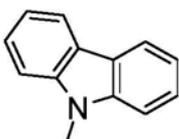
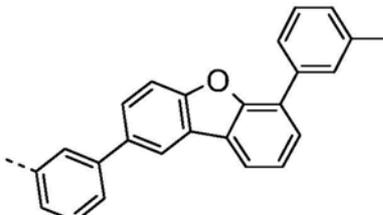
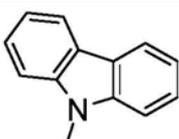
即式 (IVp) 和 (IVr) 的基团。

[0173] 优选的化合物的实例为显示在下表中的化合物 C-1 至 C-78。

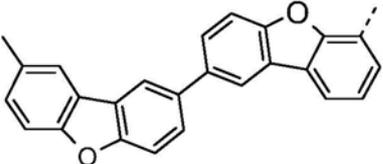
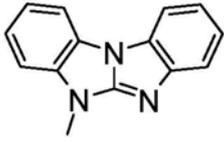
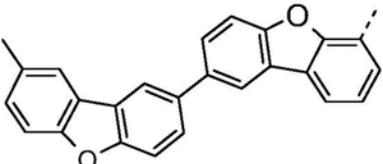
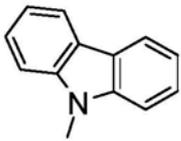
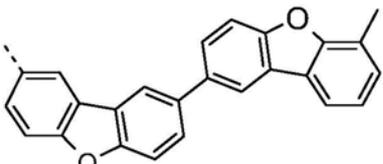
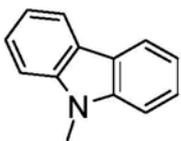
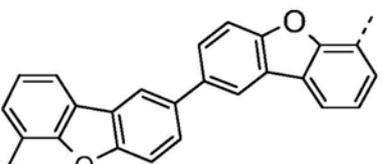
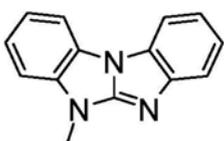
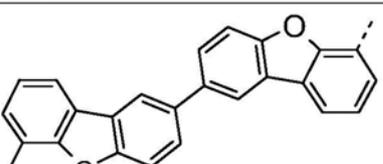
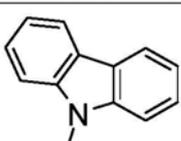
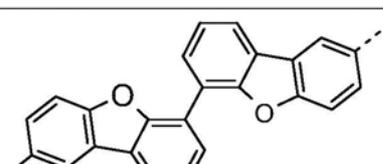
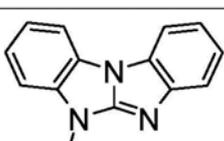
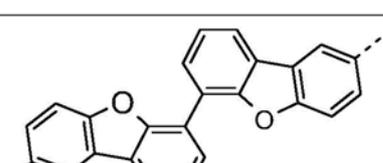
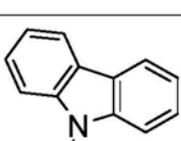
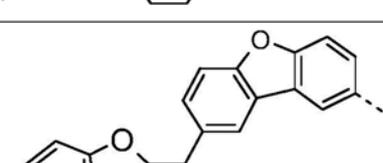
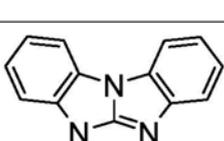
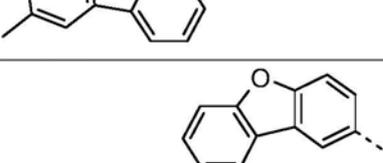
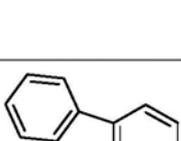


[0175]

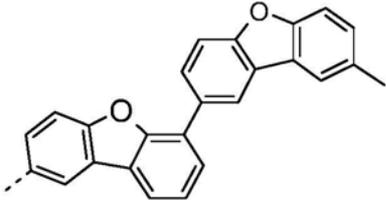
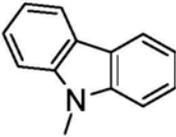
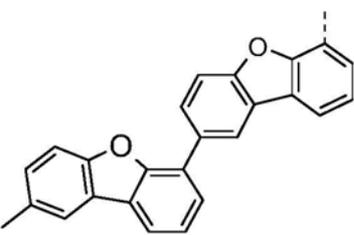
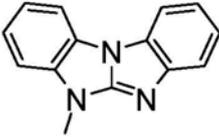
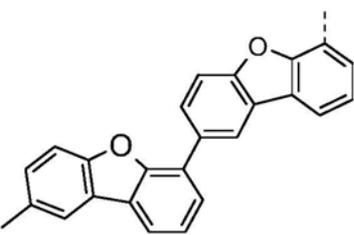
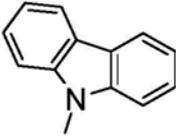
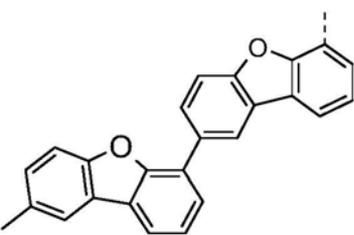
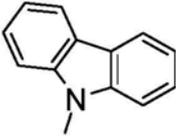
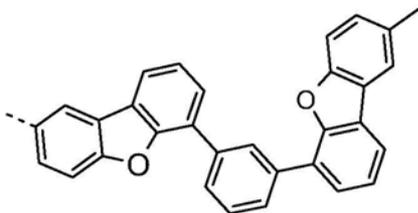
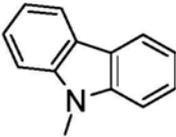
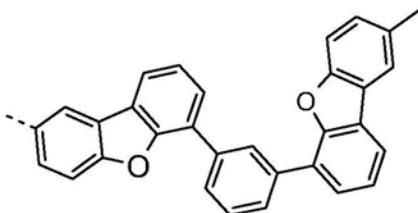
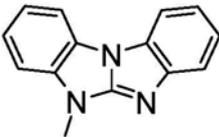
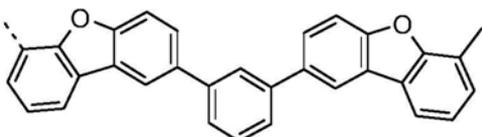
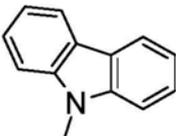
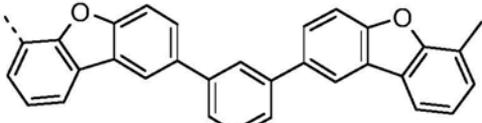
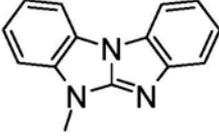
Cpd.	L ¹²⁾	R ⁶⁾
C-1		
C-2		
C-3		
C-4		

C-5		
C-6		
C-7		
C-8		
C-9		
C-10		
C-11		
C-12		

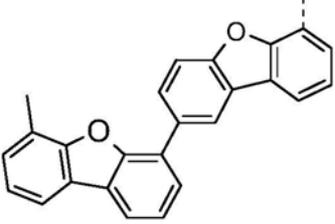
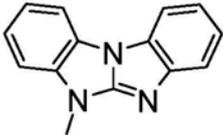
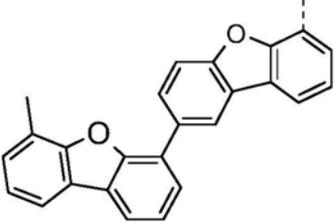
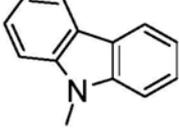
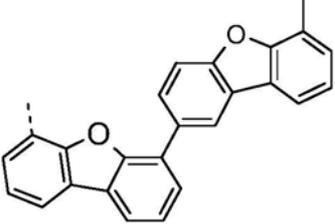
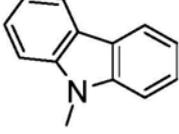
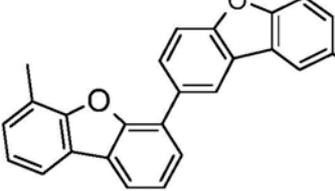
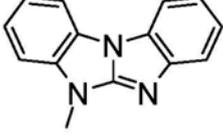
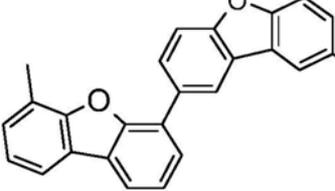
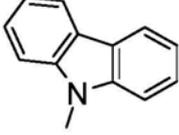
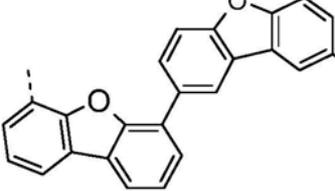
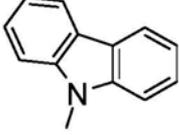
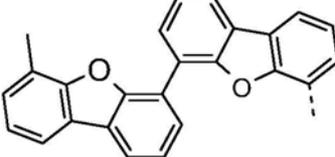
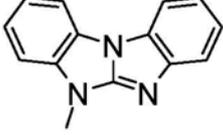
[0176]

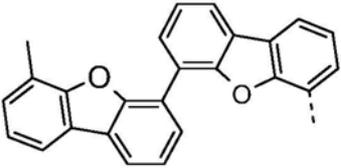
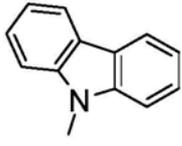
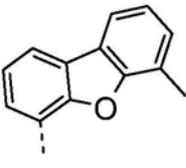
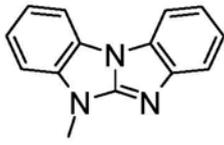
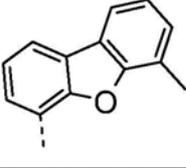
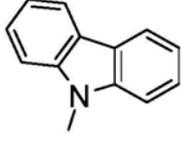
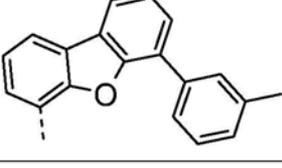
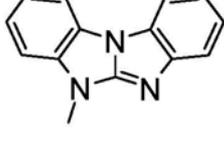
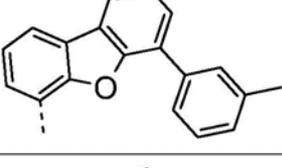
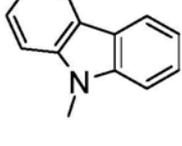
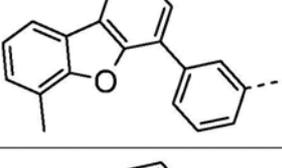
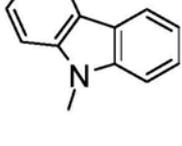
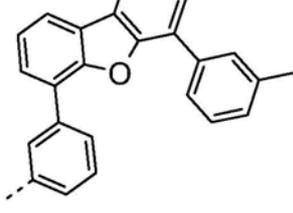
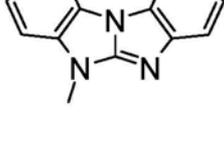
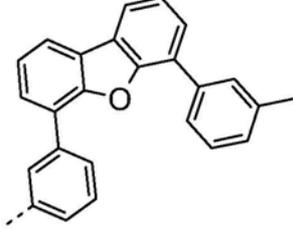
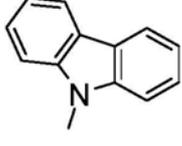
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[0177]

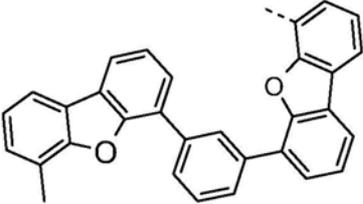
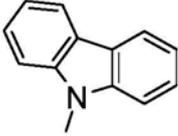
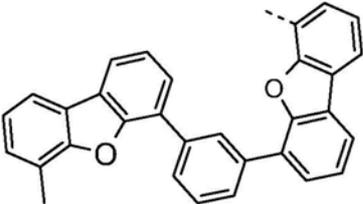
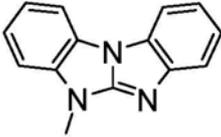
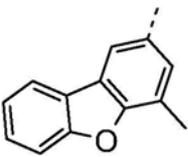
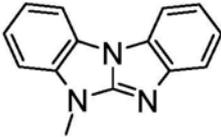
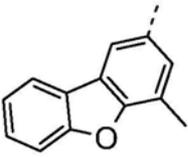
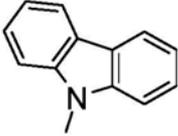
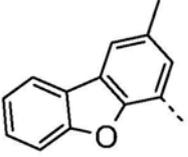
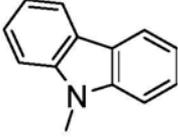
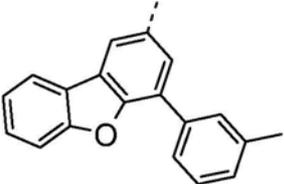
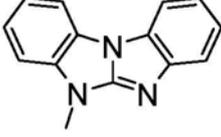
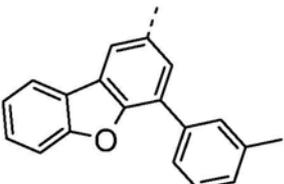
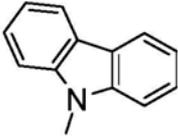
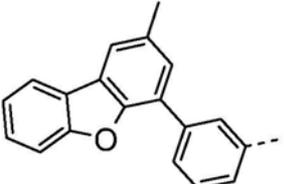
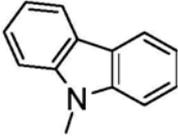
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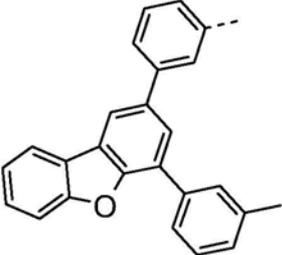
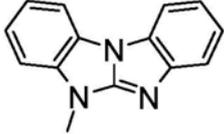
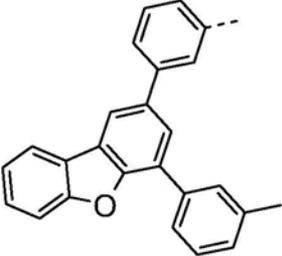
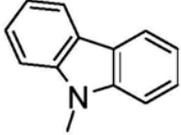
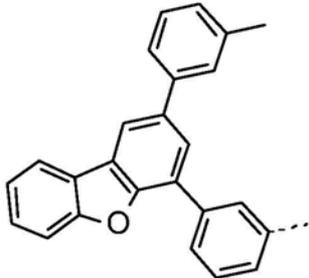
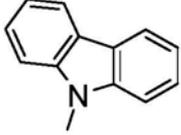
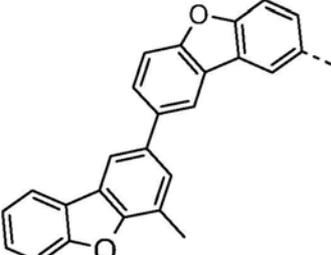
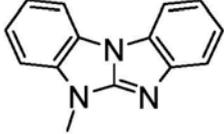
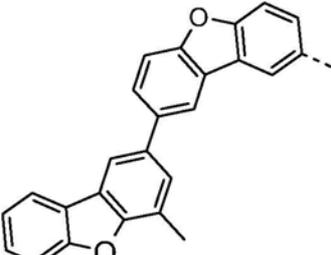
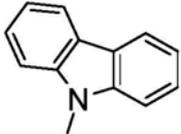
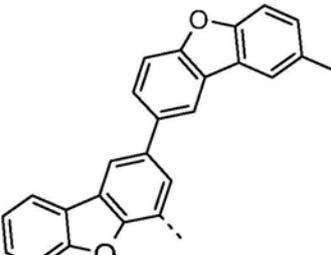
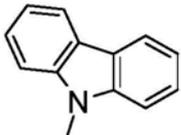
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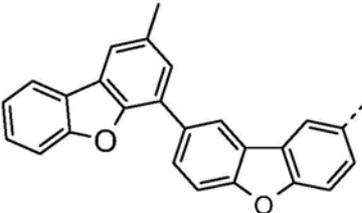
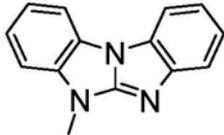
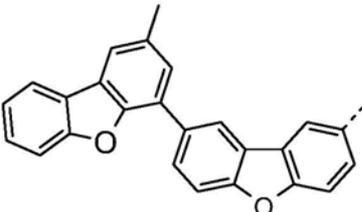
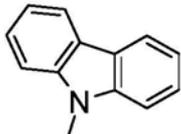
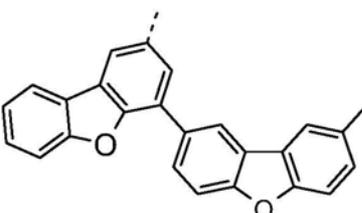
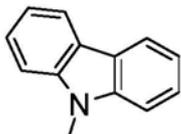
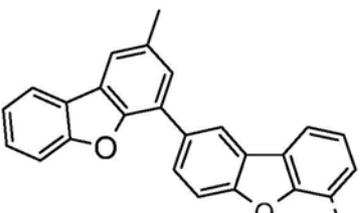
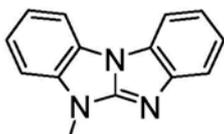
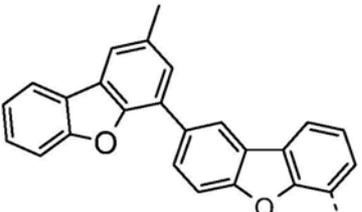
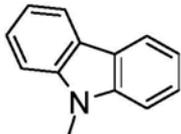
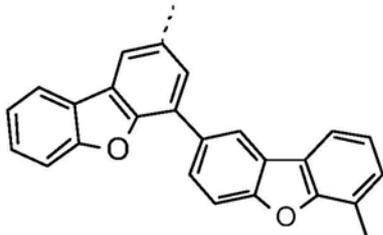
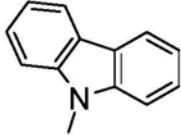
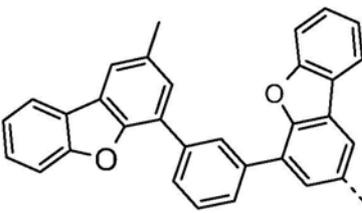
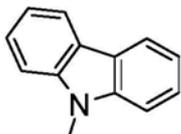
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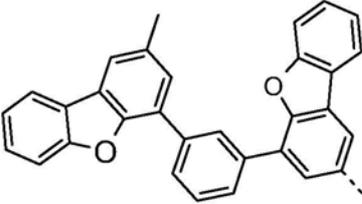
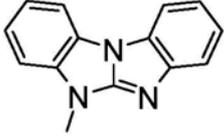
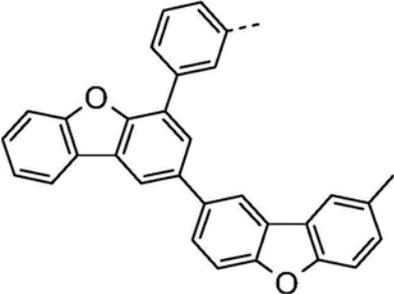
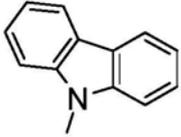
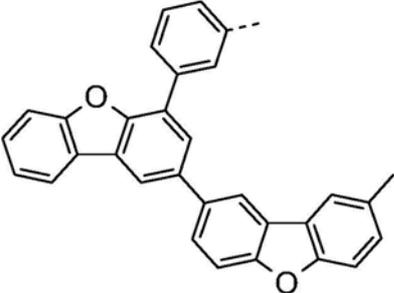
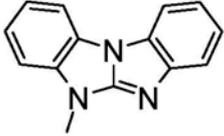
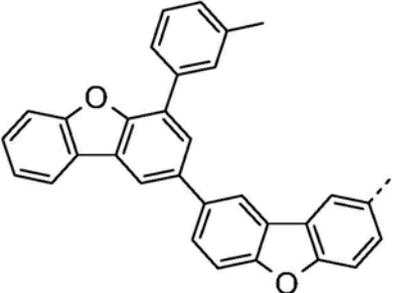
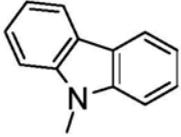
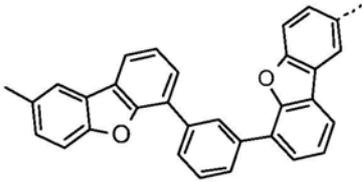
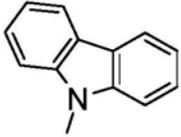
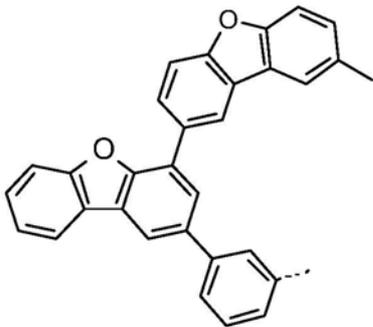
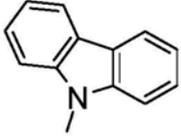
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[0182]

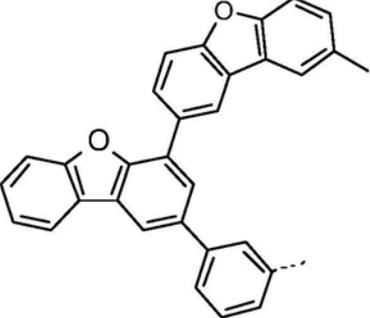
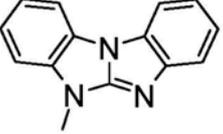
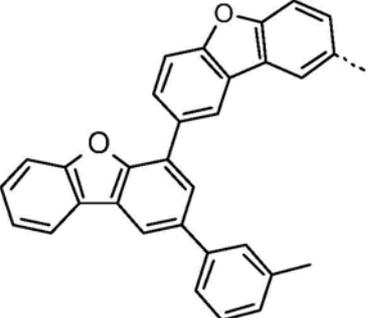
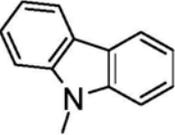
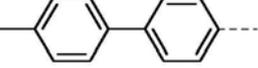
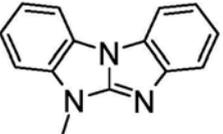
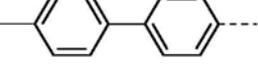
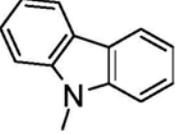
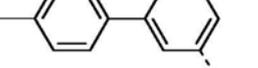
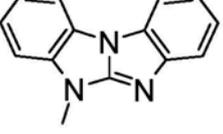
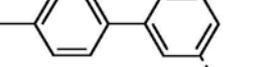
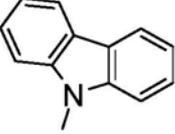
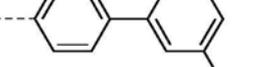
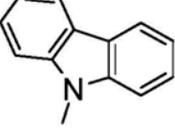
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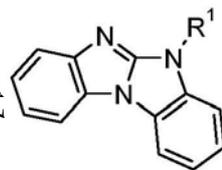
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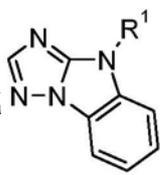
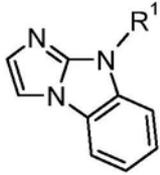
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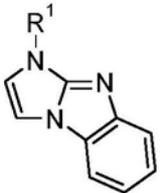
在另一优选实施方案中,本发明涉及式



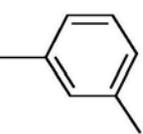
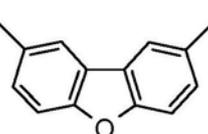
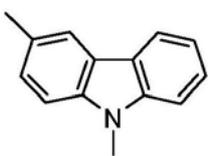
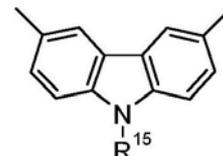
(II')

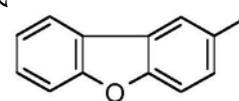
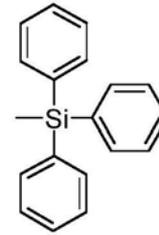
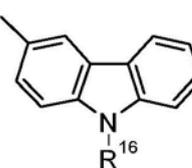
的化合物。在另一优

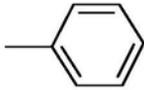
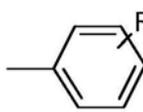
选实施方案中,本发明涉及式  (Ib')、 (Id')和

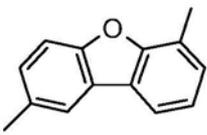
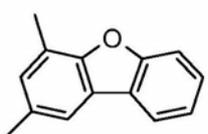
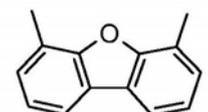
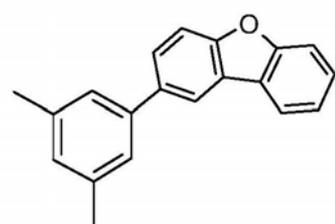
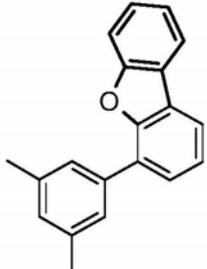
 (Id'') 的化合物。

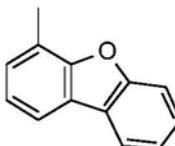
[0187] 在所述实施方案中, R¹为式 -A¹-(A²)_p-(A³)_q-(A⁴)_r-R⁶的基团,其中A¹、A²、A³和A⁴相

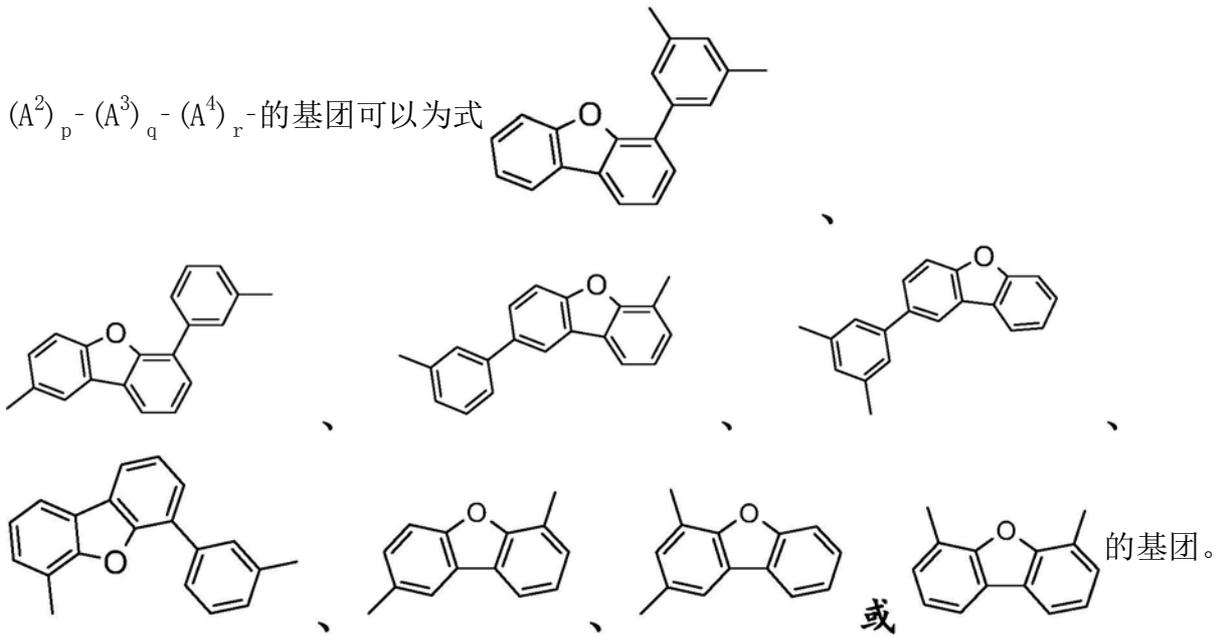
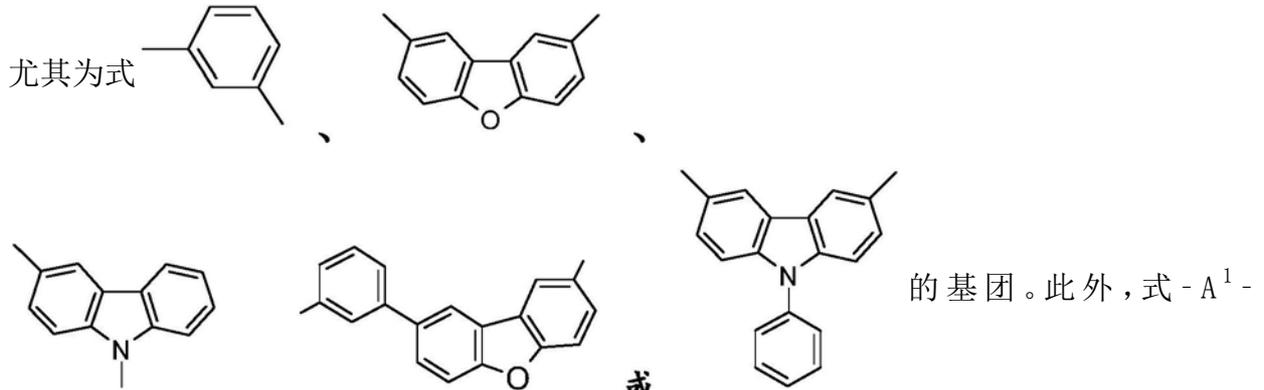
互独立地为式 、、 或  的基团,

[0188] R⁶为式 、 或  的基团;

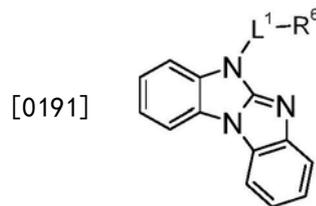
[0189] R¹⁵和R¹⁶为式  或  的基团,其中R¹⁰⁰为C₁-C₈烷基,且p、q和r

如上所定义。此外,A¹、A²、A³和A⁴可以为式 、、、 或  的基团。

此外,R⁶可为式  的基团。在所述实施方案中,式 -A¹-(A²)_p-(A³)_q-(A⁴)_r-的基团

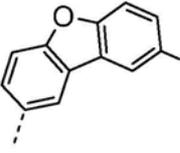
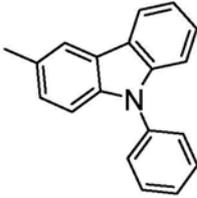
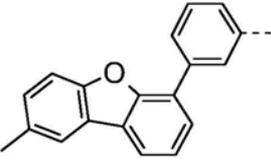
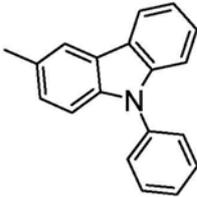
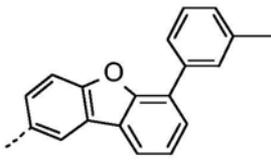
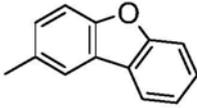
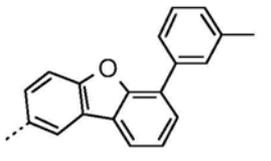
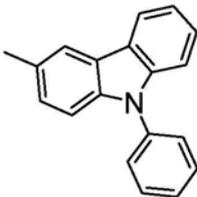
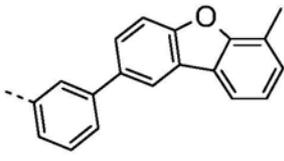
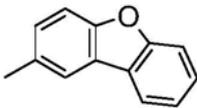
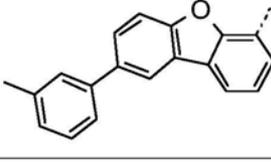
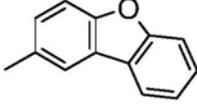
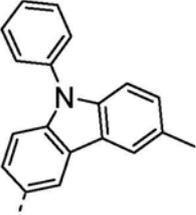
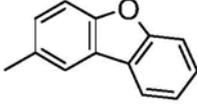
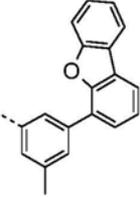
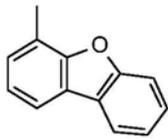


[0190] 优选的化合物的实例为化合物B-1至B-5,尤其是显示在权利要求9中的B-1至B-4,和显示在下表中的化合物B-6至B-35:

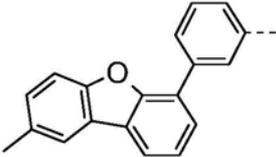
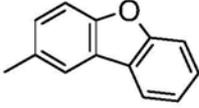
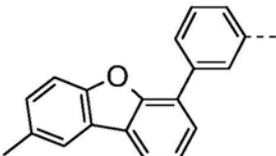
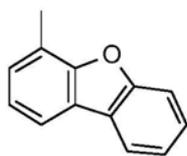
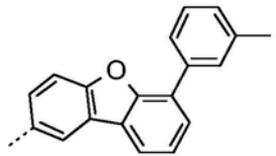
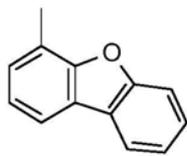
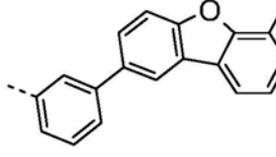
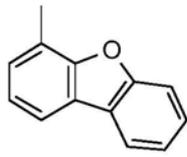
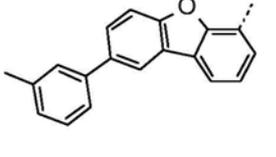
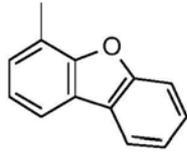
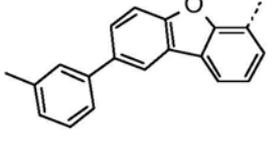
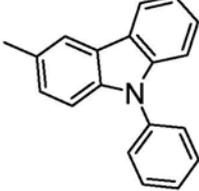
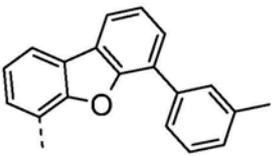
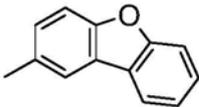
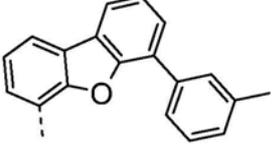
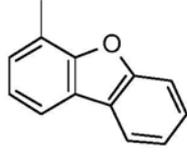
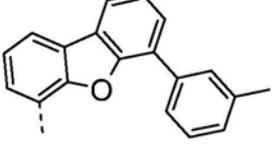
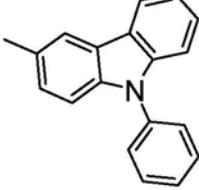


[0192]

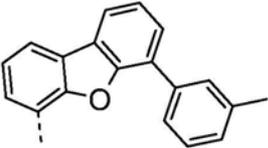
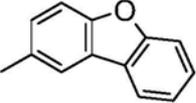
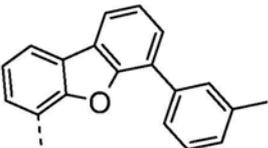
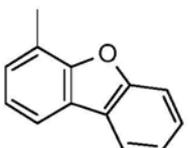
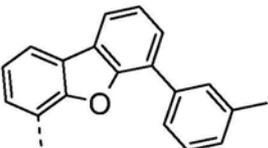
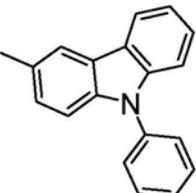
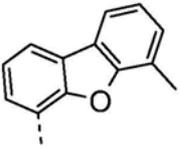
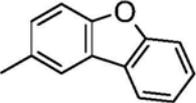
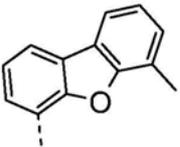
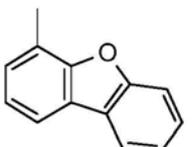
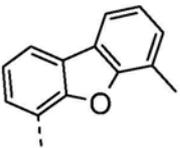
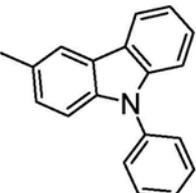
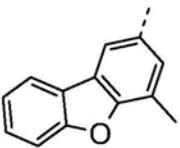
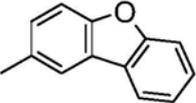
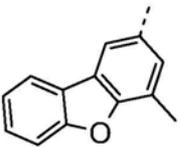
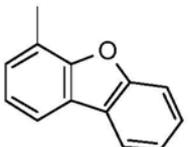
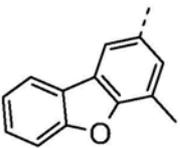
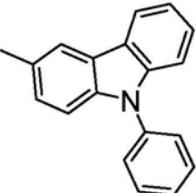
Cpd.	L ¹²⁾	R ⁶
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B-6		
B-7		
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B-9		
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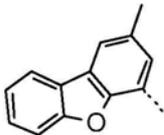
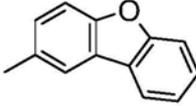
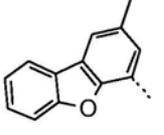
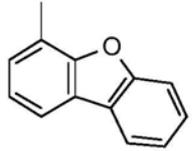
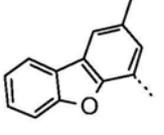
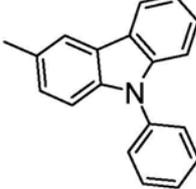
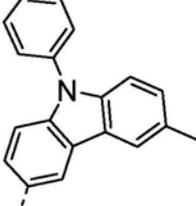
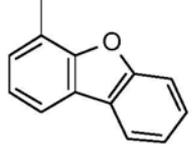
[0193]

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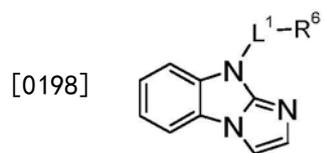
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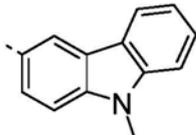
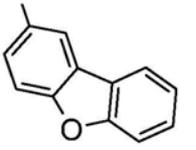
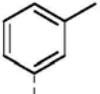
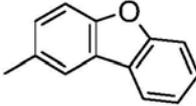
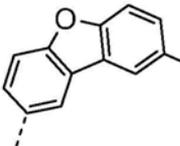
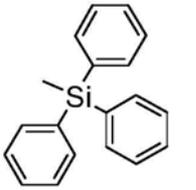
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[0195]

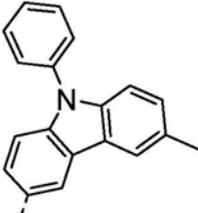
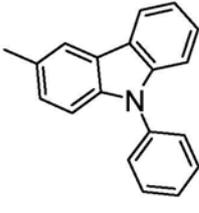
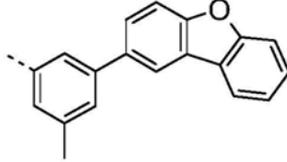
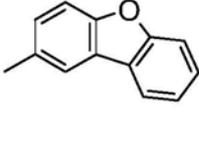
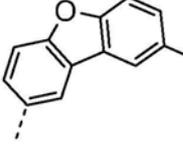
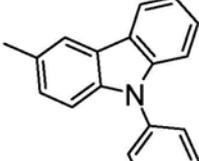
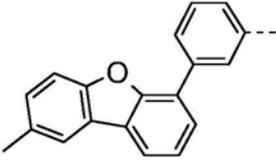
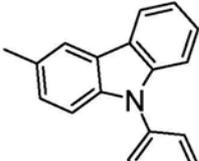
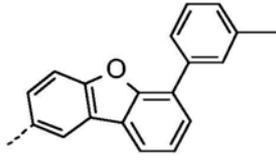
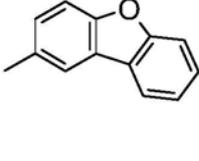
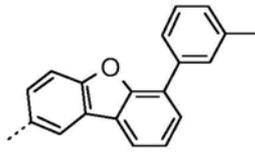
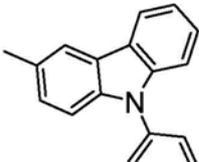
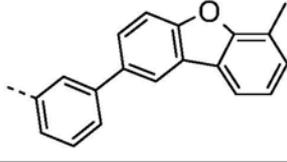
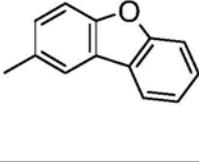
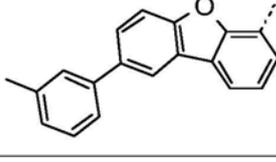
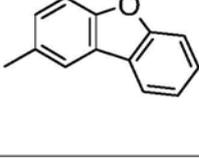
	B-32 	
	B-33 	
[0196]	B-34 	
	B-35 	

[0197] 优选化合物的额外实例为显示在下表中的化合物J-1至J-35、K-1至K-35和L-1至L-35:

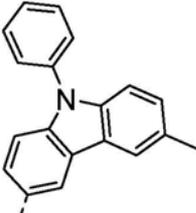
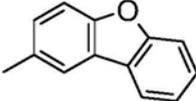
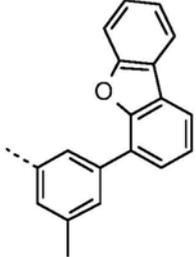
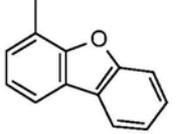
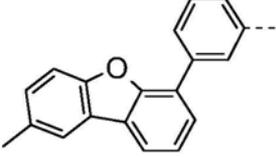
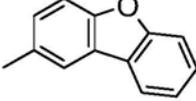
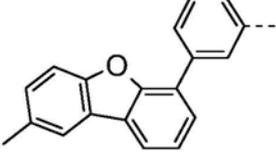
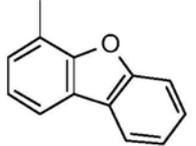
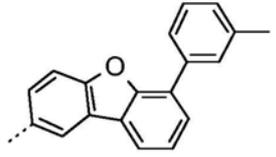
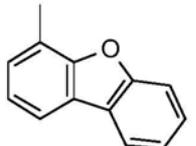
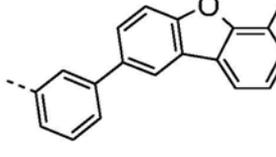
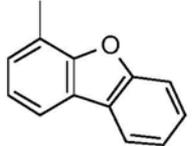
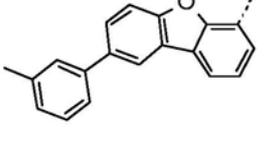
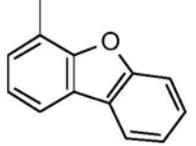
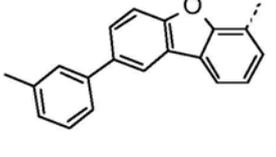
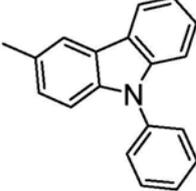


Cpd.	L ¹³⁾	R ⁶
[0199]	J-1 	
	J-2 	
	J-3 	

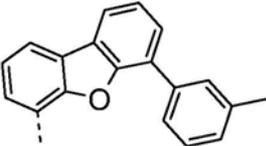
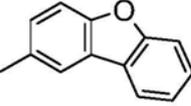
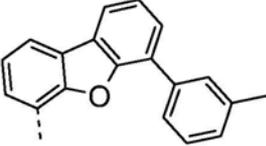
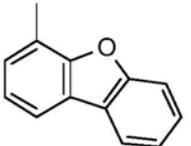
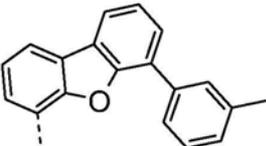
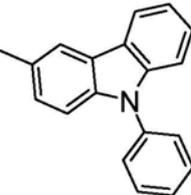
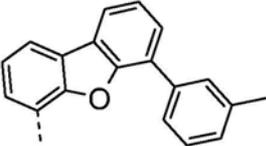
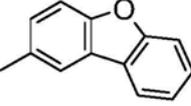
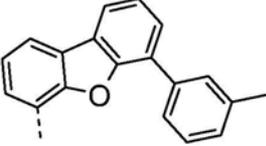
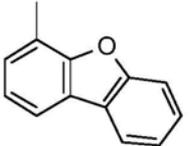
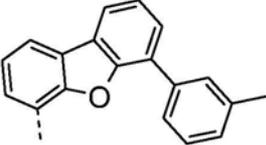
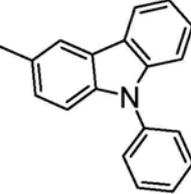
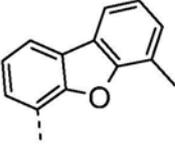
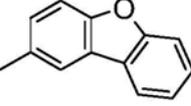
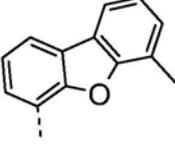
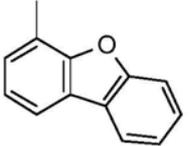
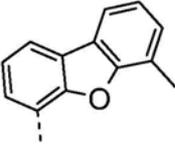
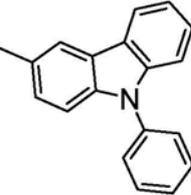
[0200]

J-4		
J-5		
J-6		
J-7		
J-8		
J-9		
J-10		
J-11		

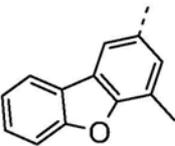
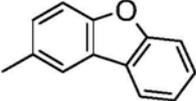
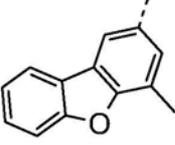
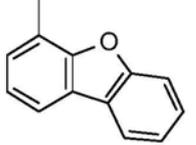
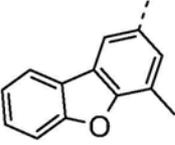
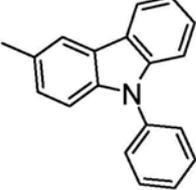
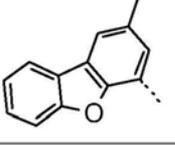
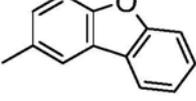
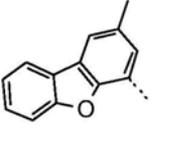
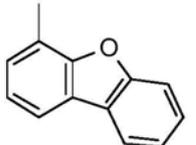
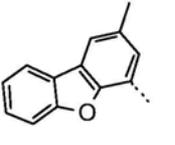
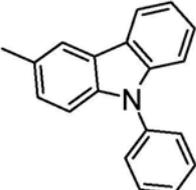
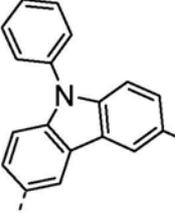
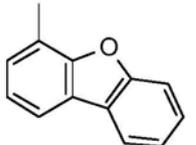
[0201]

<p>J-12</p>		
<p>J-13</p>		
<p>J-14</p>		
<p>J-15</p>		
<p>J-16</p>		
<p>J-17</p>		
<p>J-18</p>		
<p>J-19</p>		

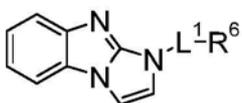
[0202]

J-20		
J-21		
J-22		
J-23		
J-24		
J-25		
J-26		
J-27		
J-28		

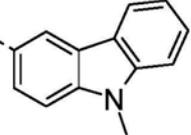
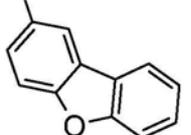
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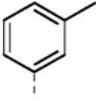
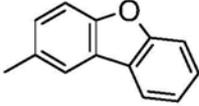
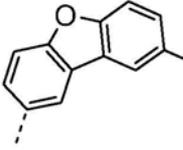
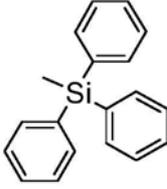
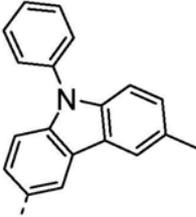
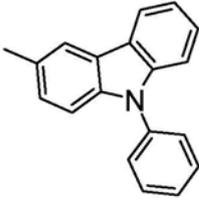
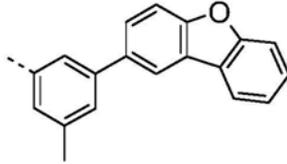
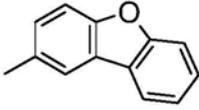
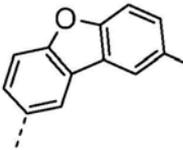
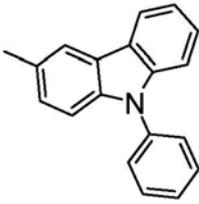
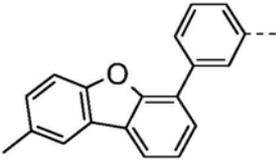
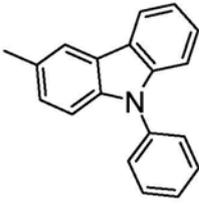
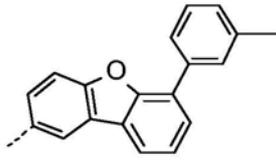
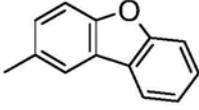
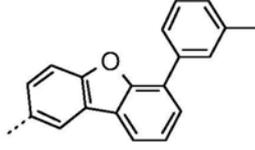
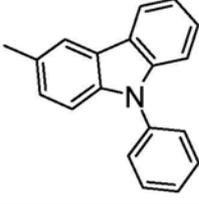
J-29		
J-30		
J-31		
J-32		
J-33		
J-34		
J-35		

[0204]

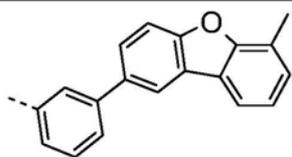
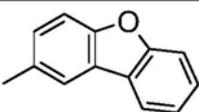
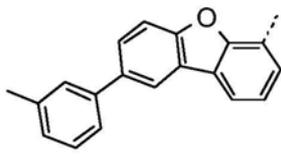
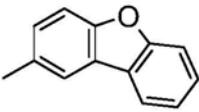
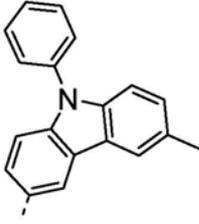
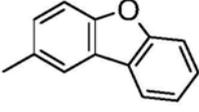
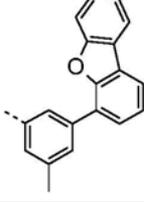
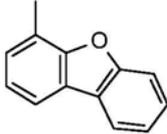
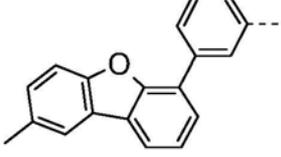
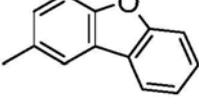
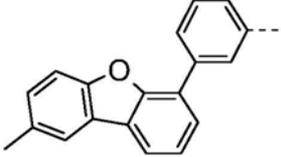
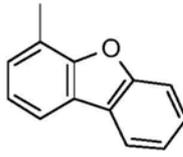
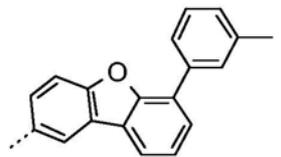
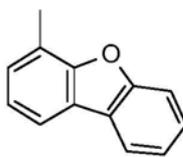
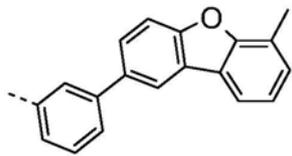
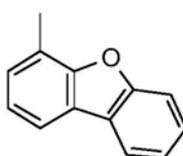
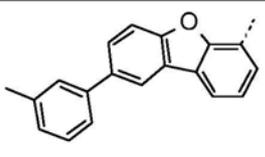
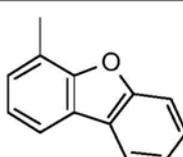


[0205]

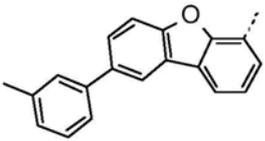
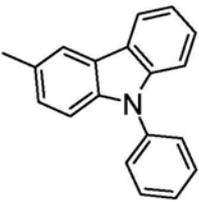
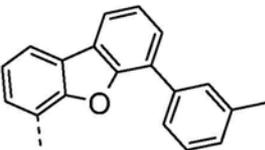
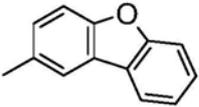
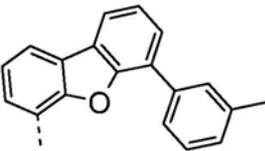
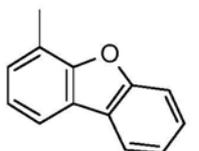
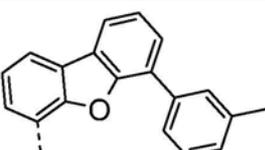
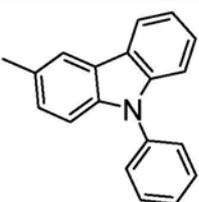
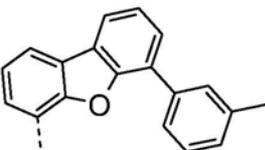
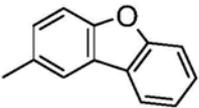
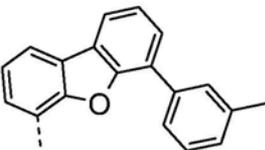
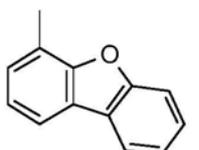
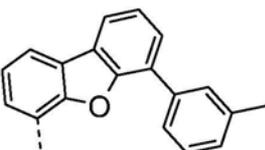
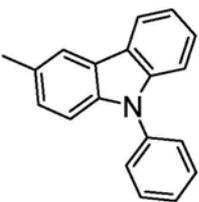
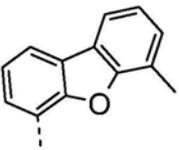
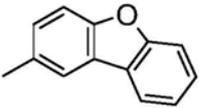
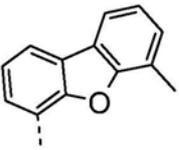
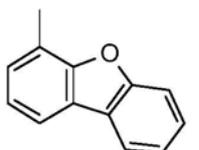
Cpd.	L¹³⁾	R⁶⁾
K-1		

K-2		
K-3		
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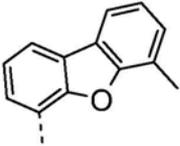
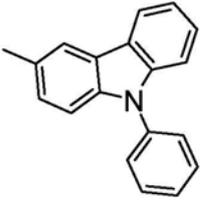
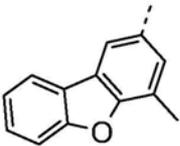
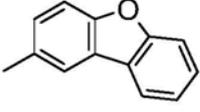
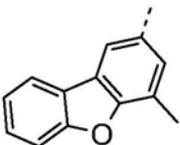
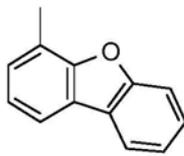
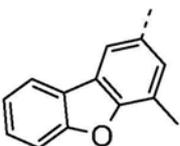
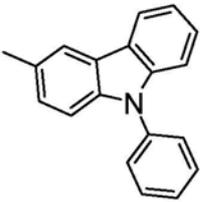
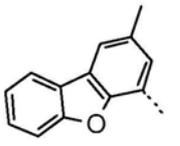
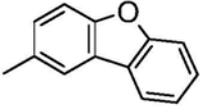
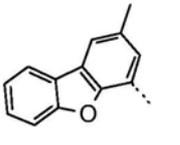
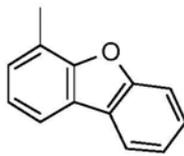
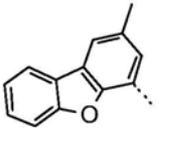
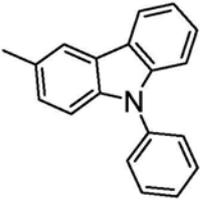
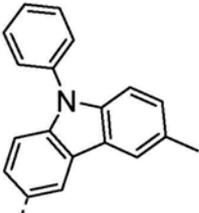
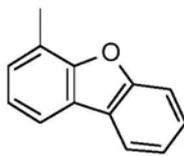
[0206]

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K-18		

[0207]

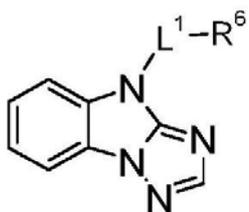
K-19		
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[0208]

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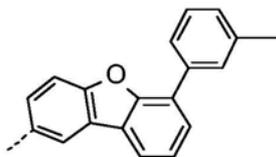
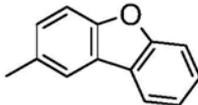
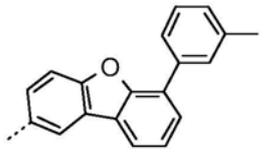
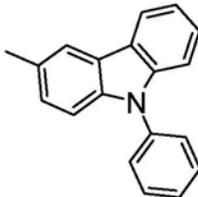
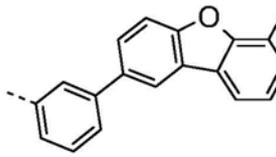
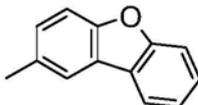
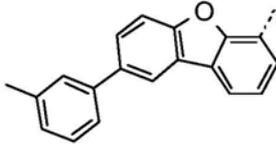
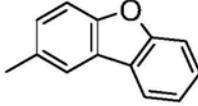
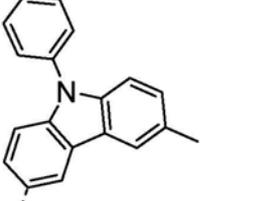
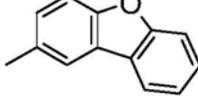
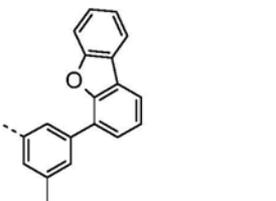
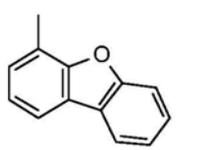
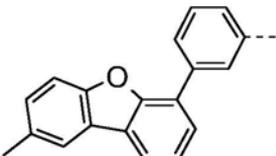
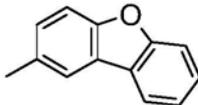
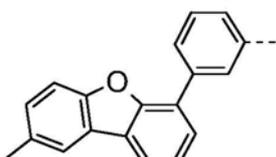
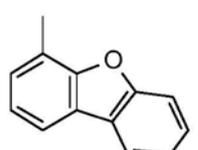
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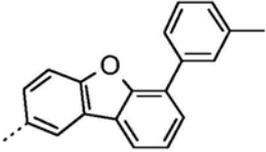
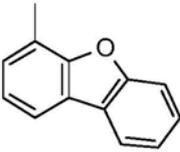
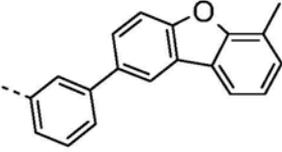
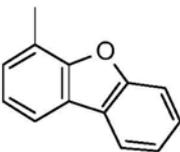
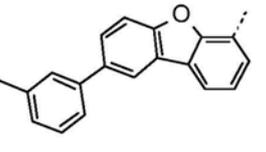
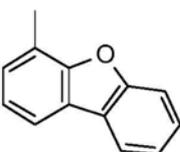
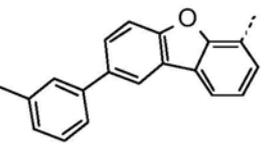
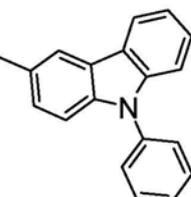
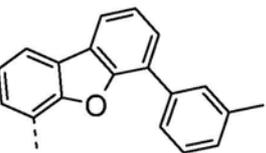
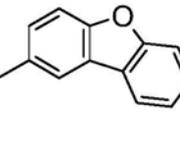
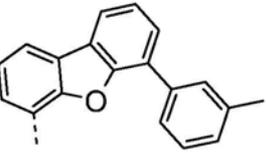
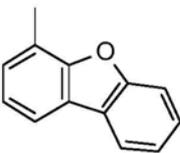
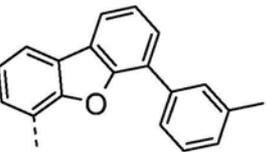
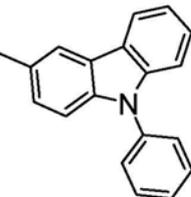
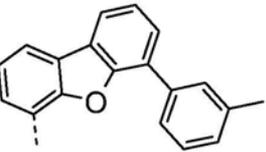
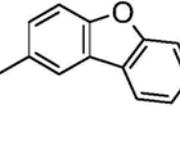
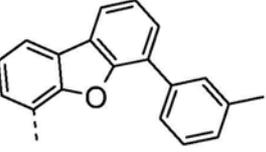
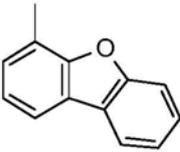
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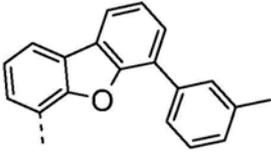
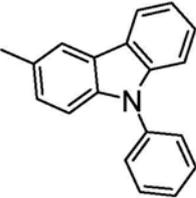
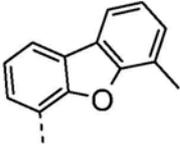
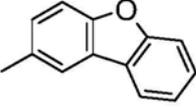
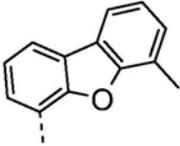
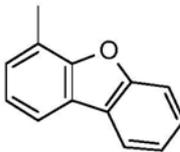
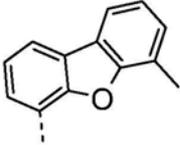
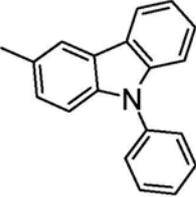
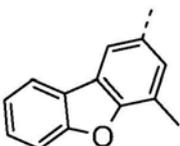
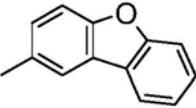
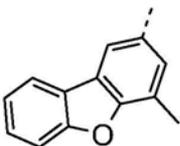
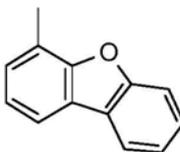
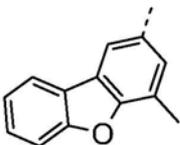
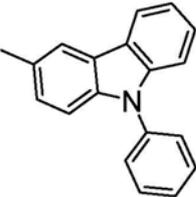
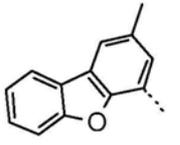
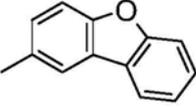
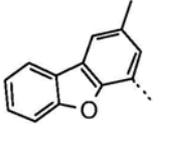
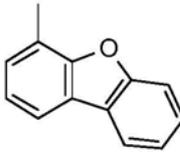
[0211]

Cpd.	L ¹⁴⁾	R ⁶
L-1		
L-2		
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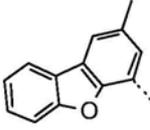
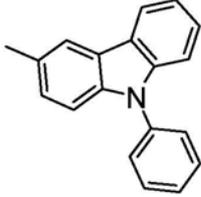
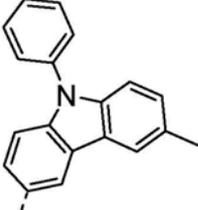
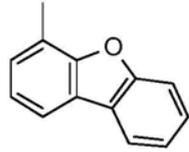
L-8		
L-9		
L-10		
L-11		
[0212] L-12		
L-13		
L-14		
L-15		

L-16		
L-17		
L-18		
L-19		
L-20		
L-21		
L-22		
L-23		
L-24		

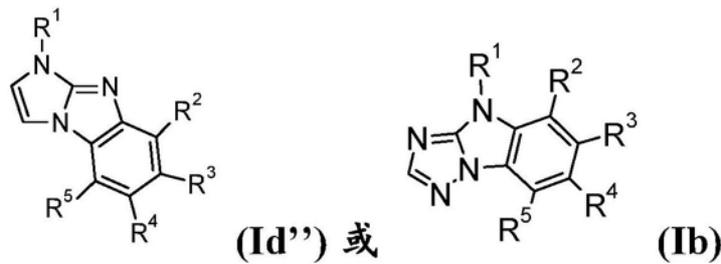
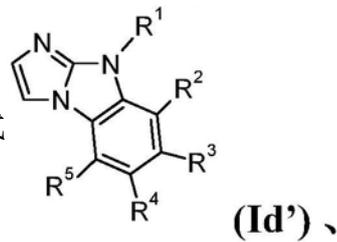
[0213]

L-25		
L-26		
L-27		
L-28		
L-29		
L-30		
L-31		
L-32		
L-33		

[0214]

<p>L-34</p>		
<p>[0215] L-35</p>		

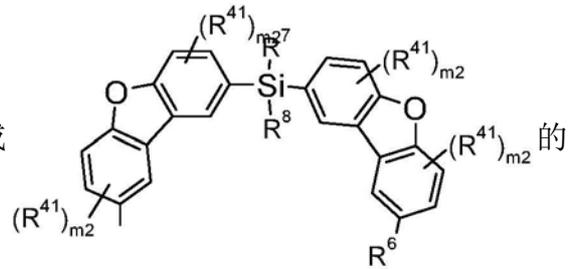
[0216] 在另一优选实施方案中,本发明涉及式



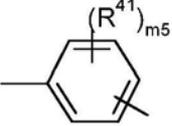
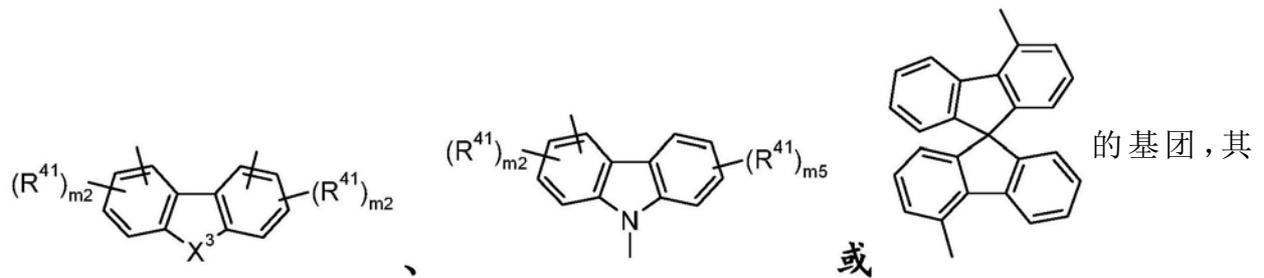
的化合物,其中R¹为式 -A¹ - (A²)_p -

(A³)_q - (A⁴)_r - R⁶的基团;和A¹、A²、A³、A⁴、R²、R³、R⁴、R⁵、p、q和r如上所定义。

[0217] R¹优选为式 -A¹ - (A²)_p - (A³)_q - (A⁴)_r - R⁶或



基团,其中A¹、A²、A³和A⁴相互独立地为式

中

[0218] m₅为0或1-4的整数,

[0219] m₂为0或整数1-3,

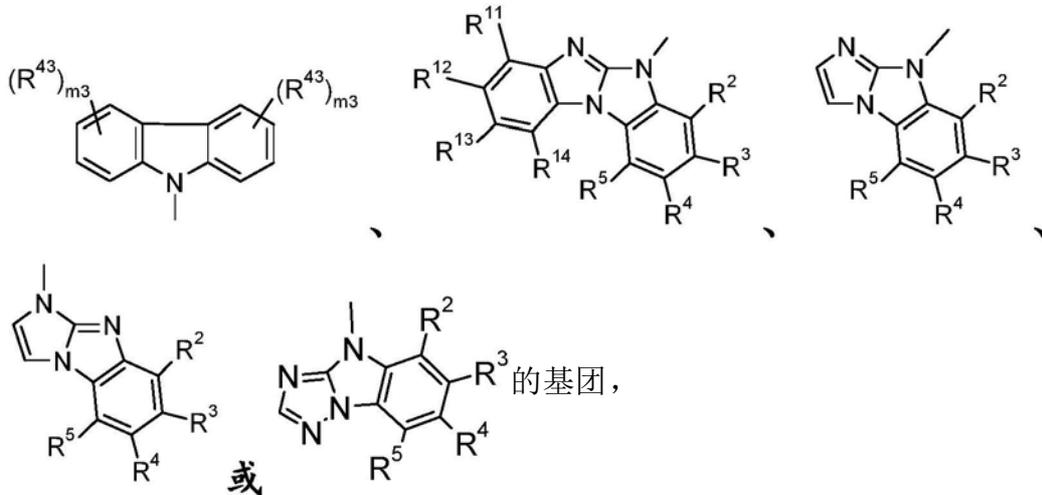
[0220] X³为-O-、-S-或-NR¹⁵-,

[0221] R⁷和R⁸为C₁-C₁₈烷基,

[0222] R¹⁵为C₁-C₁₈烷基;或被-O-间隔的C₁-C₁₈烷基;C₆-C₁₈芳基;被一个或多个C₁-C₁₈烷基或C₁-C₁₈烷氧基取代的C₆-C₁₈芳基;C₂-C₂₀杂芳基或被一个或多个C₁-C₁₈烷基取代的C₂-C₂₀杂芳基,和

[0223] R⁴¹在每次出现时可以相同或不同且为F, C₁-C₁₈烷基, 被E取代和/或被D间隔的C₁-C₁₈烷基, C₆-C₂₄芳基, 被G取代的C₆-C₂₄芳基, C₂-C₂₀杂芳基或被G取代的C₂-C₂₀杂芳基。

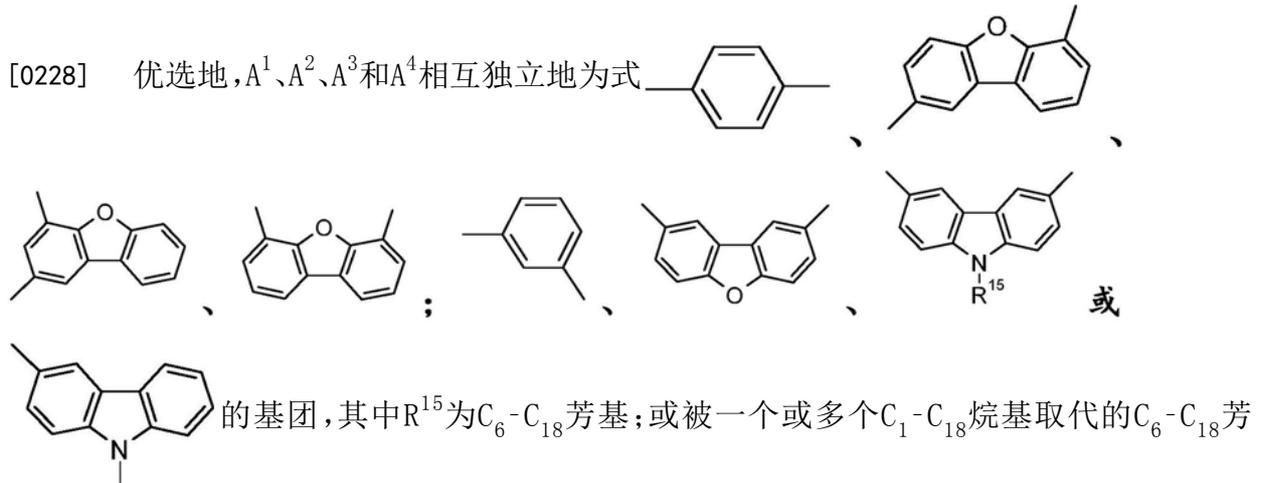
[0224] R⁶优选为式



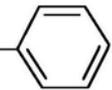
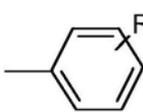
[0225] R⁴³在每次出现时可以相同或不同且为F, C₁-C₁₈烷基, 被E取代和/或被D间隔的C₁-C₁₈烷基, C₆-C₂₄芳基, 被G取代的C₆-C₂₄芳基, C₂-C₂₀杂芳基或被G取代的C₂-C₂₀杂芳基,

[0226] m₃为0或1-4的整数。E、D、G、R¹¹、R¹²、R¹³和R¹⁴如上所定义。

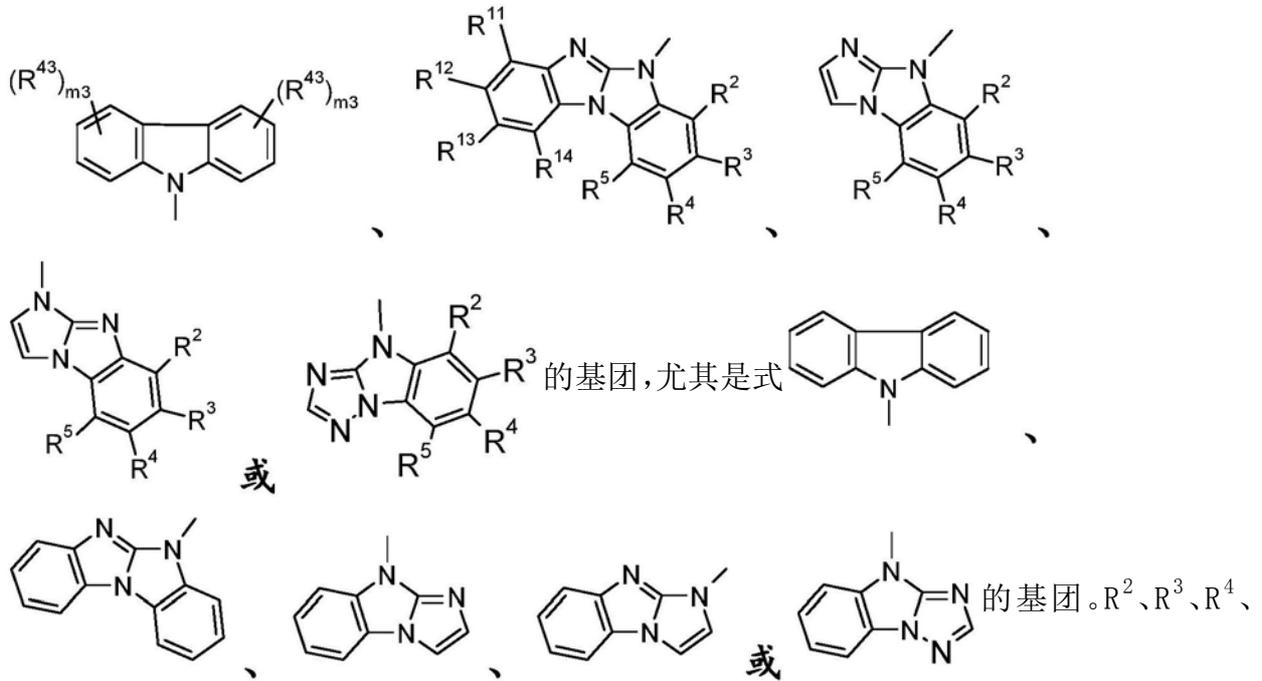
[0227] R²、R³、R⁴和R⁵优选为氢。



基。

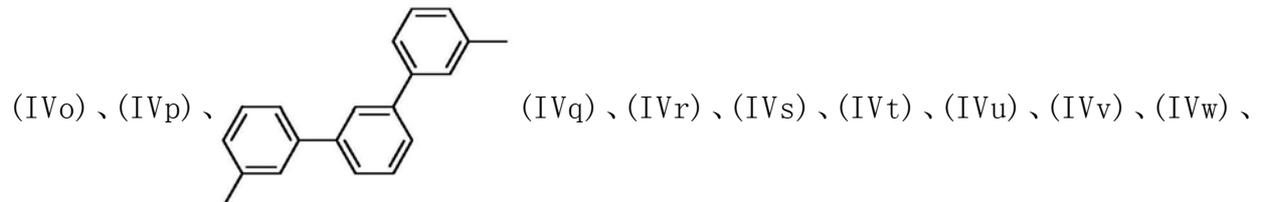
[0229] R^{15} 优选为式  或  的基团, 其中 R^{100} 为 C_1-C_8 烷基。

[0230] R^6 为式



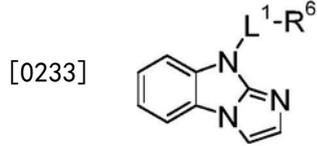
$R^5, R^{11}, R^{12}, R^{13}, R^{14}, R^{43}$ 和 m_3 如上所定义。

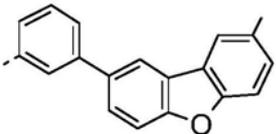
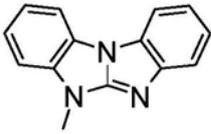
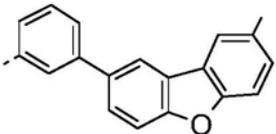
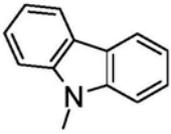
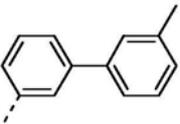
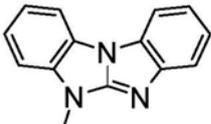
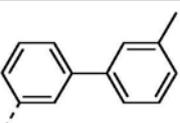
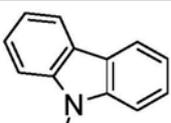
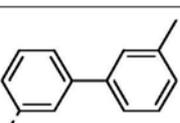
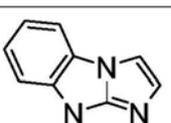
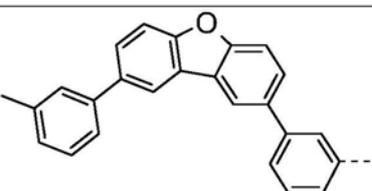
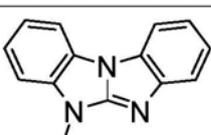
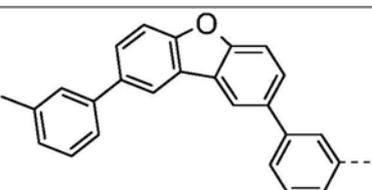
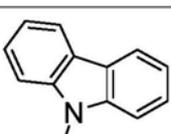
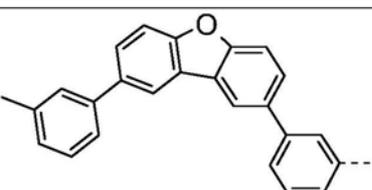
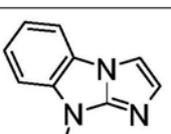
[0231] 在所述实施方案中, 式 $-A^1 - (A^2)_p - (A^3)_q - (A^4)_r -$ 的基团尤其为式 (IVa)、(IVb)、(IVc)、(IVd)、(IVe)、(IVf)、(IVg)、(IVh)、(IVi)、(IVj)、(IVk)、(IVl)、(IVm)、(IVn)、



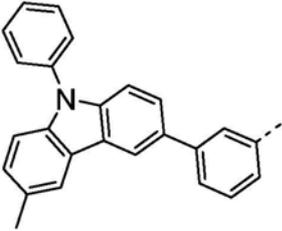
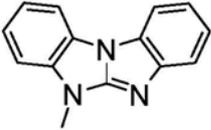
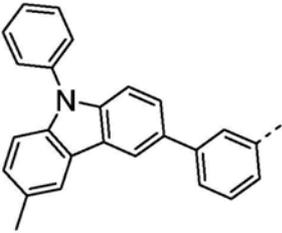
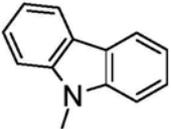
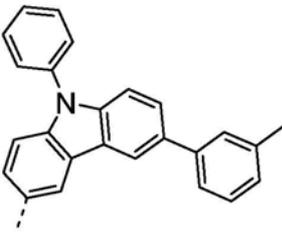
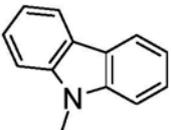
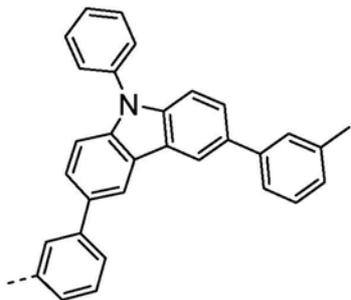
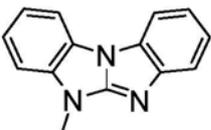
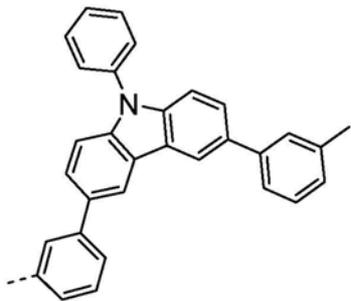
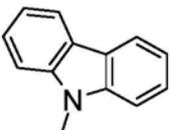
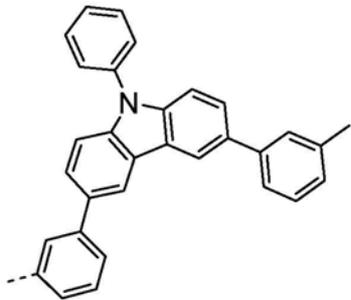
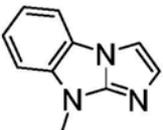
(IVx)、(IVy)、(IVz)、(VIa)、(VIb)、(VIc)、(VI d)、(VId)、(VIf)、(VIf)、(VIf)、(VIh)、(VIi)、(VIj)、(VIk)、(VI l)、(VI m)、(VI n)、(VI o)、(VI p)、(VI q)、(VI r)、(VI s)、(VI t) 或 (VI u) 的基团。目前最优选式 $-A^1 - (A^2)_p - (A^3)_q - (A^4)_r -$ 的基团为式 (IVa)、(IVb)、(IVe)、(IVl)、(IVk)、(IVs)、(IVv) 和 (VIj) 的基团。

[0232] 优选的化合物的实例为显示在下表中的化合物 D-1 至 D-144、E-1 至 E-183 以及 H-1 至 H-264。

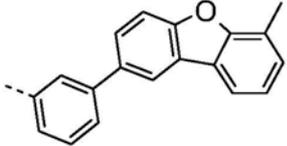
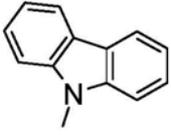
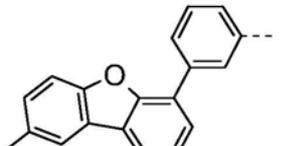
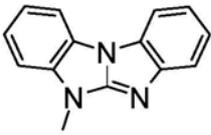
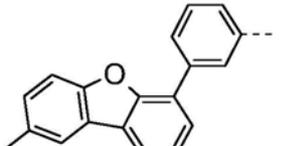
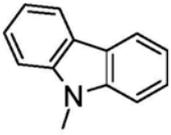
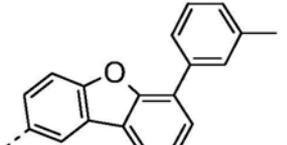
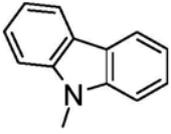
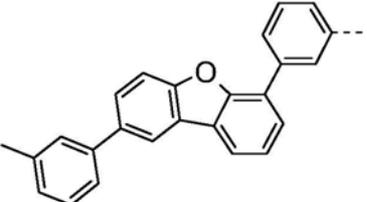
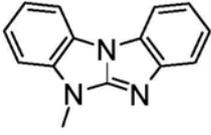
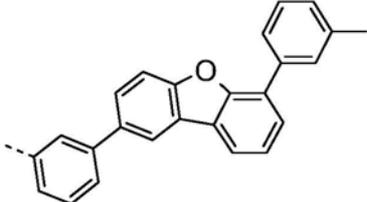
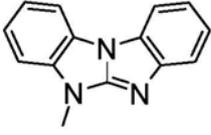
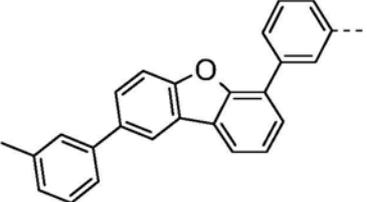
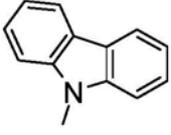
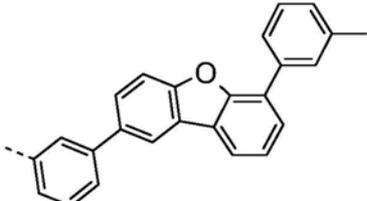
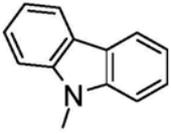


Cpd.	L ¹³⁾	R ⁶
D-1		
D-2		
D-3		
D-4		
D-5		
D-6		
D-7		
D-8		

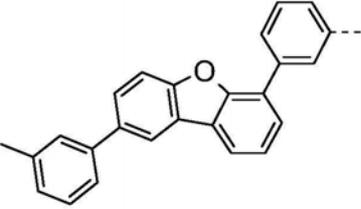
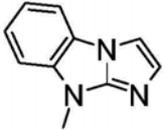
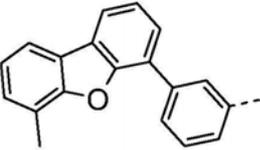
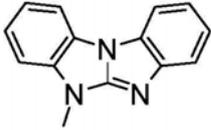
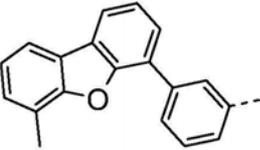
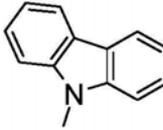
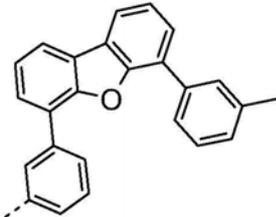
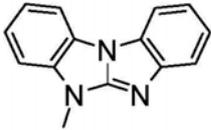
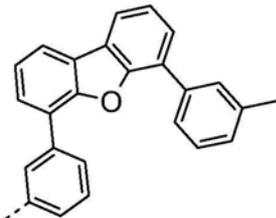
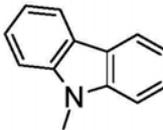
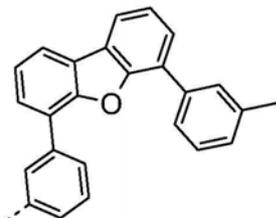
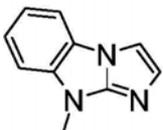
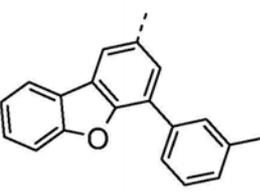
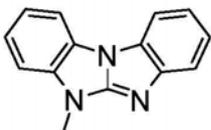
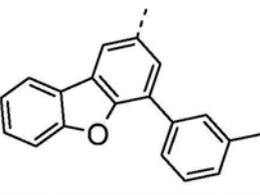
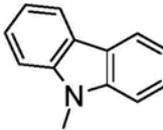
[0234]

<p>D-9</p>		
<p>D-10</p>		
<p>D-11</p>		
<p>D-12</p>		
<p>D-13</p>		
<p>D-14</p>		

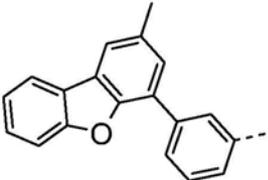
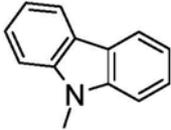
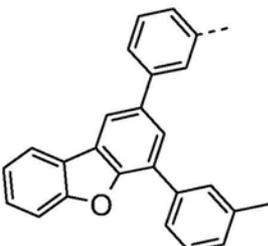
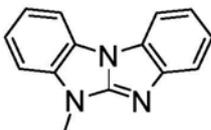
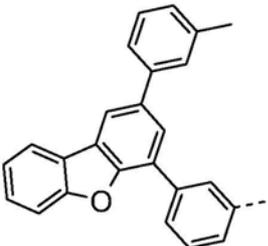
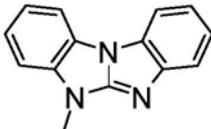
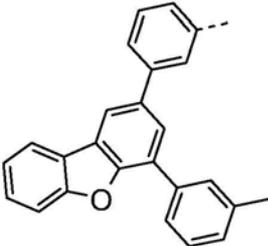
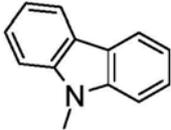
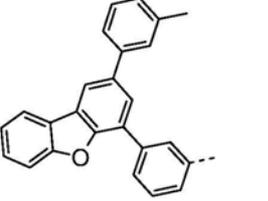
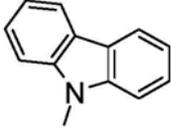
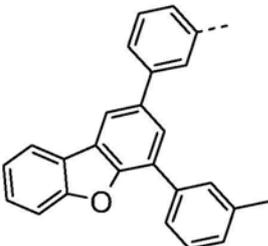
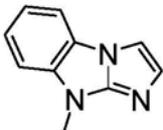
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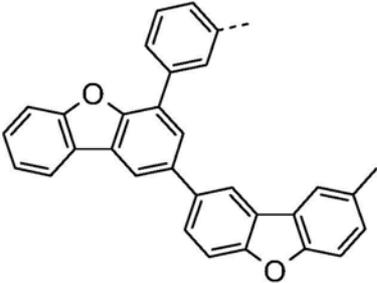
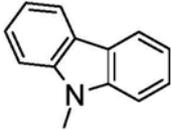
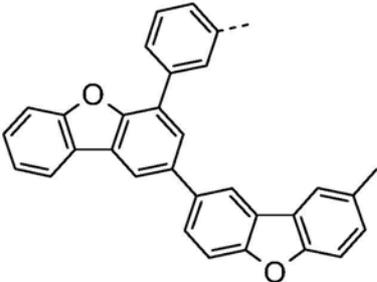
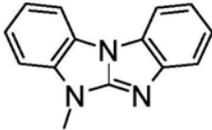
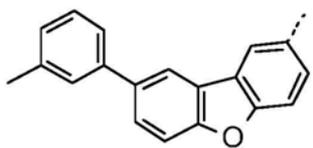
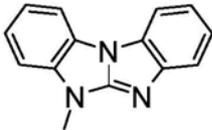
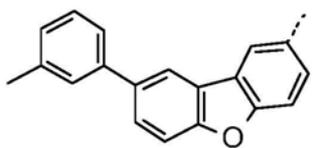
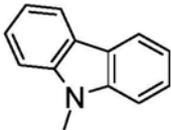
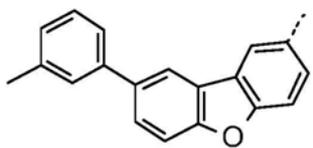
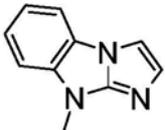
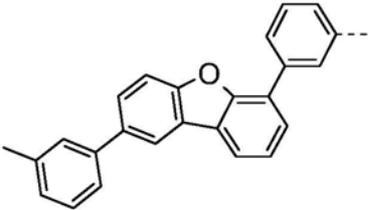
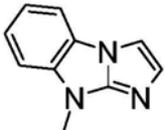
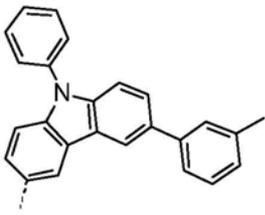
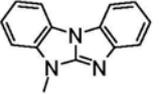
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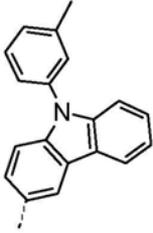
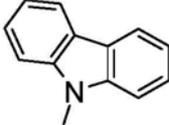
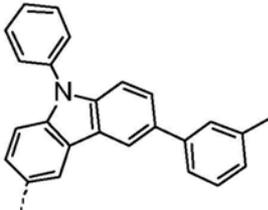
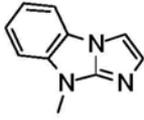
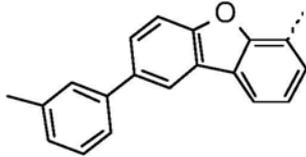
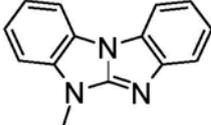
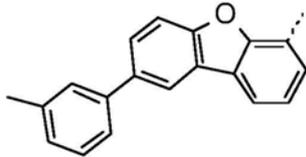
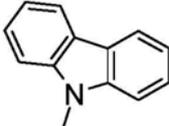
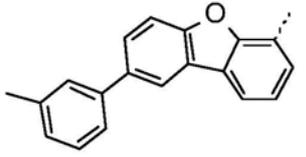
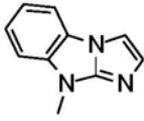
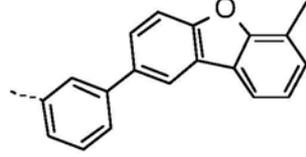
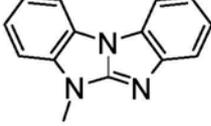
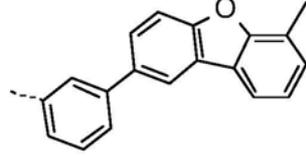
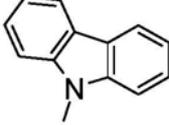
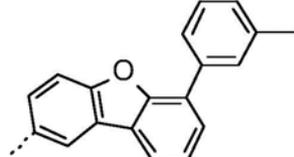
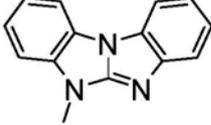
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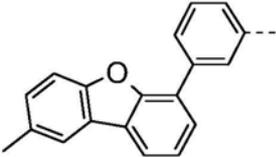
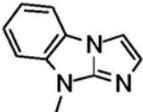
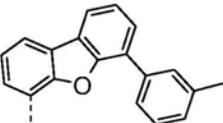
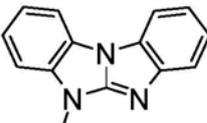
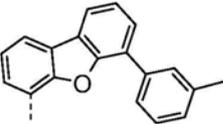
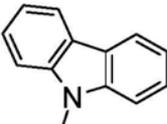
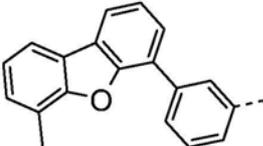
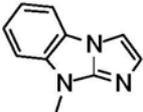
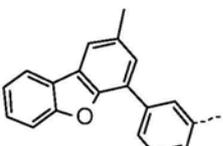
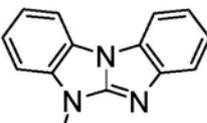
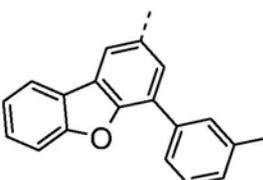
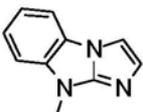
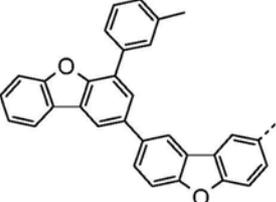
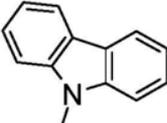
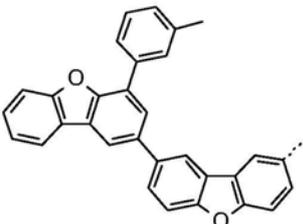
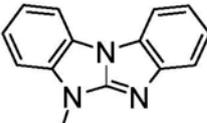
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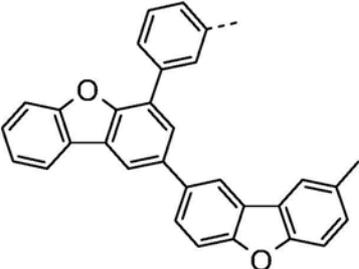
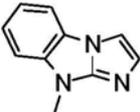
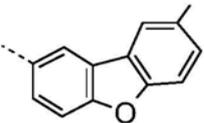
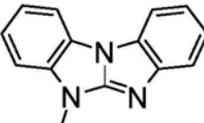
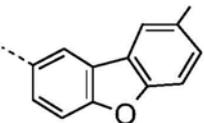
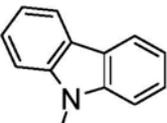
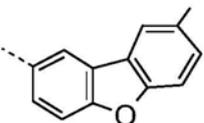
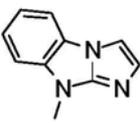
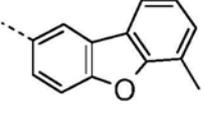
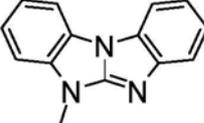
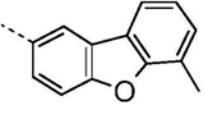
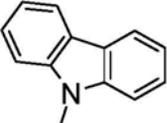
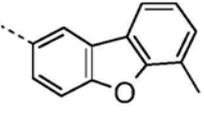
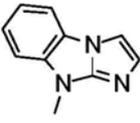
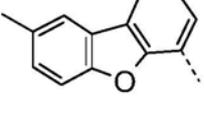
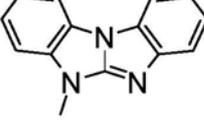
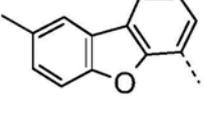
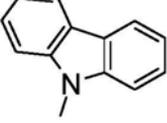
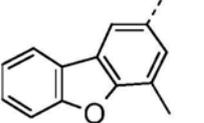
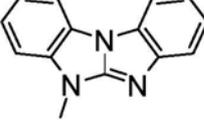
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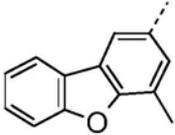
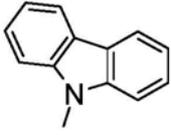
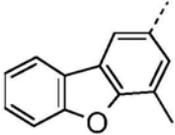
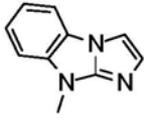
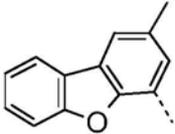
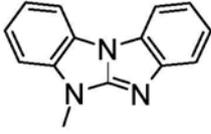
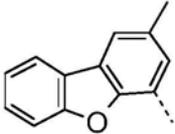
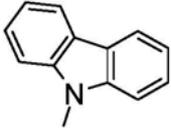
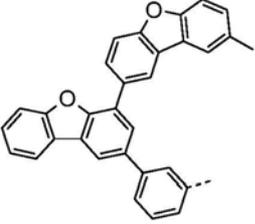
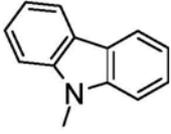
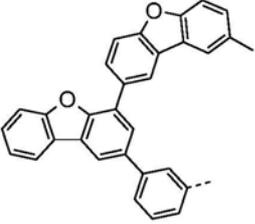
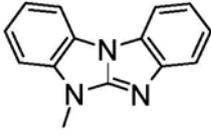
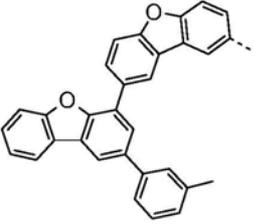
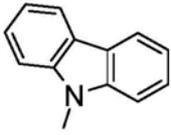
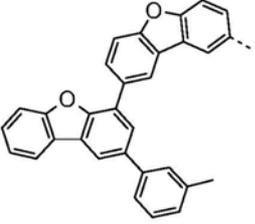
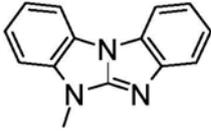
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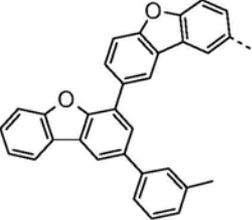
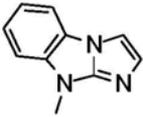
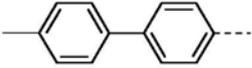
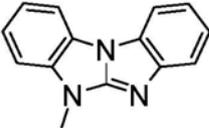
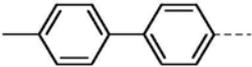
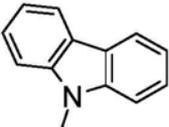
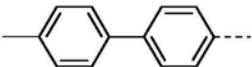
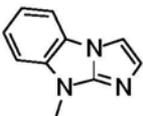
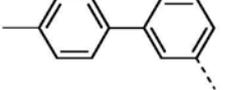
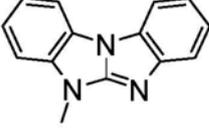
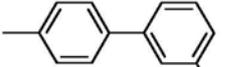
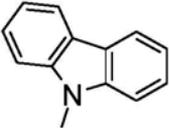
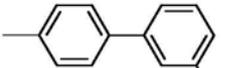
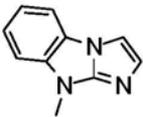
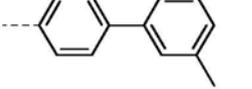
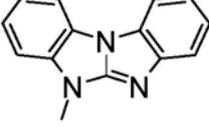
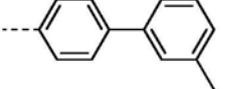
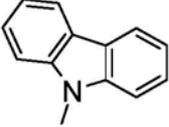
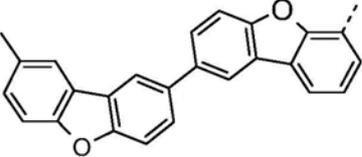
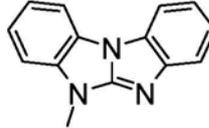
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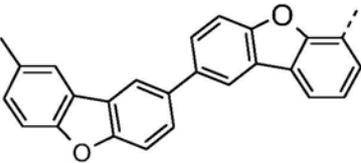
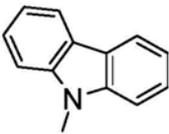
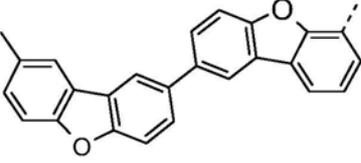
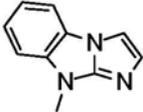
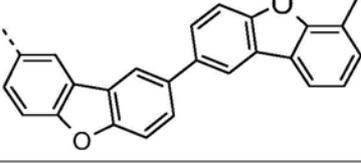
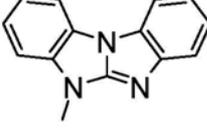
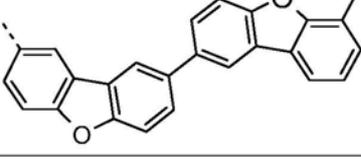
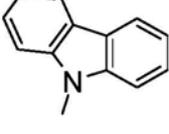
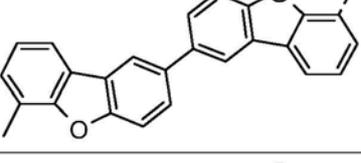
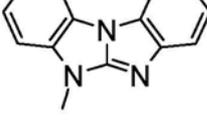
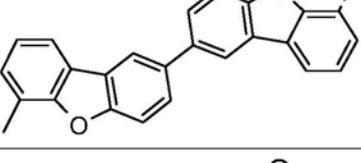
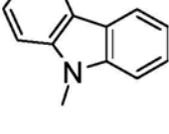
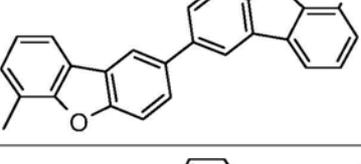
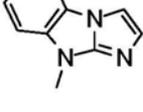
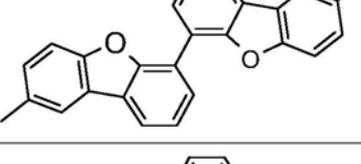
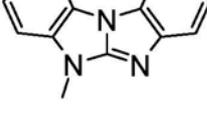
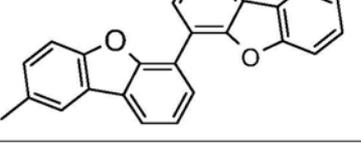
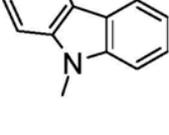
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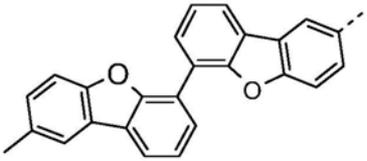
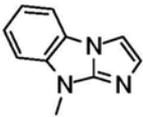
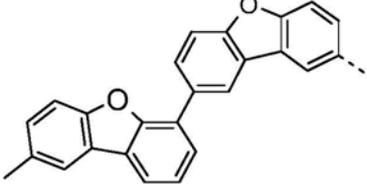
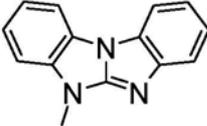
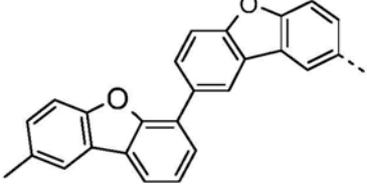
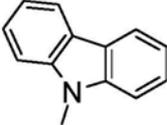
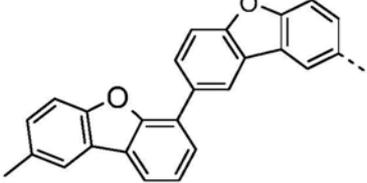
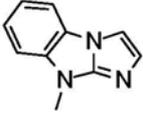
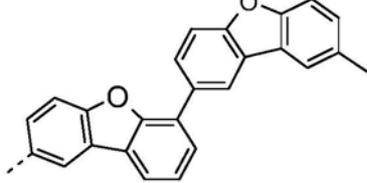
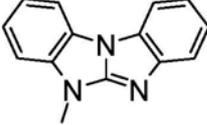
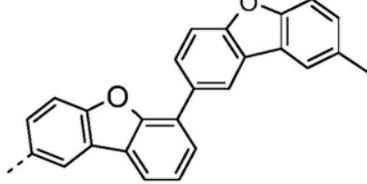
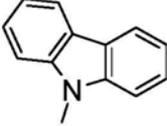
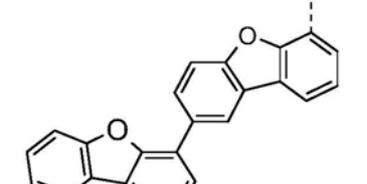
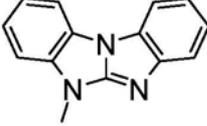
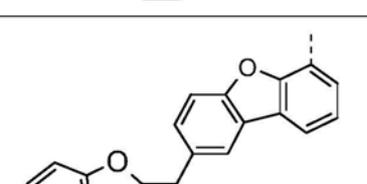
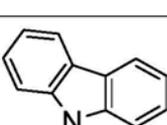
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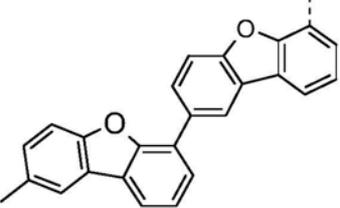
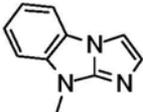
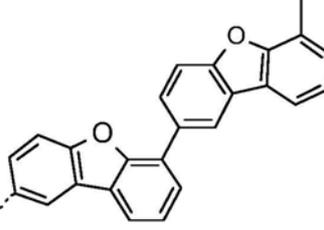
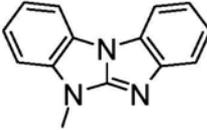
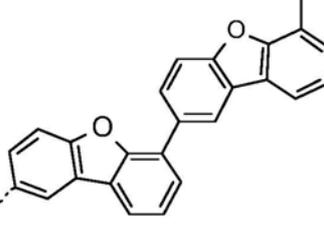
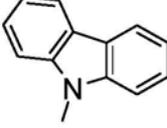
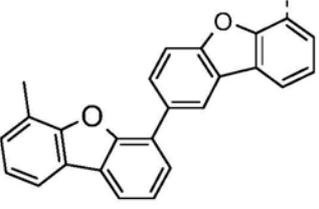
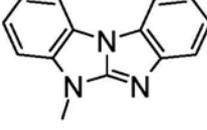
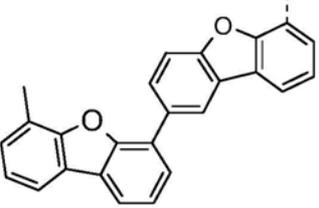
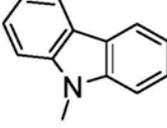
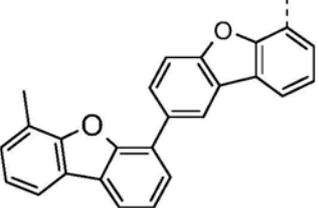
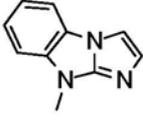
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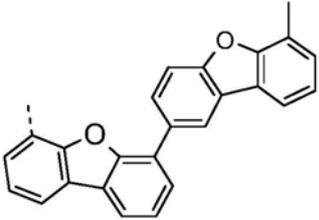
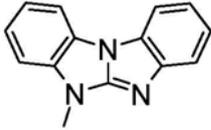
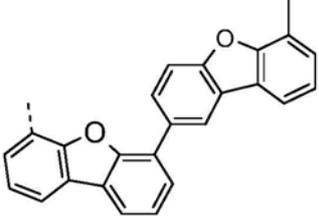
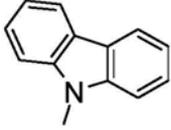
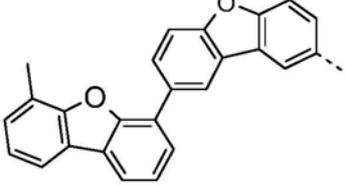
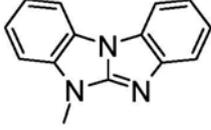
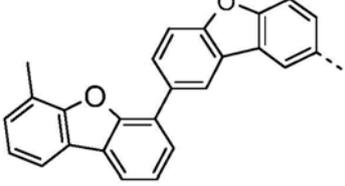
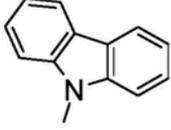
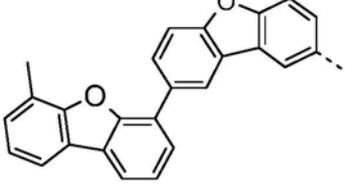
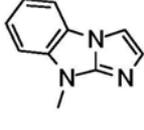
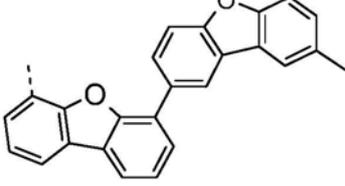
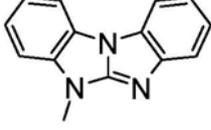
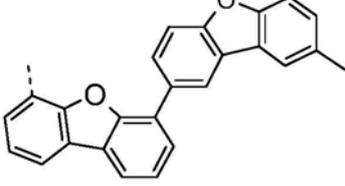
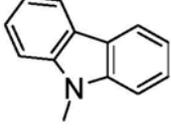
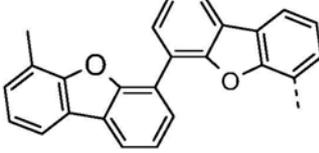
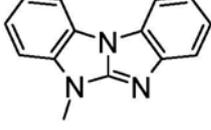
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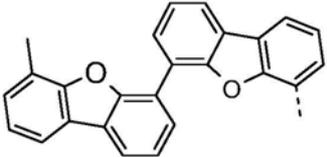
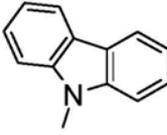
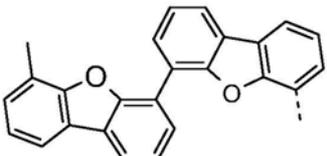
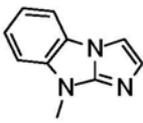
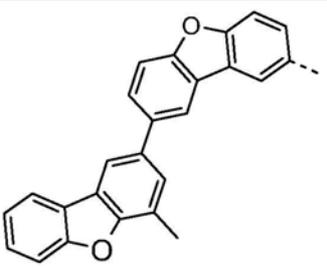
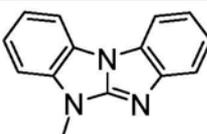
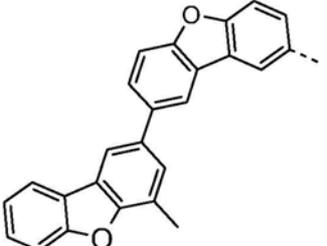
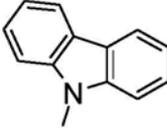
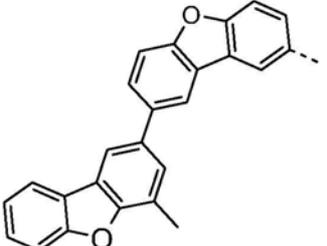
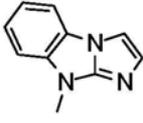
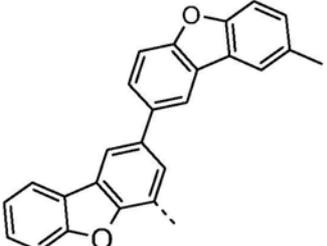
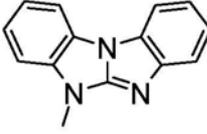
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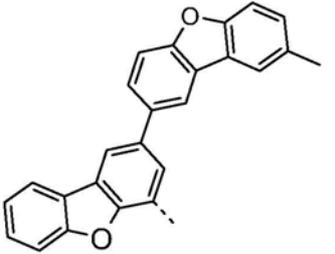
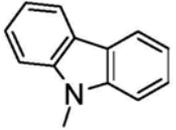
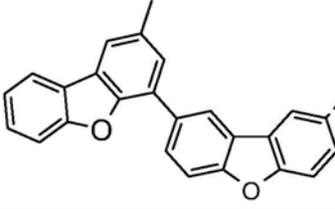
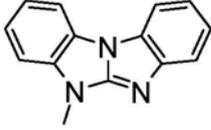
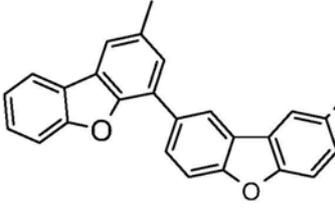
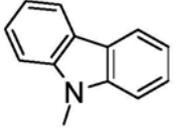
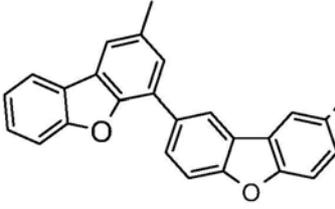
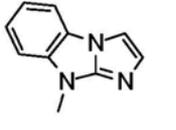
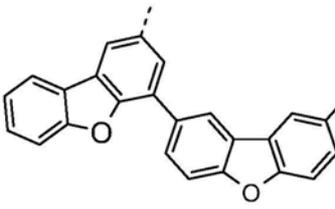
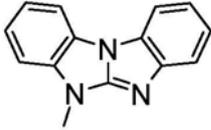
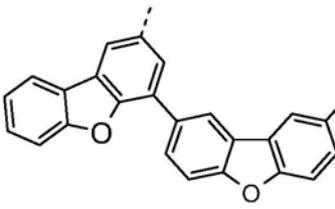
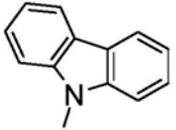
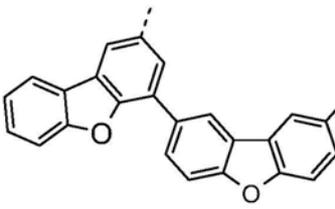
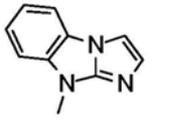
[0247]

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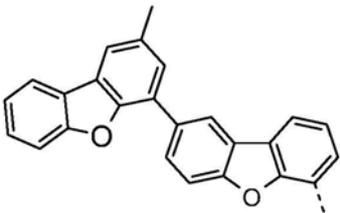
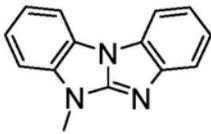
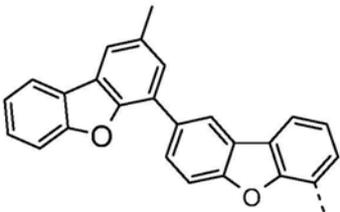
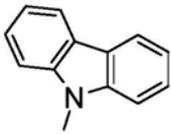
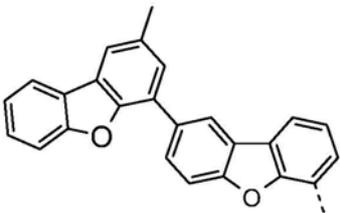
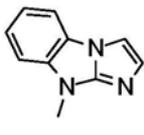
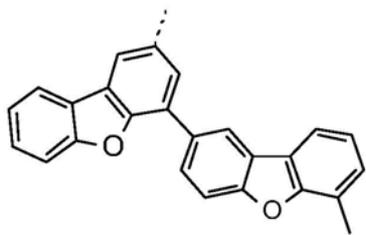
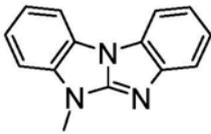
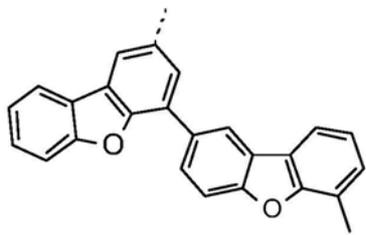
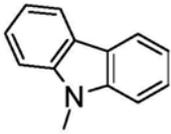
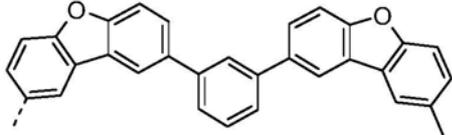
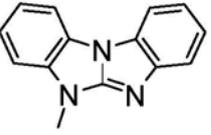
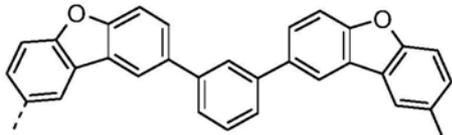
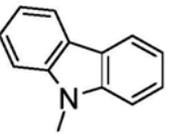
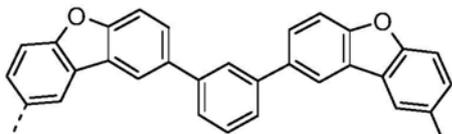
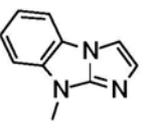
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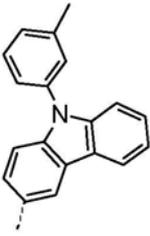
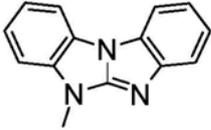
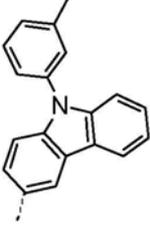
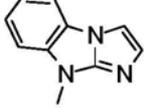
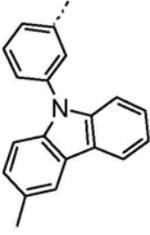
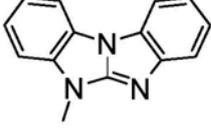
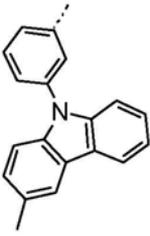
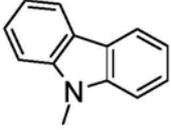
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D-125		

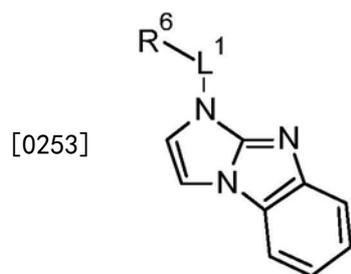
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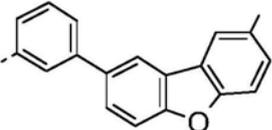
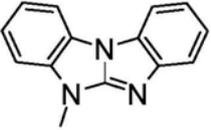
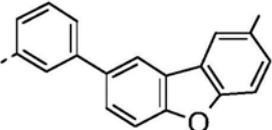
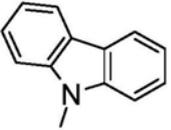
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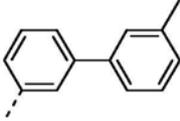
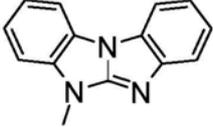
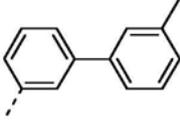
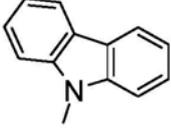
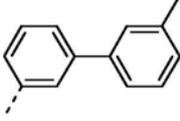
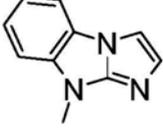
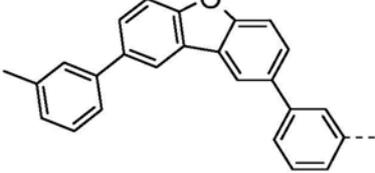
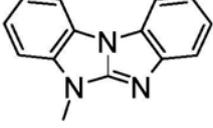
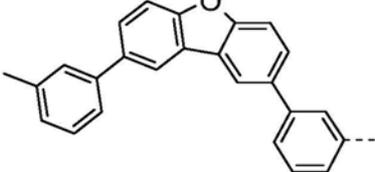
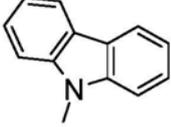
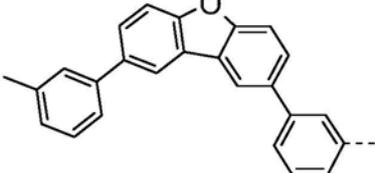
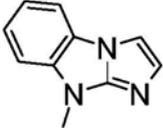
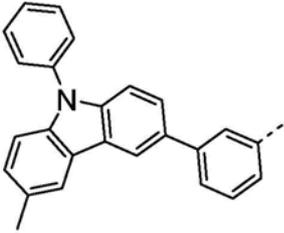
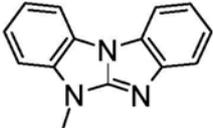
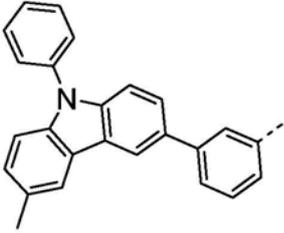
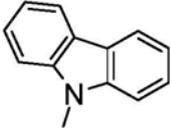
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[0252]		
		
		

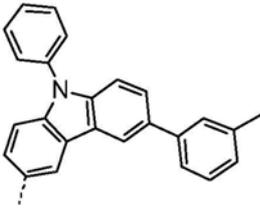
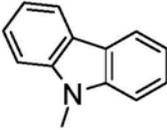
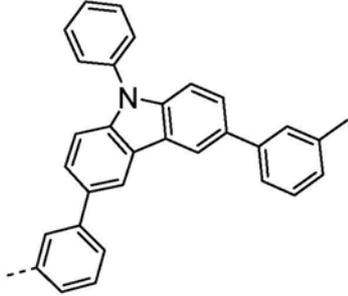
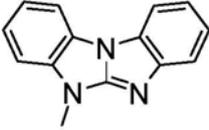
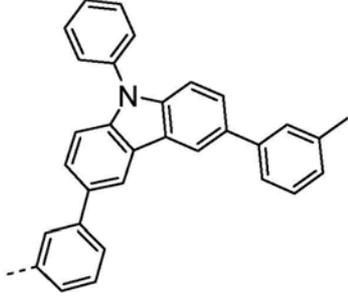
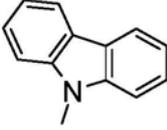
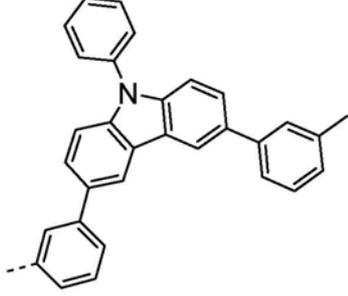
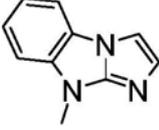
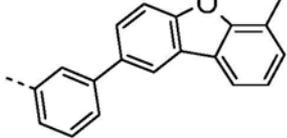
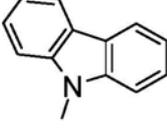
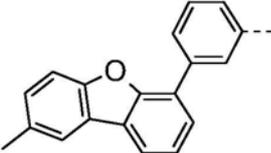
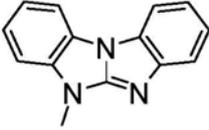


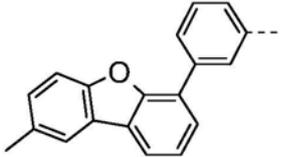
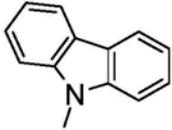
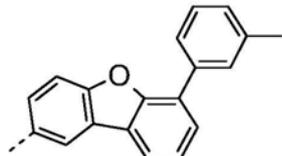
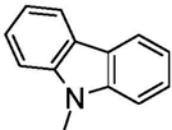
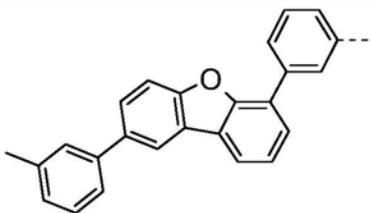
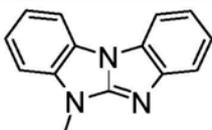
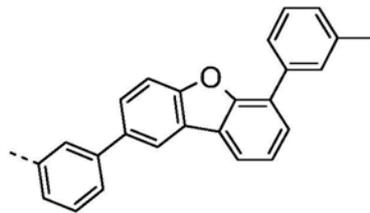
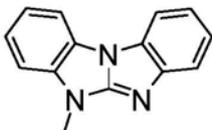
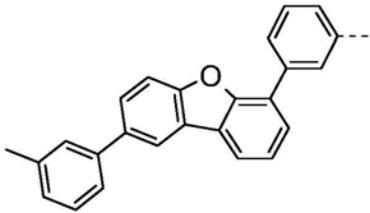
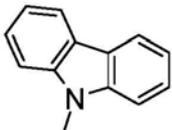
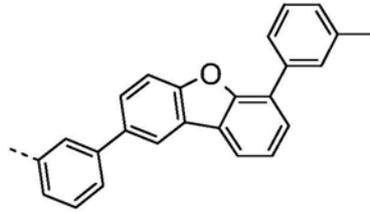
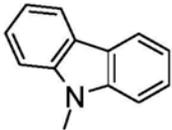
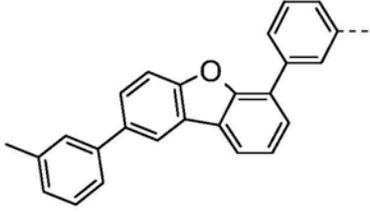
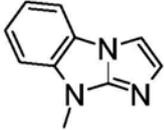
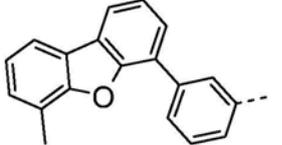
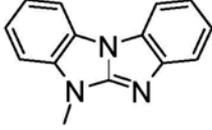
CpE.	$L^{13)}$	R^6
[0254] E-1		
E-2		

E-3		
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E-6		
E-7		
E-8		
E-9		
E-10		

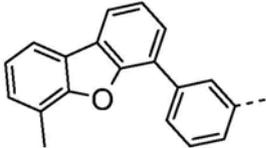
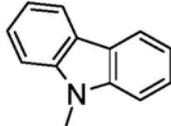
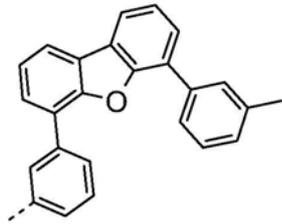
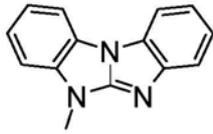
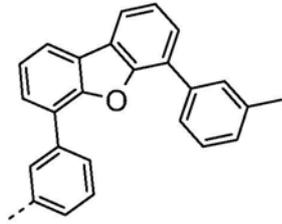
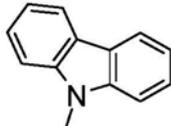
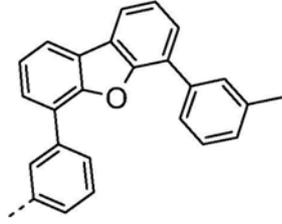
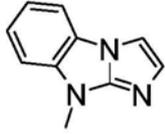
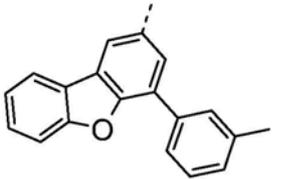
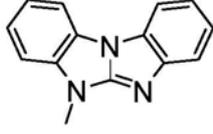
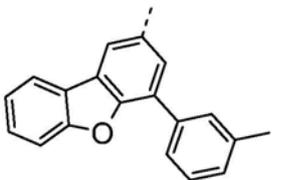
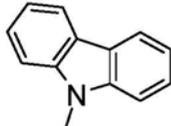
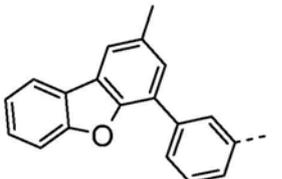
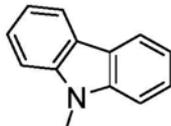
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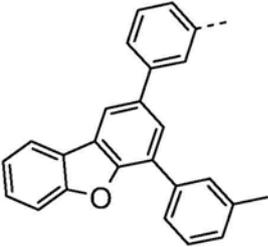
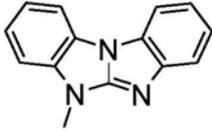
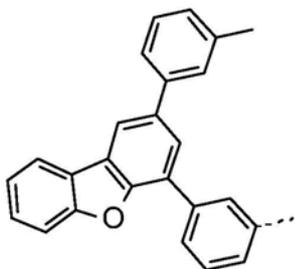
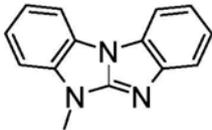
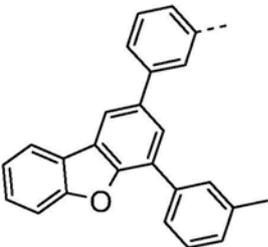
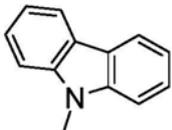
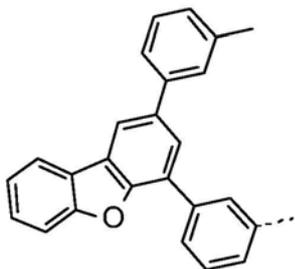
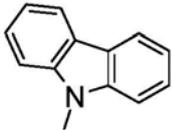
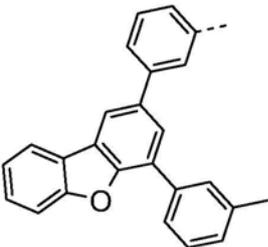
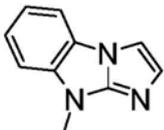
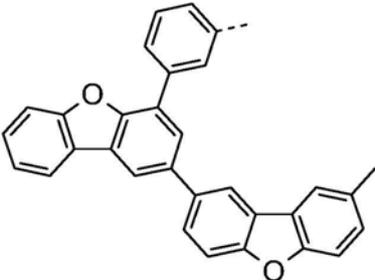
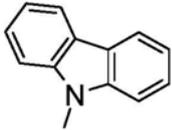
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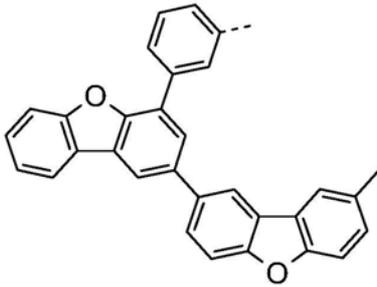
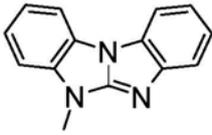
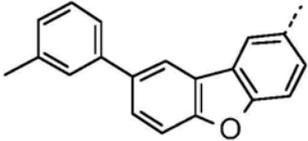
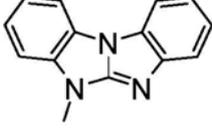
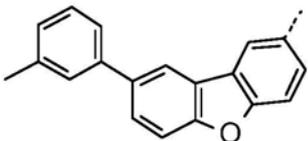
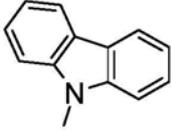
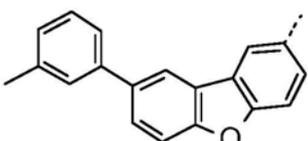
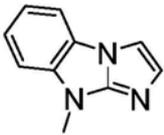
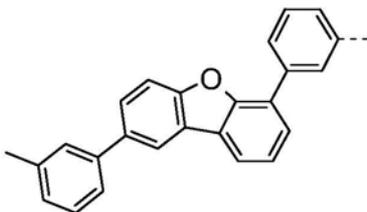
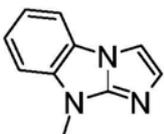
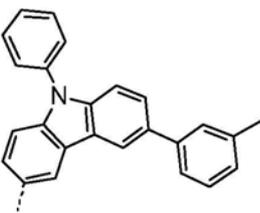
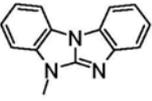
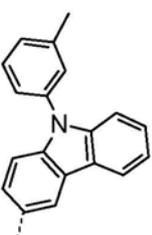
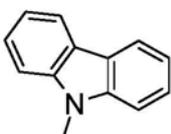
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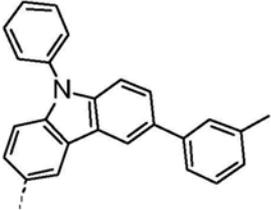
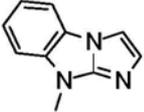
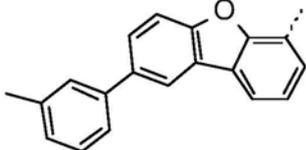
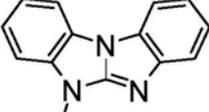
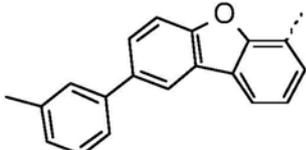
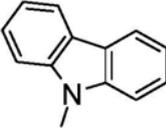
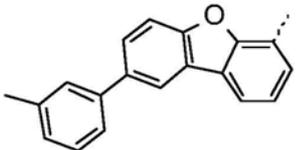
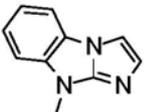
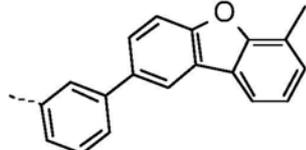
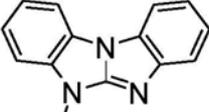
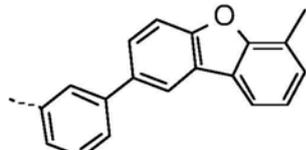
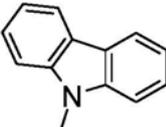
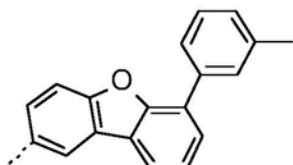
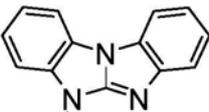
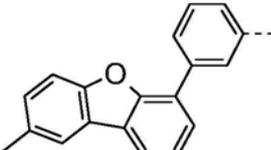
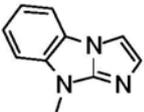
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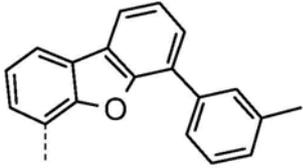
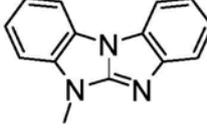
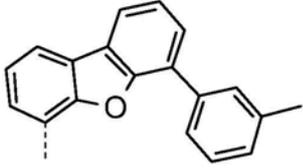
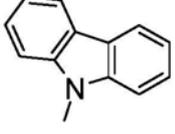
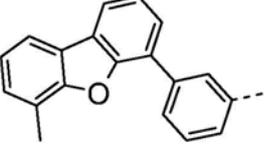
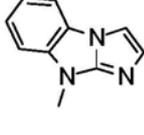
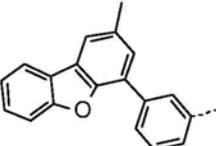
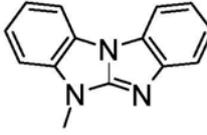
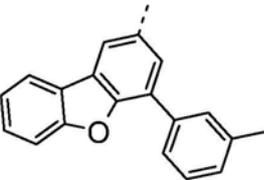
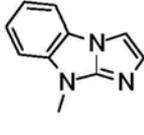
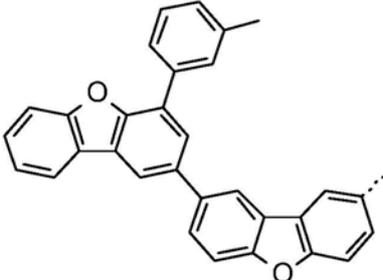
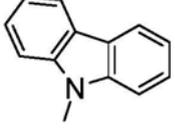
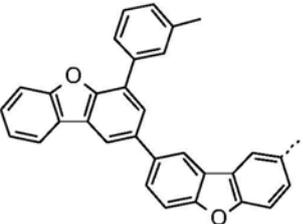
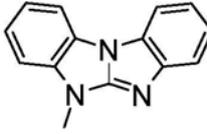
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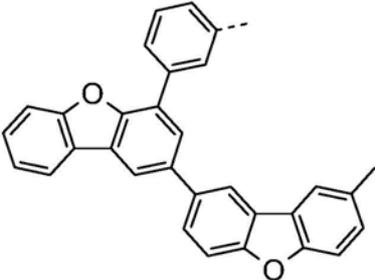
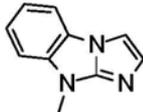
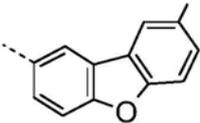
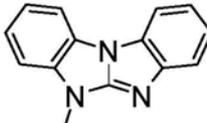
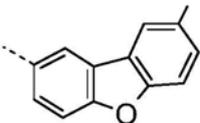
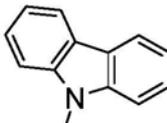
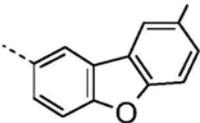
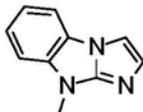
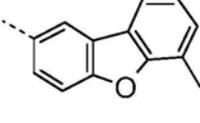
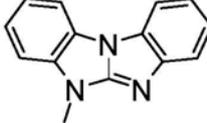
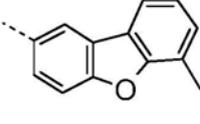
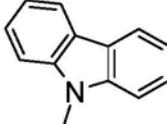
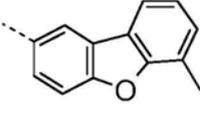
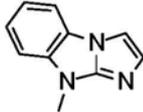
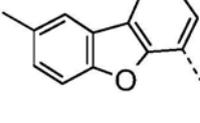
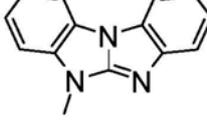
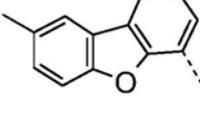
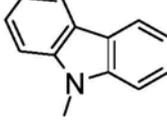
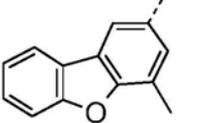
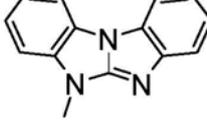
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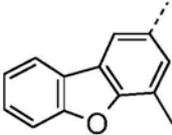
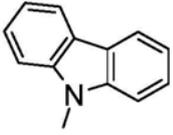
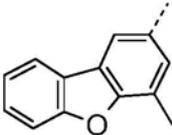
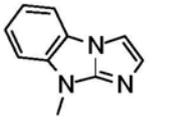
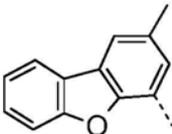
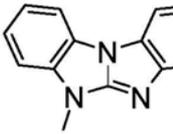
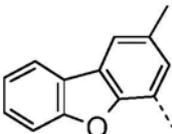
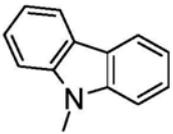
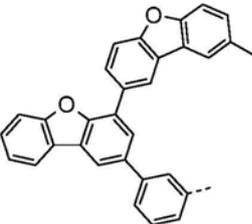
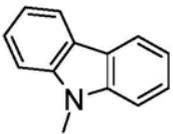
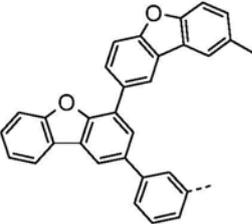
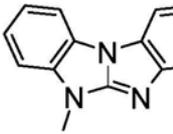
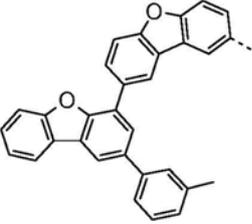
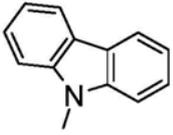
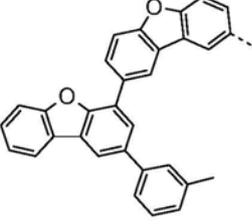
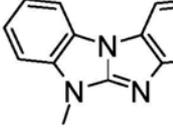
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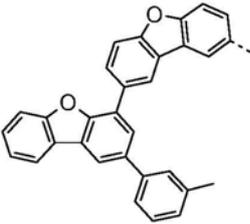
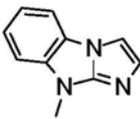
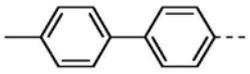
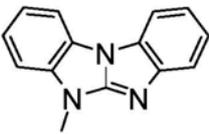
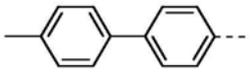
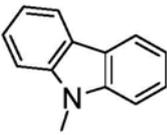
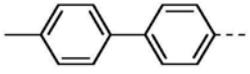
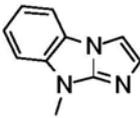
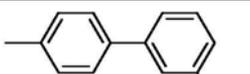
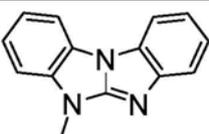
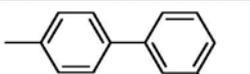
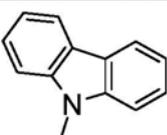
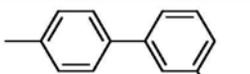
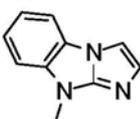
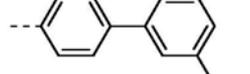
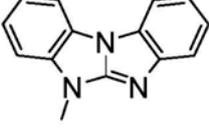
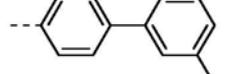
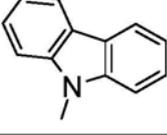
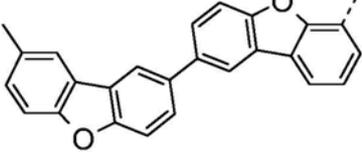
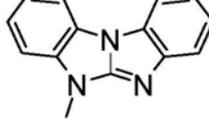
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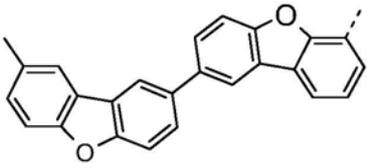
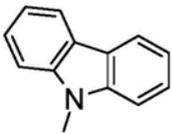
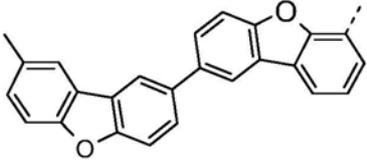
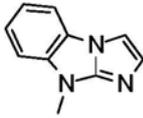
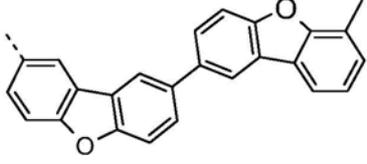
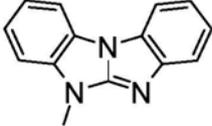
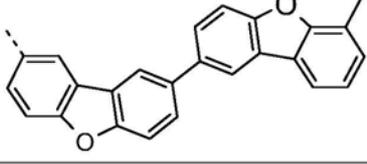
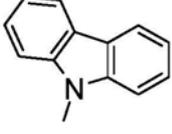
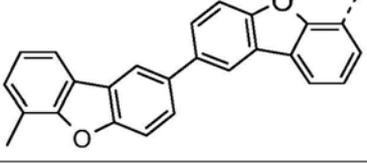
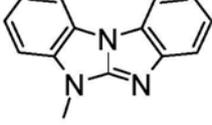
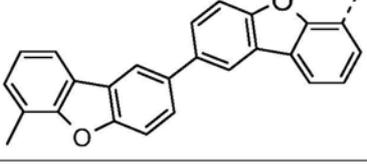
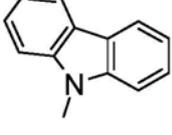
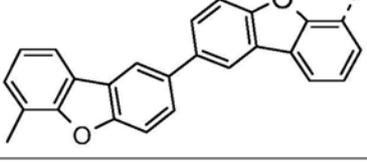
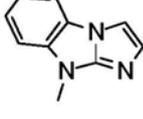
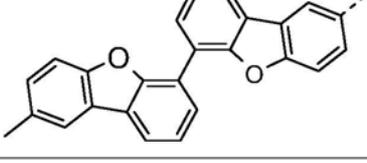
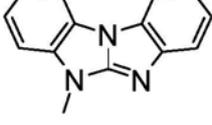
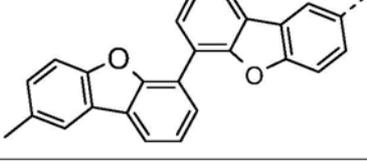
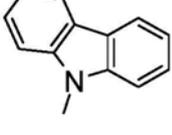
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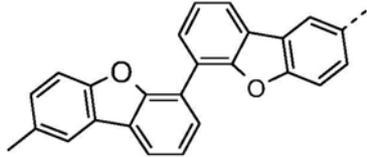
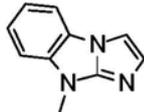
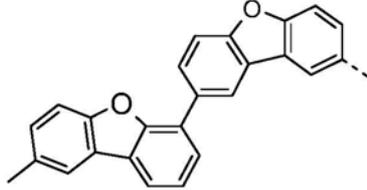
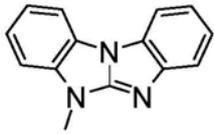
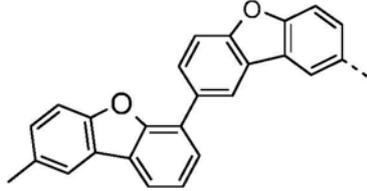
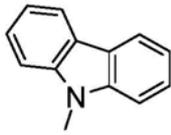
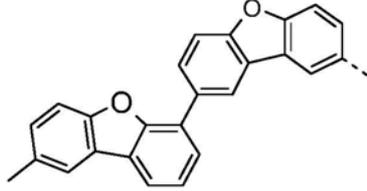
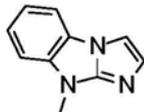
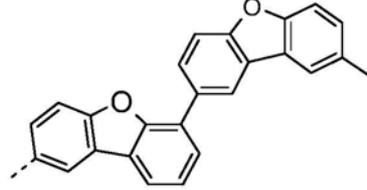
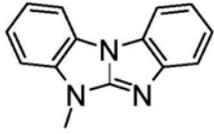
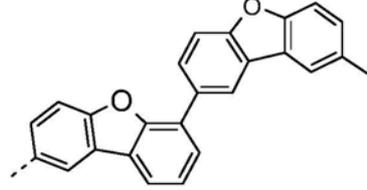
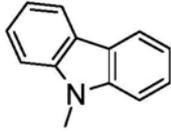
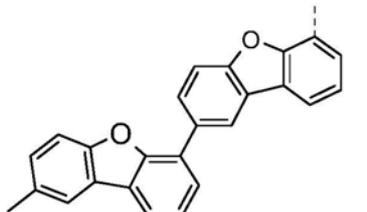
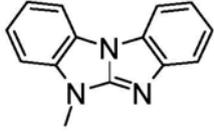
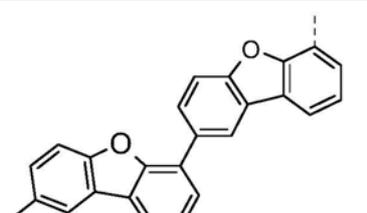
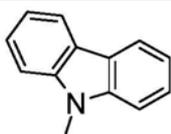
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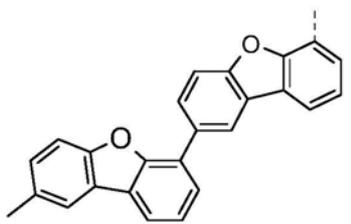
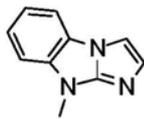
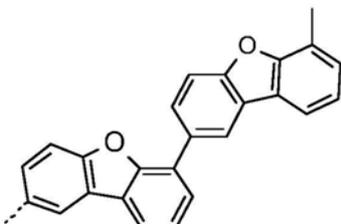
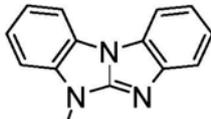
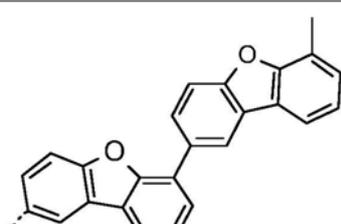
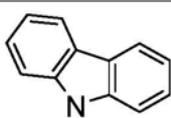
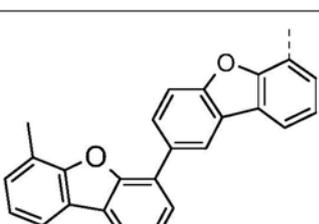
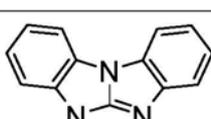
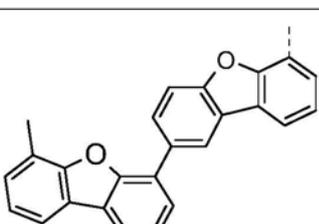
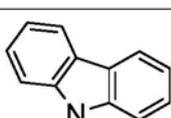
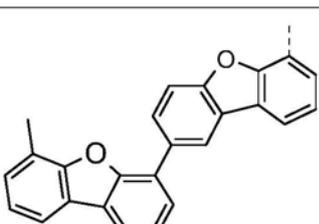
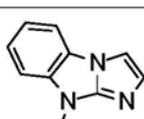
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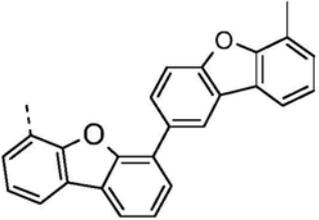
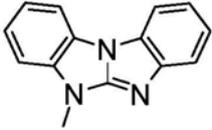
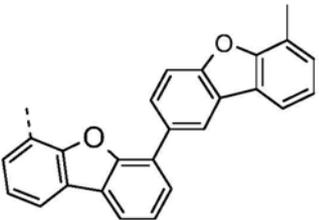
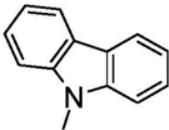
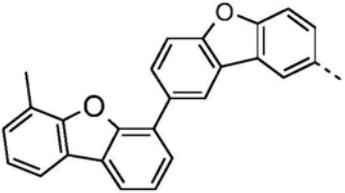
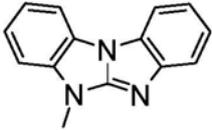
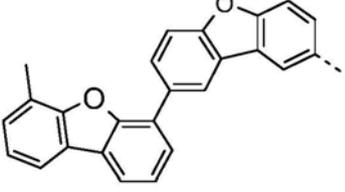
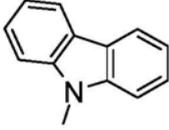
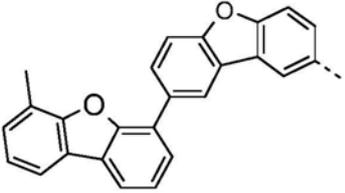
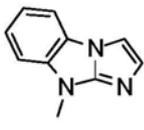
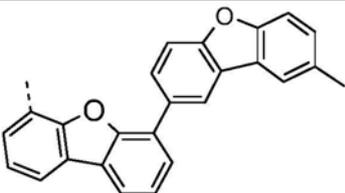
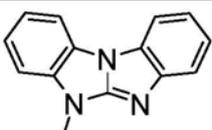
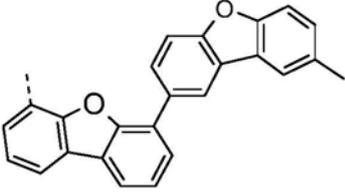
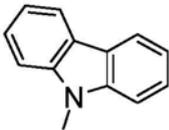
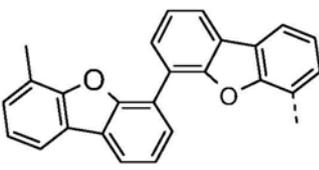
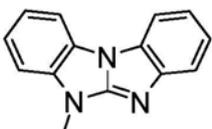
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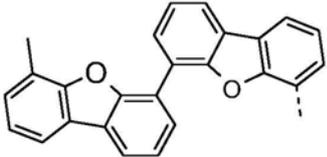
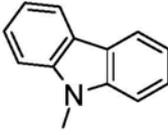
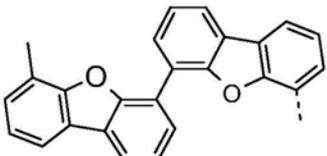
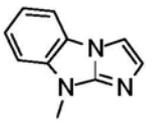
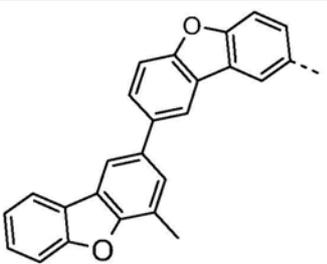
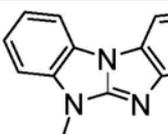
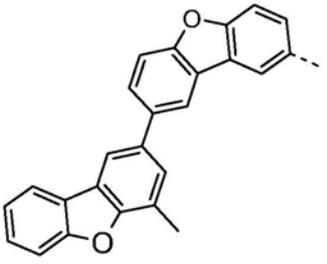
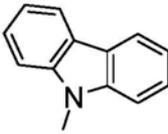
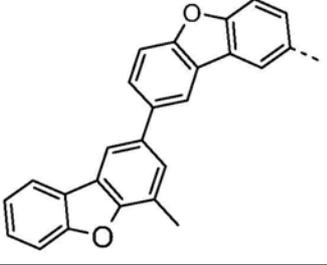
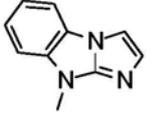
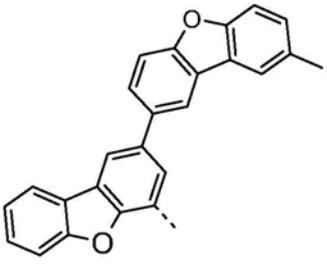
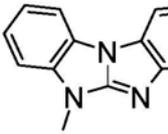
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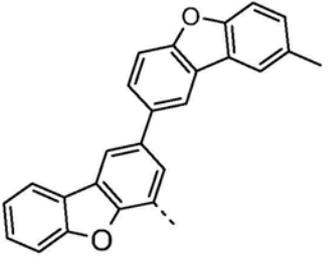
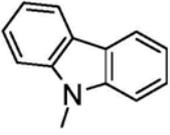
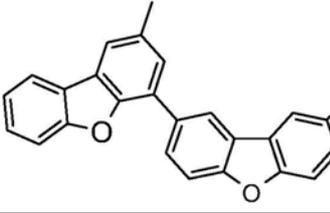
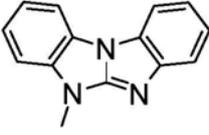
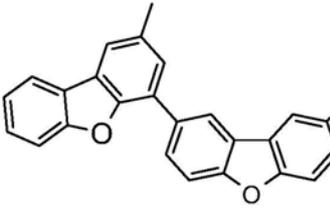
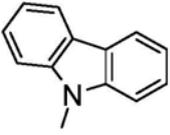
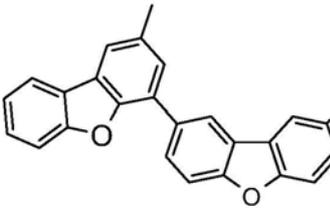
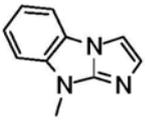
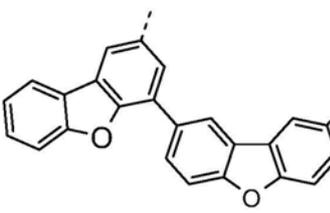
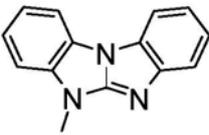
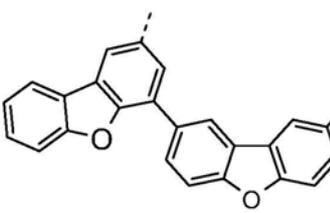
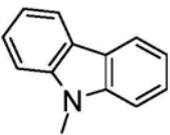
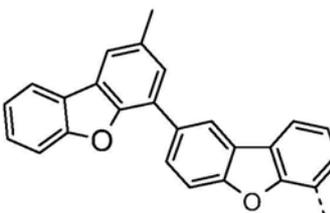
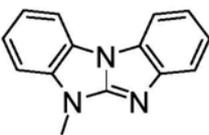
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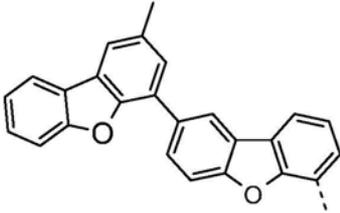
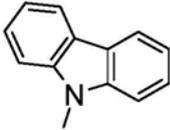
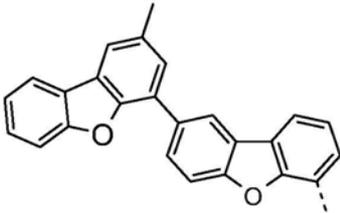
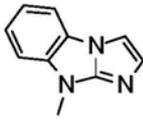
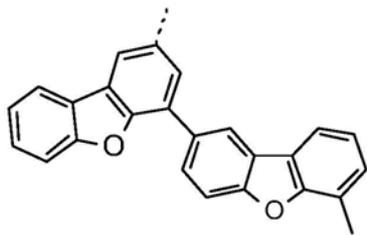
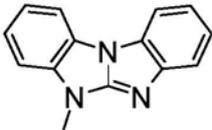
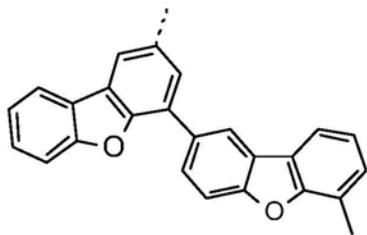
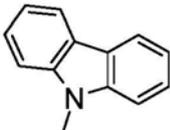
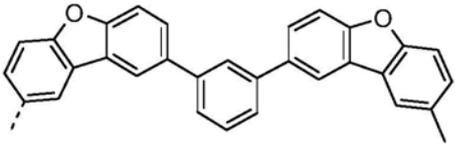
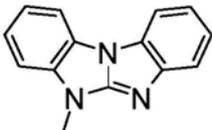
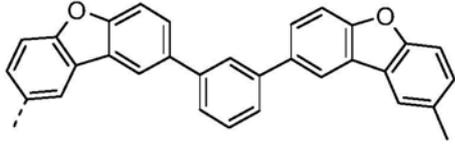
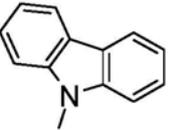
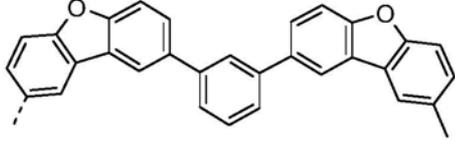
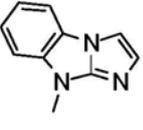
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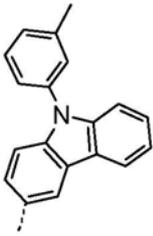
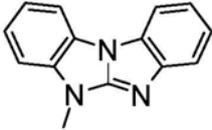
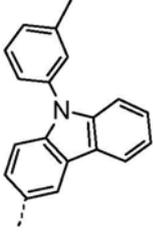
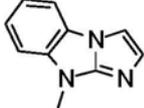
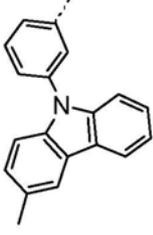
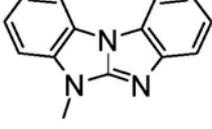
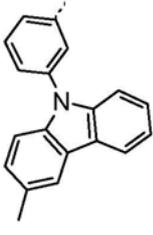
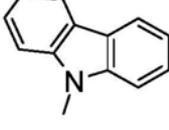
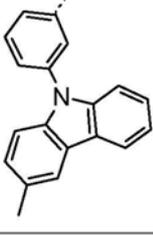
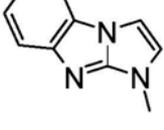
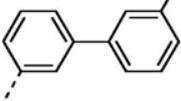
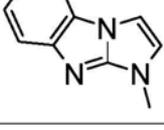
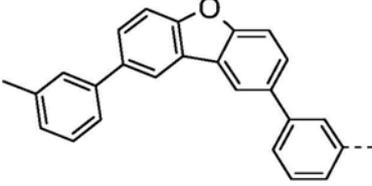
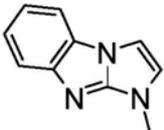
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[0271]

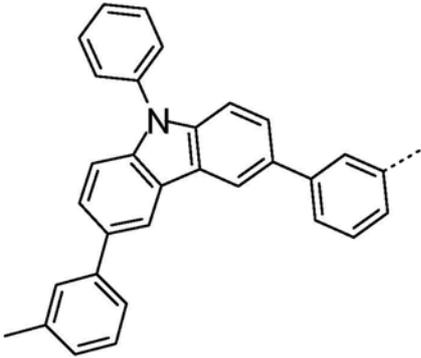
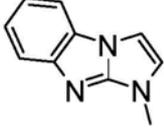
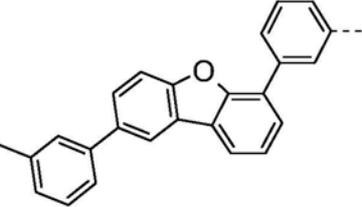
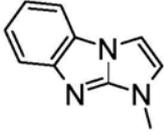
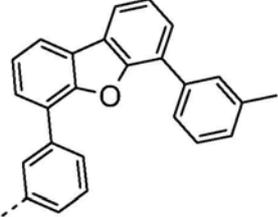
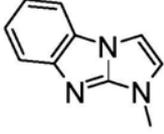
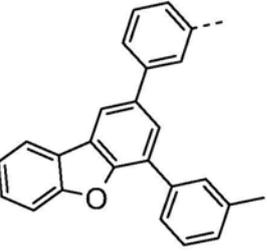
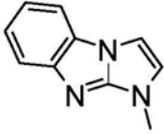
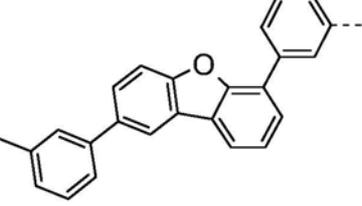
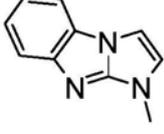
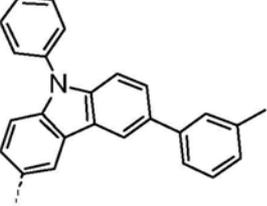
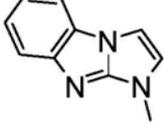
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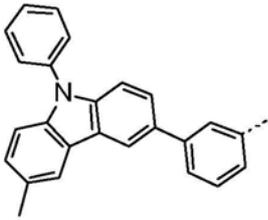
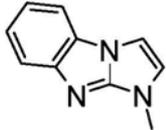
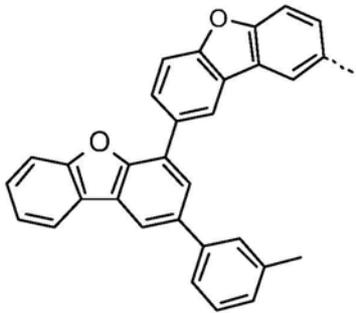
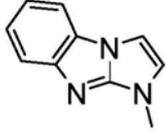
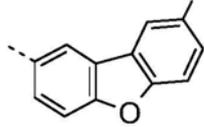
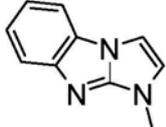
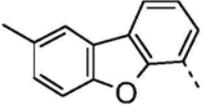
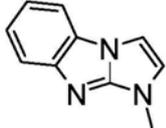
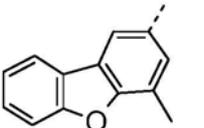
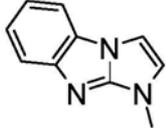
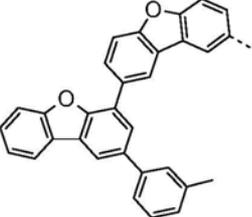
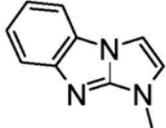
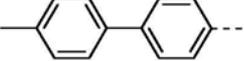
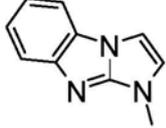
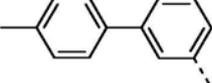
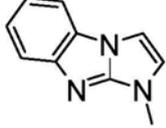
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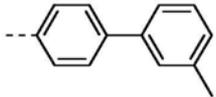
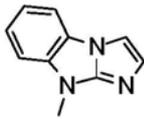
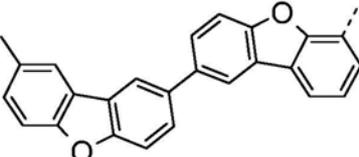
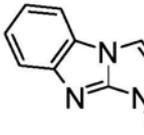
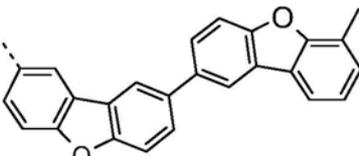
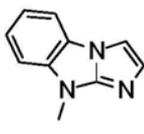
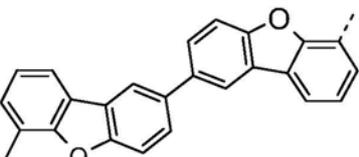
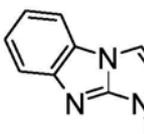
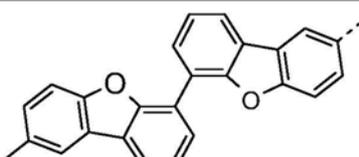
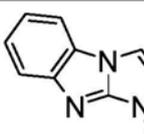
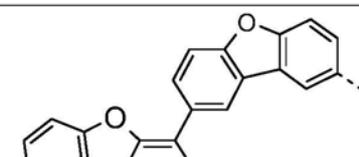
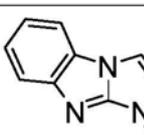
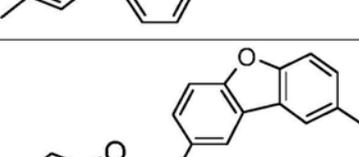
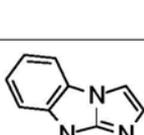
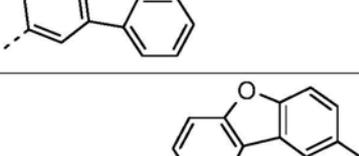
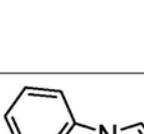
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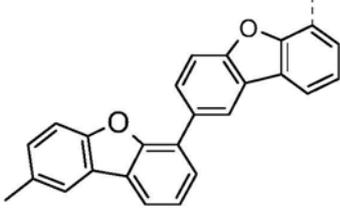
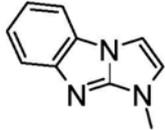
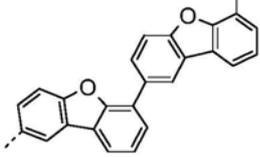
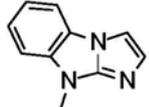
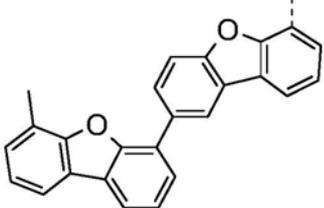
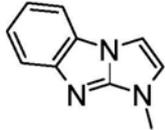
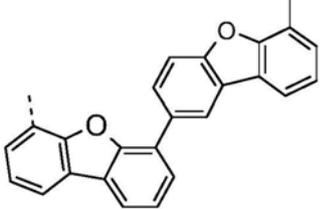
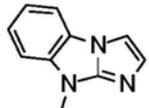
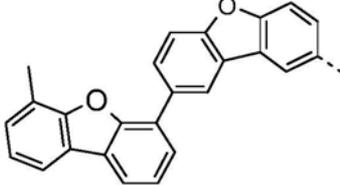
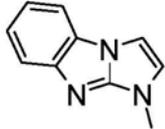
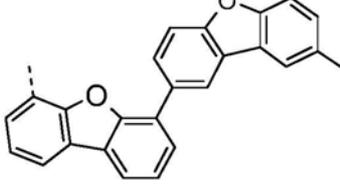
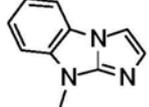
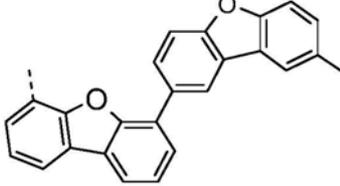
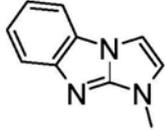
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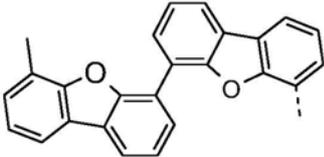
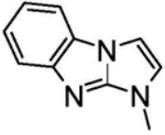
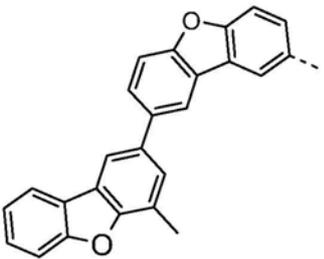
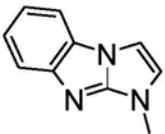
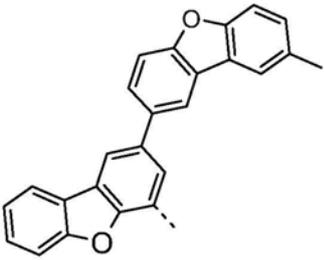
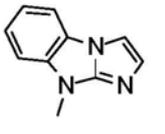
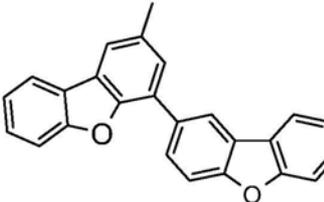
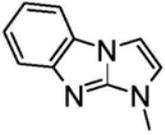
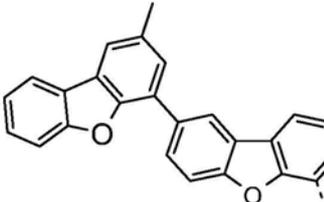
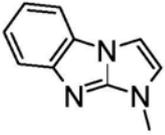
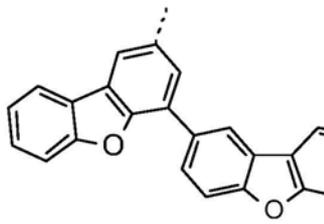
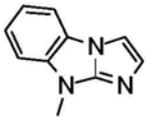
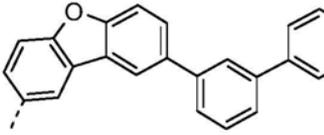
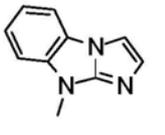
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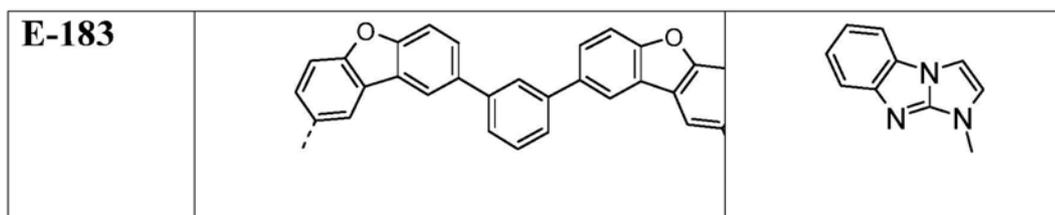
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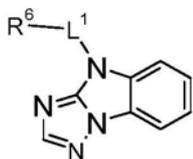
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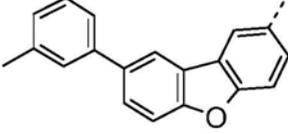
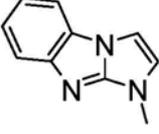
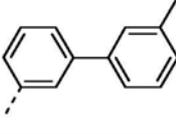
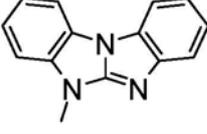
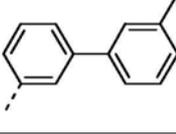
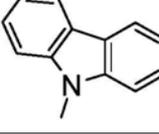
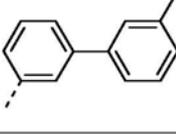
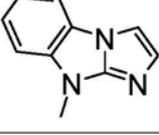
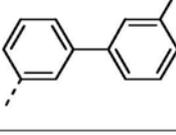
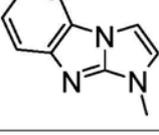
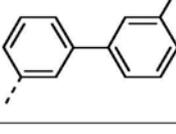
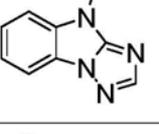
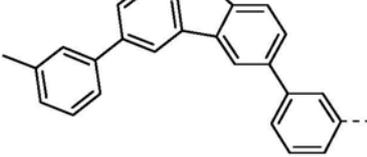
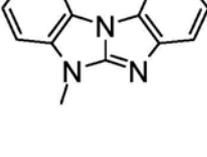
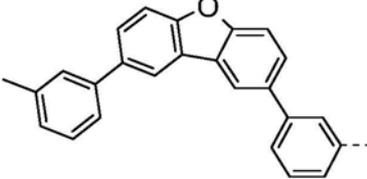
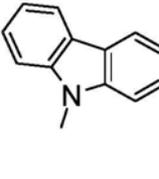
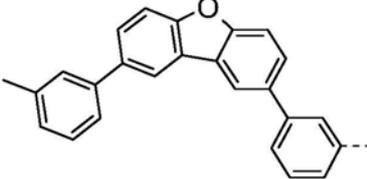
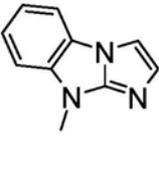
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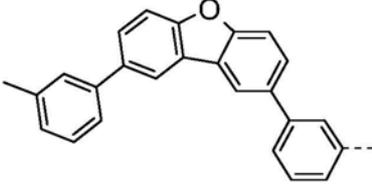
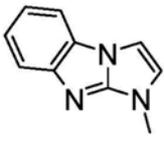
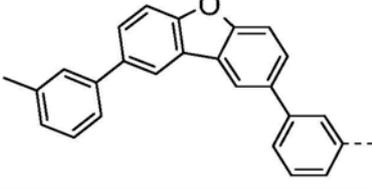
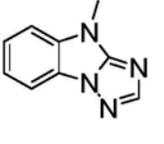
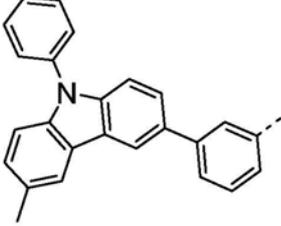
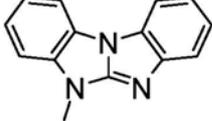
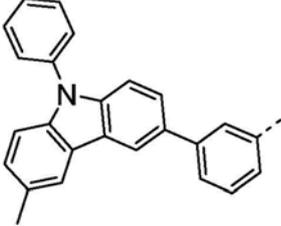
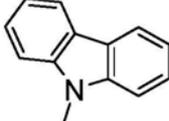
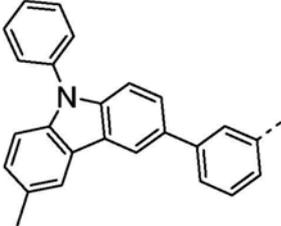
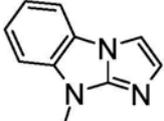
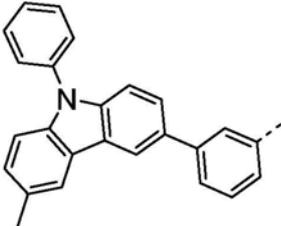
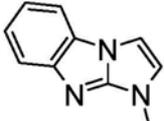
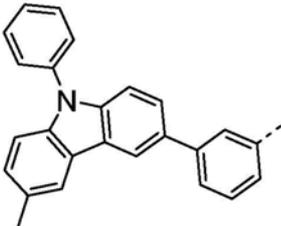
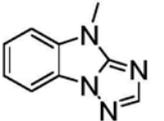


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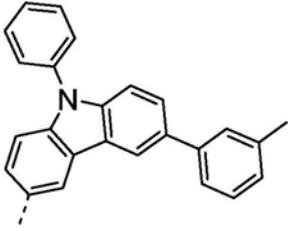
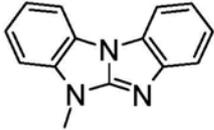
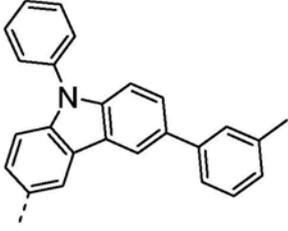
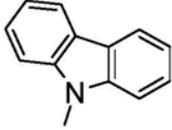
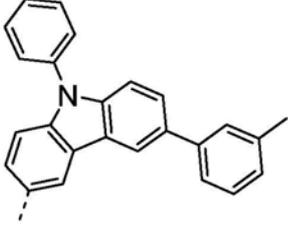
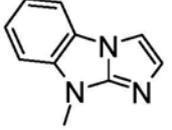
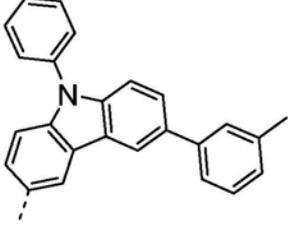
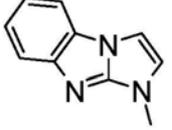
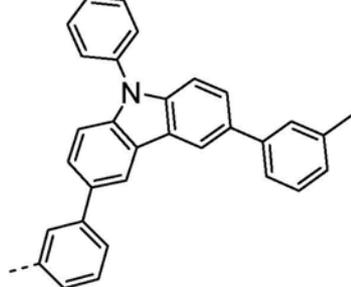
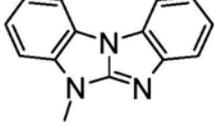
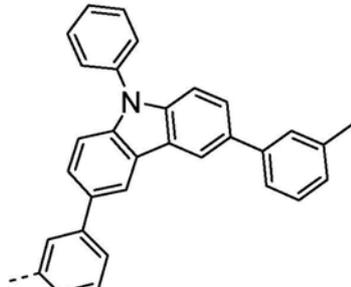
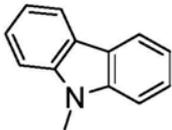
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[0282]

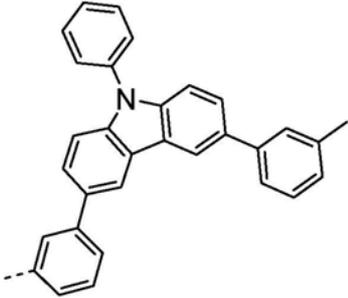
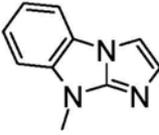
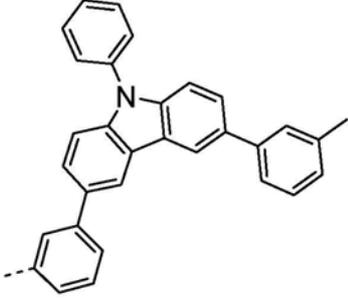
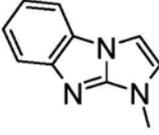
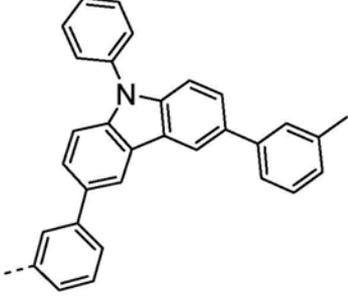
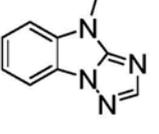
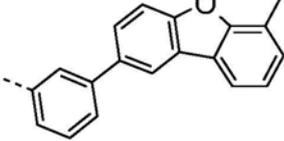
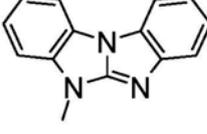
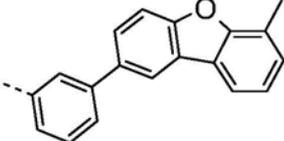
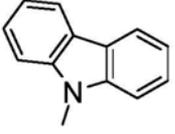
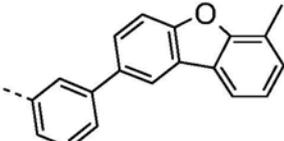
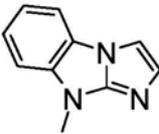
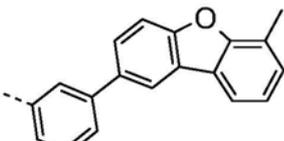
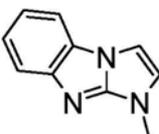
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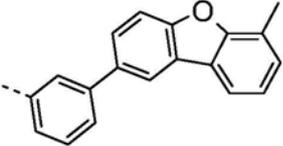
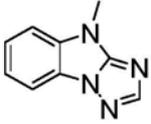
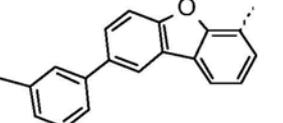
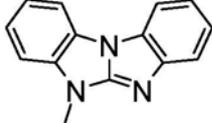
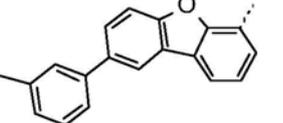
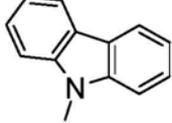
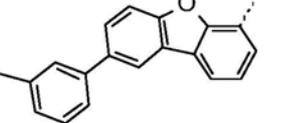
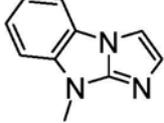
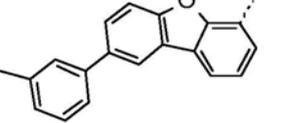
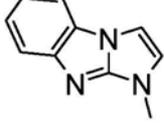
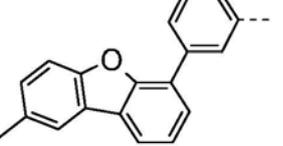
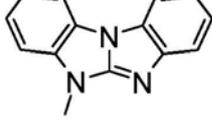
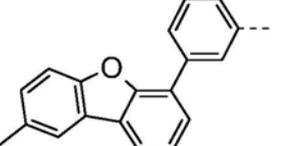
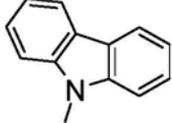
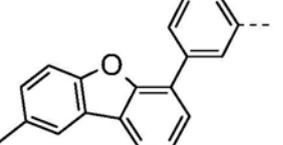
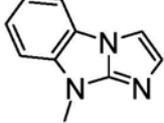
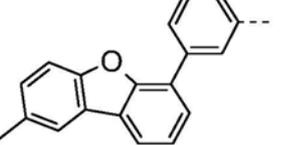
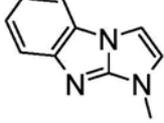
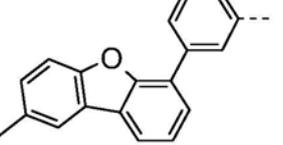
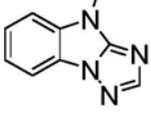
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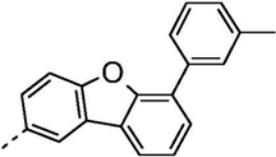
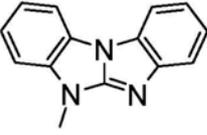
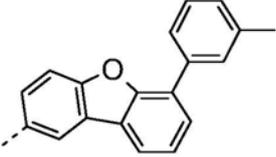
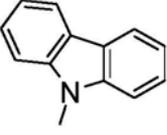
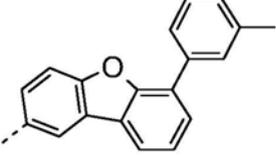
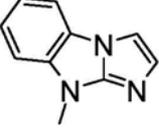
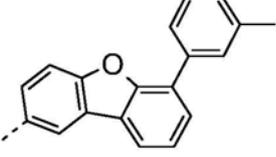
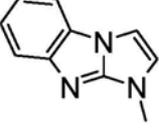
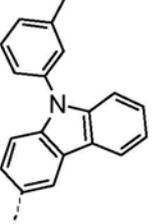
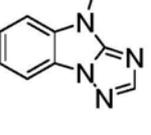
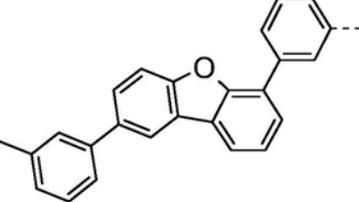
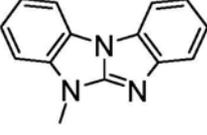
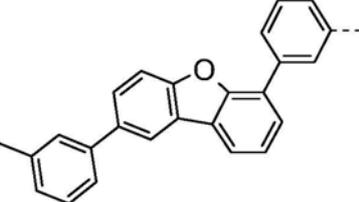
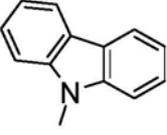
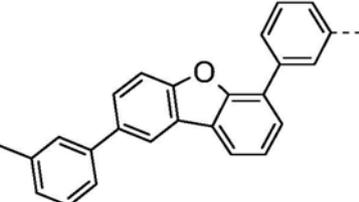
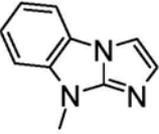
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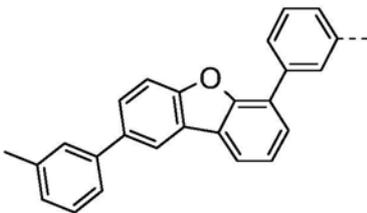
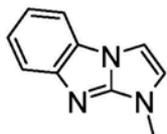
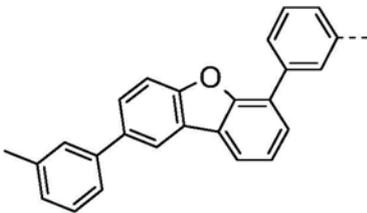
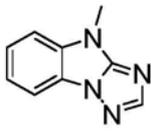
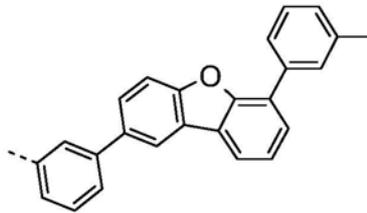
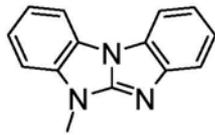
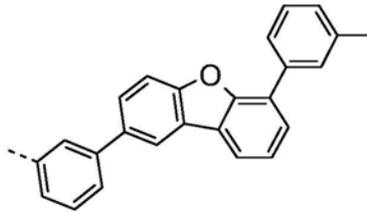
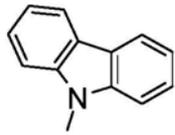
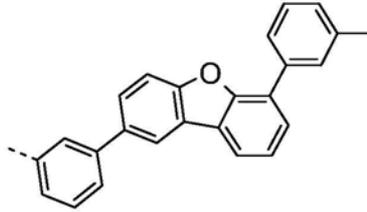
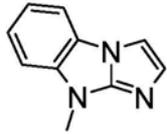
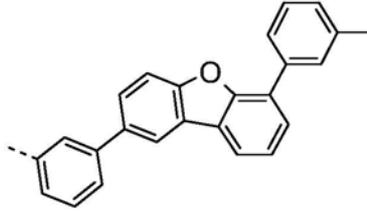
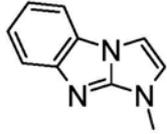
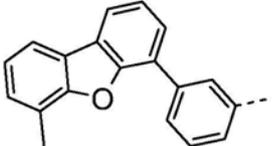
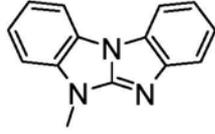
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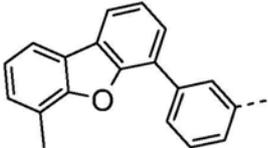
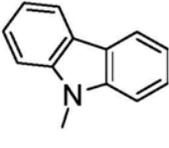
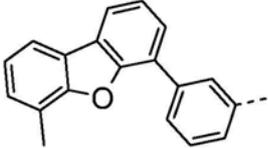
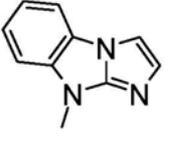
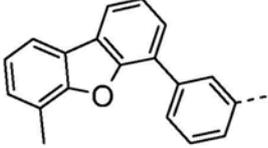
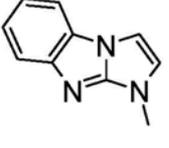
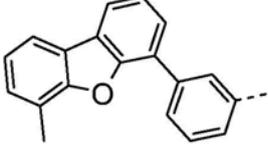
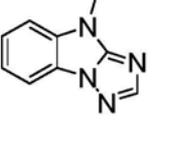
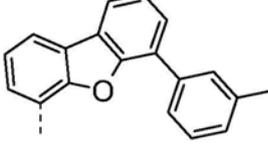
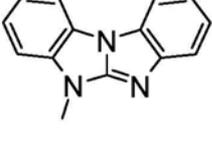
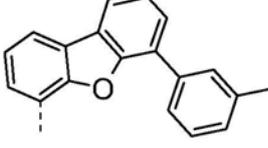
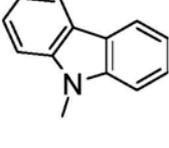
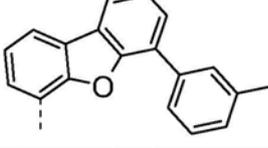
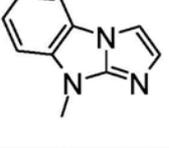
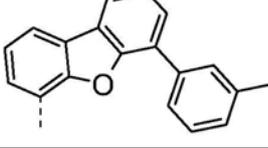
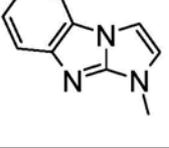
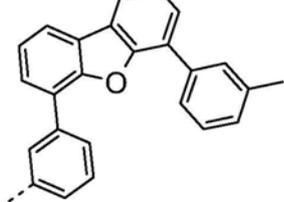
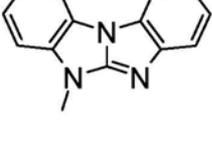
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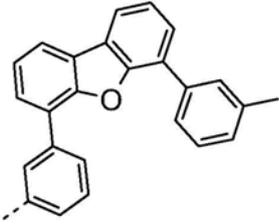
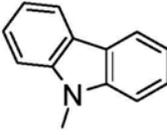
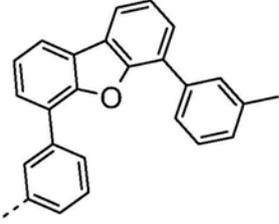
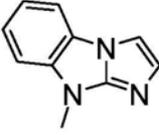
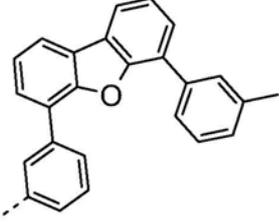
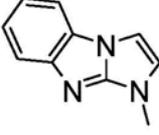
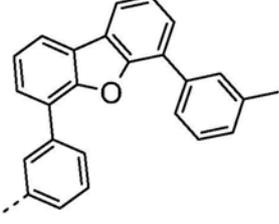
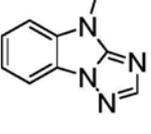
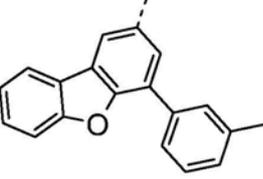
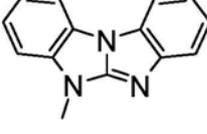
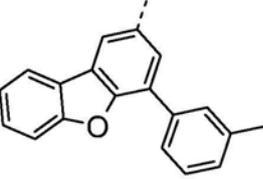
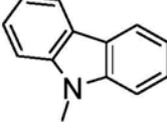
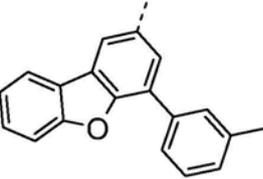
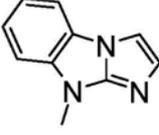
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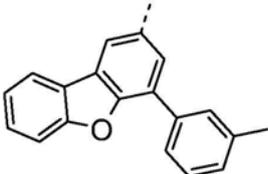
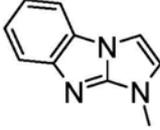
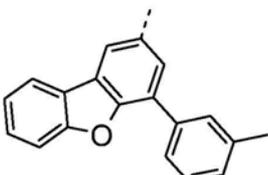
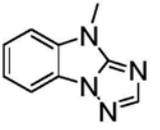
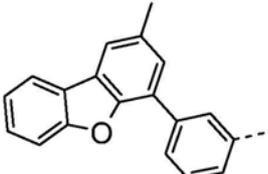
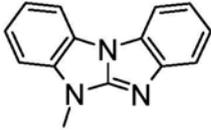
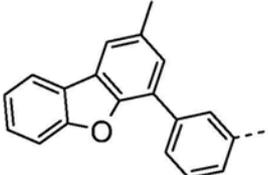
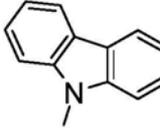
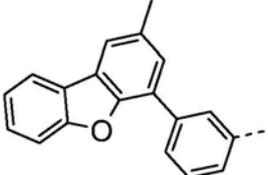
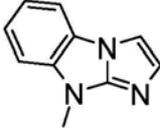
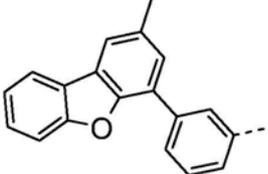
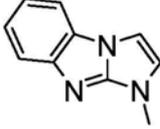
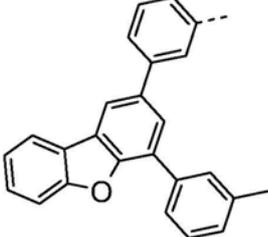
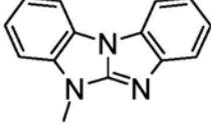
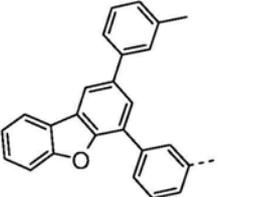
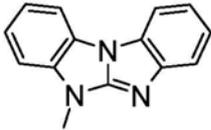
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H-69		

[0289]

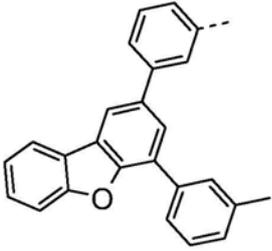
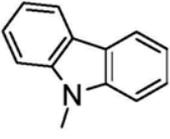
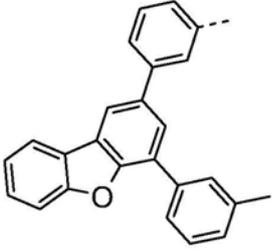
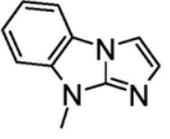
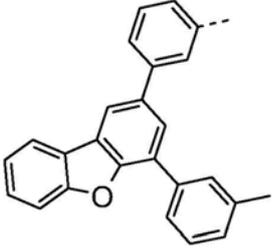
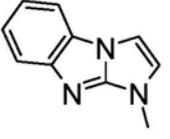
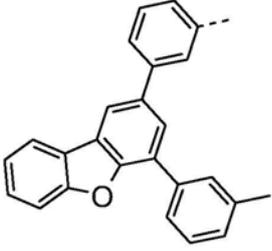
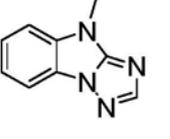
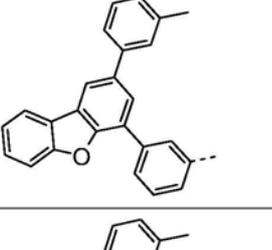
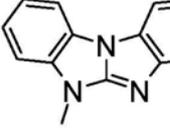
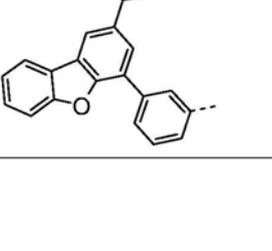
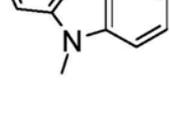
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H-76		

[0290]

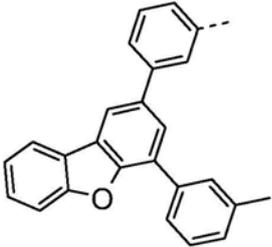
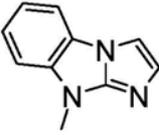
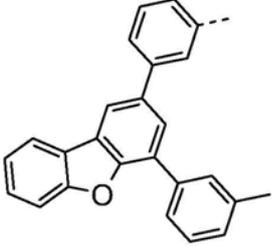
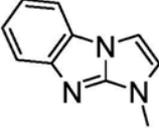
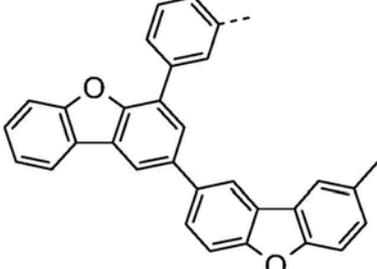
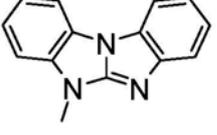
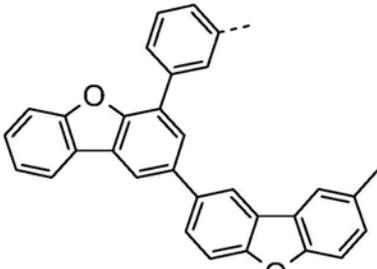
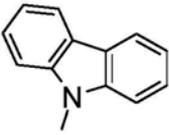
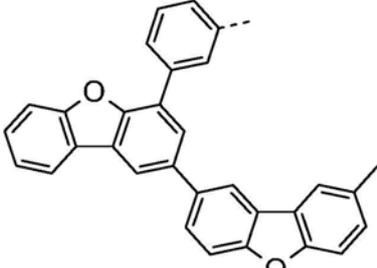
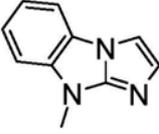
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[0291]

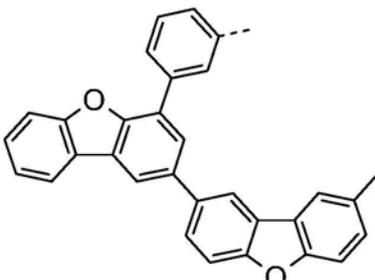
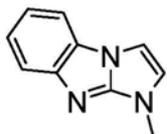
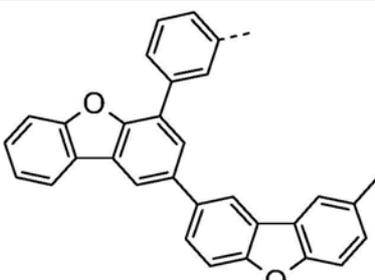
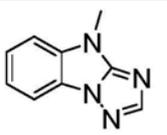
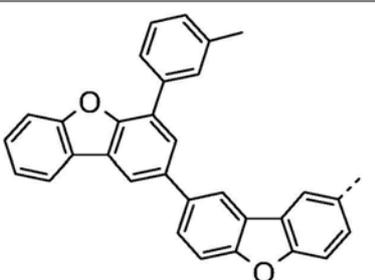
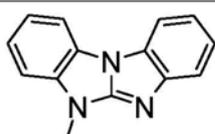
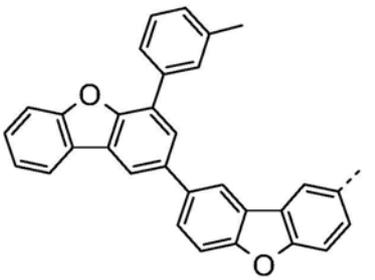
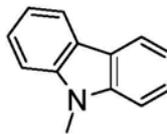
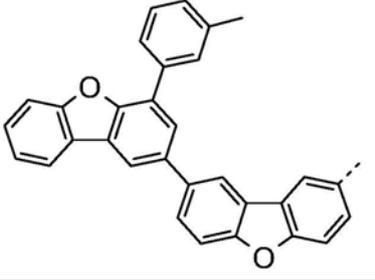
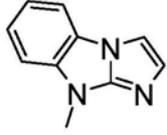
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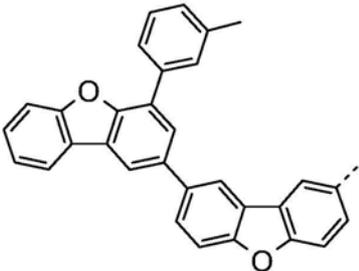
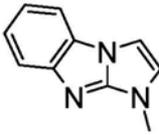
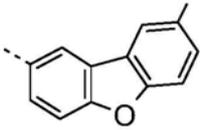
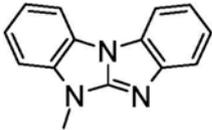
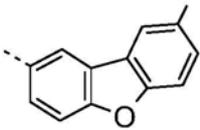
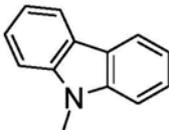
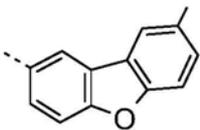
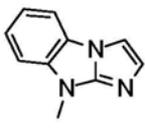
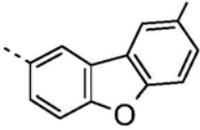
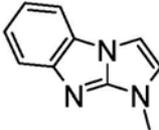
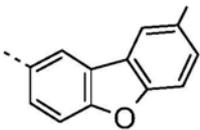
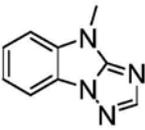
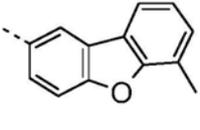
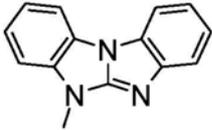
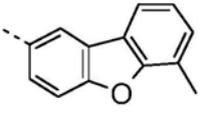
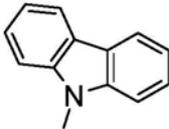
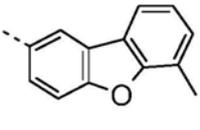
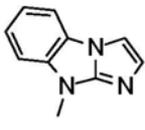
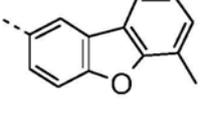
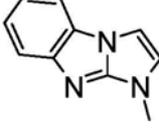
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H-90		

[0293]

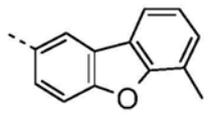
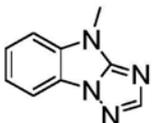
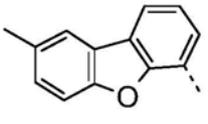
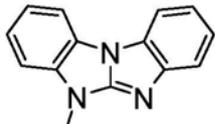
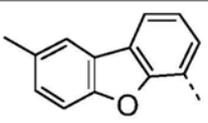
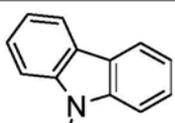
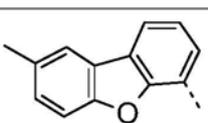
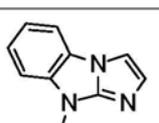
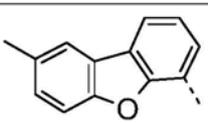
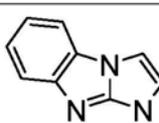
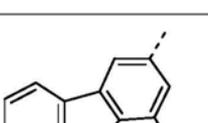
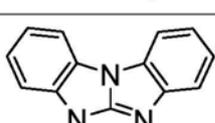
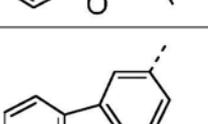
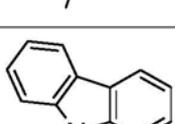
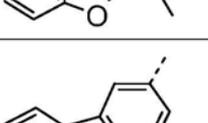
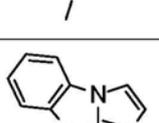
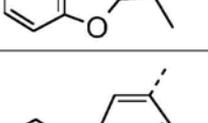
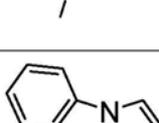
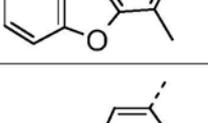
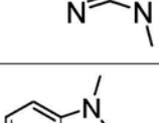
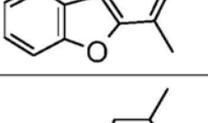
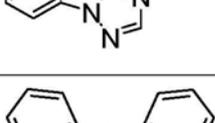
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[0294]

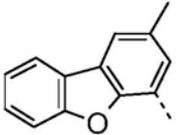
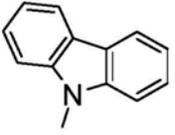
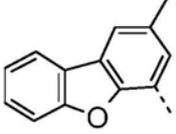
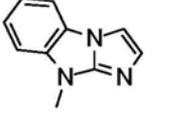
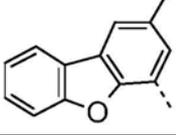
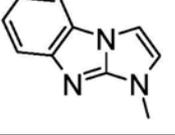
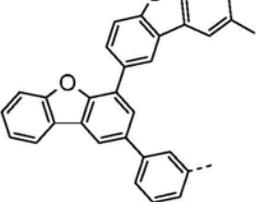
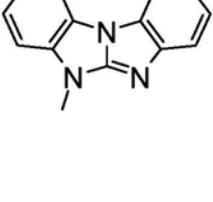
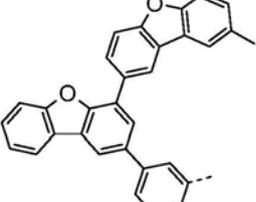
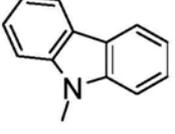
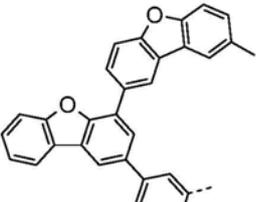
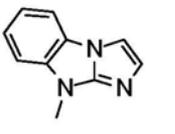
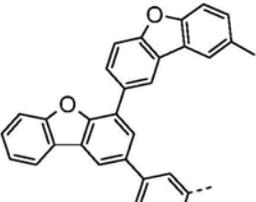
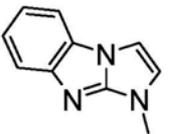
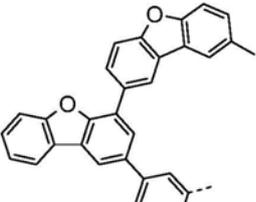
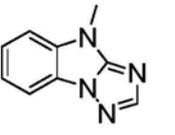
H-96		
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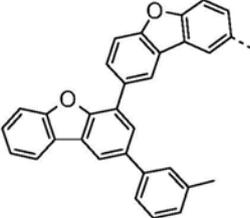
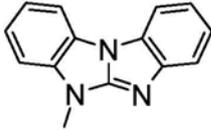
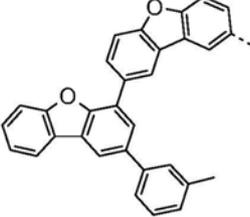
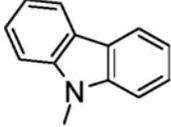
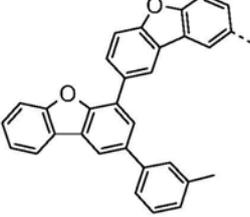
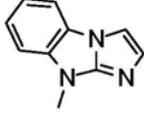
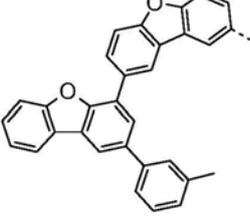
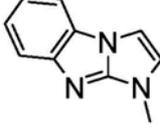
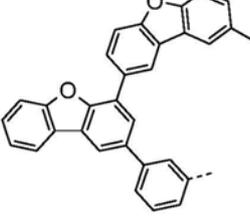
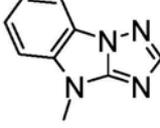
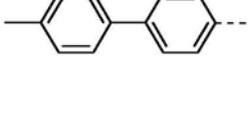
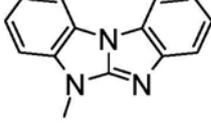
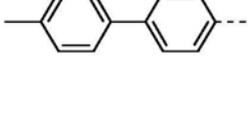
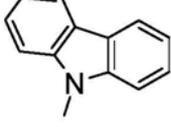
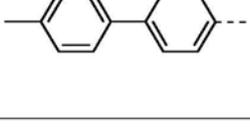
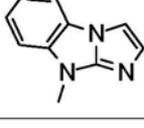
[0295]

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[0296]

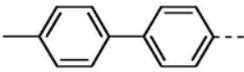
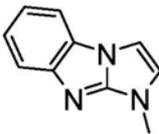
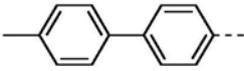
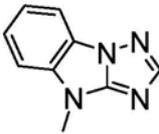
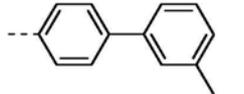
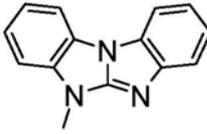
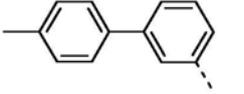
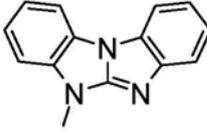
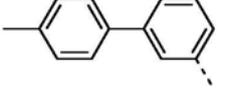
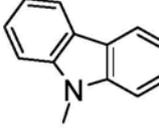
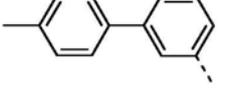
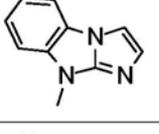
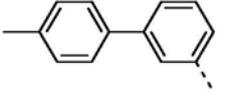
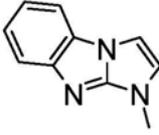
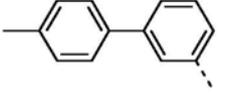
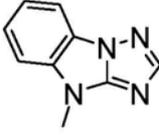
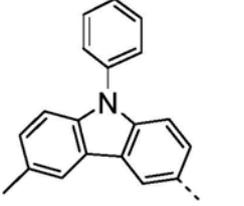
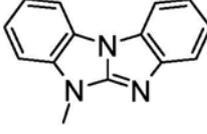
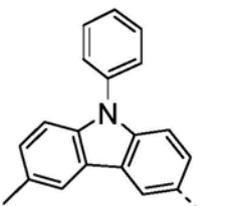
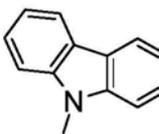
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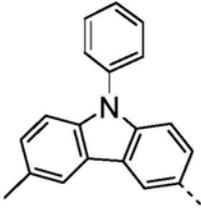
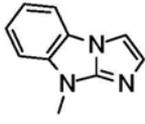
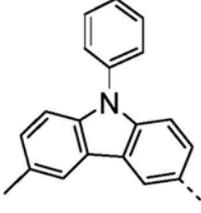
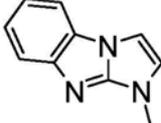
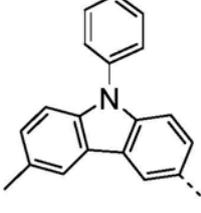
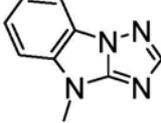
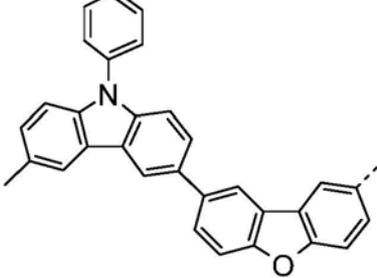
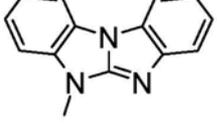
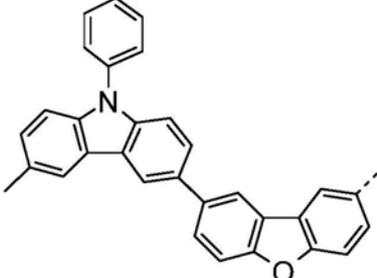
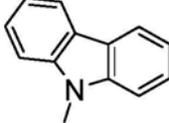
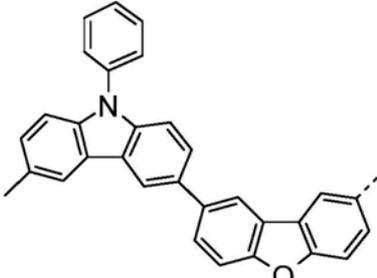
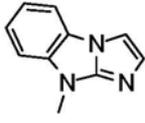
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[0298]

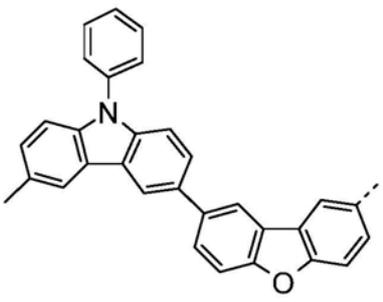
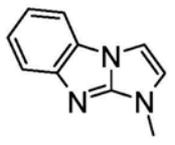
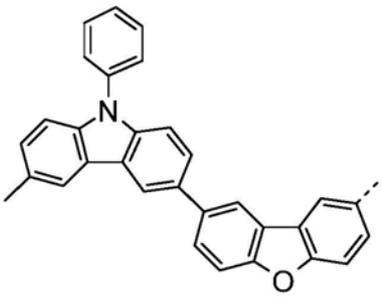
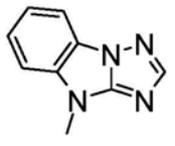
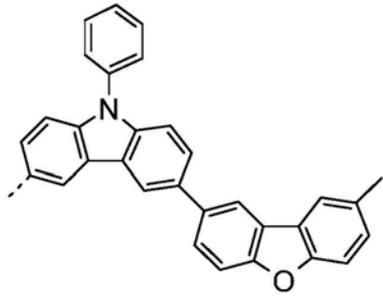
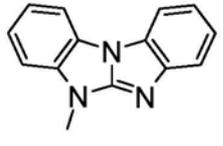
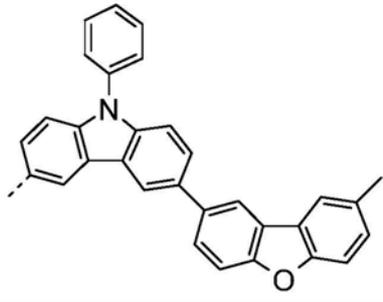
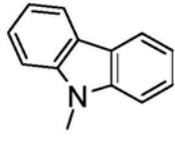
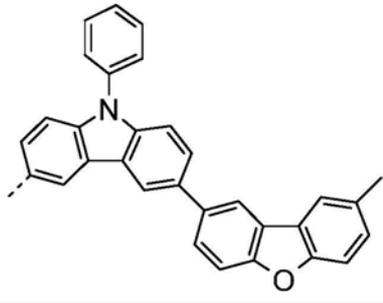
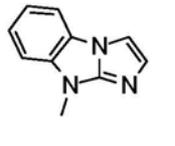
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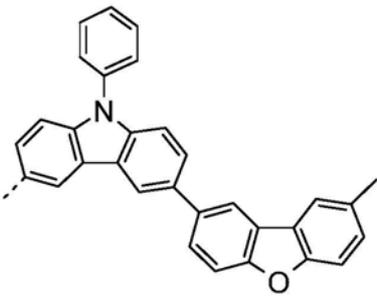
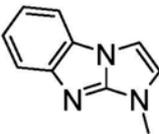
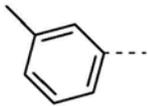
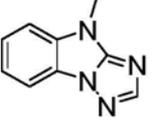
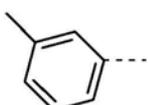
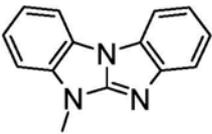
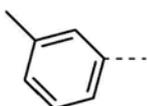
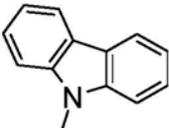
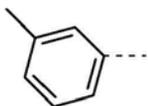
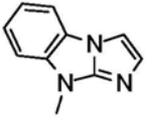
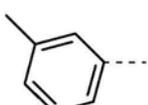
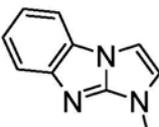
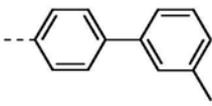
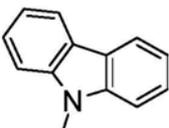
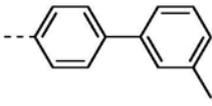
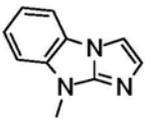
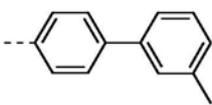
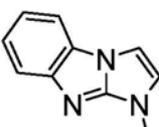
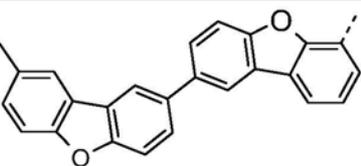
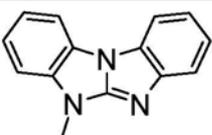
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[0300]

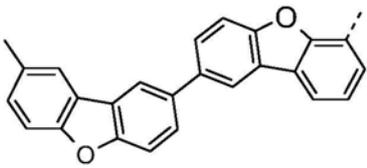
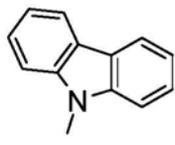
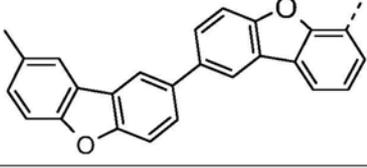
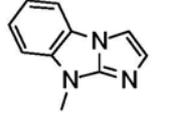
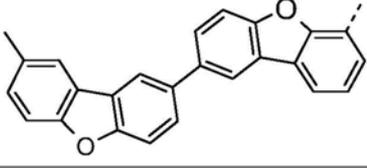
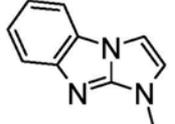
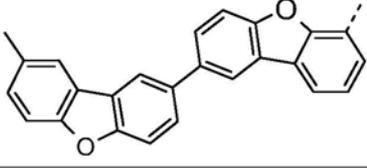
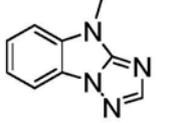
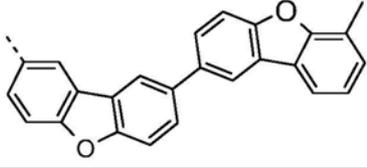
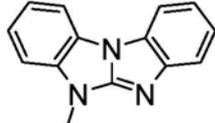
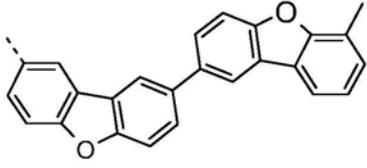
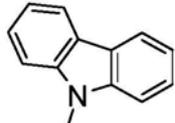
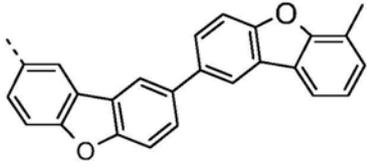
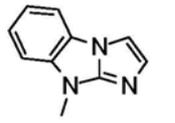
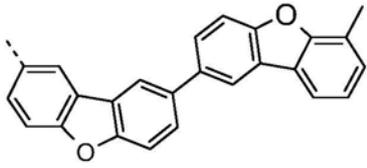
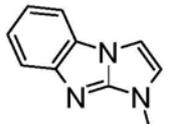
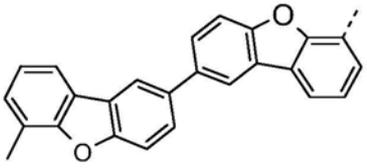
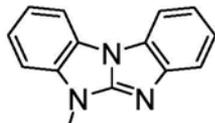
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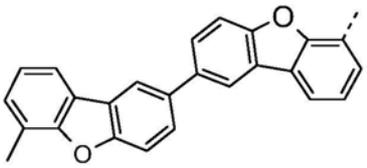
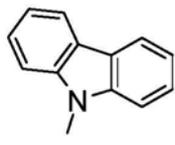
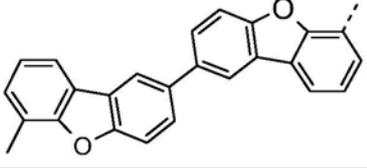
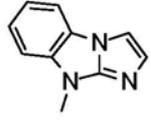
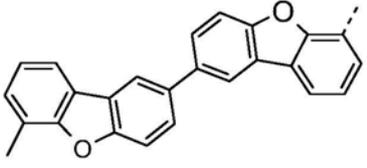
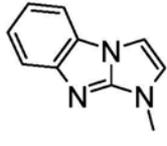
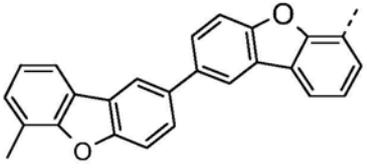
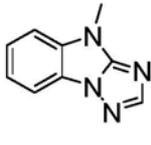
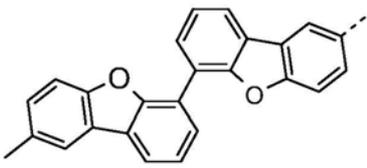
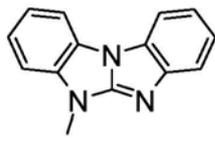
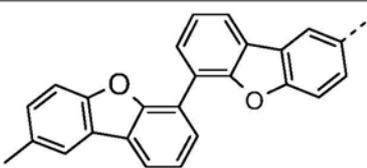
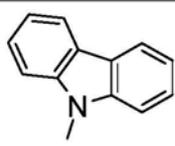
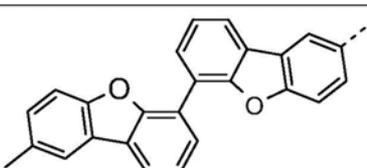
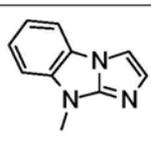
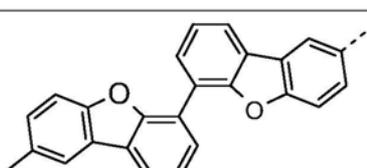
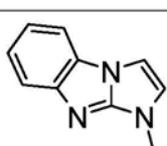
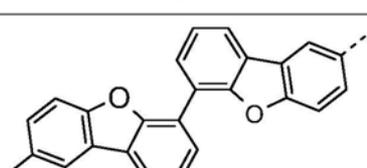
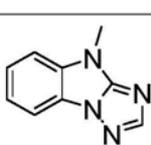
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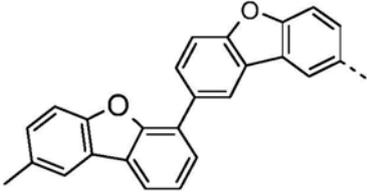
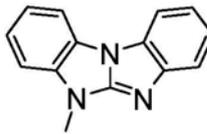
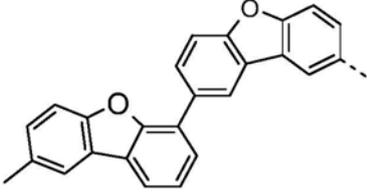
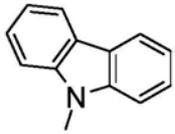
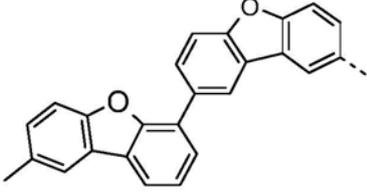
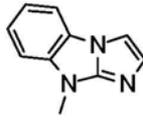
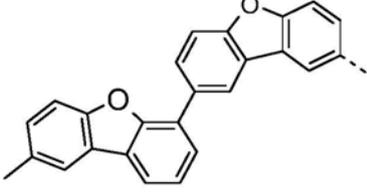
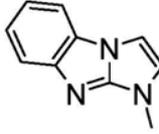
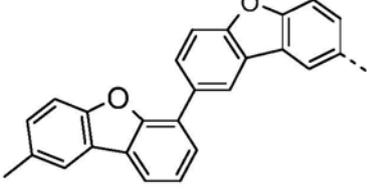
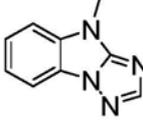
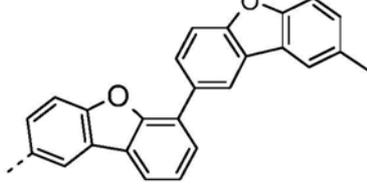
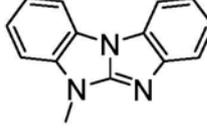
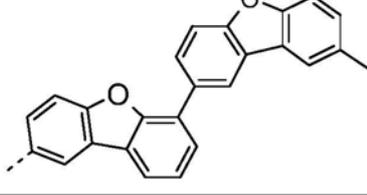
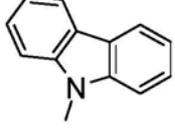
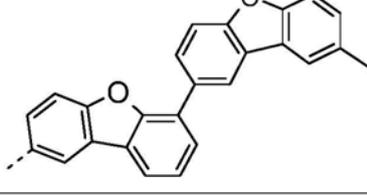
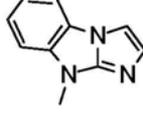
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[0303]

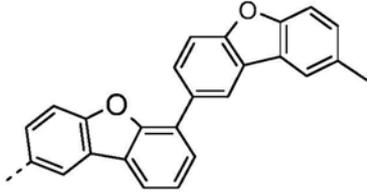
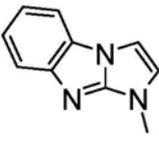
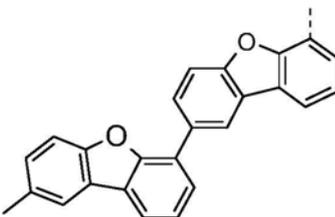
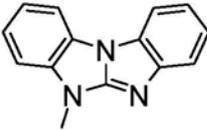
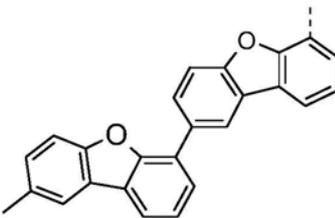
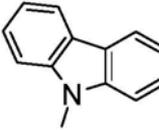
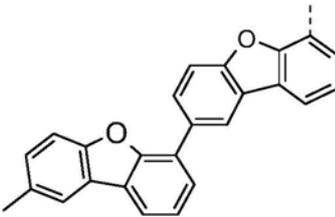
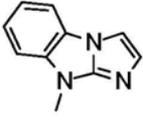
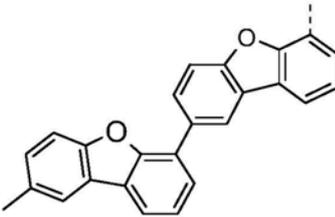
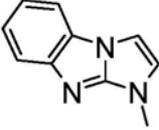
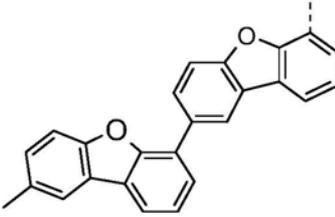
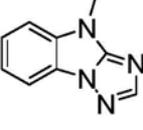
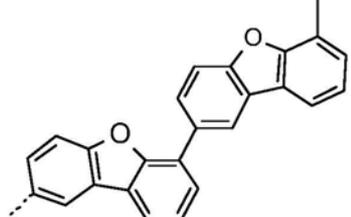
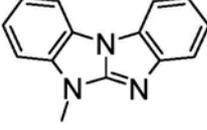
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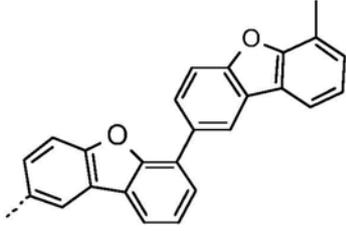
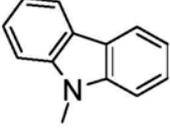
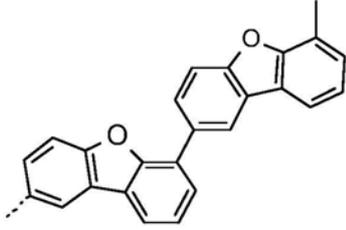
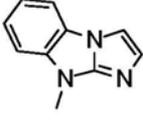
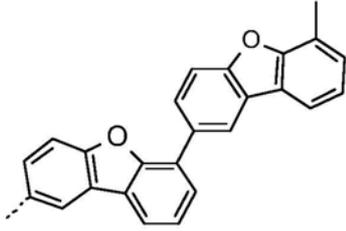
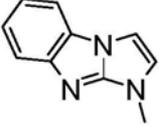
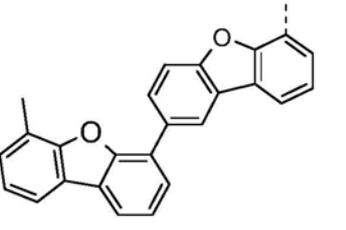
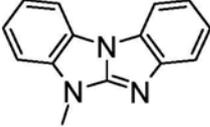
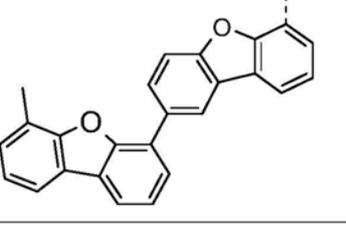
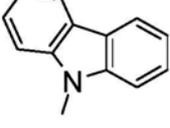
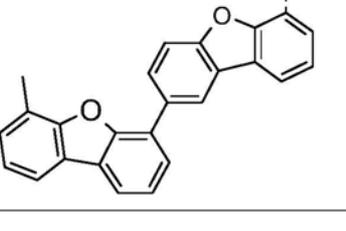
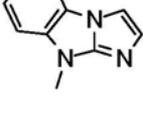
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[0305]

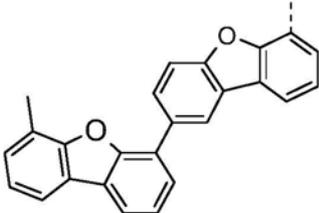
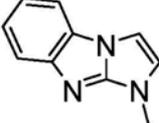
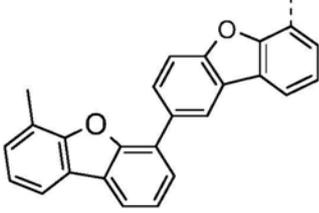
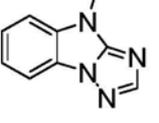
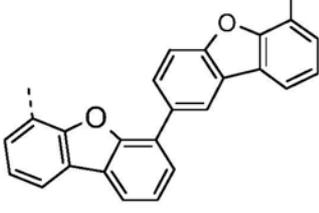
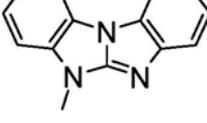
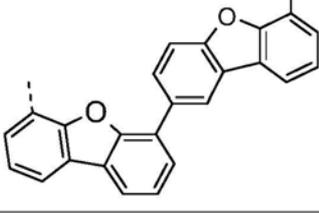
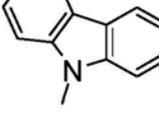
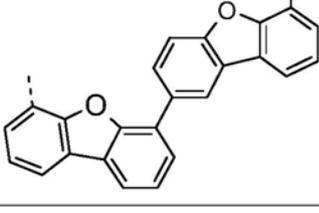
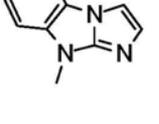
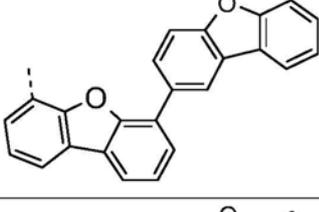
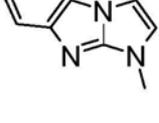
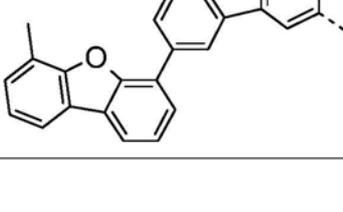
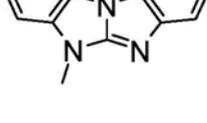
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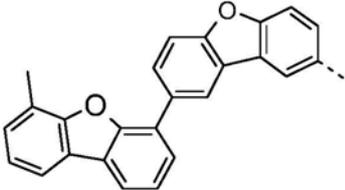
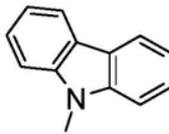
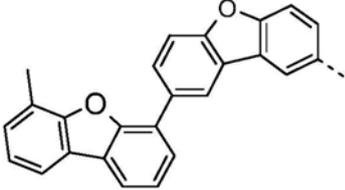
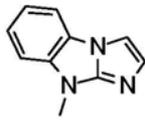
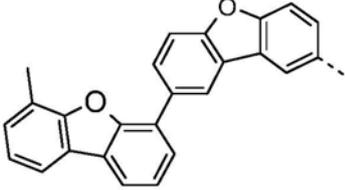
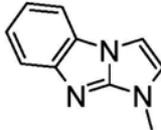
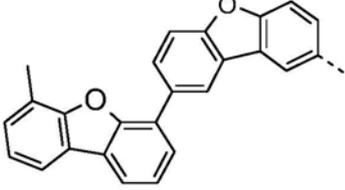
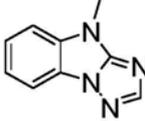
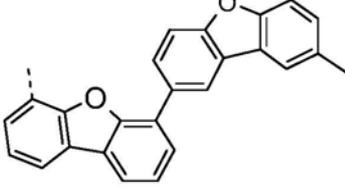
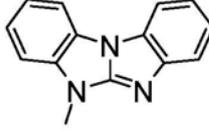
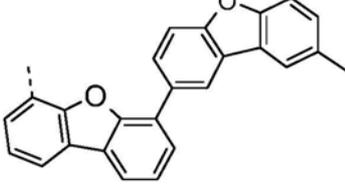
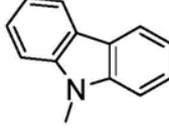
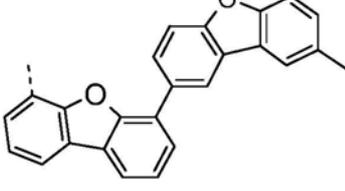
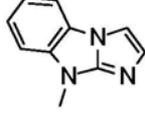
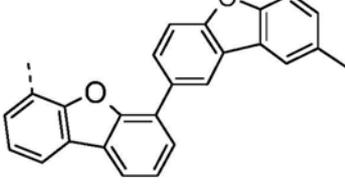
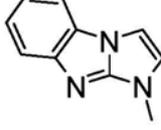
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[0307]

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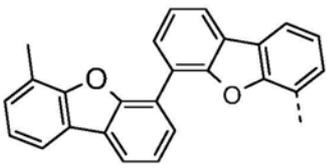
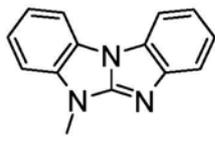
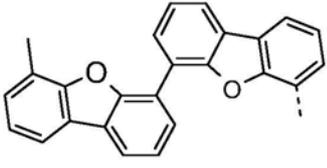
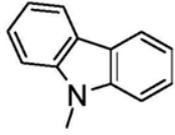
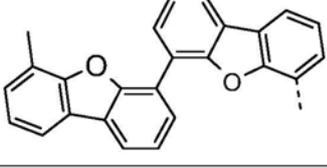
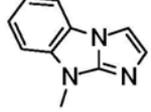
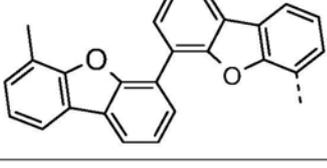
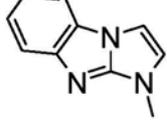
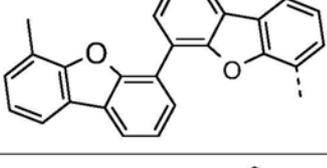
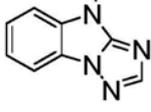
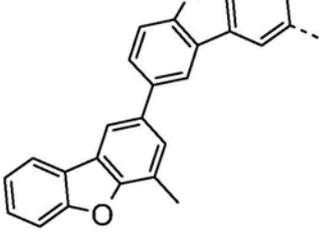
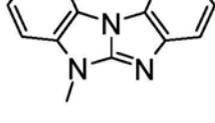
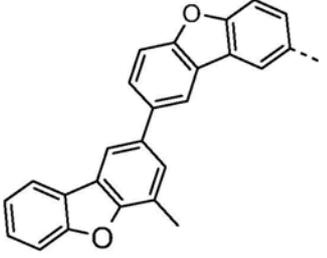
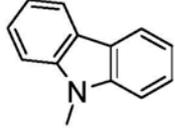
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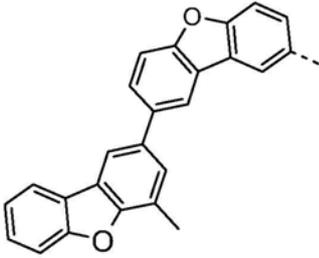
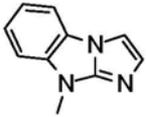
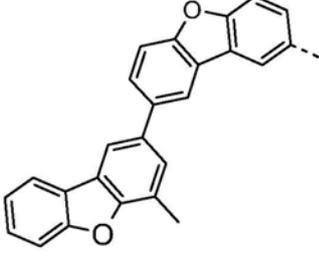
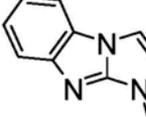
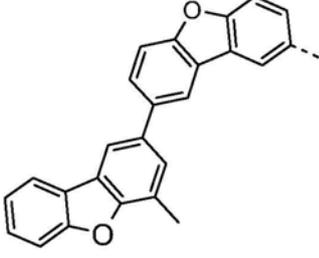
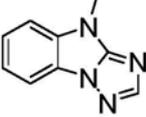
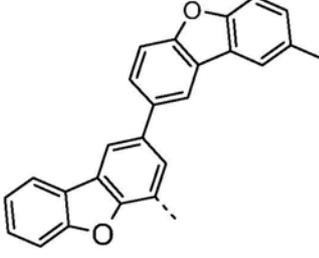
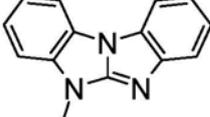
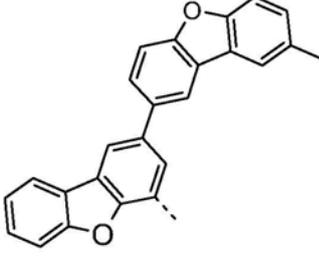
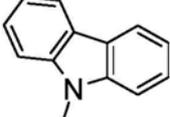
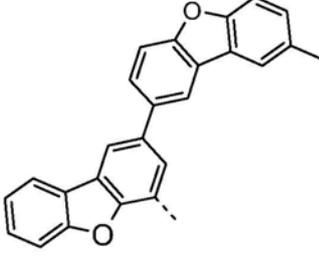
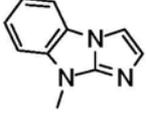
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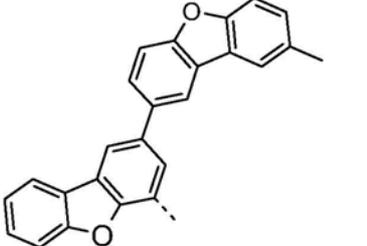
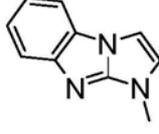
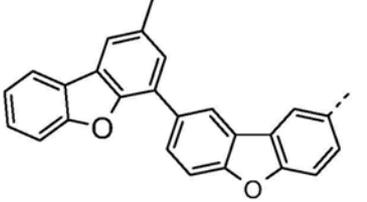
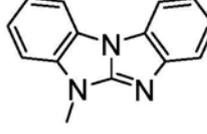
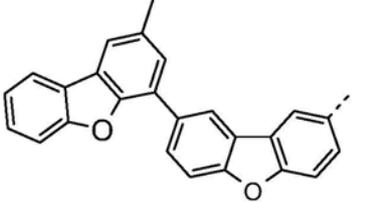
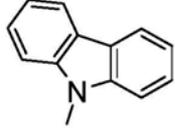
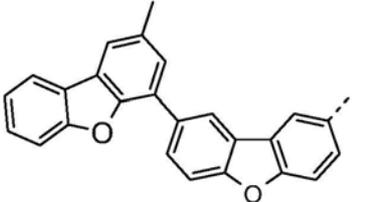
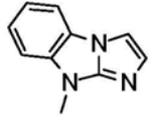
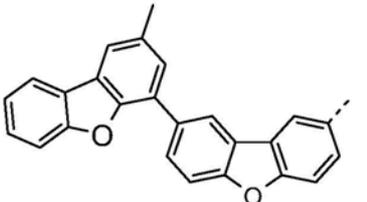
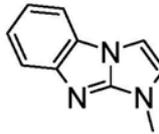
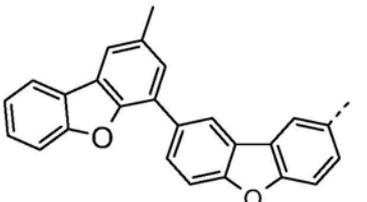
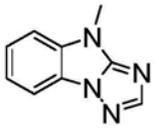
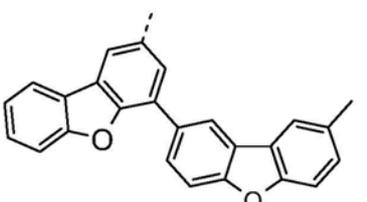
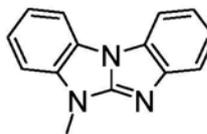
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[0310]

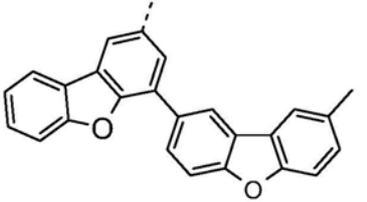
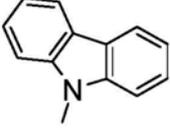
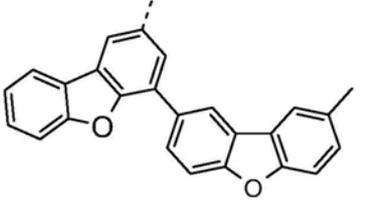
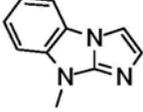
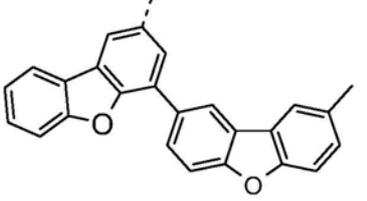
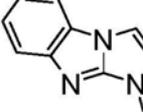
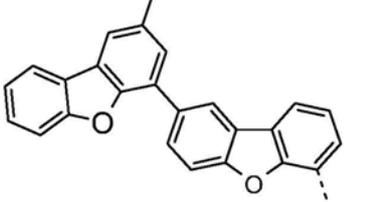
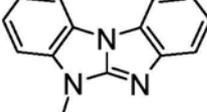
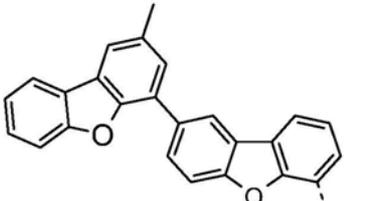
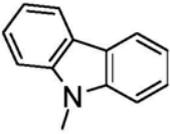
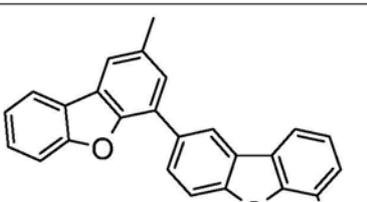
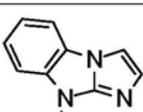
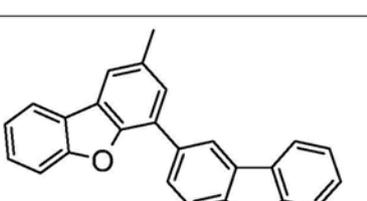
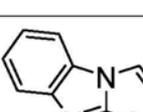
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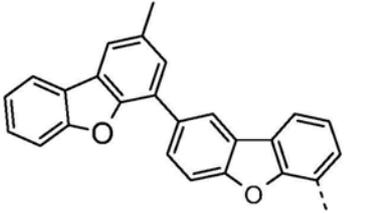
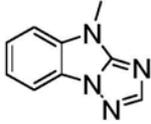
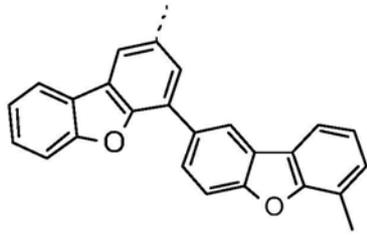
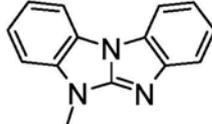
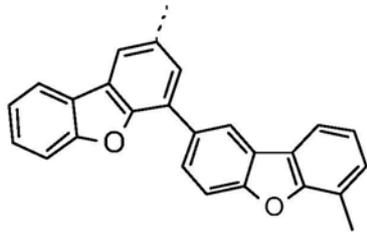
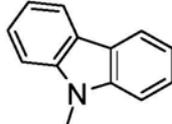
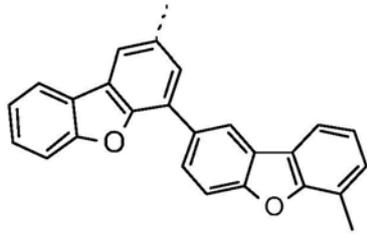
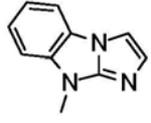
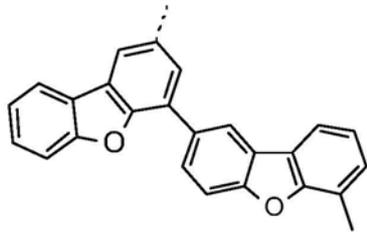
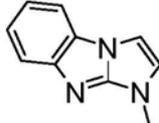
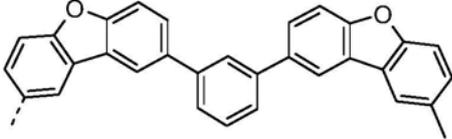
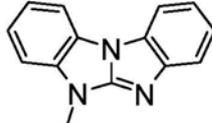
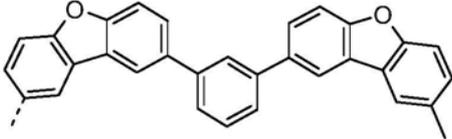
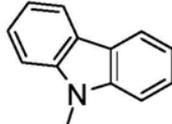
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[0312]

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[0313]

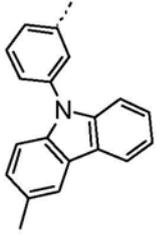
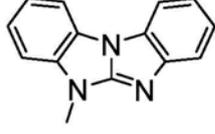
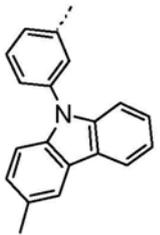
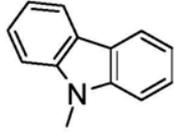
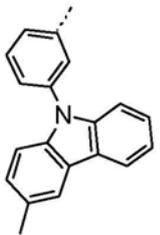
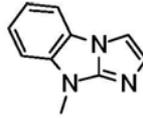
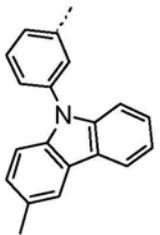
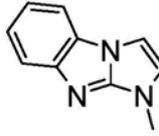
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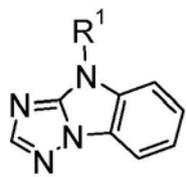
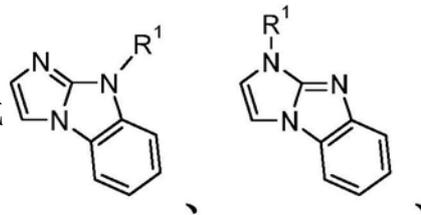
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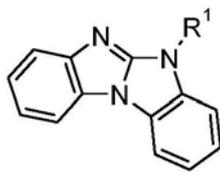
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[0317]

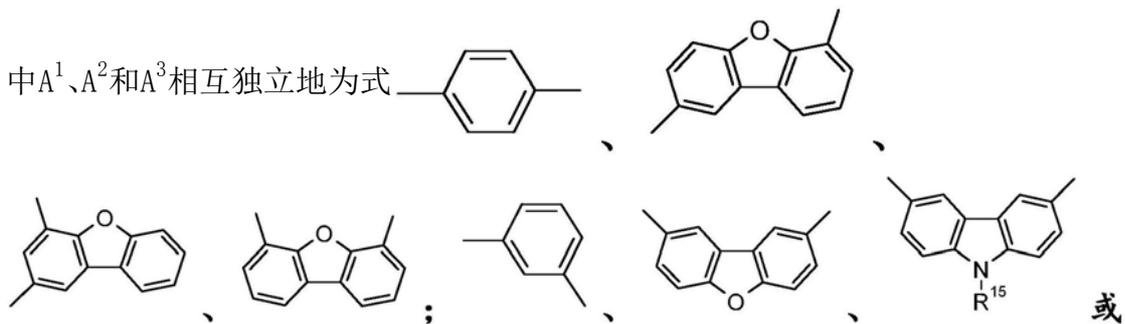
在另一优选实施方案中,本发明涉及式



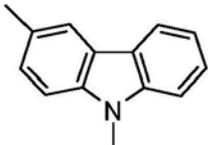
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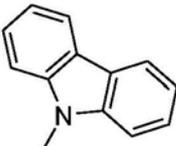
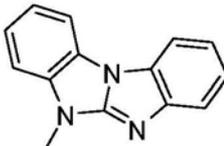


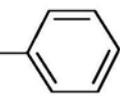
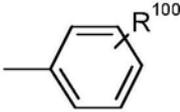
的化合物,其中R¹为式-A¹-(A²)_p-(A³)_q-A⁴-R⁶的基团;其



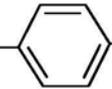
的基团,其中R¹⁵为C₆-C₁₈芳基;或被一个或多个C₁-C₁₈烷基取代的C₆-C₁₈芳基,

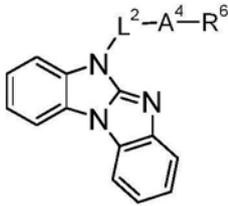
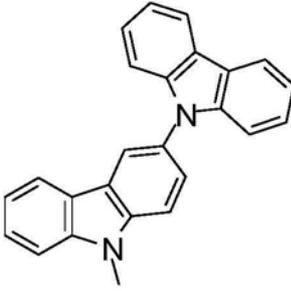
[0318] A⁴为式  的基团,其经由N原子键于A³;

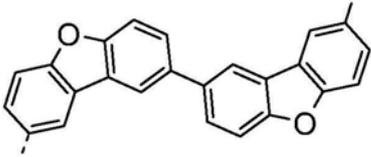
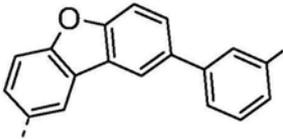
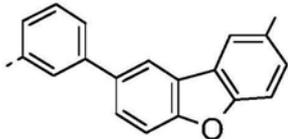
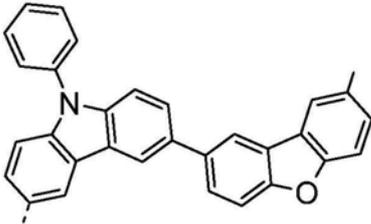
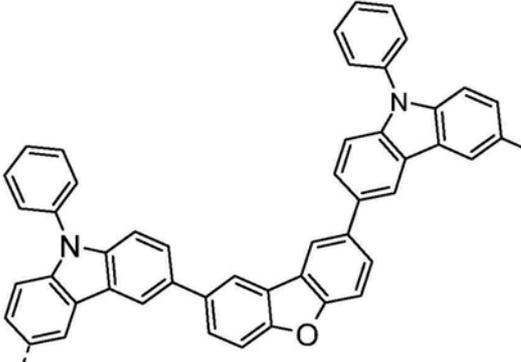
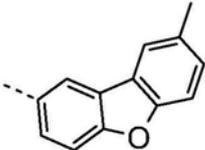
[0319] R⁶为式  或  的基团,且p和q如上所定义。

[0320] R¹⁵优选为式  或  的基团,其中R¹⁰⁰为C₁-C₈烷基。在所述实

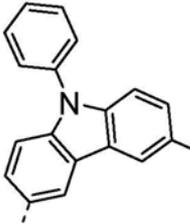
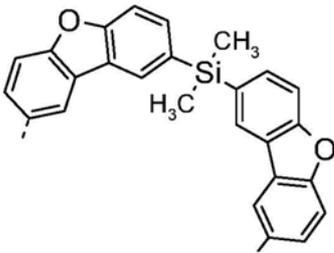
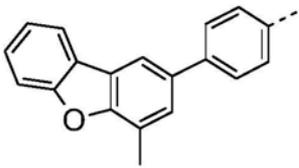
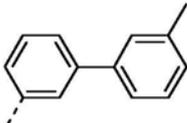
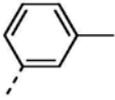
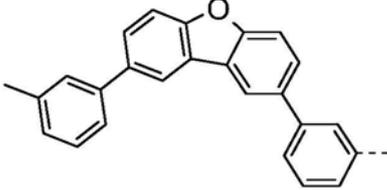
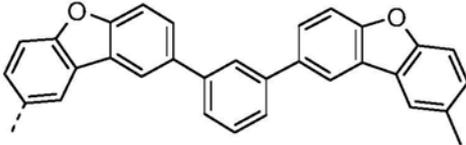
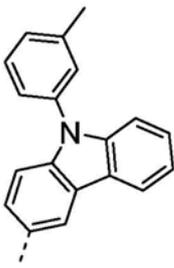
施方案中,式-A¹-(A²)_p-(A³)_q-的基团尤其为式(IVa)、(IVb)、(IVc)、(IVd)、(IVe)、(IVf)、(IVg)、(IVi)、(IVj)、(IVk)、(IVl)、(IVm)、(IVn)、(IVo)、(IVp)、(IVq)、(IVr)、(IVs)、(IVt)、(IVu)、(IVv)、(IVw)、(IVx)、(IVy)、(IVz)、(VIa)、(VIb)、(VIc)、(VI d)、(VIe)、(VIf)、(VIg)、(VIh)、(VIi)、(VIj)、(VIk)、(VI l)、(VI m)、(VI n)、(VI o)、(VI p)、(VI q)、(VI r)、(VI s)、(VI t)或(VI u)的基团。目前最优选式-A¹-(A²)_p-(A³)_q-的基团为式(IVa)、(IVb)、(IVe)、(IVl)、(IVk)、(IVs)、(IVv)和(VIj)的基团。

[0321] 在将式I化合物在蓝色或绿色磷光发光体中用作主体材料或用作电子/激子阻断材料的情况下,较不优选如下基团A¹-(A²)_p-(A³)_q-(A⁴)_r-,其含有式  的基团,即式(IVp)和(IVr)的基团。优选的化合物的实例为显示在下表中的化合物F-1至F-62以及G-1至G-62。

[0322]  (-A⁴-R⁶= )

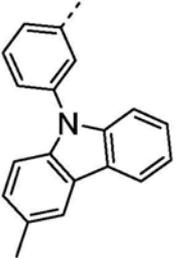
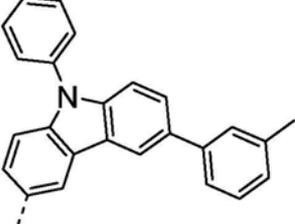
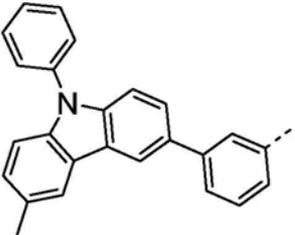
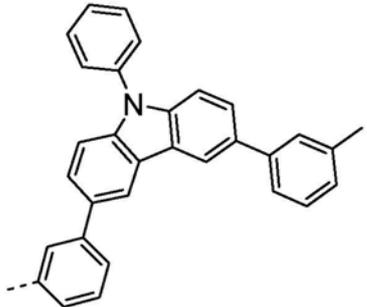
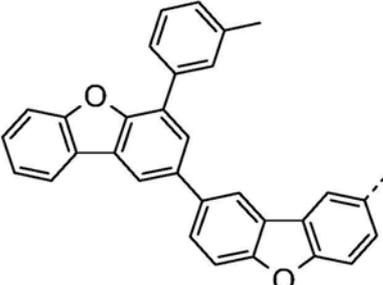
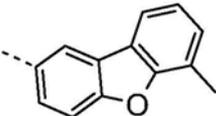
Cpd.	$L^{22)}$
F-1	
F-2	
F-3	
F-4	
F-5	
F-6	

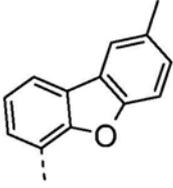
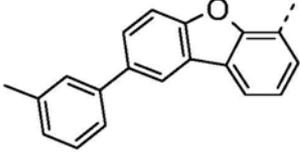
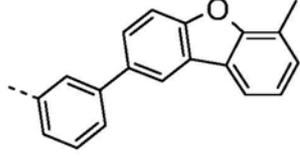
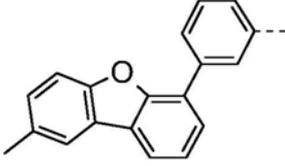
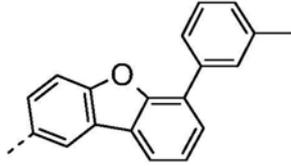
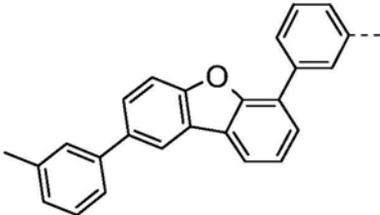
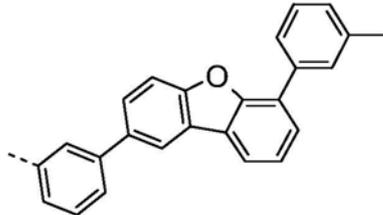
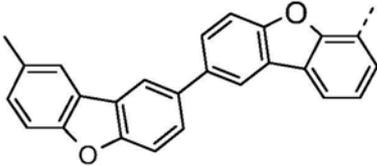
[0323]

F-7	
F-8	
F-9	
F-10	
F-11	
F-12	
F-13	
F-14	

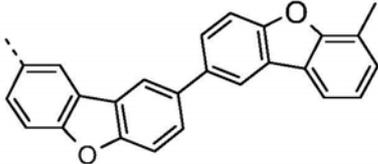
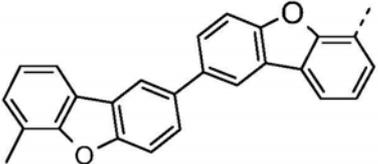
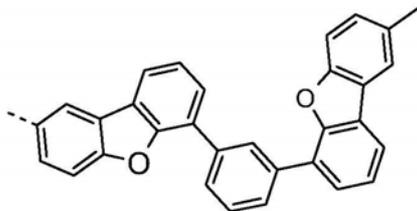
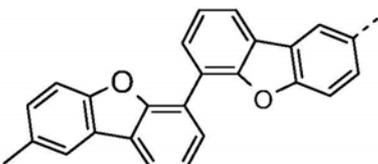
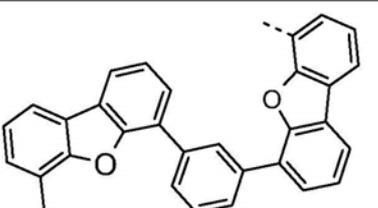
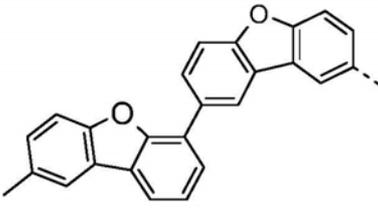
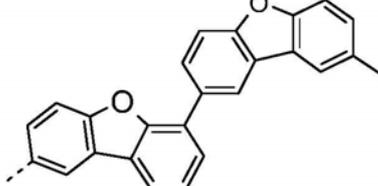
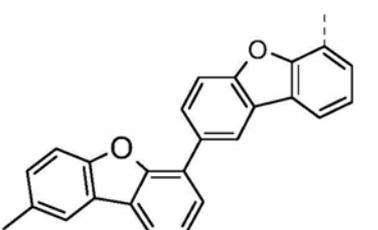
[0324]

[0325]

<p>F-15</p>	
<p>F-16</p>	
<p>F-17</p>	
<p>F-18</p>	
<p>F-19</p>	
<p>F-20</p>	

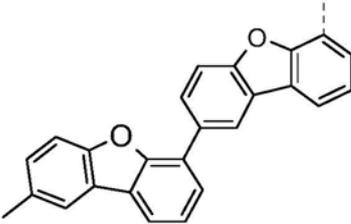
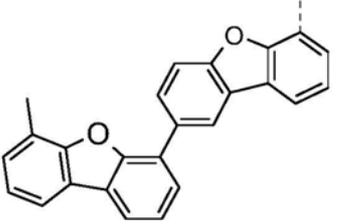
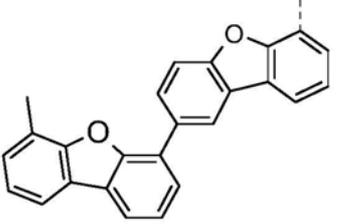
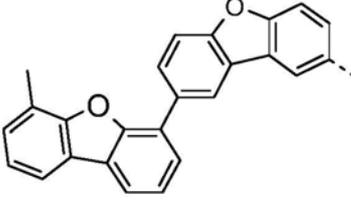
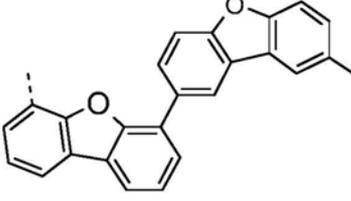
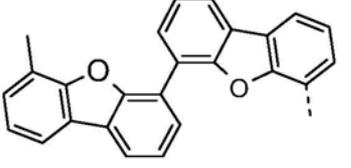
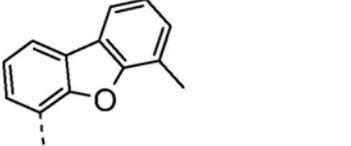
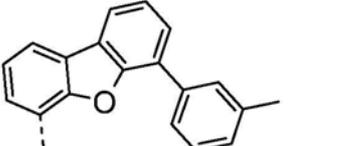
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F-22	
F-23	
F-24	
F-25	
F-26	
F-27	
F-28	

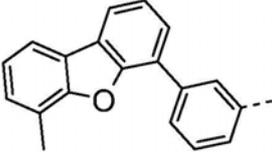
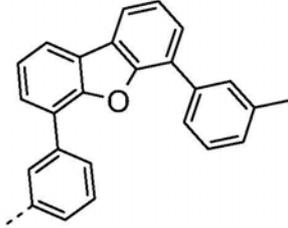
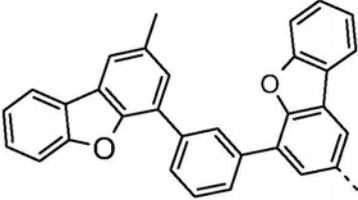
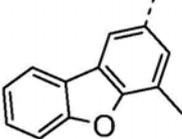
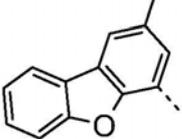
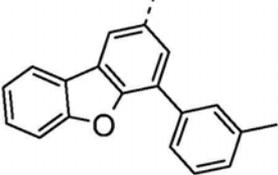
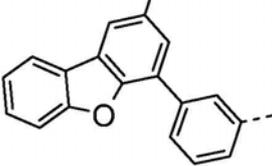
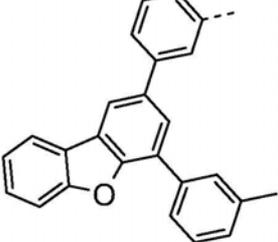
[0326]

F-29	
F-30	
F-31	
F-32	
F-33	
F-34	
F-35	
F-36	

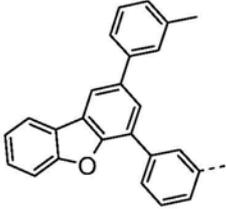
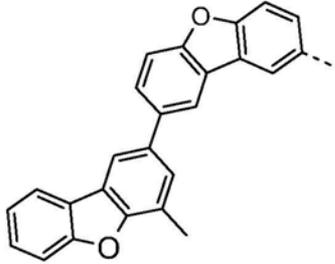
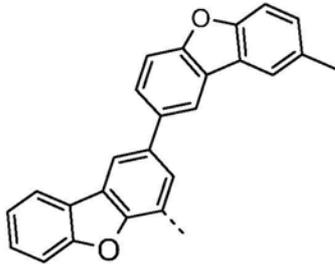
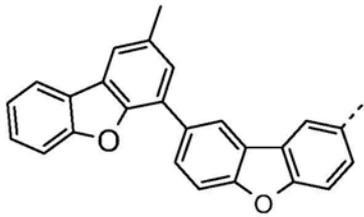
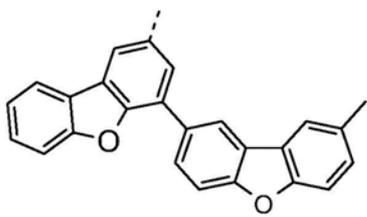
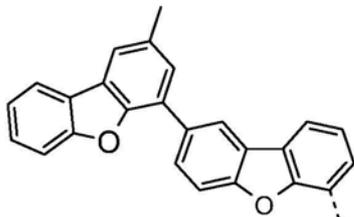
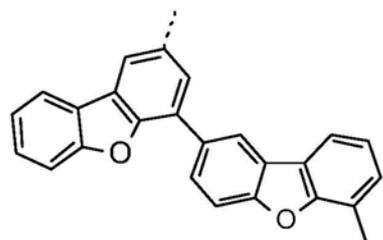
[0327]

[0328]

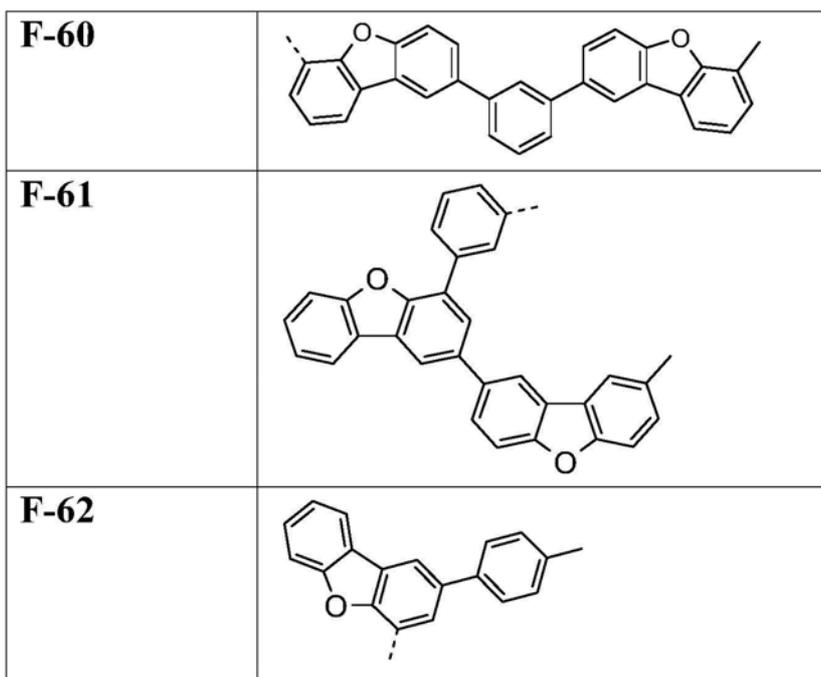
<p>F-37</p>	
<p>F-38</p>	
<p>F-39</p>	
<p>F-40</p>	
<p>F-41</p>	
<p>F-42</p>	
<p>F-43</p>	
<p>F-44</p>	

F-45	
F-46	
F-47	
F-48	
[0329] F-49	
F-50	
F-51	
F-52	

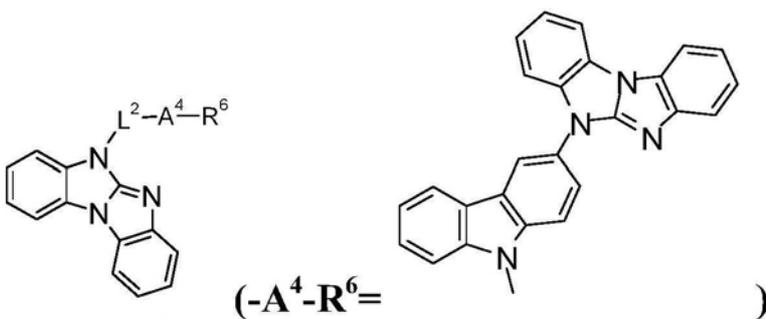
[0330]

<p>F-53</p>	
<p>F-54</p>	
<p>F-55</p>	
<p>F-56</p>	
<p>F-57</p>	
<p>F-58</p>	
<p>F-59</p>	

[0331]

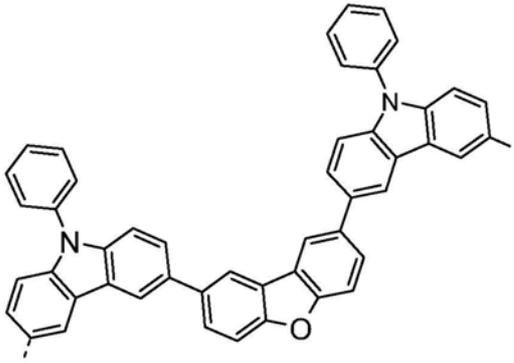
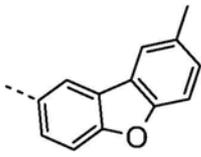
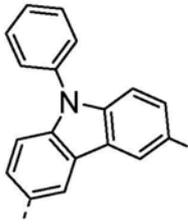
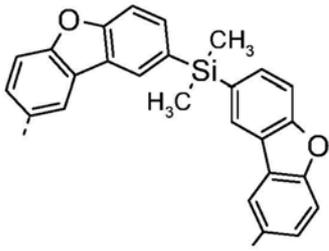
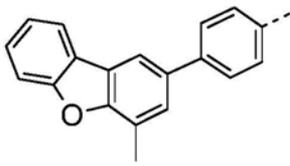
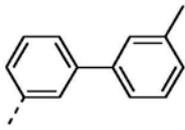
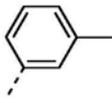
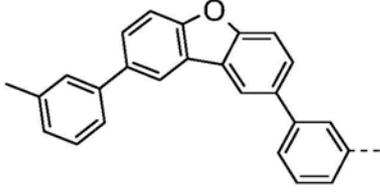


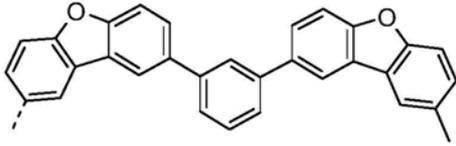
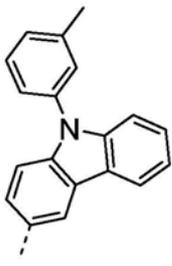
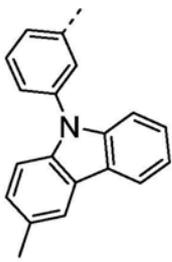
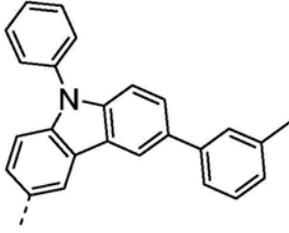
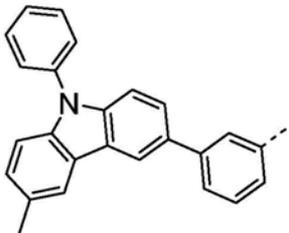
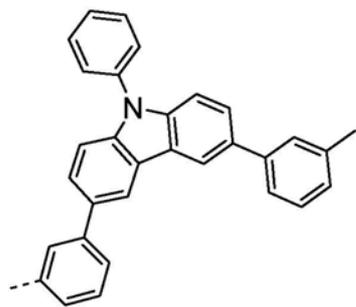
[0332]



[0333]

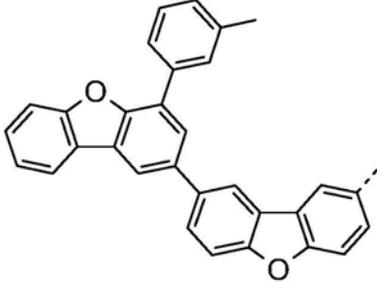
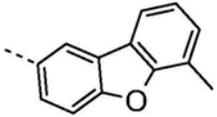
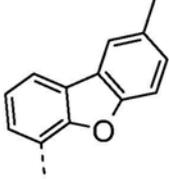
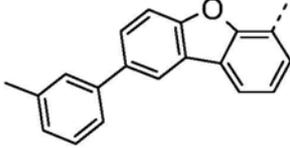
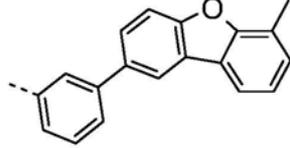
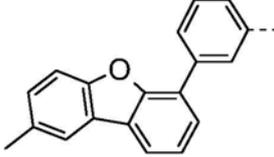
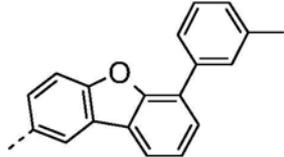
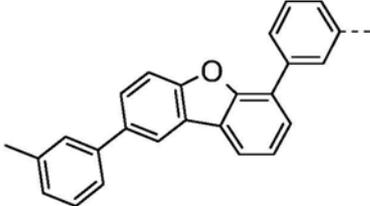
Cpd.	L ²²
G-1	
G-2	
G-3	
G-4	

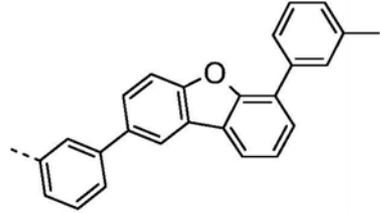
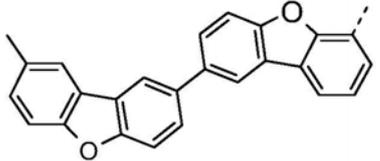
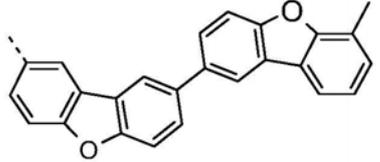
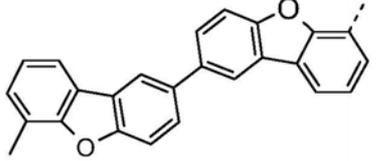
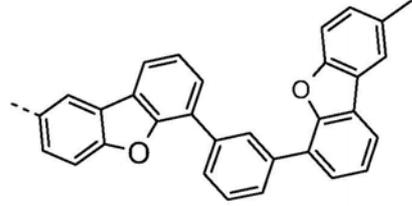
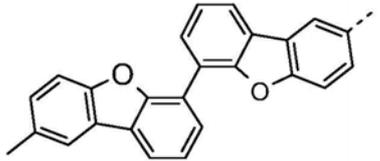
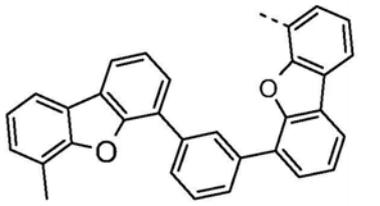
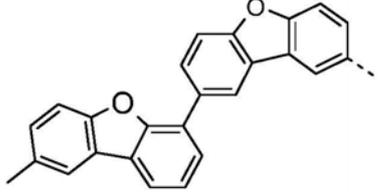
G-5	
G-6	
G-7	
[0334] G-8	
G-9	
G-10	
G-11	
G-12	

G-13	
G-14	
G-15	
G-16	
G-17	
G-18	

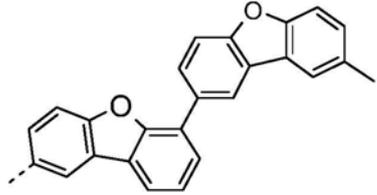
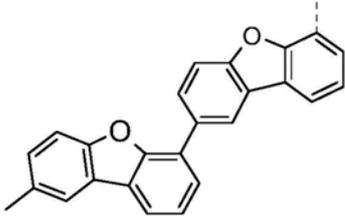
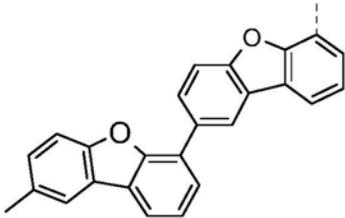
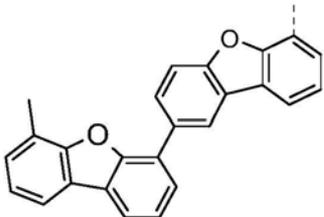
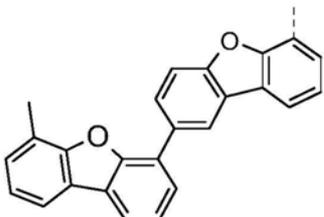
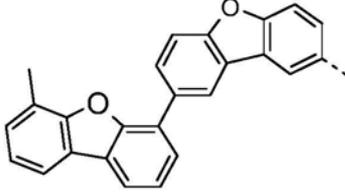
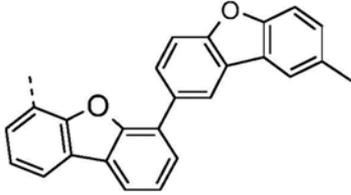
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[0336]

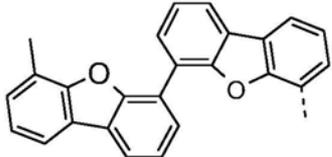
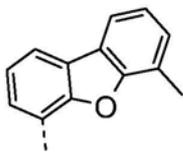
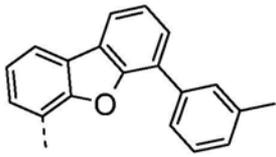
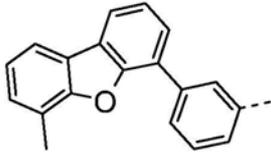
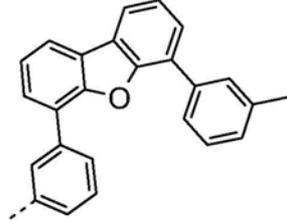
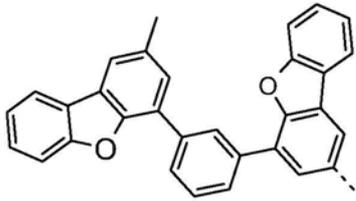
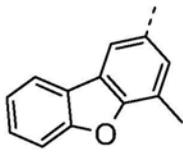
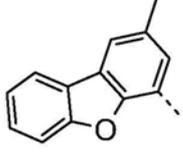
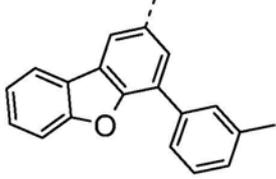
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<p>G-24</p>	
<p>G-25</p>	
<p>G-26</p>	

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G-33	
G-34	

[0337]

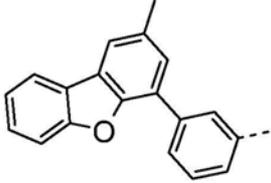
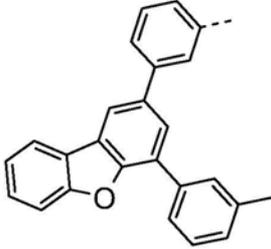
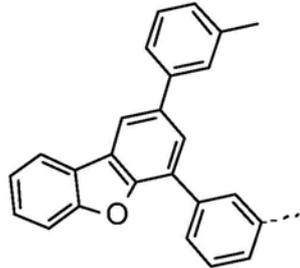
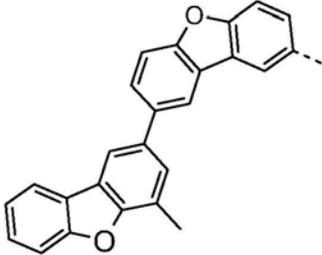
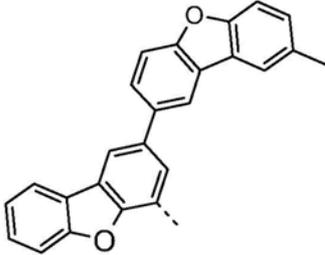
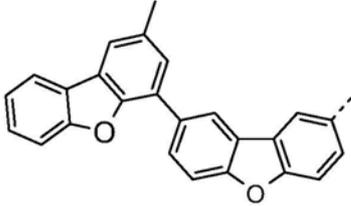
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G-37	
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G-39	
G-40	
G-41	

[0338]

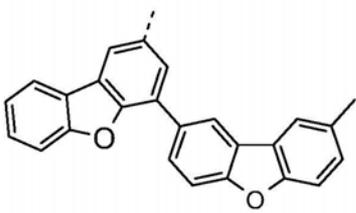
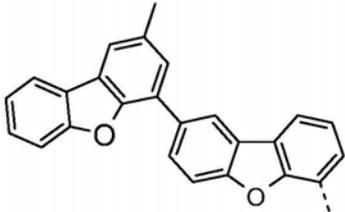
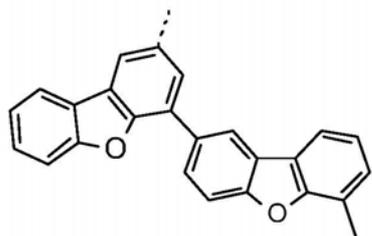
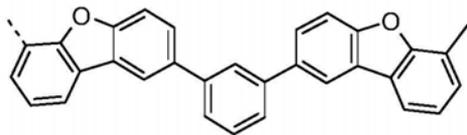
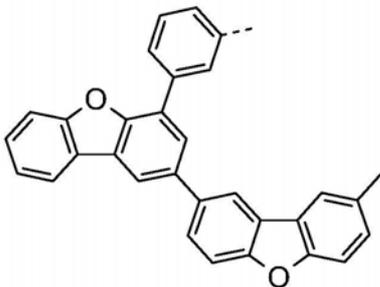
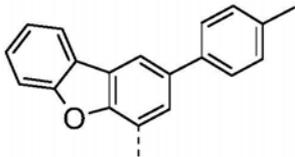
G-42	
G-43	
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G-45	
G-46	
G-47	
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G-49	
G-50	

[0339]

[0340]

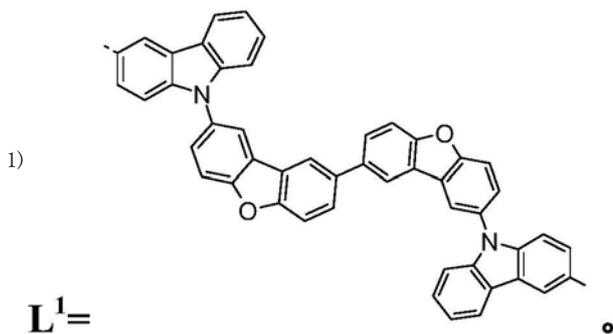
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G-56	

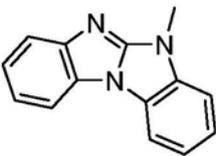
[0341]

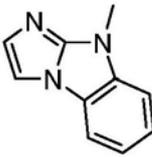
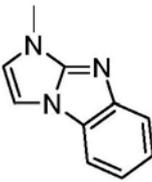
G-57	
G-58	
G-59	
G-60	
G-61	
G-62	

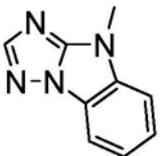
[0342] 在以上表格中,标记¹⁾、²⁾、³⁾和⁴⁾具有如下含义:

[0343]

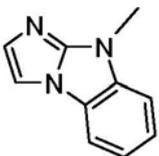
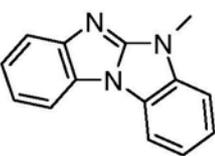


[0344] ²⁾ 虚线表示与式  的基团的键。

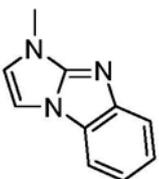
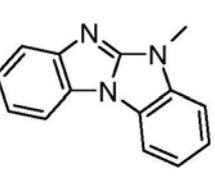
[0345] ³⁾ 虚线表示分别与式  和  的基团的键。

[0346] ⁴⁾ 虚线表示与式  的基团的键。

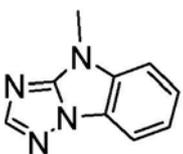
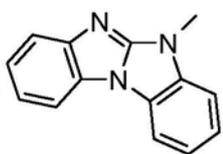
[0347] 在另一优选实施方案中,本发明涉及化合物M-1至M-62,其由化合物F-1至F-62通

过由式  的基团替换式  的基团得到。

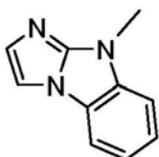
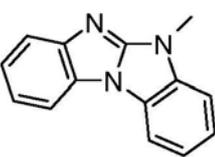
[0348] 在另一优选实施方案中,本发明涉及化合物N-1至N-62,其由化合物F-1至F-62通

过由式  的基团替换式  的基团得到。

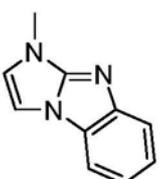
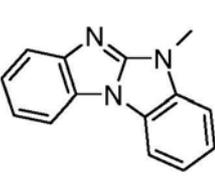
[0349] 在另一优选实施方案中,本发明涉及化合物O-1至O-62,其由化合物F-1至F-62通

过由式  的基团替换式  的基团得到。

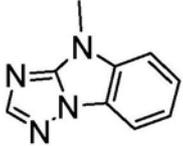
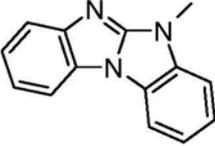
[0350] 在另一优选实施方案中,本发明涉及化合物P-1至P-62,其由化合物G-1至G-62通

过由式  的基团替换式  的基团得到。

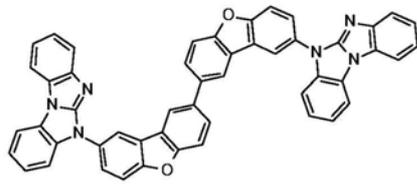
[0351] 在另一优选实施方案中,本发明涉及化合物Q-1至Q-62,其由化合物G-1至G-62通

过由式  的基团替换式  的基团得到。

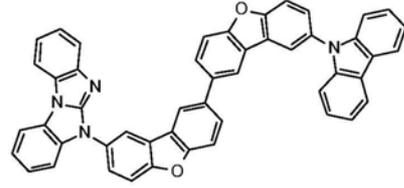
[0352] 在另一优选实施方案中,本发明涉及化合物R-1至R-62,其由化合物G-1至G-62通

过由式  的基团替换式  的基团得到。

[0353] 目前最优选化合物为如下化合物：

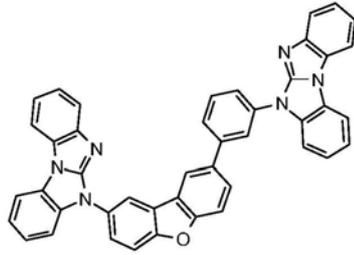


(A-1),

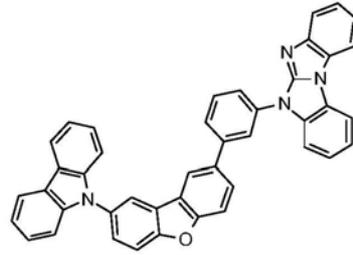


(A-2),

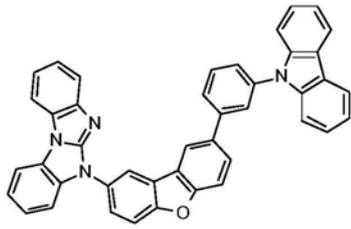
[0354]



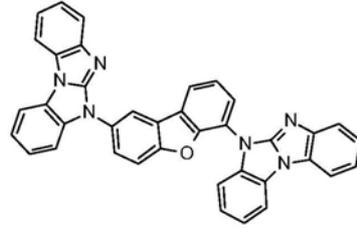
(C-4) ,



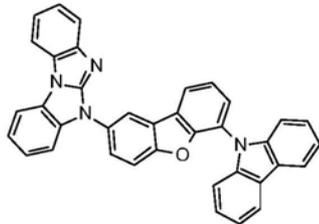
(A-5) ,



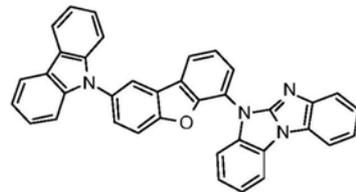
(C-9) ,



(C-1) ,

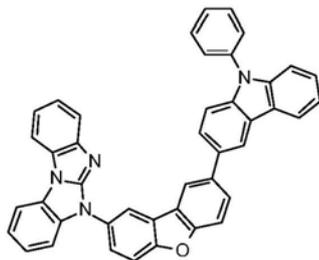


(C-2) ,



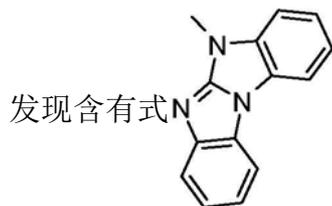
(C-3) 和

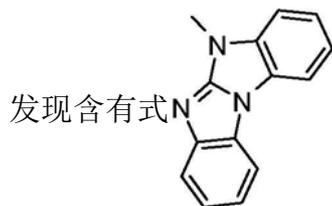
[0355]



(B-6)。

[0356]



发现含有式  咪唑结构部分的化合物特别适用于有机场致发光器件。

[0357]

卤素为氟、氯、溴和碘。

[0358]

C_1 - C_{25} 烷基 (C_1 - C_{18} 烷基) 通常为线性的或可能的话为支化的。实例是甲基、乙基、正

丙基、异丙基、正丁基、仲丁基、异丁基、叔丁基、正戊基、2-戊基、3-戊基、2,2-二甲基丙基、1,1,3,3-四甲基戊基、正己基、1-甲基己基、1,1,3,3,5,5-六甲基己基、正庚基、异庚基、1,1,3,3-四甲基丁基、1-甲基庚基、3-甲基庚基、正辛基、1,1,3,3-四甲基丁基和2-乙基己基、正壬基、癸基、十一烷基、十二烷基、十三烷基、十四烷基、十五烷基、十六烷基、十七烷基或十八烷基。 C_1-C_8 烷基通常为甲基、乙基、正丙基、异丙基、正丁基、仲丁基、异丁基、叔丁基、正戊基、2-戊基、3-戊基、2,2-二甲基丙基、正己基、正庚基、正辛基、1,1,3,3-四甲基丁基和2-乙基己基。 C_1-C_4 烷基通常为甲基、乙基、正丙基、异丙基、正丁基、仲丁基、异丁基、叔丁基。

[0359] C_1-C_{25} 烷氧基 (C_1-C_{18} 烷氧基) 是直链或支化烷氧基, 例如甲氧基、乙氧基、正丙氧基、异丙氧基、正丁氧基、仲丁氧基、叔丁氧基、戊氧基、异戊氧基或叔戊氧基、庚氧基、辛氧基、异辛氧基、壬氧基、癸氧基、十一烷氧基、十二烷氧基、十四烷氧基、十五烷氧基、十六烷氧基、十七烷氧基和十八烷氧基。 C_1-C_8 烷氧基的实例是甲氧基、乙氧基、正丙氧基、异丙氧基、正丁氧基、仲丁氧基、异丁氧基、叔丁氧基、正戊氧基、2-戊氧基、3-戊氧基、2,2-二甲基丙氧基、正己氧基、正庚氧基、正辛氧基、1,1,3,3-四甲基丁氧基和2-乙基己氧基, 优选 C_1-C_4 烷氧基, 如通常是甲氧基、乙氧基、正丙氧基、异丙氧基、正丁氧基、仲丁氧基、异丁氧基、叔丁氧基。

[0360] 术语“环烷基”通常为 C_4-C_{18} 环烷基, 尤其是 C_5-C_{12} 环烷基, 如环戊基、环己基、环庚基、环辛基、环壬基、环癸基、环十一烷基、环十二烷基, 优选环戊基、环己基、环庚基或环辛基, 它们可以未被取代或被取代。

[0361] 可以任选被取代的 C_6-C_{24} 芳基 (C_6-C_{18} 芳基) 通常为苯基, 4-甲基苯基, 4-甲氧基苯基, 萘基, 尤其是1-萘基或2-萘基, 联苯基, 三联苯基, 茈基, 2-或9-茈基, 菲基或蒽基, 其可以未被取代或被取代。

[0362] C_2-C_{30} 杂芳基表示具有5-7个环原子的环或者稠合环体系, 其中氮、氧或硫为可能的杂原子, 并且通常为具有5-30个原子且具有至少6个共轭 π -电子的杂环基团如噻吩基、苯并噻吩基、二苯并噻吩基、噻蒽基、呋喃基、糠基、2H-吡喃基、苯并呋喃基、异苯并呋喃基、氧茈基、苯氧基噻吩基、吡咯基、咪唑基、吡啶基、联吡啶基、三嗪基、嘧啶基、吡嗪基、哒嗪基、中氮茈基、异氮杂茈基、吡啶基、吡啶基、嘌呤基、喹啉基、异喹啉基、2,3-二氮杂萘基、1,5-二氮杂萘基、喹啉基、喹啉基、噌啉基、蝶啶基、咪唑基、咪唑基、苯并咪唑基、菲啶基、吡啶基、嘧啶基、菲咯啉基、吩嗪基、异噻唑基、吩噻唑基、异噻唑基、呋喃基、4-咪唑并[1,2-a]苯并咪唑基、5-苯并咪唑并[1,2-a]苯并咪唑基、咪唑基或吩噻唑基, 其可以未被取代或被取代。

[0363] C_6-C_{24} 芳基 (C_6-C_{18} 芳基) 和 C_2-C_{30} 杂芳基优选被一个或多个 C_1-C_8 烷基取代。

[0364] 术语“芳基醚基团”通常为 C_{6-24} 芳氧基, 即 $O-C_{6-24}$ 芳基, 例如苯氧基或4-甲氧基苯基。

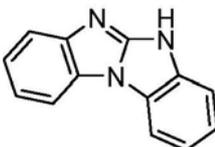
[0365] 上述基团的可能取代基是 C_1-C_8 烷基, 羟基, 巯基, C_1-C_8 烷氧基, C_1-C_8 烷硫基, 卤素, 卤代- C_1-C_8 烷基或氰基。

[0366] 若取代基如 R^{41} 在基团中不止一次出现, 则在每次出现时可能不同。

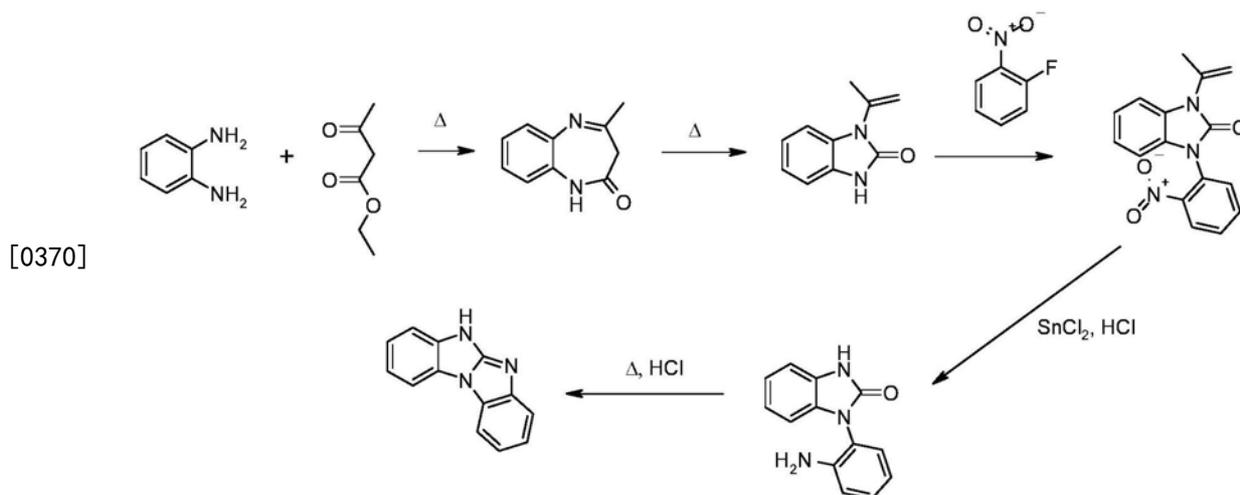
[0367] 术语“被G取代”是指可能存在一个或多个, 尤其是1-3个取代基G。

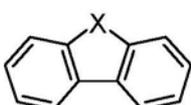
[0368] 如上所述, 上述基团可以被E取代和/或需要的话被D间隔。间隔当然仅在含有至少两个通过单键相互连接的碳原子的基团情况下是可能的; C_6-C_{18} 芳基不被间隔; 间隔的芳基

烷基在烷基结构部分中含有单元D。被一个或多个E取代和/或被一个或多个单元D间隔的C₁-C₁₈烷基例如为(CH₂CH₂O)₁₋₉-R^x,其中R^x为H或C₁-C₁₀烷基或C₂-C₁₀链烷酰基(例如CO-CH(C₂H₅)C₄H₉),CH₂-CH(OR^{y'})-CH₂-O-R^y,其中R^y为C₁-C₁₈烷基、C₅-C₁₂环烷基、苯基、C₇-C₁₅苯基烷基,以及R^{y'}包括与R^y相同的定义或为H;C₁-C₈亚烷基-COO-R^z,例如CH₂COOR^z、CH(CH₃)COOR^z、C(CH₃)₂COOR^z,其中R^z为H,C₁-C₁₈烷基,(CH₂CH₂O)₁₋₉-R^x,并且R^x包括上面所示定义;CH₂CH₂-O-CO-CH=CH₂;CH₂CH(OH)CH₂-O-CO-C(CH₃)=CH₂。

[0369]  的合成例如描述于Achour,Reddouane;Zniber,Rachid,Bulletin

des Societes Chimiques Belges 96(1987)787-92中。



[0371] 式  的合适基础骨架可市购(尤其是在X为S、O、NH的情况下)或可以通过

过本领域熟练技术人员已知的方法得到。参考W02010079051和EP1885818。

[0372] 卤化可以通过本领域熟练技术人员已知的方法进行。优选在式(II)2,8位(二苯并咪喃和二苯并噻吩)或3,6位(咪唑)的基础骨架的3和6位(二溴化)或3或6位(单溴化)溴化或碘化。

[0373] 任选取代的二苯并咪喃、二苯并噻吩和咪唑可以用溴或NBS在冰乙酸或氯仿中进行2,8位(二苯并咪喃和二苯并噻吩)或3,6位(咪唑)的二溴化。例如,用Br₂溴化可以在冰乙酸或氯仿中在低温如0°C下进行。合适的方法例如描述于M.Park,J.R.Buck,C.J.Rizzo,Tetrahedron,54(1998)12707-12714(对于X=NPh)和W.Yang等,J.Mater.Chem.13(2003)1351(对于X=S)。此外,3,6-二溴咪唑、3,6-二溴-9-苯基咪唑、2,8-二溴二苯并噻吩、2,8-二溴二苯并咪喃、2-溴咪唑、3-溴二苯并噻吩、3-溴二苯并咪喃、3-溴咪唑、2-溴二苯并噻吩和2-溴二苯并咪喃可市购。

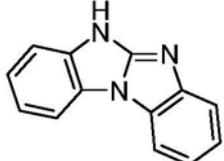
[0374] 在二苯并咪喃(和类似地对二苯并噻吩)的4位的单溴化例如描述于J.Am.Chem.Soc.1984,106,7150。二苯并咪喃(二苯并噻吩)可以在3位通过本领域熟练技术人员已知包括硝化、还原和随后的Sandmeyer反应的顺序进行单溴化。

[0375] 在二苯并咪喃或二苯并噻吩的2位的单溴化和在咪唑的3位的单溴化类似于二溴

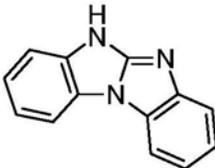
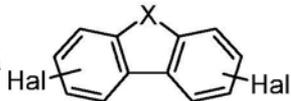
化进行,不同在于仅加入1当量溴或NBS。

[0376] 或者,还可以使用碘化的二苯并呋喃、二苯并噻吩和咪唑。制备尤其描述于Tetrahedron.Lett.47(2006)6957-6960,Eur.J.Inorg.Chem.24(2005)4976-4984,J.heterocyclic Chem.39(2002)933-941,J.Am.Chem.Soc.124(2002)11900-11907,J.heterocyclic Chem,38(2001)77-87。

[0377] 对于亲核取代,需要Cl-或F-取代的二苯并呋喃、二苯并噻吩和咪唑。氯化尤其描述于J.heterocyclic Chemistry,34(1997)891-900或g.Lett.,6(2004)3501-3504;J.Chem.Soc.[Section]C:Organic,16(1971)2775-7,Tetrahedron Lett.25(1984)5363-6,J.Org.Chem.69(2004)8177-8182。氟化描述于J.Org.Chem.63(1998)878-880和J.Chem.Soc.,Perkin Trans.2,5(2002)953-957。

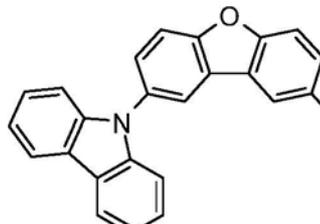
[0378] 基团  的引入在碱的存在下进行。合适的碱对本领域熟练技术人员

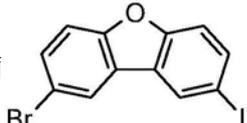
已知且优选选自碱金属和碱土金属的氢氧化物,例如NaOH、KOH、Ca(OH)₂,碱金属氢化物,例如NaH、KH,碱金属酰胺化物,例如NaNH₂,碱金属或碱土金属的碳酸盐,例如K₂CO₃或Cs₂CO₃,和碱金属醇盐,例如NaOMe、NaOEt。此外,上述碱的混合物是合适的。特别优选NaOH、KOH、NaH或K₂CO₃。

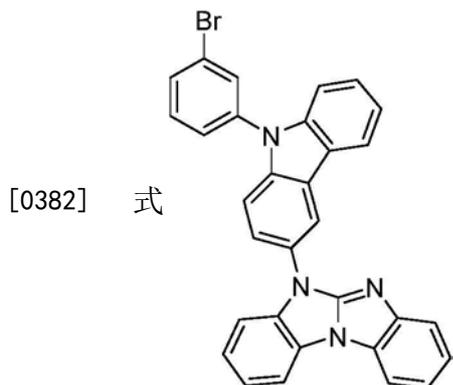
[0379] 杂芳基化可以通过使  铜催化偶联为卤化式  的化合物进行(Ullmann反应)。

的化合物进行(Ullmann反应)。

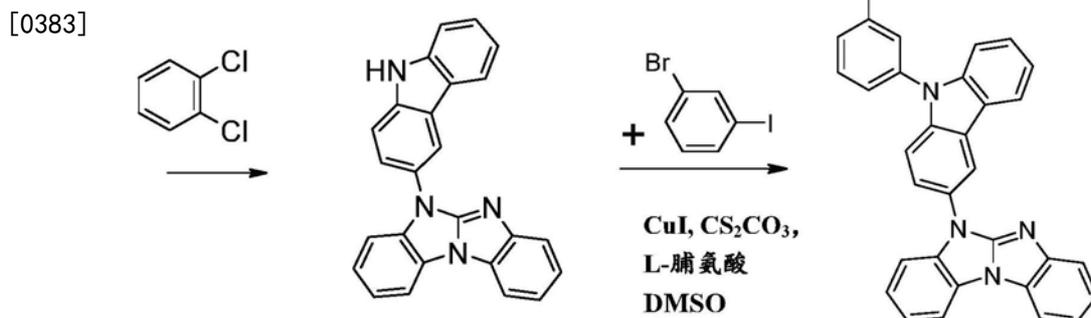
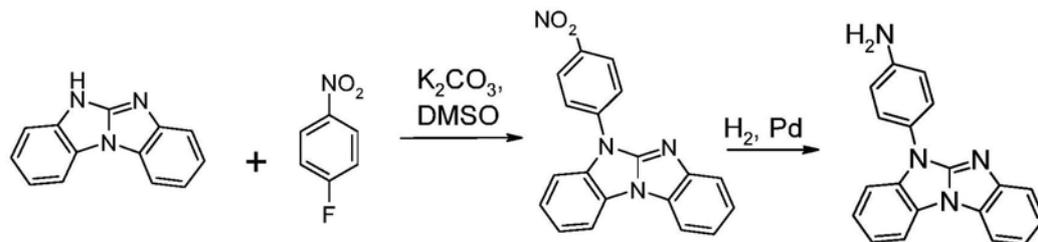
[0380] N-芳基化例如描述于H.Gilman和D.A.Shirley,J.Am.Chem.Soc.66(1944)888;D.Li等,Dyes and Pigments 49(2001)181-186和Eur.J.Org.Chem.(2007)2147-2151。反应可以在溶剂中或以熔体进行。合适的溶剂例如为(极性)非质子性溶剂如二甲亚砜、二甲基甲酰胺、NMP、十三烷或醇。

[0381] 9-(8-溴二苯并呋喃-2-基)咪唑  Br的合成描述于

W02010079051中。2-溴-8-碘二苯并呋喃  的合成描述于EP1885818中。



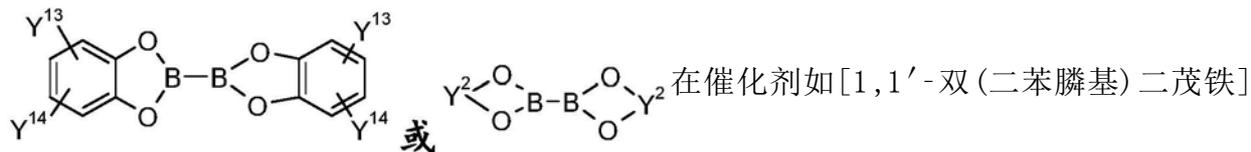
化合物的可能合成路径显示在以下图解中：



[0384] 参考Angew.Chem.Int.Ed.46(2007)1627-1629。

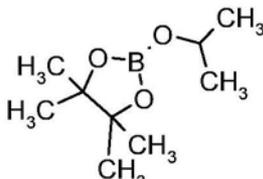
[0385] 含有二硼酸或二硼酸盐基团的二苯并咪喃、二苯并噻吩和咔唑可以通过增加数量的路径制备。合成路径的概述例如在Angew.Chem.Int.Ed.48(2009)9240-9261中给出。

[0386] 通过一种常见路径,含有二硼酸或二硼酸盐基团的二苯并咪喃、二苯并噻吩和咔唑可以通过使卤化二苯并咪喃、二苯并噻吩和咔唑与 $(Y^1O)_2B-B(OY^1)_2$ 、



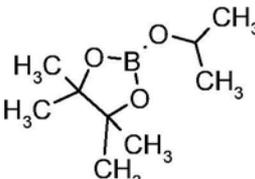
二氯化钯(II)、配合物 $(Pd(Cl)_2(dppf))$ 和碱如乙酸钾存在下在溶剂如二甲基甲酰胺、二甲亚砜、二噁烷和/或甲苯中反应而制备(参见Prasad Appukkuttan等,Synlett 8(2003)1204),其中 Y^1 在每次出现时独立地为 C_1-C_{18} 烷基且 Y^2 在每次出现时独立地为 C_2-C_{10} 亚烷基,例如 $-CY^3Y^4-CY^5Y^6-$ 或 $-CY^7Y^8-CY^9Y^{10}-CY^{11}Y^{12}-$, 其中 $Y^3, Y^4, Y^5, Y^6, Y^7, Y^8, Y^9, Y^{10}, Y^{11}$ 和 Y^{12} 相互独立地为氢或 C_1-C_{18} 烷基,尤其是 $-C(CH_3)_2C(CH_3)_2-$ 、 $-C(CH_3)_2CH_2C(CH_3)_2-$ 或 $-CH_2C(CH_3)_2CH_2-$, 且 Y^{13} 和 Y^{14} 相互独立地为氢或 C_1-C_{18} 烷基。

[0387] 含有二硼酸或二硼酸盐基团的二苯并咪喃、二苯并噻吩和咔唑还可以通过使卤化二苯并咪喃、二苯并噻吩和咔唑与烷基锂试剂,例如正丁基锂或叔丁基锂反应并随后与硼

酸酯例如B(异丙氧基)₃、B(甲氧基)₃或 反应而制备(参见Synthesis

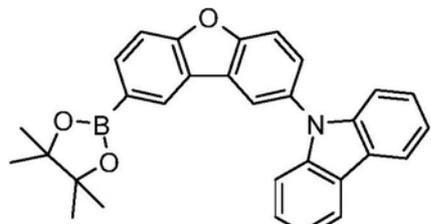
(2000) 442-446)。

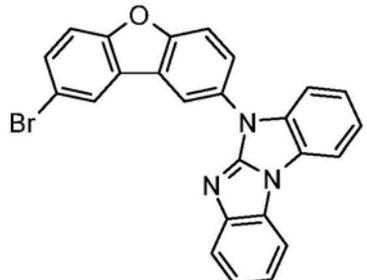
[0388] 含有二硼酸或二硼酸盐基团的二苯并呋喃、二苯并噻吩和咪唑还可以通过使二苯并呋喃、二苯并噻吩和咪唑与氨基锂如二异丙基氨基锂(LDA)反应并随后与硼酸酯例如B

(异丙氧基)₃、B(甲氧基)₃或 反应而制备(J.Org.Chem.73(2008)2176-

2181)。

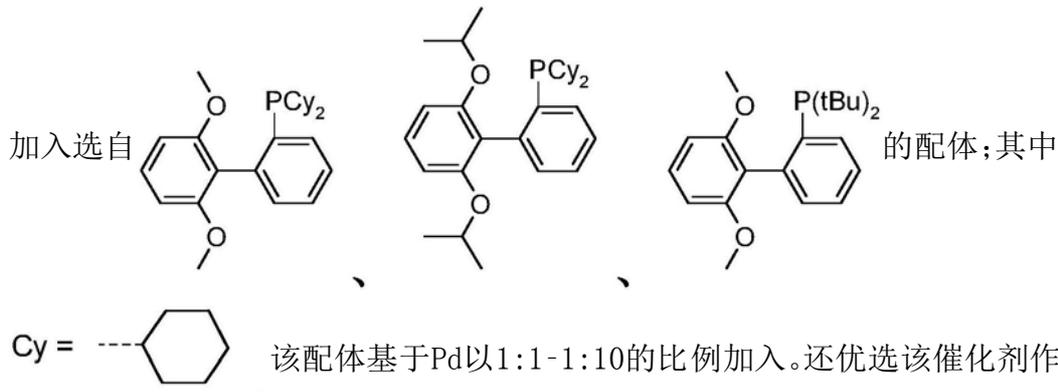
[0389] 可以使含有二硼酸或二硼酸盐基团的二苯并呋喃、二苯并噻吩和咪唑,例如

 与等摩尔量卤化二苯并呋喃、二苯并噻吩和咪唑,例如

 在溶剂中和在催化剂存在下反应。催化剂可为μ-卤化(三异丙基

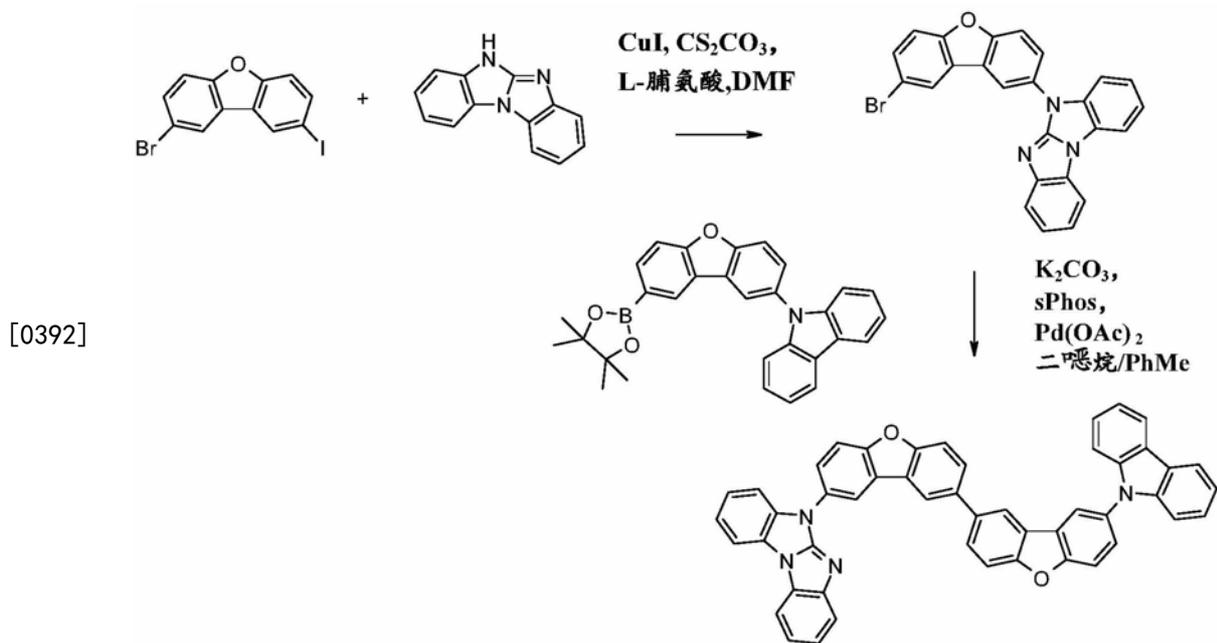
膦)(η³-烯丙基)钯(II)类型的一种(例如参见W099/47474)。

[0390] 优选地,Suzuki反应在有机溶剂如芳族烃或常规极性有机溶剂如苯、甲苯、二甲苯、四氢呋喃或二噁烷或其混合物,最优选甲苯存在下进行。溶剂的量通常在1-10L/mol硼酸衍生物范围内选择。还优选该反应在惰性气氛如氮气或氩气下进行。此外,优选在水性碱如碱金属氢氧化物或碳酸盐如NaOH、KOH、Na₂CO₃、K₂CO₃、Cs₂CO₃等存在下进行该反应,优选选择K₂CO₃水溶液。该碱与硼酸或硼酸酯衍生物的摩尔比通常在0.5:1-50:1范围内选择,非常尤其为1:1。反应温度通常在40-180℃范围内选择,优选在回流条件下。优选反应时间在1-80小时,更优选20-72小时范围内选择。在一个优选实施方案中,使用偶联反应或缩聚反应常用的催化剂,优选W02007/101820中所述的Pd基催化剂。钯化合物基于待闭合的键数目以1:10000-1:50,优选1:5000-1:200的比例加入。优选例如使用钯(II)盐如PdAc₂或Pd₂dba₃并

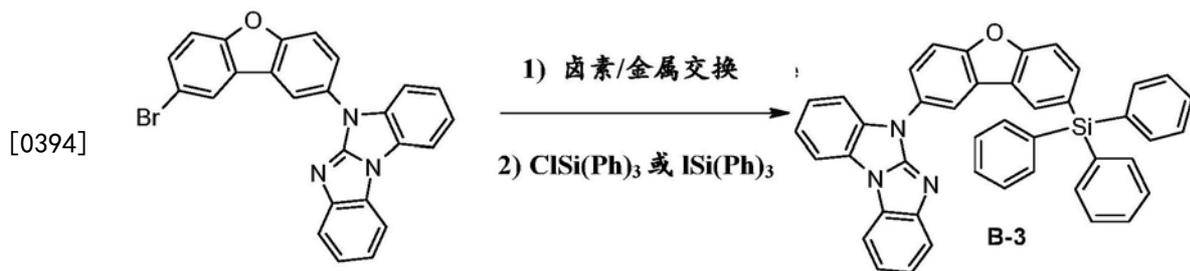


液加入。优选使用合适的有机溶剂如上述那些, 优选苯、甲苯、二甲苯、THF、二噁烷, 更优选甲苯, 或其混合物。溶剂的量通常在1-10L/mol硼酸衍生物范围内选择。有机碱如四烷基氢氧化铵和相转移催化剂如TBAB可提高硼的活性(例如参见Leadbeater&Marco; Angew.Chem.Int.Ed.Eng. 42 (2003) 1407和其中所引用的文献)。其他反应条件的变体由T.I.Wallow和B.M.Novak, J.Org.Chem. 59 (1994) 5034-5037; 和M.Remmers, M.Schulze, G.Wegner, Macromol.Rapid Commun. 17 (1996) 239-252和G.A.Molander und B.Canturk, Angew.Chem., 121 (2009) 9404-9425中给出。

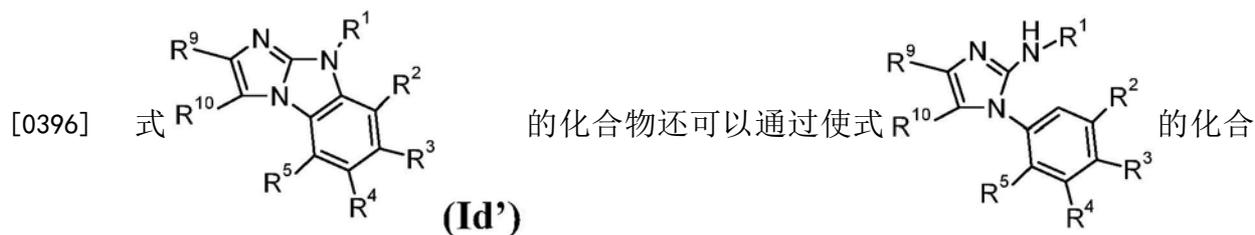
[0391] 化合物A-1的可能合成路径显示于以下反应图解中:



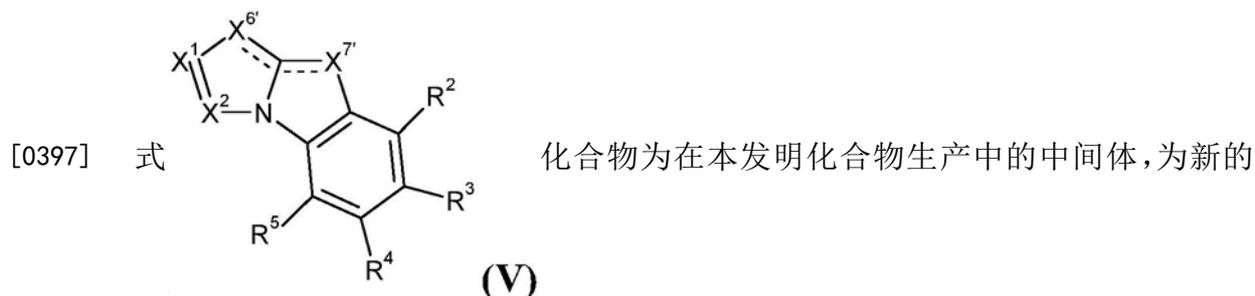
[0393] 化合物B-3的可能合成路径显示于以下反应图解中:



[0395] 卤素/金属交换使用nBuLi/THF在-78℃下或使用tBuLi/THF在-78℃下进行。参考W02010/079051,其中描述了该类化合物的合成。



物在催化剂如乙酸铜(Cu(OAc)₂)；配体如PPh₃和1,10-菲咯啉；碱如乙酸钠(NaOAc)、碳酸钠、碳酸钾、碳酸铯、磷酸钾和碳酸氢钠；溶剂如邻-、间-、和对二甲苯,和氧气(1atm)在升高的温度下,尤其是在100-160℃的温度下反应而合成。参考X.Wang等,Org.Lett.14(2012)452-455[网上公布:12月29日,2011]。



且构成本发明的另一主题。 X^6 为-N=且 X^7 为-NR¹-或 X^7 为=N-且 X^6 为-NR¹-,

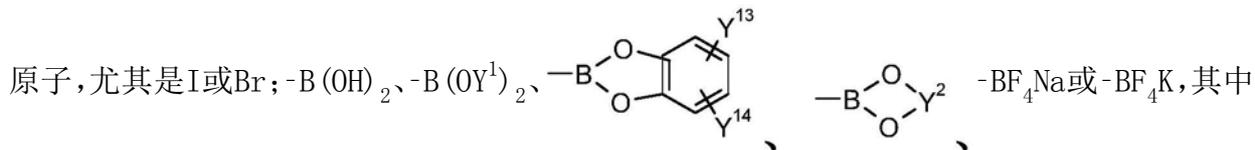
[0398] R¹为式-A¹-(A²)_p-(A³)_q-(A⁴)_r-(R⁶)_t的基团,t为1或2,尤其是1;

[0399] p、q、r、A¹、A²、A³、A⁴、X¹、X²、R²、R³、R⁴和R⁵如上所定义。

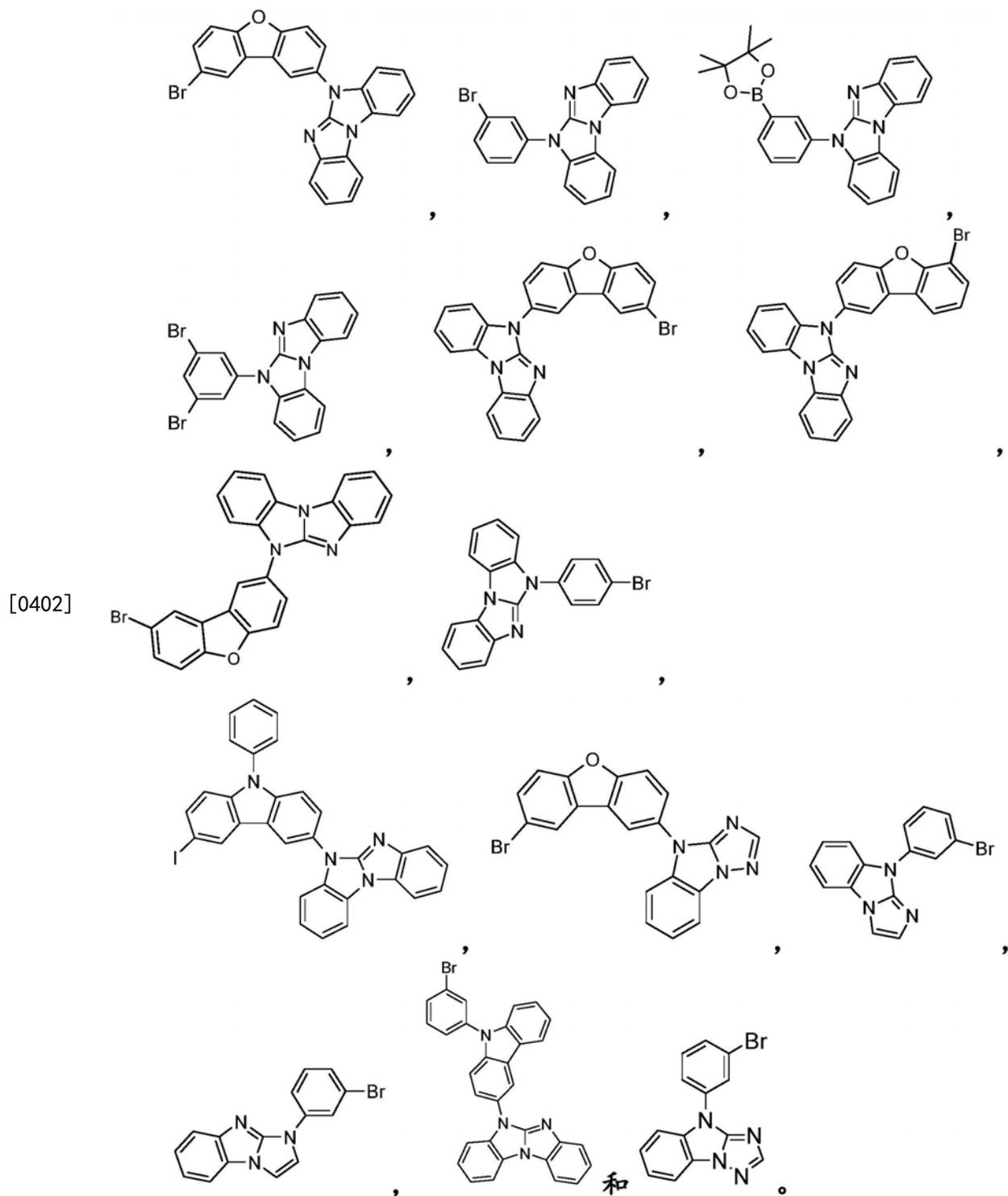
[0400] 在一个实施方案中,R⁶为卤素,尤其是Cl、Br或J;-OS(O)₂CF₃,-OS(O)₂-芳基,尤其是

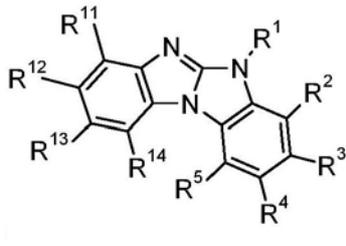


[0401] 在另一实施方案中,R⁶为ZnX¹²;-SnR²⁰⁷R²⁰⁸R²⁰⁹,其中R²⁰⁷、R²⁰⁸和R²⁰⁹相同或不同且为H或C₁-C₆烷基,其中两个基团任选形成公用环且这些基团任选支化或未支化;和X¹²为卤



Y¹在每次出现时独立地为C₁-C₁₀烷基且Y²在每次出现时独立地为C₂-C₁₀亚烷基,例如-CY³Y⁴-CY⁵Y⁶-或-CY⁷Y⁸-CY⁹Y¹⁰-CY¹¹Y¹²-,其中Y³、Y⁴、Y⁵、Y⁶、Y⁷、Y⁸、Y⁹、Y¹⁰、Y¹¹和Y¹²相互独立地为氢或C₁-C₁₀烷基,尤其是-C(CH₃)₂C(CH₃)₂-、-C(CH₃)₂CH₂C(CH₃)₂-或-CH₂C(CH₃)₂CH₂-,和Y¹³和Y¹⁴相互独立地为氢或C₁-C₁₀烷基。就p、q、r、A¹、A²、A³、A⁴、X¹、X²、R²、R³、R⁴和R⁵而言,适用与式I化合物相同的优选情形。中间体的实例如下所示:

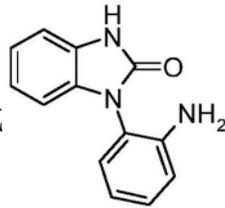




(II)

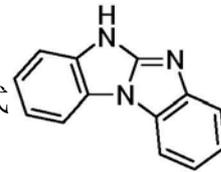
的化合物的方法可包括(a)加热在H₃PO₄、多磷酸、CH₃SO₃H/

P₂O₅、CH₃SO₃H或硫酸中的式



(X)

的化合物以得到式



(XI)

的

化合物;和(b)使式XI化合物反应而得到式II化合物。上文说明了步骤b)的各种实例。在步骤a)中,可存在沸点在140℃以上的溶剂或溶剂混合物,例如二甲苯或**菜**。式X的化合物在惰性气体如氮气或氩气的气氛下在140℃以上,优选160℃以上,更优选180℃以上的温度下搅拌30分钟至3周,优选1-48h。

[0405] 已发现式I化合物特别适合于其中要求载流子导电性的应用,尤其是用于有机电子应用,例如选自开关元件如有机晶体管,例如有机FET和有机TFT,有机太阳能电池和有机发光二极管(OLED)的有机电子应用,其中式I化合物特别适合在OLED中在发光层中用作基体材料和/或用作空穴和/或激子阻断剂材料和/或用作电子和/或激子阻断剂材料,尤其是与磷光发光体组合。在将本发明式I化合物用于OLED的情况下,得到了具有良好效率和长使用寿命且可以尤其是在低使用和操作电压下操作的OLED。本发明式I化合物特别适合在蓝色和绿色发光体如浅蓝色或深蓝色发光体中用作基体和/或空穴/激子阻断剂材料,这些尤其是磷光发光体。此外,式I化合物可在选自开关元件和有机太阳能电池的有机电子应用中用于导体/补充材料。

[0406] 式I化合物可在有机电子应用,尤其是OLED中用作基体材料和/或空穴/激子阻断剂材料和/或电子/激子阻断剂材料和/或空穴注入材料和/或电子注入材料和/或空穴导体材料(空穴传输材料)和/或电子导体材料(电子传输材料),优选用作基体材料和/或电子/激子阻断剂和/或空穴传输材料。本发明式I化合物更优选在有机电子应用,尤其是OLED中用作基体材料。

[0407] 在OLED的发光层或发光层之一中,可以将发光剂材料与式I化合物的基体材料以及具有例如良好空穴导体(空穴传输)性能的其他基体材料组合。这实现了该发光层的高量子效率。

[0408] 当将式I化合物在发光层中用作基体材料和额外用作空穴/激子阻断剂材料和/或电子/激子阻断剂材料时,由于所述材料的化学相同或类似性,在发光层和相邻空穴/激子阻断剂材料和/或电子/激子阻断剂材料之间获得改善的界面,这可以导致在相同亮度下电压的降低和OLED使用寿命的延长。此外,将相同材料用于空穴/激子阻断剂材料和/或电子/激子阻断剂材料和发光层的基体可以使OLED的生产工艺简化,这是因为可以将同一来源用于式I化合物之一的材料的气相沉积工艺。

[0409] 有机电子器件的合适结构对本领域熟练技术人员是已知的且如下所述。

[0410] 有机晶体管通常包括由具有空穴传输能力和/或电子传输能力的有机层形成的半导体层;由导电层形成的栅极电极;以及在该半导体层和该导电层之间引入的绝缘层。在该排列上安装源电极和漏电极以产生该晶体管元件。此外,在该有机晶体管中可以存在本领域熟练技术人员已知的其他层。

[0411] 该有机太阳能(光电转换元件)通常包括存在于两个平行设置的板式电极之间的有机层。该有机层可以构造在梳形电极上。对于该有机层的位置没有特殊限制且对于电极材料没有特殊限制。然而,当使用平行设置的板式电极时,至少一个电极优选由透明电极,例如ITO电极或氟掺杂的氧化锡电极形成。该有机层由两个子层,即具有p型半导体性能或空穴传输能力的层和具有n型半导体性能或电子传输能力的层形成。此外,在该有机太阳能电池中可以存在本领域熟练技术人员已知的其他层。具有空穴传输能力的层可包含式I化合物。

[0412] 同样可能的是式I化合物存在于发光层(优选作为基体材料)和电子阻断层(作为电子/激子阻断剂)。

[0413] 本发明进一步涉及一种有机发光二极管,其包含阳极An和阴极Ka,设置在阳极An和阴极Ka之间的发光层E,以及合适的话至少一个选自至少一个空穴/激子阻断层、至少一个电子/激子阻断层、至少一个空穴注入层、至少一个空穴导体层、至少一个电子注入层和至少一个电子导体层的其他层,其中所述至少一种式I化合物存在于发光层E和/或其他层中至少一个中。所述至少一种式I化合物存在于发光层和/或空穴阻断层。

[0414] 本申请进一步涉及包含至少一种式I化合物的发光层。

[0415] 本发明OLED的结构

[0416] 本发明有机发光二极管(OLED)因此通常具有下列结构:

[0417] 阳极(An)和阴极(Ka),设置在阳极(An)和阴极(Ka)之间的发光层E。

[0418] 本发明OLED例如在一个优选实施方案中可以由下列层形成:

[0419] 1. 阳极

[0420] 2. 空穴导体层

[0421] 3. 发光层

[0422] 4. 空穴/激子阻断层

[0423] 5. 电子导体层

[0424] 6. 阴极

[0425] 不同于上述结构的层顺序也是可能的并且为本领域熟练技术人员已知。例如,可能的是该OLED不具有所有所述层;例如,同样合适的为具有层(1)(阳极)、(3)(发光层)和(6)(阴极)的OLED,在这种情况下,相邻层呈层(2)(空穴导体层)和(4)(空穴/激子阻断层)以及(5)(电子导体层)的功能。同样合适的为具有层(1)、(2)、(3)和(6)或层(1)、(3)、(4)、(5)和(6)的OLED。此外,OLED可以在空穴导体层(2)和发光层(3)之间具有电子/激子阻断层。

[0426] 额外可能的是将多个上述功能(电子/激子阻断剂、空穴/激子阻断剂、空穴注入、空穴传导、电子注入、电子传导)组合在一层中并且例如由存在于该层中的单一材料呈现。例如,在一个实施方案中,用于空穴导体层中的材料可以同时阻断激子和/或电子。

[0427] 此外,在上面所述那些中的OLED的各单层又可以由两个或更多个层形成。例如空

穴导体层可以由将空穴由电极注入其中的层和将空穴由该空穴注入层传输至发光层的层形成。电子导体层同样可以由多个层构成,例如由在其中由电极注入电子的层和接收来自电子注入层的电子并将它们传输至发光层的层构成。所述这些层各自根据诸如能级、耐热性和载流子迁移率的因素以及还有就有机层或金属电极的所述各层的能量差而选择。本领域熟练技术人员能够选择OLED的结构以使它与按照本发明用作发光体物质的有机化合物最佳匹配。

[0428] 为了获得特别有效的OLED,例如该空穴导体层的HOMO(最高被占分子轨道)应与阳极的功函数相匹配且该电子导体层的LUMO(最低未占分子轨道)应与阴极的功函数相匹配,条件是上述各层存在于本发明OLED中。

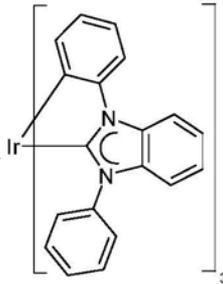
[0429] 阳极(1)为提供正载流子的电极。它例如可以由包括金属、各种金属的混合物、金属合金、金属氧化物或各种金属氧化物的混合物的材料形成。或者,阳极可以为导电性聚合物。合适的金属包括主族、过渡族和镧系金属及其合金,尤其是元素周期表第Ib、IVa、Va和VIa族金属和第VIIIa族过渡金属及其合金。当阳极呈透明时,通常使用元素周期表(IUPAC版)第IIb、IIIb和IVb族的混合金属氧化物,例如氧化铟锡(ITO)。同样可能的是阳极(1)包含有机材料,例如聚苯胺,例如如Nature,第357卷,第477-479页(1992年6月11日)所述。至少阳极或阴极应至少部分透明以能够发射形成的光。用于阳极(1)的材料优选为ITO。

[0430] 适合本发明OLED的层(2)的空穴导体材料例如公开于Kirk-Othmer Encyclopedia of Chemical Technology,第4版,第18卷,第837-860页,1996中。空穴传输分子和聚合物均可以用作空穴传输材料。通常使用的空穴传输分子选自三[N-(1-萘基)-N-(苯基氨基)]三苯基胺(1-NaphDATA),4,4'-二[N-(1-萘基)-N-苯基氨基]联苯(α -NPD),N,N'-二苯基-N,N'-二(3-甲基苯基)-[1,1'-联苯基]-4,4'-二胺(TPD),1,1-二[(二-4-甲苯基氨基)苯基]环己烷(TAPC),N,N'-二(4-甲基苯基)-N,N'-二(4-乙基苯基)-[1,1'-(3,3'-二甲基)联苯基]-4,4'-二胺(ETPD),四(3-甲基苯基)-N,N,N',N'-2,5-苯二胺(PDA), α -苯基-4-N,N-二苯基氨基苯乙烯(TPS),对(二乙基氨基)苯甲醛二苯基脒(DEH),三苯基胺(TPA),二[4-(N,N-二乙基氨基)-2-甲基苯基](4-甲基苯基)甲烷(MPMP),1-苯基-3-[对(二乙基氨基)苯乙烯基]-5-[对(二乙基氨基)苯基]吡啶啉(PPR或DEASP),1,2-反式-二(9H-咪唑-9-基)环丁烷(DCZB),N,N,N',N'-四(4-甲基苯基)-(1,1'-联苯基)-4,4'-二胺(TTB),4,4',4''-三(N,N-二苯基氨基)三苯基胺(TDTA),4,4',4''-三(N-咪唑基)三苯基胺(TCTA),N,N'-二(萘-2-基)-N,N'-二(苯基)联苯胺(β -NPB),N,N'-二(3-甲基苯基)-N,N'-二(苯基)-9,9-螺二茱(Spiro-TPD),N,N'-二(萘-1-基)-N,N'-二(苯基)-9,9-螺二茱(Spiro-NPB),N,N'-二(3-甲基苯基)-N,N'-二(苯基)-9,9-二甲基茱(DMFL-TPD),二[4-(N,N-二甲苯基氨基)苯基]环己烷,N,N'-二(萘-1-基)-N,N'-二(苯基)-9,9-二甲基茱,N,N'-二(萘-1-基)-N,N'-二(苯基)-2,2-二甲基联苯胺,N,N'-二(萘-1-基)-N,N'-二(苯基)联苯胺,N,N'-二(3-甲基苯基)-N,N'-二(苯基)联苯胺,2,3,5,6-四氟-7,7,8,8-四氰基醌二甲烷(F4-TCNQ),4,4',4''-三(N-3-甲基苯基-N-苯基氨基)三苯基胺,4,4',4''-三(N-(2-萘基)-N-苯基氨基)三苯基胺,吡嗪并[2,3-f][1,10]菲咯啉-2,3-二甲腈(PPDN),N,N,N',N'-四(4-甲氧基苯基)联苯胺(MeO-TPD),2,7-二[N,N-二(4-甲氧基苯基)氨基]-9,9-螺二茱(MeO-Spiro-TPD),2,2'-二[N,N-二(4-甲氧基苯基)氨基]-9,9-螺二茱(2,2'-MeO-Spiro-TPD),N,N'-二苯基-N,N'-二[4-(N,N-二甲苯基氨基)苯基]联苯胺(NTNPB),N,N'-二苯基-N,N'-二[4-(N,N-二苯基氨基

基)苯基]联苯胺(NPNPB),N,N'-二(萘-2-基)-N,N'-二苯基苯-1,4-二胺(β -NPP),N,N'-二(3-甲基苯基)-N,N'-二(苯基)-9,9-二苯基芴(DPFL-TPD),N,N'-二(萘-1-基)-N,N'-二(苯基)-9,9-二苯基芴(DPFL-NPB),2,2',7,7'-四(N,N-二苯基氨基)-9,9'-螺二芴(Spiro-TAD),9,9-二[4-(N,N-二(联苯-4-基)氨基)苯基]-9H-芴(BPAPF),9,9-二[4-(N,N-二(萘-2-基)氨基)苯基]-9H-芴(NPAPF),9,9-二[4-(N,N-二(萘-2-基)-N,N'-二苯基氨基)苯基]-9H-芴(NPBAPF),2,2',7,7'-四[N-萘基(苯基)氨基]-9,9'-螺二芴(Spiro-2NPB),N,N'-二(菲-9-基)-N,N'-二(苯基)联苯胺(PAPB),2,7-二[N,N-二(9,9-螺二芴-2-基)氨基]-9,9-螺二芴(Spiro-5),2,2'-二[N,N-二(联苯-4-基)氨基]-9,9-螺二芴(2,2'-Spiro-DBP),2,2'-二(N,N-二苯基氨基)-9,9-螺二芴(Spiro-BPA),2,2',7,7'-四(N,N-二甲苯基)氨基螺二芴(Spiro-TTB),N,N,N',N'-四萘-2-基联苯胺(TNB),卟啉化合物以及酞菁类如铜酞菁和氧化钛酞菁。通常使用的空穴传输聚合物选自聚乙烯基吡啶、(苯基甲基)聚硅烷和聚苯胺。同样可以通过将空穴传输分子掺杂到聚合物如聚苯乙烯和聚碳酸酯中获得空穴传输聚合物。合适的空穴传输分子是已经在上面提到的分子。

[0431] 此外,在一个实施方案中可以使用卡宾配合物作为空穴导体材料,该至少一种空穴导体材料的带隙通常大于所用发光体材料的带隙。在本申请上下文中,“带隙”应理解为指三线态能量。合适的卡宾配合物例如为WO 2005/019373A2、WO 2006/056418A2、WO 2005/113704、WO 2007/115970、WO 2007/115981和WO 2008/000727中所述的卡宾配合物。合适卡

宾配合物的一个实例是例如公开于WO 2005/019373中式I的 $\text{Ir}(\text{dpbic})_3$ 。原



则上,空穴导体层可包含至少一种式I化合物作为空穴导体材料。

[0432] 发光层(3)包含至少一种发光体材料。它原则上可以是荧光或磷光发光体,合适的发光体材料为本领域熟练技术人员已知。该至少一种发光体材料优选为磷光发光体。优选使用的磷光发光体化合物基于金属配合物,尤其是金属Ru、Rh、Ir、Pd和Pt的配合物,特别是Ir的配合物具有重要性。式I化合物可以在发光层中用作基体。

[0433] 适合用于本发明OLED中的金属配合物例如描述于文献WO 02/60910 A1、US 2001/0015432 A1、US 2001/0019782 A1、US 2002/0055014 A1、US 2002/0024293 A1、US 2002/0048689 A1、EP 1 191 612 A2、EP 1 191 613 A2、EP 1 211 257 A2、US 2002/0094453 A1、WO 02/02714 A2、WO 00/70655 A2、WO 01/41512 A1、WO 02/15645 A1、WO 2005/019373 A2、WO 2005/113704 A2、WO 2006/115301 A1、WO 2006/067074 A1、WO 2006/056418、WO 2006121811 A1、WO 2007095118 A2、WO 2007/115970、WO 2007/115981、WO 2008/000727、WO2010129323、WO2010056669和WO10086089中。

[0434] 其他合适的金属配合物为市售金属配合物三(2-苯基吡啶)铱(III)、三(2-(4-甲基苯基)吡啶根合-N,C^{2'})铱(III)、双(2-苯基吡啶)(乙酰丙酮根合)铱(III)、三(1-苯基异喹啉)铱(III)、双(2,2'-苯并噻吩基)吡啶根合-N,C^{3'})铱(III)(乙酰丙酮化物)、三(2-苯基喹啉)铱(III)、双(2-(4,6-二氟苯基)吡啶根合-N,C^{2'})皮考啉铱(III)、双(1-苯基异喹啉)铱

(III) (乙酰丙酮化物)、双(2-苯基喹啉)(乙酰丙酮根合)铱(III)、双(二苯并[f,h]喹啉)铱(III) (乙酰丙酮化物)、双(2-甲基二苯并[f,h]喹啉)铱(III) (乙酰丙酮化物)和三(3-甲基-1-苯基-4-三甲基乙酰基-5-吡唑啉基)铱(III)、双[1-(9,9-二甲基-9H-芴-2-基)异喹啉](乙酰丙酮根合)铱(III)、双(2-苯基苯并噻唑根合)(乙酰丙酮根合)铱(III)、双(2-(9,9-二己基芴基)-1-吡啶)(乙酰丙酮根合)铱(III)、双(2-苯并[b]噻吩-2-基-吡啶)(乙酰丙酮根合)铱(III)。

[0435] 另外,以下市售物质是合适的:三(二苯甲酰丙酮根合)单(菲咯啉)钿(III)、三(二苯甲酰甲烷)单(菲咯啉)钿(III)、三(二苯甲酰甲烷)单(5-氨基菲咯啉)钿(III)、三(二-2-萘酰甲烷)单(菲咯啉)钿(III)、三(4-溴苯甲酰甲烷)单(菲咯啉)钿(III)、三(二(联苯)甲烷)单(菲咯啉)钿(III)、三(二苯甲酰甲烷)单(4,7-二苯基菲咯啉)钿(III)、三(二苯甲酰甲烷)单(4,7-二甲基菲咯啉)钿(III)、三(二苯甲酰甲烷)单(4,7-二甲基菲咯啉二磺酸)钿(III)二钠盐、三[二(4-(2-(2-乙氧基乙氧基)乙氧基)苯甲酰甲烷)]单(菲咯啉)钿(III)和三[二[4-(2-(2-乙氧基乙氧基)乙氧基)苯甲酰甲烷)]单(5-氨基菲咯啉)钿(III)、双(3-三氟甲基-5-(4-叔丁基吡啶基)-1,2,4-三唑根合)二苯基甲基膦钨(II)、双(3-三氟甲基-5-(2-吡啶基)-1,2,4-三唑)二甲基苯基膦钨(II)、双(3-三氟甲基)-5-(4-叔丁基吡啶基)-1,2,4-三唑根合)二甲基苯基膦钨(II)、双(3-三氟甲基)-5-(2-吡啶基)吡唑根合)二甲基苯基膦钨(II)、三[4,4'-二叔丁基(2,2')-联吡啶]钿(III)、双(2-(9,9-二丁基芴基)-1-异喹啉)钨(II) (乙酰丙酮化物)。

[0436] 发光层优选包含式
$$\begin{array}{c} [L]_m \\ \diagdown \\ M^1 \\ \diagup \\ [K]_o \end{array} [\text{碳烯}]_{n1} \quad (IX)$$
 的化合物,其描述于WO 2005/019373A2中,其中各符号具有下列含义:

[0437] M为选自Co、Rh、Ir、Nb、Pd、Pt、Fe、Ru、Os、Cr、Mo、W、Mn、Tc、Re、Cu、Ag和Au的金属原子,其处于相应金属原子的任何可能氧化态;

[0438] 碳烯为可以不带电荷的或者一价阴离子的并且单齿、二齿或三齿的碳烯配体,其中该碳烯配体还可以是双碳烯或三碳烯配体;

[0439] L为一价阴离子或二价阴离子配体,其可以是单齿或二齿的;

[0440] K为不带电荷的单齿或二齿配体,其选自膦;膦酸酯和其衍生物;砷酸酯和其衍生物;亚磷酸酯;CO;吡啶;腈类以及与M¹形成π配合物的共轭二烯;

[0441] n₁为碳烯配体的数量,其中n₁至少为1,并且当n₁>1时,式I配合物中的碳烯配体可以相同或不同;

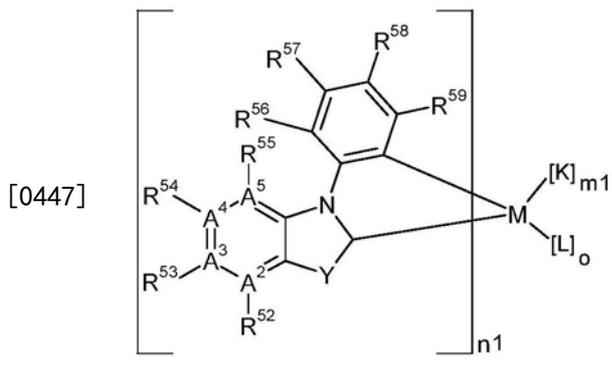
[0442] m₁为配体L的数量,其中m₁可以为0或≥1,并且当m₁>1时,配体L可以相同或不同;

[0443] o为配体K的数量,其中o可以为0或≥1,并且当o>1时,配体K可以相同或不同;

[0444] 其中n₁+m₁+o之和取决于金属原子的氧化态和配位数,取决于配体碳烯、L和K的齿数以及取决于配体,碳烯和L上的电荷,条件是n₁至少为1。

[0445] 为合适的三线态发光体的卡宾配合物例如描述于WO 2006/056418A2、WO 2005/113704、WO 2007/115970、WO 2007/115981和WO 2008/000727、WO2009050281、WO2009050290、WO2011051404和WO2011073149。

[0446] 更优选如下通式的金属-碳烯配合物:



其描述于美国专利申请号61/286046、

61/323885和欧洲专利申请10187176.2 (PCT/EP2010/069541) 中,其中M、n1、Y、A²、A³、A⁴、A⁵、R⁵¹、R⁵²、R⁵³、R⁵⁴、R⁵⁵、R⁵⁶、R⁵⁷、R⁵⁸、R⁵⁹、K、L、m1和o各自定义如下:

[0448] M为Ir或Pt,

[0449] n1为选自1、2和3的整数,

[0450] Y为NR⁵¹、O、S或C(R²⁵)₂,

[0451] A²、A³、A⁴和A⁵各自独立地为N或C,其中2A=氮原子且至少一个碳原子存在于环中两个氮原子之间,

[0452] R⁵¹为线性或支化烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有1-20个碳原子;环烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有3-20个碳原子;取代或未取代的芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有6-30个碳原子;取代或未取代的杂芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有总共5-18个碳原子和/或杂原子,

[0453] 如果A²、A³、A⁴和/或A⁵为N,则R⁵²、R⁵³、R⁵⁴和R⁵⁵各自为游离电子对,或者如果A²、A³、A⁴和/或A⁵为C,则各自独立地为氢;线性或支化烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有1-20个碳原子;环烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有3-20个碳原子;取代或未取代的芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有6-30个碳原子;取代或未取代的杂芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有总共5-18个碳原子和/或杂原子;具有给体或受体作用的基团,或者R⁵³和R⁵⁴与A³和A⁴一起形成任选取代的不饱和环,其任选被至少一个其他杂原子间隔且具有总共5-18个碳原子和/或杂原子,

[0454] R⁵⁶、R⁵⁷、R⁵⁸和R⁵⁹各自独立地为氢;线性或支化烷基,其任选被至少一个其他杂原子间隔,任选带有至少一个官能团且具有1-20个碳原子;环烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有3-20个碳原子;环杂烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有3-20个碳原子;取代或未取代的芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有6-30个碳原子;取代或未取代的杂芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有总共5-18个碳原子和/或杂原子;具有给体或受体作用的基团,或者

[0455] R⁵⁶和R⁵⁷、R⁵⁷和R⁵⁸或R⁵⁸和R⁵⁹与它们所键合的碳原子一起形成饱和、不饱和或芳族、任选取代的环,其任选被至少一个杂原子间隔且具有总共5-18个碳原子和/或杂原子,和/或

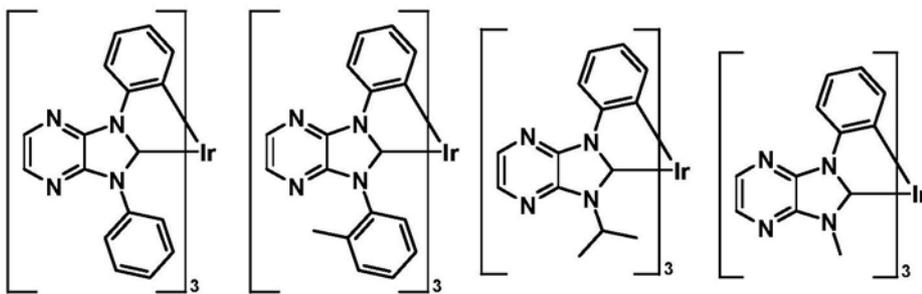
[0456] 如果A⁵为C,则R⁵⁵和R⁵⁶一起形成饱和或不饱和、线性或支化桥,其任选包含杂原子、芳族单元、杂芳族单元和/或官能团且具有总共1-30个碳原子和/或杂原子,任选稠合有包含碳原子和/或杂原子的取代或未取代的5-8员环,

[0457] R²⁵独立地为线性或支化烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有1-20个碳原子;环烷基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有3-20个碳原子;取代或未取代的芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有6-30个碳原子;取代或未取代的杂芳基,其任选被至少一个杂原子间隔,任选带有至少一个官能团且具有总共5-18个碳原子和/或杂原子,K为不带电荷的单齿或二齿配体,

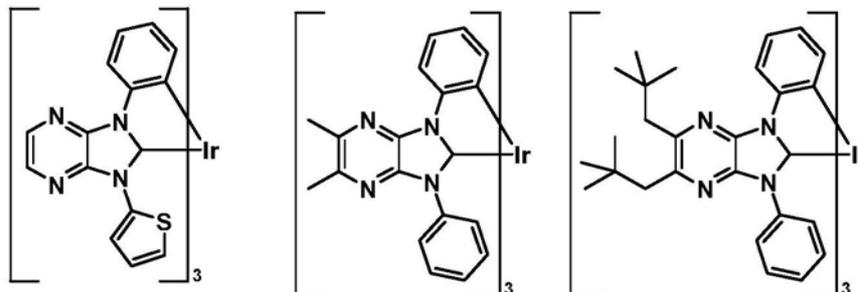
[0458] L为一价阴离子或二价阴离子配体,优选一价阴离子配体,其可以是单齿或二齿的,

[0459] m1为0、1或2,其中当m1为2时,配体K可以相同或不同,o为0、1或2,其中当o为2时,配体L可以相同或不同。

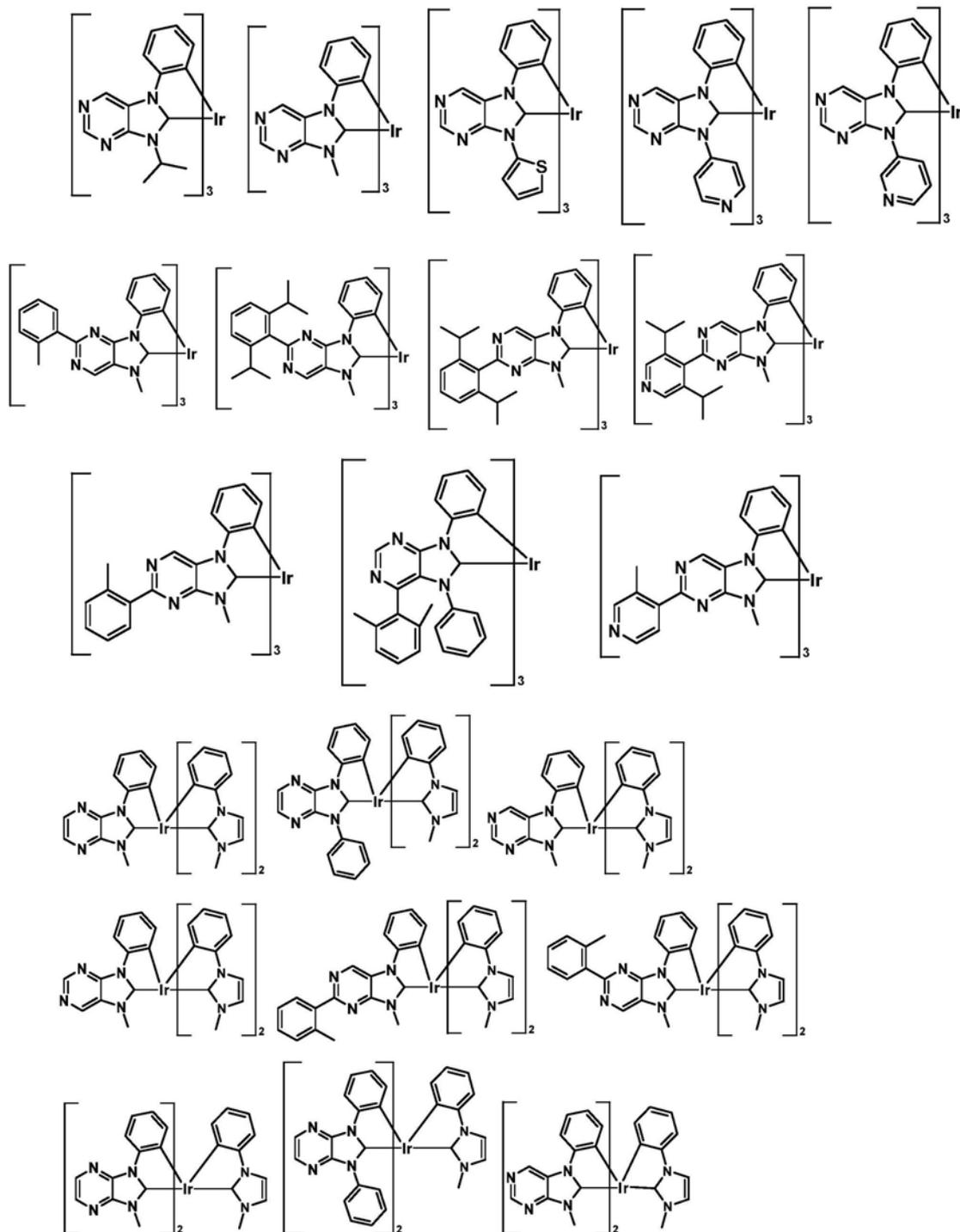
[0460] 式IX化合物优选为下式化合物:

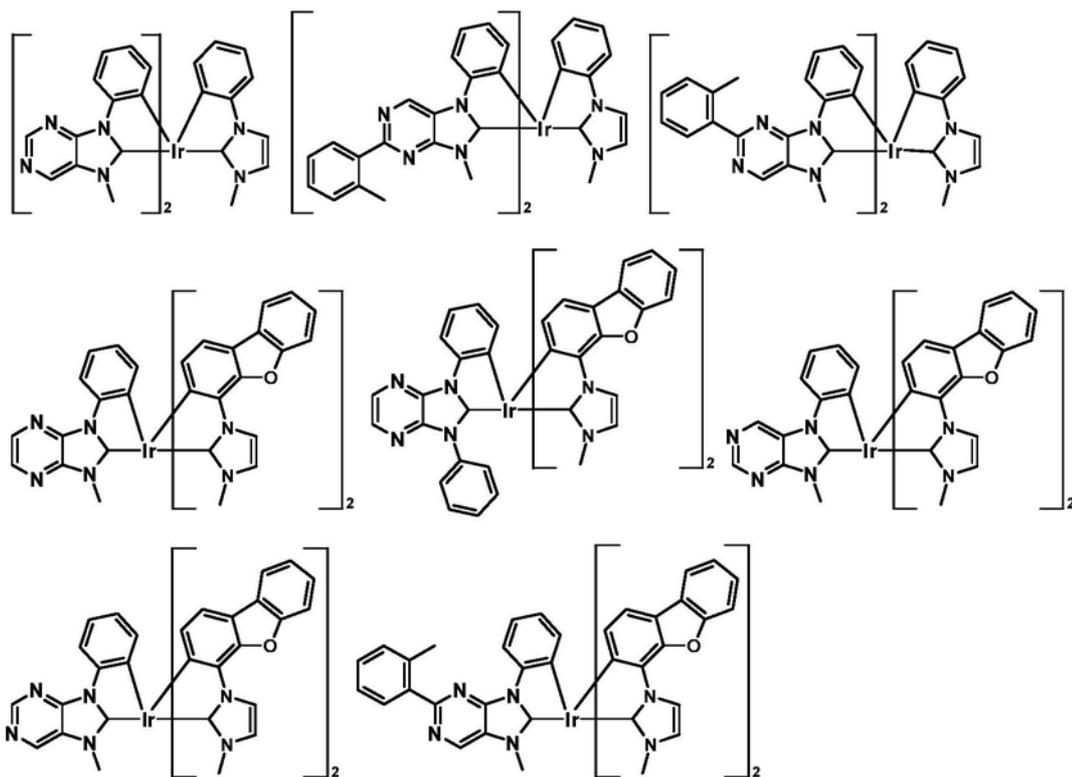


[0461]

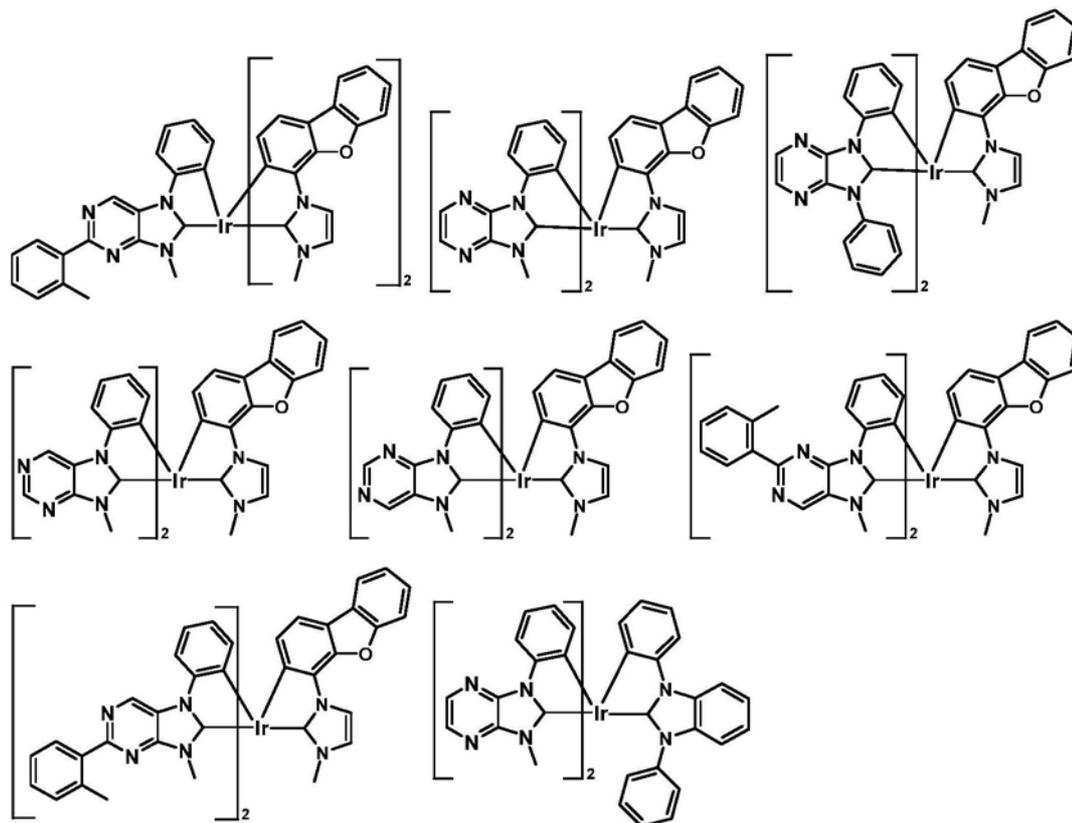


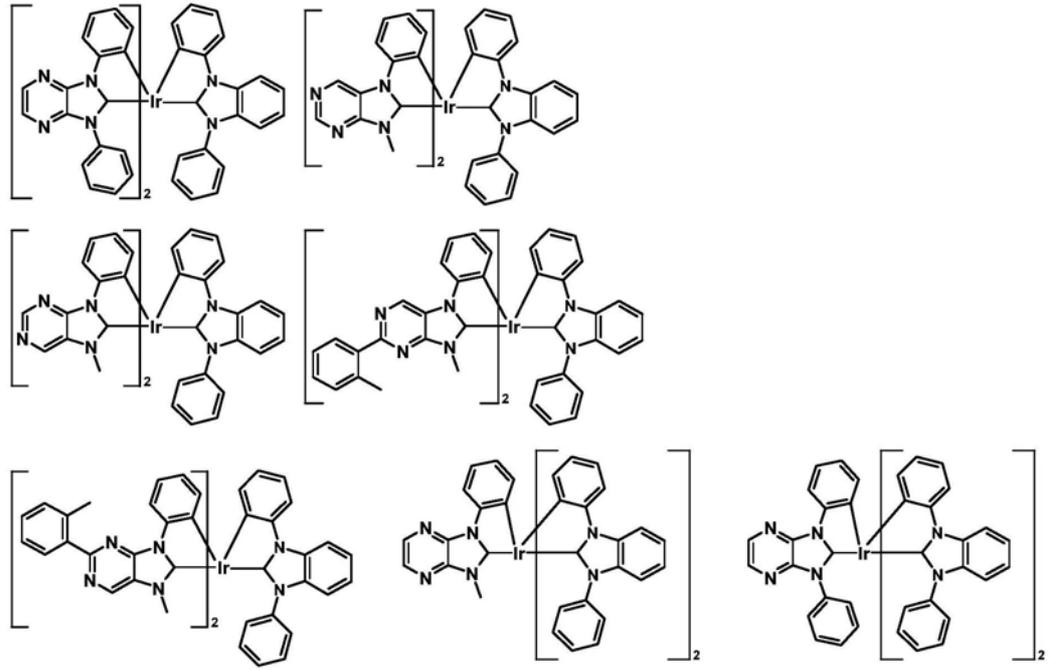
[0463]



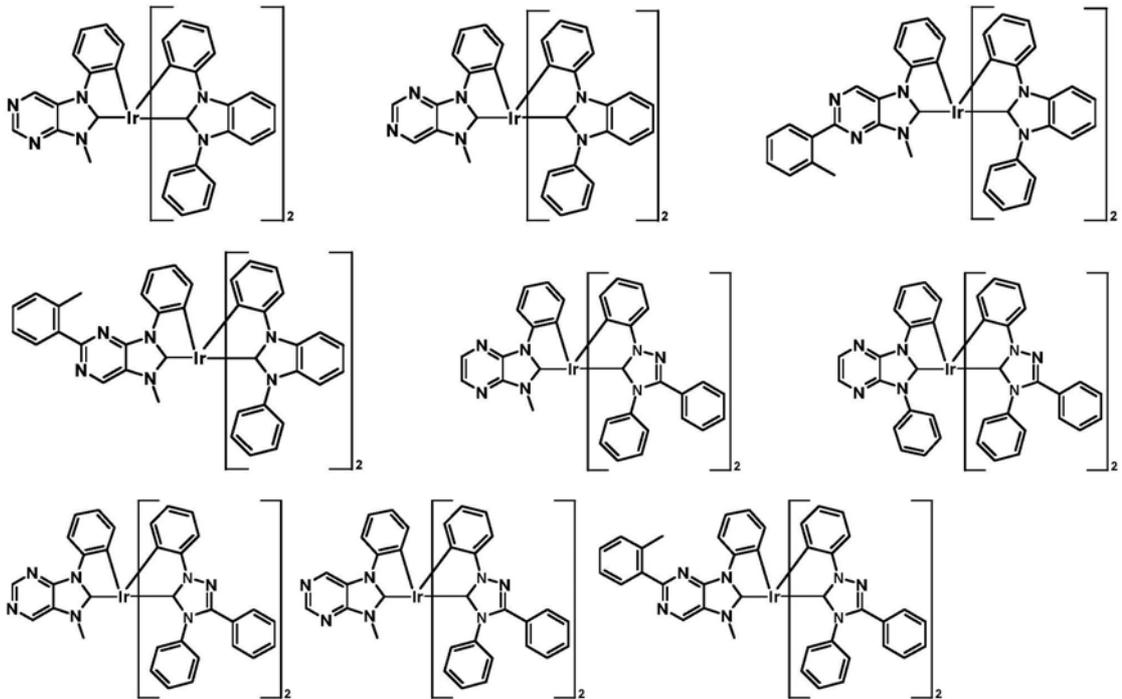


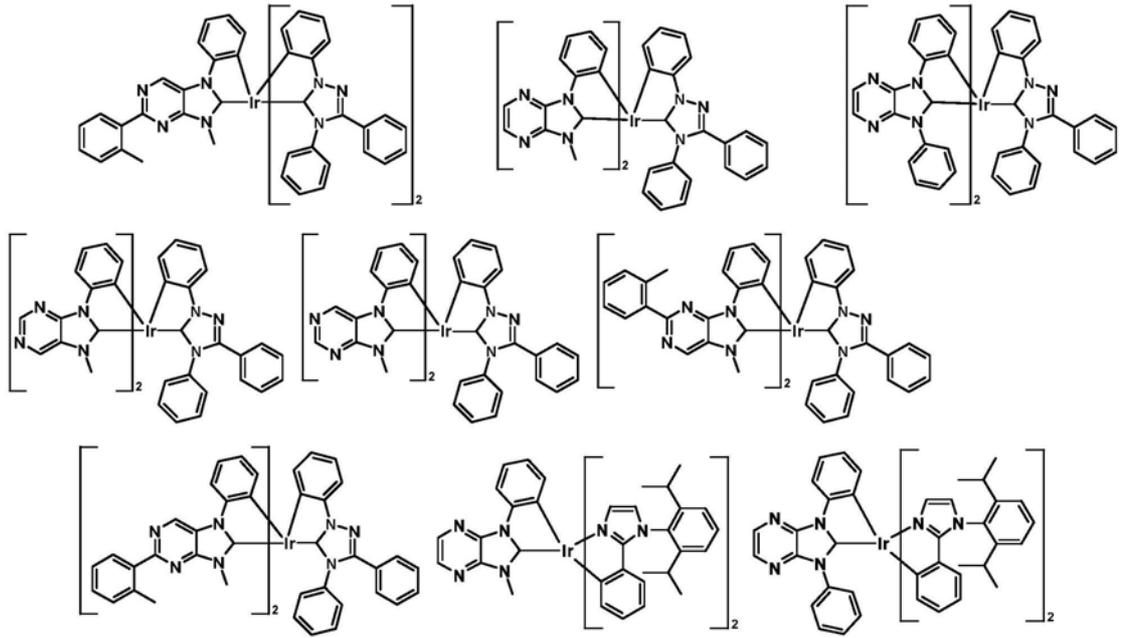
[0464]



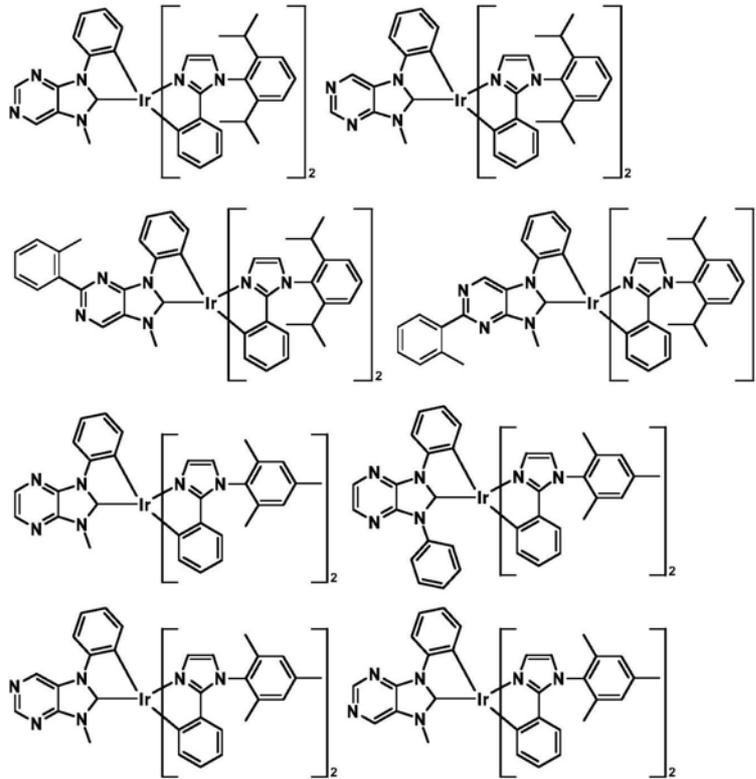


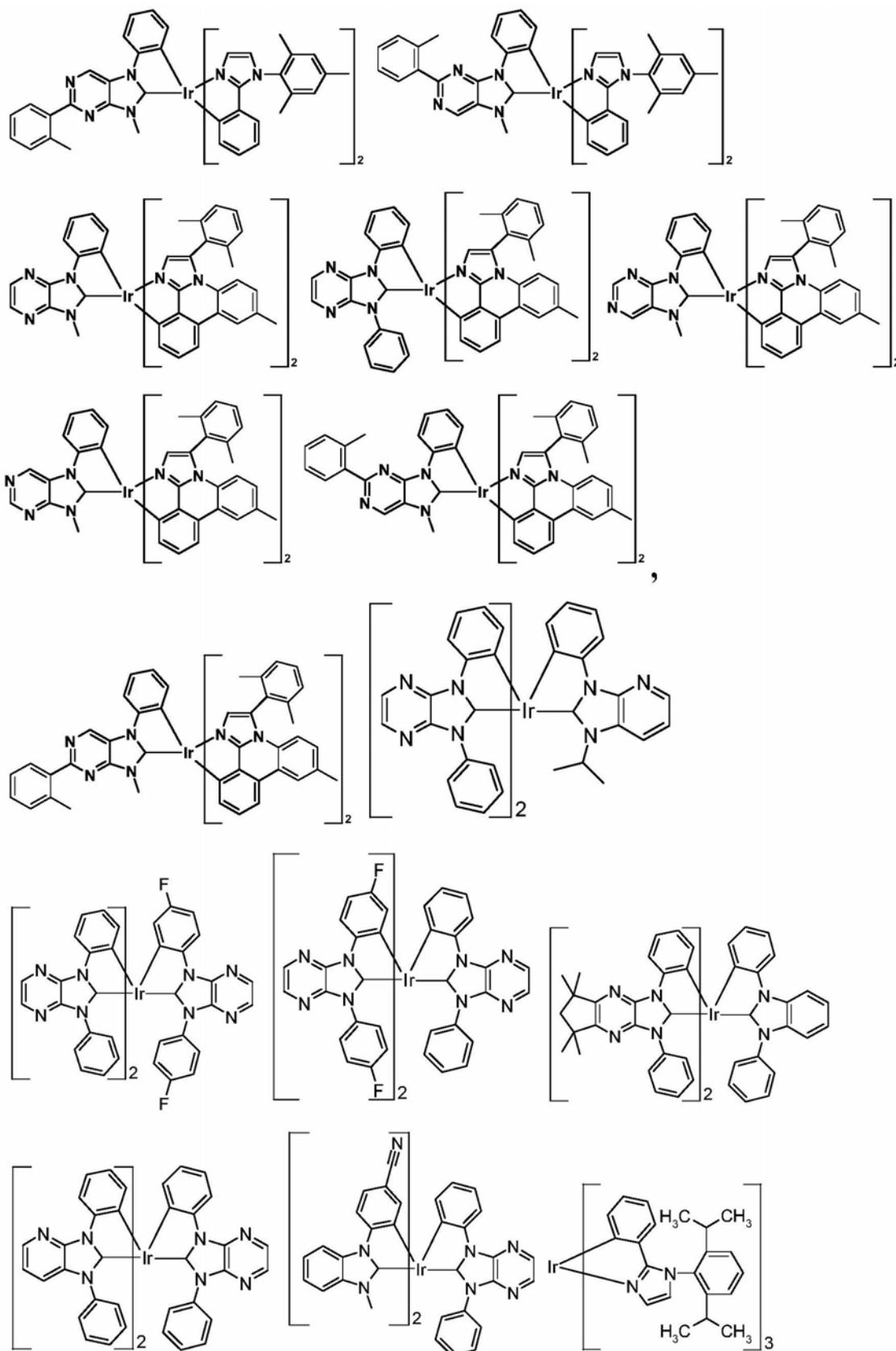
[0465]

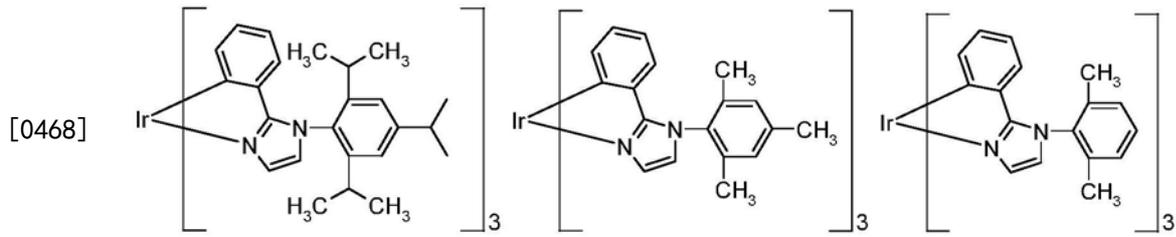




[0466]







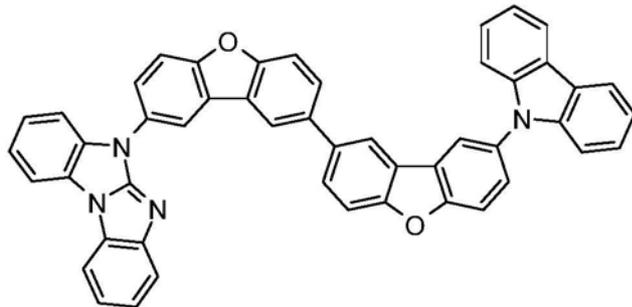
[0469] 均配型 (homoleptic) 金属-碳烯配合物可以面式异构体或子午线式异构体形式存在, 优选面式异构体。

[0470] 在杂配型 (heteroleptic) 金属-碳烯配合物的情况下, 可存在四种不同异构体, 优选假-面式异构体。

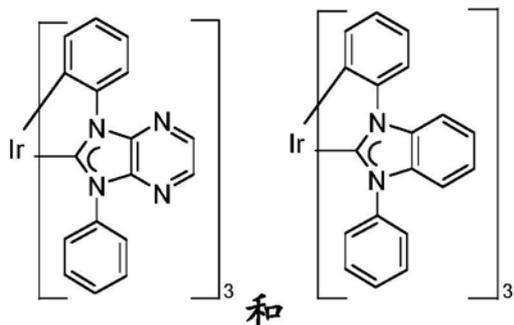
[0471] 除发光体材料外, 发光层可包含其他组分。例如, 发光层中可存在荧光染料以改变发光体材料的发射颜色。此外, 在一个优选实施方案中, 可使用基体材料。该基体材料可为聚合物如聚(N-乙烯基咪唑) 或聚硅烷。然而, 基体材料可为小分子, 例如4,4'-N,N'-二咪唑联苯 (CDP=CBP) 或叔芳族胺, 例如TCTA。在本发明的一个优选实施方案中, 将至少一种式I化合物用作基体材料。

[0472] 在一个优选实施方案中, 发光层由2-40重量%, 优选5-35重量%至少一种上述发光体材料和60-98重量%, 优选75-95重量%至少一种上述基体材料-在一个实施方案中至少一种式I化合物-形成, 其中发光体材料和基体材料的总和合计为100重量%。

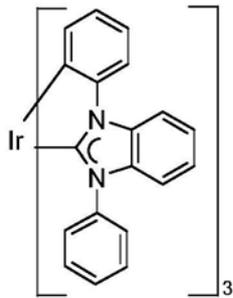
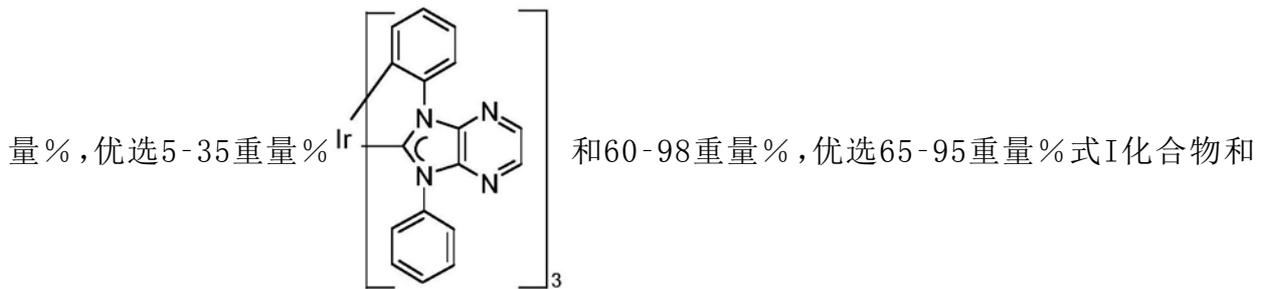
[0473] 在特别优选的实施方案中, 发光层包含式I化合物, 例如



和两种卡宾配合物, 优选式



的卡宾配合物。在所述实施方案中, 发光层由2-40重



形成，其中卡宾配合物和式I化合物的总和合计为100重量%。

[0474] 因此，适于与式I化合物一起在OLED中用作基体材料和/或空穴/激子阻断剂材料和/或电子/激子阻断剂材料和/或空穴注入材料和/或电子注入材料和/或空穴导体材料和/或电子导体材料，优选作为基体材料和/或空穴/激子阻断剂材料的金属配合物例如有如WO 2005/019373A2、WO 2006/056418A2、WO 2005/113704、WO 2007/115970、WO 2007/115981和WO 2008/000727所述的碳烯配合物。此处明确参考所引用WO申请的公开内容，应认为将这些公开内容并入本申请的内容中。

[0475] 如果空穴/激子阻断层(4)不含任何式I化合物，则OLED具有-如果空穴阻断层存在的话-常用OLED于中的空穴阻断剂材料例如为2,6-二(N-咔唑基)吡啶(mCPy)，2,9-二甲基-4,7-二苯基-1,10-菲咯啉(bathocuproin, (BCP))，二(2-甲基-8-喹啉根合)-4-苯基苯酚根合)铝(III)(BAIq)，吩噻嗪S,S-二氧化物衍生物和1,3,5-三(N-苯基-2-苄基咪唑基)苯(TPBI)，TPBI也适合作为电子传导材料。其他合适的空穴阻断剂和/或电子导体材料是2,2',2''-(1,3,5-苯三基)三(1-苯基-1-H-苯并咪唑)，2-(4-联苯基)-5-(4-叔丁基苯基)-1,3,4-噁二唑，8-羟基喹啉根合锂，4-(萘-1-基)-3,5-二苯基-4H-1,2,4-三唑，1,3-二[2-(2,2'-联吡啶-6-基)-1,3,4-噁二唑-5-基]苯，4,7-二苯基-1,10-菲咯啉，3-(4-联苯基)-4-苯基-5-叔丁基苯基-1,2,4-三唑，6,6'-二[5-(联苯-4-基)-1,3,4-噁二唑-2-基]-2,2'-联吡啶，2-苯基-9,10-二(萘-2-基)蒽，2,7-二[2-(2,2'-联吡啶-6-基)-1,3,4-噁二唑-5-基]-9,9-二甲基芴，1,3-二[2-(4-叔丁基苯基)-1,3,4-噁二唑-5-基]苯，2-(萘-2-基)-4,7-二苯基-1,10-菲咯啉，三(2,4,6-三甲基-3-(吡啶-3-基)苯基)硼烷，2,9-二(萘-2-基)-4,7-二苯基-1,10-菲咯啉，1-甲基-2-(4-(萘-2-基)苯基)-1H-咪唑并[4,5-f][1,10]菲咯啉。在另一实施方案中，可以在发光层(3)中使用WO 2006/100298中所公开的包含经由含羰基的基团连接的芳族或杂芳族环的化合物，例如如在本申请的优先权日还未公布的PCT申请PCT/EP2008/058207和PCT/EP2008/058106中所述的选自二甲硅烷基咪唑、二甲硅烷基苯并呋喃、二甲硅烷基苯并噻吩、二甲硅烷基苯并磷杂环戊二烯(disilylbenzophosphole)、二甲硅烷基苯并噻吩S-氧化物和二甲硅烷基苯并噻吩S,S-二氧化物的二甲硅烷基化合物以及如WO2008/034758中所公开的二甲硅烷基化合物作为空

穴/激子阻断层(4)或作为基体材料。

[0476] 适合本发明OLED的层(5)的电子导体材料包括与喔星类(oxinoid)化合物螯合的金属,例如2,2',2''-(1,3,5-亚苯基)三[1-苯基-1H-苯并咪唑](TPBI),三(8-喹啉根合)铝(Alq_3),基于菲咯啉的化合物如2,9-二甲基-4,7-二苯基-1,10-菲咯啉(DDPA=BCP)或4,7-二苯基-1,10-菲咯啉(DPA),以及唑类化合物如2-(4-联苯基)-5-(4-叔丁基苯基)-1,3,4-噁二唑(PBD)和3-(4-联苯基)-4-苯基-5-(4-叔丁基苯基)-1,2,4-三唑(TAZ),8-羟基喹啉根合锂(Liq),4,7-二苯基-1,10-菲咯啉(BPhen),二(2-甲基-8-喹啉根合)-4-(苯基苯酚根和)铝($BAlq$),1,3-二[2-(2,2'-联吡啶-6-基)-1,3,4-噁二唑-5-基]苯(Bpy-OXD),6,6'-二[5-(联苯-4-基)-1,3,4-噁二唑-2-基]-2,2'-联吡啶(BP-OXD-Bpy),4-(萘-1-基)-3,5-二苯基-4H-1,2,4-三唑(NTAZ),2,9-二(萘-2-基)-4,7-二苯基-1,10-菲咯啉(NBphen),2,7-二[2-(2,2'-联吡啶-6-基)-1,3,4-噁二唑-5-基]-9,9-二甲基芴(Bby-FOXD),1,3-二[2-(4-叔丁基苯基)-1,3,4-噁二唑-5-基]苯(OXD-7),三(2,4,6-三甲基-3-(吡啶-3-基)苯基)硼烷(3TPYMB),1-甲基-2-(4-(萘-2-基)苯基)-1H-咪唑并[4,5-f][1,10]菲咯啉(2-NPIP),2-苯基-9,10-二(萘-2-基)蒽(PADN),2-(萘-2-基)-4,7-二苯基-1,10-菲咯啉(HNBphen)。该层(5)既可以用于促进电子传输又可以用作缓冲层或阻隔层以防止激子在该OLED各层的界面处猝灭。该层(5)优选改善电子的迁移率并降低激子的猝灭。在一个优选实施方案中,将TBPI用作电子导体材料。在另一优选实施方案中,将BCP用作电子导体材料。原则上可能的是该电子导体层包含至少一种式I化合物作为电子导体材料。

[0477] 在上面作为空穴导体材料和电子导体材料提到的材料中,一些可能满足几个功能。例如,一些电子导体材料在具有低HOMO时同时也是空穴阻断材料。这些例如可以用于空穴/激子阻断层(4)中。然而,同样可以的是也采用层(5)作为空穴/激子阻断剂,使得可以省去层(4)。

[0478] 电荷传输层也可以是电子掺杂的,以改善所用材料的传输性能,首先使层厚更大(避免针孔/短路),其次使该器件的操作电压最小化。例如,该空穴导体材料可以用电子受体掺杂;例如,酞菁或芳基胺如TPD或TDTA可以用四氟四氰基醌二甲烷(F4-TCNQ)或 MoO_3 或 WO_3 掺杂。电子导体材料可以例如掺杂有碱金属,例如含有锂的 Alq_3 。此外,电子导体可以掺杂有盐,例如 Cs_2CO_3 或8-羟基喹啉根合锂(Liq)。电子掺杂为本领域熟练技术人员已知且例如公开于W.Gao,A.Kahn,J.Appl.Phys.,第94卷,第1期,2003年7月1日(p-掺杂有机层);A.G.Werner,F.Li,K.Harada,M.Pfeiffer,T.Fritz,K.Leo.Appl.Phys.Lett.,第82卷,第25期,2003年6月23日以及Pfeiffer等,Organic Electronics 2003,4,89-103中。例如,该空穴导体层除了卡宾配合物如 $Ir(dpbic)_3$ 外还可以被 MoO_3 或 WO_3 掺杂。例如,电子导体层可包含掺杂有 Cs_2CO_3 的BCP。

[0479] 阴极(6)为用于引入电子或负载流子的电极。适合阴极的材料选自元素周期表(老的IUPAC版)第Ia族碱金属,例如Li、Cs,第IIa族碱土金属,例如钙、钡或镁,第IIb族金属,包括镧系元素和锕系元素,例如钐。此外,还可以使用金属如铝或铟,以及所有所述金属的组合。此外,可以在该有机层和阴极之间施加含有碱金属,尤其是含有锂的有机金属化合物或碱金属氟化物,例如LiF、CsF或KF,以降低操作电压。

[0480] 本发明的OLED可以额外包括本领域熟练技术人员已知的其他层。例如,可以在层

(2) 和发光层 (3) 之间施加促进正电荷传输和/或使各层的带隙相互匹配的层。或者, 该其他层可以用作保护层。以类似方式, 可以在发光层 (3) 和层 (4) 之间存在额外层, 以促进负电荷传输和/或使各层之间的带隙相互匹配。或者, 该层可以用作保护层。

[0481] 在一个优选实施方案中, 本发明 OLED 除了层 (1) - (6) 外还包括下文所述下列层中的至少一个:

[0482] - 厚度为 2-100nm, 优选 5-50nm 的在阳极 (1) 和空穴传输层 (2) 之间的空穴注入层;

[0483] - 在空穴传输层 (2) 和发光层 (3) 之间的电子阻断层;

[0484] - 在电子传输层 (5) 和阴极 (6) 之间的电子注入层。

[0485] 用于空穴注入层的材料可以选自铜酞菁, 4,4',4''-三(N-3-甲基苯基-N-苯基氨基)三苯基胺(m-MTDATA), 4,4',4''-三(N-(2-萘基)-N-苯基氨基)三苯基胺(2T-NATA), 4,4',4''-三(N-(1-萘基)-N-苯基氨基)三苯基胺(1T-NATA), 4,4',4''-三(N,N-二苯基氨基)三苯基胺(NATA), 氧化钛酞菁, 2,3,5,6-四氟-7,7,8,8-四氰基醌二甲烷(F4-TCNQ), 吡嗪并[2,3-f][1,10]菲咯啉-2,3-二甲腈(PPDN), N,N,N',N'-四(4-甲氧基苯基)联苯胺(MeO-TPD), 2,7-二[N,N-二(4-甲氧基苯基)氨基]-9,9-螺二芴(MeO-Spiro-TPD), 2,2'-二[N,N-二(4-甲氧基苯基)氨基]-9,9-螺二芴(2,2'-MeO-Spiro-TPD), N,N'-二苯基-N,N'-二-[4-(N,N-二-甲苯基氨基)苯基]联苯胺(NTNPB), N,N'-二苯-N,N'-二-[4-(N,N-二苯基氨基)苯基]联苯胺(NPNPB), N,N'-二(萘-2-基)-N,N'-二苯基苯-1,4-二胺(α -NPP)。原则上可能的是该空穴注入层包含至少一种式 I 化合物作为空穴注入材料。此外, 可以使用聚合物空穴注入材料, 如聚(N-乙烯基吡啶)(PVK), 聚噻吩, 聚吡咯, 聚苯胺, 自掺杂聚合物如磺化聚(噻吩-3-[2[(2-甲氧基乙氧基)乙氧基]-2,5-二基]) (**Plexcore**[®] OC Conducting Inks, 由 Plextronics 市购) 以及共聚物如也称为 PEDOT/PSS 的聚(3,4-亚乙二氧基噻吩)/聚(4-苯乙烯磺酸酯)。

[0486] 作为电子注入层的材料, 例如可以选择 LiF。原则上, 电子注入层可以包含至少一种式 I 化合物作为电子注入材料、

[0487] 本领域熟练技术人员知晓(例如基于电化学研究) 如何选择合适材料。适合各层的材料对本领域熟练技术人员而言是已知的且例如公开于 W000/70655 中。

[0488] 此外, 可能的是一些用于本发明 OLED 中的层已经进行表面处理, 以提高载流子传输效率。对所述各层的材料选择优选通过获得具有高效率和使用寿命的 OLED 决定。

[0489] 本发明 OLED 可以通过本领域熟练技术人员已知的方法生产。通常而言, 本发明 OLED 通过在合适基底上依次气相沉积各层而生产。合适的基底例如为玻璃, 无机半导体, 或聚合物薄膜。对于气相沉积, 可以使用常规技术, 如热蒸发、化学气相沉积(CVD)、物理气相沉积(PVD) 以及其他技术。在一种替换方法中, 可以使用本领域熟练技术人员已知的涂敷技术由在合适溶剂中的溶液或分散体施加该 OLED 的有机层。

[0490] 不同的层通常具有下列厚度: 阳极 (1) 50-500nm, 优选 100-200nm; 空穴传导层 (2) 5-100nm, 优选 20-80nm, 发光层 (3) 1-100nm, 优选 10-80nm, 空穴/激子阻断层 (4) 2-100nm, 优选 5-50nm, 电子传导层 (5) 5-100nm, 优选 20-80nm, 阴极 (6) 20-1000nm, 优选 30-500nm。空穴和电子在本发明 OLED 中的再结合区相对于阴极的相对位置以及因此该 OLED 的发光光谱尤其可以由各层的相对厚度影响。这意味着电子传输层的厚度应优选选择得使再结合区的位置与该二极管的光学共振器性能匹配并因此与该发光体的发光波长匹配。该 OLED 中各层的

层厚比例取决于所用材料。所用任何额外层的层厚为本领域熟练技术人员已知。可能的是电子传导层和/或空穴传导层在电掺杂时具有的厚度大于所述层厚度。

[0491] 将式I化合物用于OLED的至少一个层,优选在发光层(优选作为基体材料)中和/或在用于空穴/激子的阻断层中可以获得具有高效率以及低使用和操作电压的OLED。通过使用式I化合物获得的OLED通常额外具有高使用寿命。该OLED的效率可以通过优化该OLED的其他层而额外改善。例如,可以使用高效率阴极如Ca或Ba,如果合适的话,与LiF的中间层组合。成型基底和导致操作电压降低或量子效率提高的新型空穴传输材料同样可以用于本发明OLED中。此外,额外层可以存在于该OLED中以调节不同层的能级并促进场致发光。

[0492] 该OLED可以进一步包括至少一个第二发光层。该OLED的总发光可以包含至少两个发光层的发光且还可以包括白光。

[0493] 该OLED可以用于所有其中场致发光有用的设备中。合适的器件优选选自固定和移动可视显示单元以及照明单元。固定可视显示单元例如为计算机、电视的可视显示单元,打印机、厨房用具和广告板、照明和信息板中的可视显示单元。移动可视显示单元例如为移动电话、台式PC、便携式电脑、数字相机、MP3播放器、车辆以及公共汽车和火车上的目的地显示中的可视显示单元。其中可以使用本发明OLED的其他器件例如为键盘;衣物;家具;壁纸。

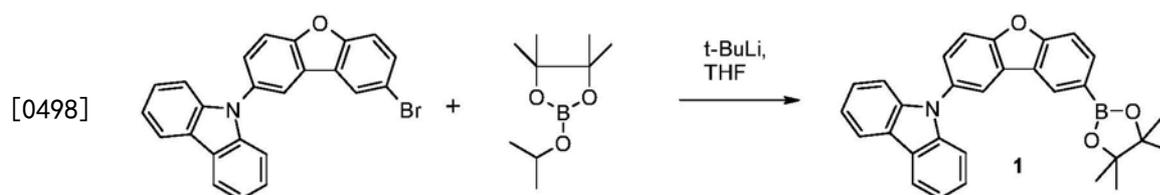
[0494] 此外,本发明涉及选自如下的器件:固定可视显示单元如计算机、电视的可视显示单元,打印机、厨房用具和广告板、照明、信息板中的可视显示单元以及移动可视显示单元如移动电话、台式PC、便携式电脑、数字相机、MP3播放器、车辆以及公共汽车和火车上的目的地显示中的可视显示单元;照明单元;键盘;衣物;家具;壁纸,其包含至少一种本发明有机发光二极管或至少一种本发明发光层。

具体实施方式

[0495] 下列实施例仅以说明目的包括在此且不限权利要求书的范围。除非另有指明,所有份数和百分数按重量计。

[0496] 实施例

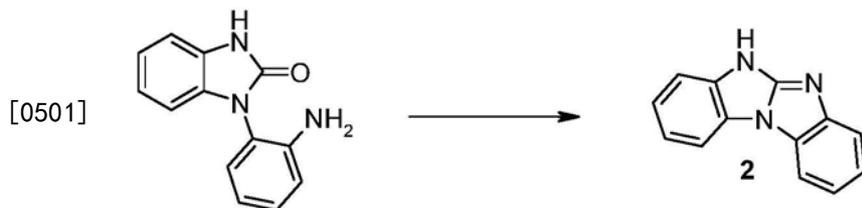
[0497] 实施例1



[0499] a) 在 -78°C 下在氩气下将16.4ml (27.9mmol) 在戊烷中的叔丁基锂加入5.00g (12.1mmol) 9-(8-溴二苯并呋喃-2-基)咔唑(其合成描述于W02010079051)在30ml无水四氢呋喃(THF)中的溶液中。在15分钟之后,缓慢加入2.93g (15.8mmol) 2-异丙氧基-4,4,5,5-四甲基-1,3,2-二氧化杂戊硼烷。将反应混合物在 -78°C 下在氩气下搅拌3h,倾入水中并将水相用乙醚萃取。使有机相用硫酸镁干燥并除去溶剂。由醚结晶得到2.57g化合物1(产率:46%)。

[0500] ^1H NMR (400MHz, CDCl_3): δ 8.50 (s, 1H), 8.18-8.23 (m, 3H), 8.05 (dd, $J=8.3\text{Hz}$, $J=1.3\text{Hz}$, 1H), 7.80-7.82 (m, 1H), 7.64-7.70 (m, 2H), 7.43-7.49 (m, 4H), 7.28-7.37 (m, 2H),

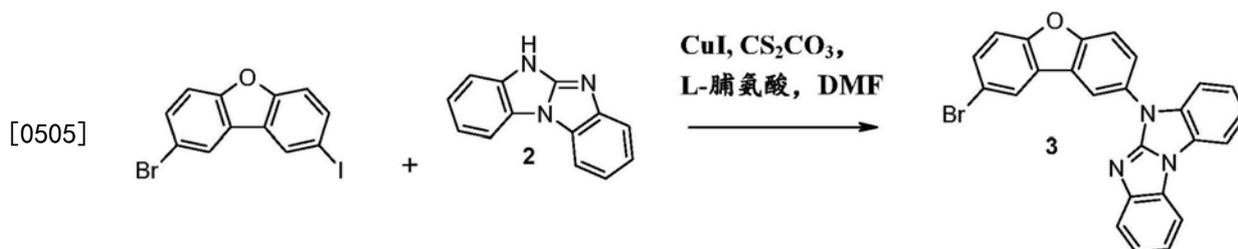
1.43 (s, 12H) .



[0502] b) 在180℃下将11.3g (50.0mmol) 3-(2-氨基苯基)-1H-苯并咪唑-2-酮加入50g多磷酸。将反应混合物在220℃下在氮气下搅拌3h并倾入水中。将产物滤出并用水和甲醇洗涤。将50ml 30%氢氧化钠溶液加入产物在200ml THF中的悬浮液。将该混合物搅拌30分钟并使有机相分离,使用硫酸镁干燥并蒸除溶剂。获得9.26g化合物2 (产率:89%)。

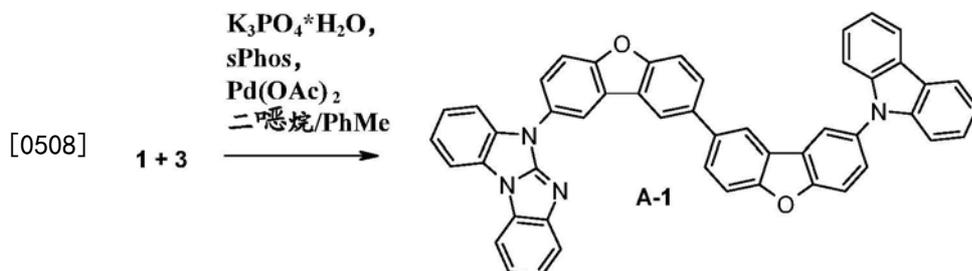
[0503] ^1H NMR (400MHz, DMSO-d₆) : δ 7.88 (d, J=7.7Hz, 2H) , 7.39 (d, J=8.0Hz, 2H) , 7.12-7.16 (m, 2H) , 6.97-7.01 (m, 2H) .

[0504] 5H-苯并咪唑并[1,2-a]苯并咪唑和3-(2-氨基苯基)-1H-苯并咪唑-2-酮的合成描述于Bull.Soc.Chem.Belg.96 (1987) 787中。



[0506] c) 在氮气下将5.00g (13.4mmol) 2-溴-8-碘二苯并呋喃 (其合成描述于EP1885818) 、8.74g (26.8mmol) 碳酸铯、255mg (1.34mmol) 碘化铜(I) 和309mg (2.68mmol) L-脯氨酸加入2.78g (13.4mmol) 在75ml 二甲基甲酰胺中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在150℃下加热19h并在Hyflo Super Cel (R) 介质,Fluka 56678,CAS[91053-39-3]上使用THF过滤。用水洗涤有机相。蒸除溶剂。用甲苯/乙酸乙酯19/1在硅胶上柱层析得到化合物3 (产率:2.29g (37.7%))。

[0507] ^1H NMR (400MHz, THF-d₈) : δ 8.66 (s, 1H) , 8.41 (s, 1H) , 8.01-8.16 (m, 3H) , 7.89 (d, J=8.8Hz, 1H) , 7.63-7.75 (m, 4H) , 7.25-7.49 (m, 4H) .



[0509] d) 将1.98g (3.41mmol) 化合物1和4.02g (16.6mmol) 磷酸三钾单水合物 (potassium phosphate tribasic monohydrate) 、15ml 二噁烷、60ml 甲苯和12ml 水加入1.50g (3.32mmol) 化合物3中。将该混合物用氩气脱气。加入81mg (0.199mmol) 2-二环己基膦基-2',6'-二甲氧基联苯 (sPhos) 和74mg (0.033mmol) 乙酸钾 (II) 。将反应混合物用氩气脱气并在氩气下在100℃下搅拌4.5h。加入40ml 氰化钠的1% 溶液并将反应混合物回流1小时。加入

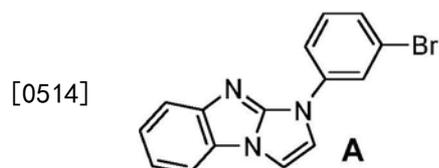
二氯甲烷,将有机相用水洗涤并用硫酸镁干燥。用甲苯和然后甲苯/乙酸乙酯9/1在硅胶上柱层析得到化合物A-1(产率:1.42g(61%))。¹H NMR(400MHz,THF-d₈): δ 8.71(s,1H),8.54-8.58(m,2H),8.41(s,1H),8.20(d,J=7.8Hz,2H),7.62-8.11(m,12H),7.26-7.31(m,10H)。

[0510] 实施例2

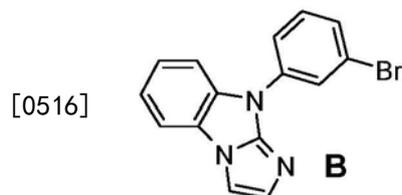
[0511] 4H-咪唑并[1,2-a]苯并咪唑的合成描述于ARKIVOC 2002(v)48-61中。



[0513] a) 在氮气下将8.02g(51.0mmol)4H-咪唑并[1,2-a]苯并咪唑、15.9g(56.1mmol)1-溴-3-碘苯、33.2g(102mmol)碳酸铯、1.94g(10.2mmol)碘化铜(I)和2.35g(20.4mmol)L-脯氨酸在200ml二甲亚砜(DMSO)中在100℃下搅拌24h。将固体滤出并用二氯甲烷洗涤。将有机相用水洗涤并用硫酸镁干燥。蒸除溶剂。用甲苯和然后甲苯/乙酸乙酯(20/1)柱层析得到产物混合物(3.89g(24%)异构体A和4.46g(28%)异构体B)。两种异构体的分离通过用甲苯/乙酸乙酯(甲苯100%,甲苯/乙酸乙酯95/5,甲苯/乙酸乙酯90/10和乙酸乙酯100%)梯度柱层析实现。

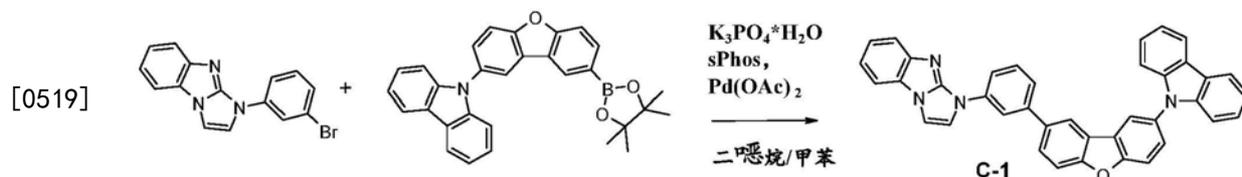


[0515] ¹H NMR(400MHz,THF-d₈): δ 8.54-8.56(m,1H),8.33(dd,J=7.8Hz,J=1.4Hz,1H),7.80(d,J=2.8Hz,1H),7.76(d,J=2.8Hz,1H),7.79(d,J=8.6Hz,2H),7.39-7.46(m,2H),7.20-7.29(m,1H),7.12-7.16(m,1H)。



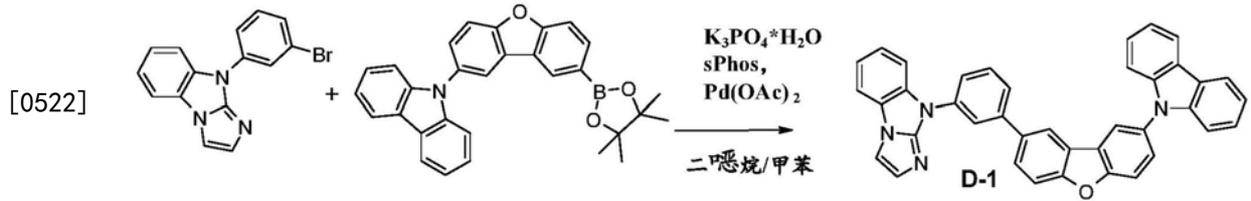
[0517] ¹H NMR(400MHz,THF-d₈): δ 8.23(s,1H),7.95-7.97(m,1H),7.70-7.74(m,2H),7.56(s,1H),7.45-7.53(m,2H),7.24-7.33(m,2H),7.17(s,1H)。

[0518] b) 合成化合物C-1类似于合成化合物A-1进行。



[0520] ¹H NMR(400MHz,THF-d₈): δ 8.63(s,1H),8.52(s,1H),8.42(s,1H),8.32-8.35(m,1H),8.20-8.22(m,2H),7.59-8.02(m,10H),7.40-7.47(m,4H),7.09-7.20(m,4H)。

[0521] c) 实施例2c)的产物的合成类似于合成化合物A-1进行。

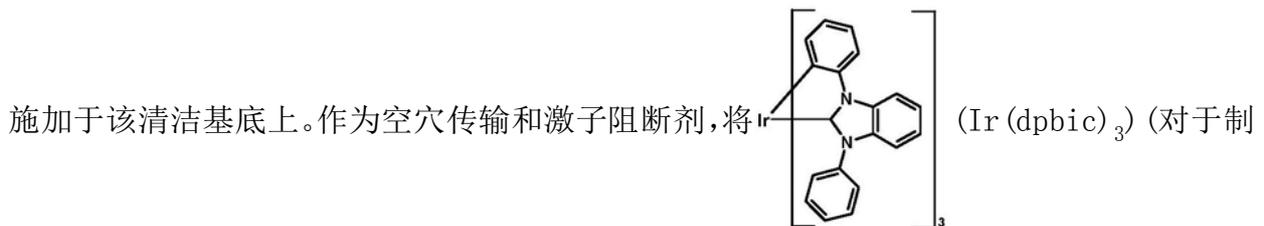


[0523] ¹H NMR (400MHz, THF-d8) : δ = 8.51 (d, J = 1.7Hz, 1H), 8.41 (d, J = 2.1Hz, 1H), 8.37-8.39 (m, 1H), 8.21 (s, 1H), 8.19 (s, 1H), 7.98 (dd, J = 8.6Hz, J = 1.9Hz, 1H), 7.91-7.94 (m, 2H), 7.65-7.82 (m, 6H), 7.57 (d, J = 1.5Hz, 1H), 7.37-7.44 (m, 4H), 7.24-7.34 (m, 4H), 7.12 (d, J = 1.5Hz, 1H).

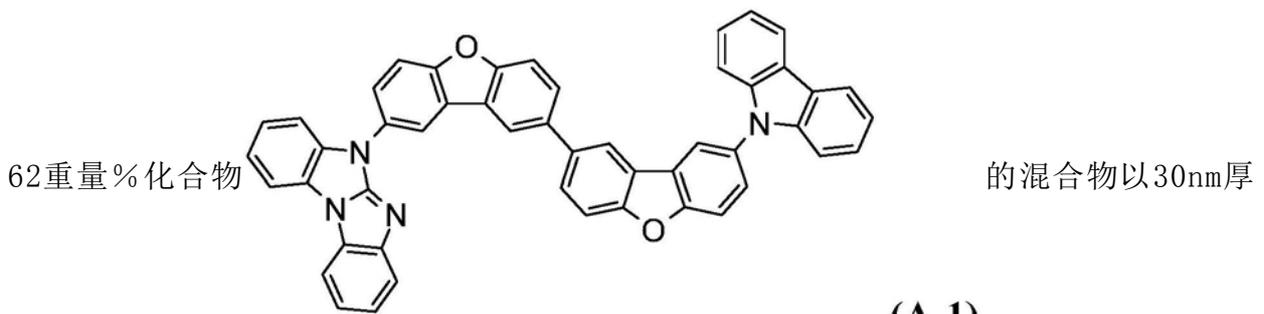
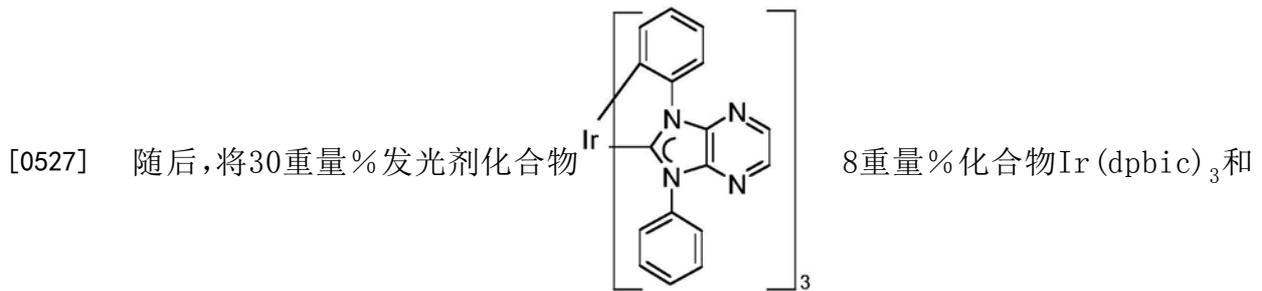
[0524] 应用实施例1-混合基体

[0525] 首先将用作阳极的ITO基底用LCD生产用市售清洁剂 (**Deconex**[®] 20NS和25 **ORGAN-ACID**[®]中和剂) 清洁,然后在丙酮/异丙醇混合物中在超声浴中清洁。为了消除任何可能的有机残余物,在臭氧炉中将该基底暴露于连续臭氧流另外25分钟。该处理也改进该ITO的空穴注入性能。然后旋涂 **Plexcore**[®] OC AJ20-1000 (由Plextronics Inc.市购) 并干燥,形成空穴注入层 (~40nm)。

[0526] 然后在约10⁻⁷-10⁻⁹毫巴下通过气相沉积将下述有机材料以约0.5-5nm/min的速率

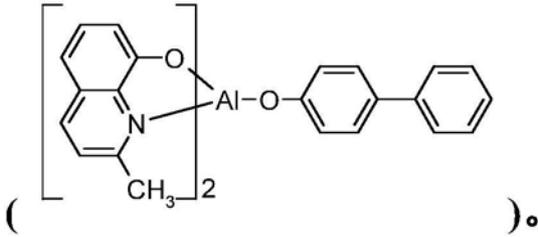


备见申请WO 2005/019373中的Ir配合物(7))以20nm的厚度施加于该基底上,其中最初10nm用MoO_x (~10%) 掺杂以改善导电率。

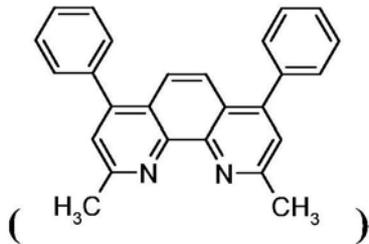


度通过气相沉积施加。

[0528] 然后通过气相沉积以5nm的厚度施加作为阻断剂的材料BA1q



[0529] 接下来通过气相沉积以20nm的厚度施加Cs₂CO₃掺杂的BCP

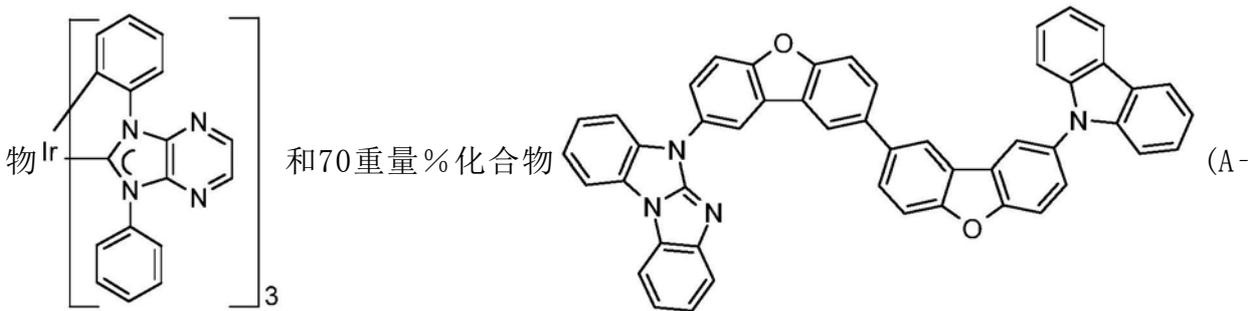


层作为电子传输层,最后施加100nm厚的Al电极完成器件。

[0530] 所有制造部件用玻璃盖和吸气剂密封在惰性氮气气氛中。

[0531] 应用实施例2-单基体

[0532] OLED的生产和构造按照应用实施例1中进行,不同的是发光层仅由30重量%化合



1) 构成,即不包含化合物Ir (dpbic)₃。

[0533] 为了表征该OLED,在各种电流和电压下记录场致发光光谱。此外,与发射的光输出组合测量电流-电压特性。光输出可以通过用光度计校正而转化成测光参数。为了测定使用寿命,使该OLED在恒定电流密度下操作并记录光输出降低。使用寿命定义为直到亮度降低至初始亮度的一半所流逝的时间。

	EML	电压 @300nits[V]	EQE ¹⁾ @300nits[%]	使用寿命 @4000nits[h]	CIE
[0534] 应用实施例 1	混合基体	3.8 V	14.7%	125h	0.17/0.33
应用实施例 2	单基体	3.6V	14.2%	65h	0.17/0.34

[0535] ¹⁾ 外部量子效率 (EQE) 为由物质或器件逃逸所产生的光子#/流过它的电子#。

[0536] 实施例3

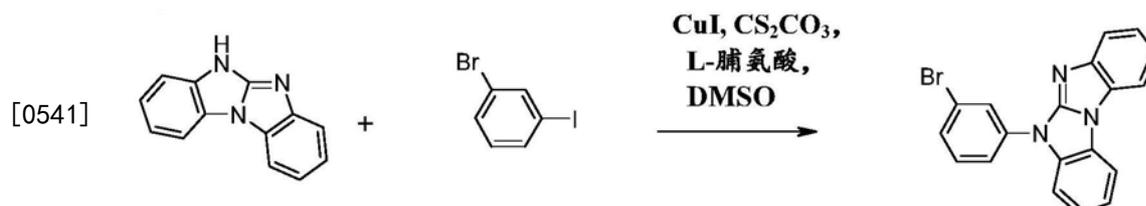
[0537]



[0538] 在氮气下将3.30g (10mmol) 1,3-二碘苯、13.0g (40.0mmol) 碳酸铯、1.90g (1.00mmol) 碘化铜(I) 和2.30g (20.0mmol) L-脯氨酸加入4.56g (22.0mmol) mmol) 在100ml 二甲亚砜(DMSO) 中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在100℃下搅拌5h。将反应混合物倾入水中并滤出产物。将产物由甲苯结晶两次。产率为1.6g (48%)。MS (APCI (pos) : $m/z=489 (M^+)$.

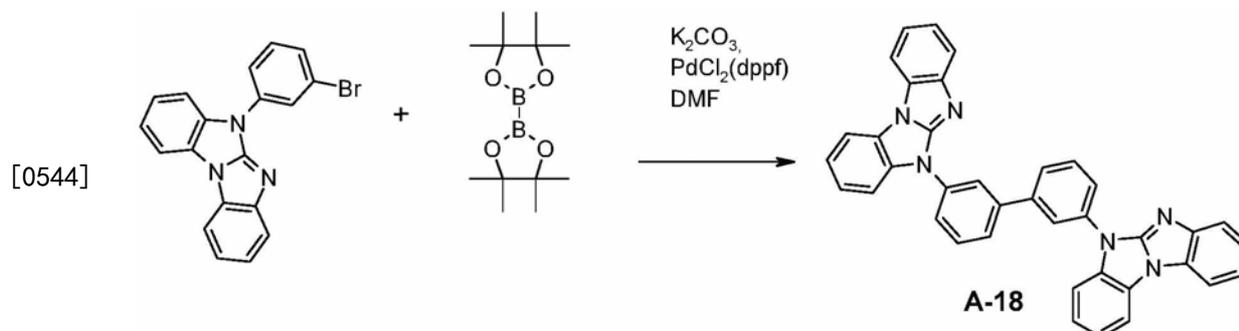
[0539] $^1\text{H NMR}$ (400MHz, THF-d8) : δ 8.79 (s, 1H) , 8.22 (d, $J=8.4\text{Hz}$, 2H) , 8.15-8.18 (m, 2H) , 8.00-8.06 (m, 4H) , 7.88 (t, $J=8.1\text{Hz}$, 1H) 7.71 (d, $J=7.9\text{Hz}$, 2H) , 7.41-7.49 (m, 4H) , 7.25-7.34 (m, 4H) .

[0540] 实施例4



[0542] a) 在氮气下将7.78g (25mmol) 1-溴-3-碘苯、16.3g (50.0mmol) 碳酸铯、1.24g (6.50mmol) 碘化铜(I) 和1.50g (13.0mmol) L-脯氨酸加入5.18g (25.0mmol) 在100ml 二甲亚砜(DMSO) 中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在100℃下搅拌18h。将反应混合物倾入水中。将有机相用二氯甲烷萃取。将有机相用硫酸镁干燥。蒸除溶剂。用甲苯在硅胶上柱层析得到产物。产率为8.35g (92%)。

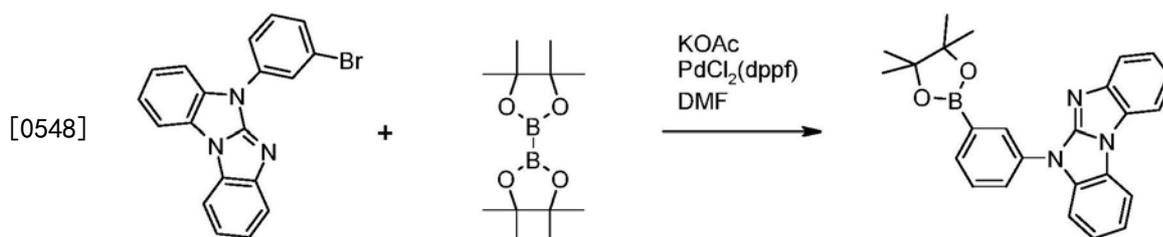
[0543] $^1\text{H NMR}$ (400MHz, CDCl_3) : δ 8.25 (s, 1H) , 7.90-8.05 (m, 3H) , 7.95-8.05 (m, 3H) , 7.71 (d, $J=7.9\text{Hz}$, 1H) , 7.65 (d, $J=7.9\text{Hz}$, 1H) . 7.50-7.65 (m, 2H) , 7.26-7.45 (m, 4H) .



[0545] b) 将1.09g (3.00mmol) 实施例4a) 的产物、690mg (2.70mmol) 4,4,5,5-四甲基-2-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基)-1,3,2-二氧杂戊硼烷、5.86g (1.80mmol) 在20ml DMF中的碳酸钾用氩气脱气。加入1,1'-双(二苯膦基)二茂铁二氯化钯(II) 并将反应混合物用氩气脱气。将反应混合物在80℃下搅拌18h。滤出产物并用二甲基甲酰胺(DMF)、水和甲醇洗涤。产率为370mg (44%)。

[0546] $^1\text{H NMR}$ (400MHz, CDCl_3) : δ 8.43 (s, 2H) , 7.95-8.10 (m, 6H) 7.70-7.90 (m, 6H) , 7.63 (d, $J=7.6\text{Hz}$, 2H) , 7.20-7.45 (m, 8H) .

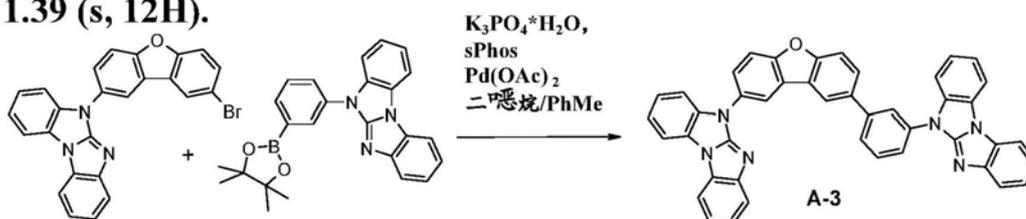
[0547] 实施例5



[0549] a) 将5.78g (16.0mmol) 实施例4a) 的产物、12.16g (47.8mmol) 4,4,5,5-四甲基-2-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基)-1,3,2-二氧杂戊硼烷、12.5g (0.128mol) 乙酸钾在50ml DMF中用氩气脱气。加入1,1'-双(二苯膦基)二茂铁)二氯化钯(II)并将反应混合物用氩气脱气。将反应混合物在60℃下搅拌22h并倾入氯化钠的饱和水溶液。将水相用四氢呋喃(THF)萃取,将有机相用硫酸镁干燥并蒸除溶剂。将产物由乙醚和环己烷结晶。产率为3.62g (59%)。

[0550] ^1H NMR (400MHz, THF-d8) : δ 8.26 (s, 1H) , 8.09-8.10 (m, 1H) , 8.07-8.09 (m, 2H) , 7.86 (s, J=7.6Hz, 1H) , 7.60-7.67 (m, 3H) , 7.28-7.42 (m,

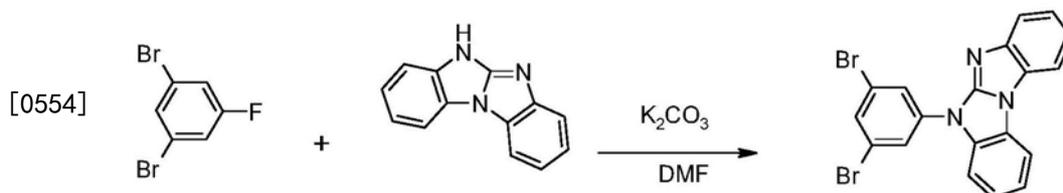
4H), 1.39 (s, 12H).



[0551] b) 2.72g (6.01mmol) 5-(8-溴二苯并咪唑-2-基)苯并咪唑并[1,2-a]苯并咪唑和6.92g (3.01mmol) 磷酸三钾单水合物、27ml 二噁烷、100ml 甲苯和21ml 水加入3.20g (7.82mmol) 5-[3-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基)苯基]苯并咪唑并[1,2-a]苯并咪唑。将混合物用氩气脱气。加入148mg (0.361mmol) 2-二环己基膦基-2',6'-二甲氧基联苯(sPhos)和135mg (0.060mmol) 乙酸钯(II)。将反应混合物用氩气脱气并在氩气下在100℃下搅拌22h。加入110ml 氰化钠的1%溶液并将反应混合物回流1小时。加入二氯甲烷,将有机相用水洗涤并用硫酸镁干燥。产物用甲醇煎煮(产率:1.62g (41%))。

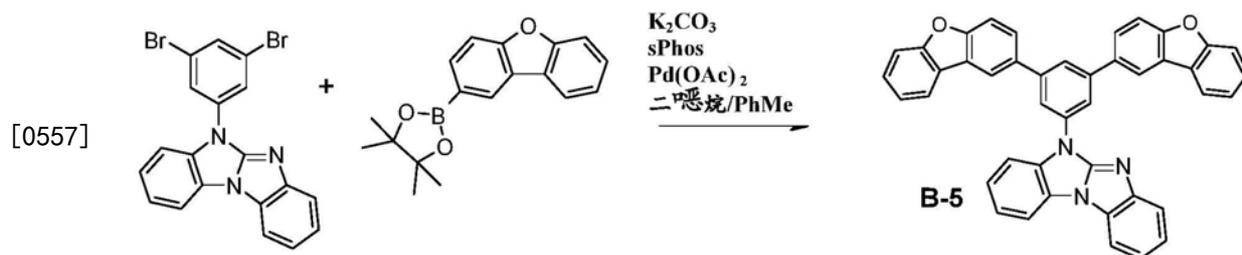
[0552] ^1H NMR (400MHz, THF-d8) : δ 8.69 (d, J=1.1Hz, 1H) , 8.57 (d, J=1.6Hz, 1H) , 8.42 (s, 1H) , 8.05-8.08 (m, 1H) , 7.89-7.99 (m, 6H) , 7.80-7.85 (m, 2H) , 7.76-7.75 (m, 4H) , 7.57-7.61 (m, 2H) , 7.18-7.37 (m, 8H) .

[0553] 实施例6



[0555] a) 将20.0g (78.8mmol) 1,3-二溴-5-氟苯、16.3g (78.8mmol) 6H-苯并咪唑并[1,2-a]苯并咪唑和43.5g (0.315mmol) 碳酸钾在200ml DMF中在170℃下搅拌17h。将反应混合物趁热过滤并将来自母液的沉淀物在冷却之后过滤。将产物用水和乙醇洗涤并用乙醚和乙醇煎煮。产率为21.2g (61%)。

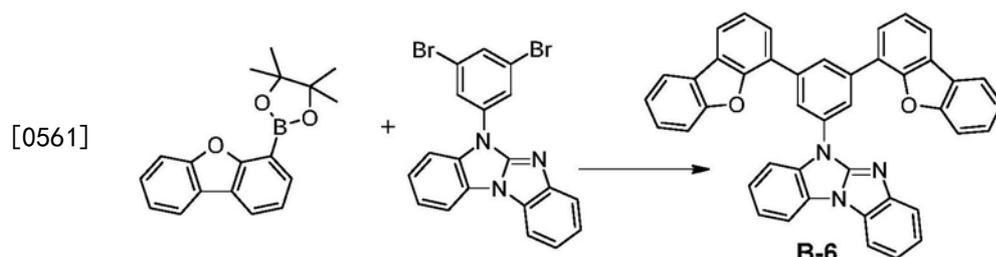
[0556] ^1H NMR (400MHz, THF-d8) : δ 8.21-8.26 (m, 4H) , 7.98-7.8.00 (m, 1H) , 7.68-7.73 (m, 2H) , 7.31-7.49 (m, 4H) .



[0558] b) 将2.00g (4.53mmol) 5-(3,5-二溴苯基)苯并咪唑并[1,2-a]苯并咪唑和4.15g (3.00mmol) 碳酸钾、27ml 二噁烷、100ml 甲苯和21ml 水加入3.20g (7.82mmol) 2-二苯并咪喃-2-基-4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷。将混合物用氩气脱气。加入37mg (0.090mmol) 2-二环己基磷基-2',6'-二甲氧基联苯 (sPhos) 和10mg (0.0045mmol) 乙酸钯 (II)。将反应混合物用氩气脱气并在氩气下在120℃下搅拌19h。加入110ml 氰化钠的1%溶液并将反应混合物回流1小时。蒸除溶剂。加入30ml 甲苯, 滤出产物, 用水和环己烷洗涤并由甲基丁基酮 (MEK) 结晶。产率为1.84g (66%)。

[0559] ^1H NMR (400MHz, THF-d8) : δ 8.21-8.26 (m, 4H) , 7.98-7.8.00 (m, 1H) , 7.68-7.73 (m, 2H) , 7.31-7.49 (m, 4H) .

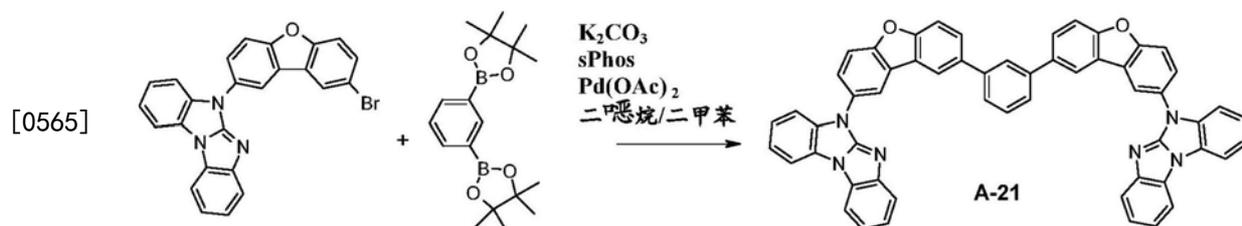
[0560] 实施例7



[0562] 实施例7的产物类似于实施例6中描述的制备。

[0563] ^1H NMR (400MHz, DMSO-d6) : δ 8.59 (d, J=1.5Hz, 2H) , 8.46-8.47 (m, 1H) , 8.24-8.33 (m, 6H) , 8.13 (d, J=8.0Hz, 1H) , 7.99-8.01 (m, 2H) , 7.78 (d, J=8.2Hz, 2H) , 7.37-7.68 (m, 9H) , 7.29-7.37 (m, 2H)

[0564] 实施例8

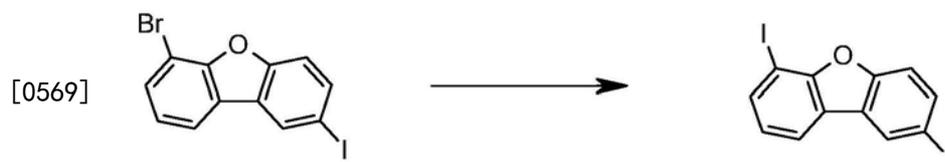


[0566] 将3.85g (8.51mmol) 5-(8-溴二苯并咪喃-2-基)苯并咪唑并[1,2-a]苯并咪唑和10.3g (4.26mmol) 磷酸三钾单水合物、20ml 二噁烷、80ml 甲苯和16ml 水加入2.01g (4.09mmol) 4,4,5,5-四甲基-2-[3-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基)苯基]-1,3,2-二氧杂戊硼烷。将反应混合物用氩气脱气。加入210mg (0.511mmol) 2-二环己基磷基-2',6'-二甲氧基联苯 (sPhos) 和191mg (0.085mmol) 乙酸钯 (II)。将反应混合物用氩气脱气

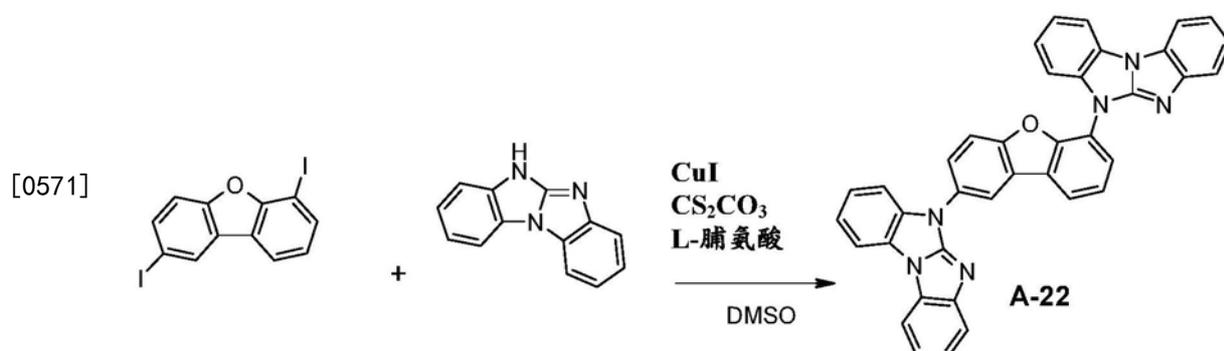
并在氩气下在100℃下搅拌22h。加入110ml氰化钠的1%溶液并将反应混合物回流1小时。加入二氯甲烷,将有机相用水洗涤并用硫酸镁干燥。产物用乙醚煎煮。产率为1.31g (39%)。

[0567] ^1H NMR (400MHz, DMF-d₇): δ 8.90 (d, J=1.7Hz, 2H), 8.86 (d, J=2.2Hz, 2H), 8.37 (s, 1H), 8.26-8.31 (m, 4H), 8.14-8.21 (m, 4H), 8.08-8.11 (m, 2H), 7.94-7.96 (m, 2H), 7.89-7.93 (m, 2H), 7.76-7.78 (m, 2H), 7.66-7.71 (m, 3H), 7.32-7.49 (m, 8H)

[0568] 实施例9

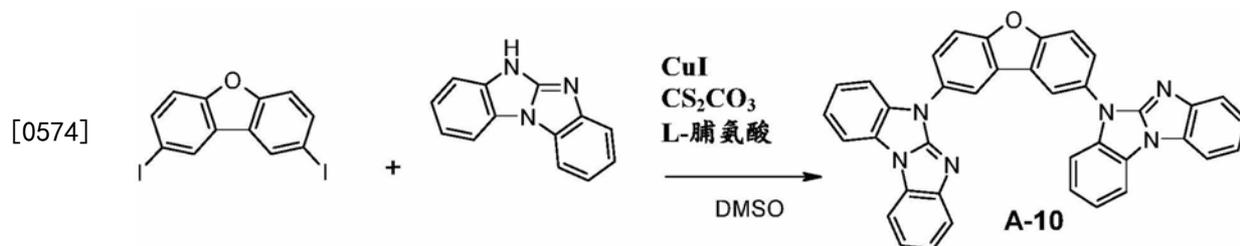


[0570] a) 2,6-二碘二苯并呋喃根据W02011/111423的实施例13制备并通过由环己烷结晶纯化。



[0572] b) 实施例9的产物类似于实施例5中描述的程序制备。

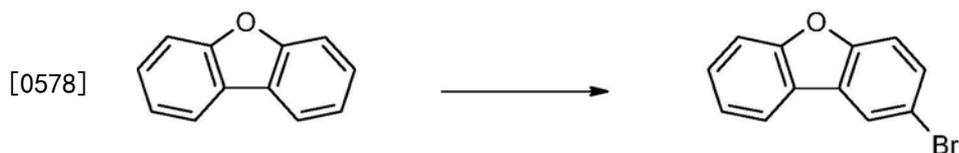
[0573] 实施例10



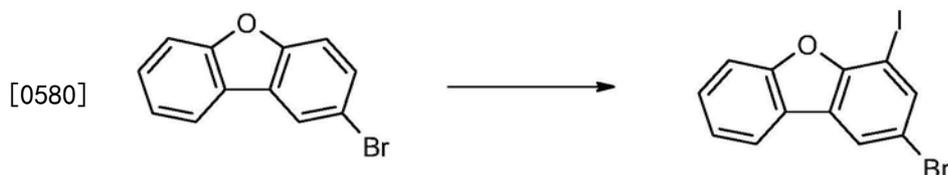
[0575] 在氮气下将4.20g (10mmol) 2,8-二碘二苯并呋喃、13.0g (40.0mmol) 碳酸铯、1.90g (1.00mmol) 碘化铜(I)和2.30g (20.0mmol) L-脯氨酸加入4.56g (22.0mmol) 在100ml二甲亚砜(DMSO)中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在100℃下搅拌24h,过滤并用二氯甲烷洗涤。将有机相用硫酸镁干燥并蒸除溶剂。将产物由醚结晶。产率为4.7g (81%)。

[0576] ^1H NMR (400MHz, THF-d₈): δ 8.73 (d, J=1.2Hz, 2H), 8.14 (d, J=2.3Hz, J=8.8Hz, 2H), 7.96-8.02 (m, 4H), 7.92 (d, J=8.8Hz, 2H), 7.70-7.73 (m, 2H), 7.62 (d, J=7.1Hz, 2H), 7.25-7.40 (m, 8H)。

[0577] 实施例11



[0579] a) 2-溴二苯并呋喃根据E.Hand, J.Org.Chem.62(1997)1348制备并通过由叔丁基甲基醚(TBME)结晶而纯化。



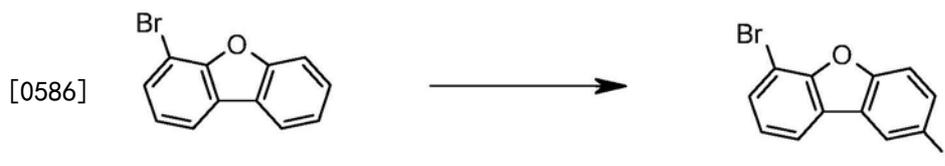
[0581] b) 在氩气下将47.27g (0.199mol) 2-溴二苯并呋喃溶于440ml干燥THF并冷却至-78℃。在1h内,将二异丙基氨基锂的溶液(LDA;由81.2ml (0.219mol) 正丁基锂(2.7M,在庚烷中)和20.18g (0.199mol) 二异丙基胺在250ml干燥THF中制备)加入,保持温度在-73℃以下。将所得黄色溶液在-78℃下搅拌2h。然后将50.6g (0.199mol) 溶于150ml干燥THF中的碘溶液在50分钟内加入,保持温度在-73℃以下。将所得褐色溶液温热至室温,倾入500ml缓冲溶液pH=7并用2N HCl中和至pH=7。将有机溶剂蒸发并将水相用乙酸乙酯萃取3次。将合并的有机相用水洗涤3次,用硫酸镁干燥,过滤并将溶剂蒸发。由环己烷/TBME=1:1结晶两次得到35.0g 2-溴-4-碘二苯并呋喃(产率:45.6%)。

[0582] $^1\text{H NMR}$ (400MHz, CDCl_3): δ 7.99 (d, $J=1.8\text{Hz}$, 1H), 7.95 (d, $J=1.8\text{Hz}$, 1H), 7.85 (d, $J=8\text{Hz}$, 1H), 7.63 (d, $J=8\text{Hz}$, 1H), 7.51 (t, $J=8\text{Hz}$, 1H), 7.37 (t, $J=8\text{Hz}$, 1H)。

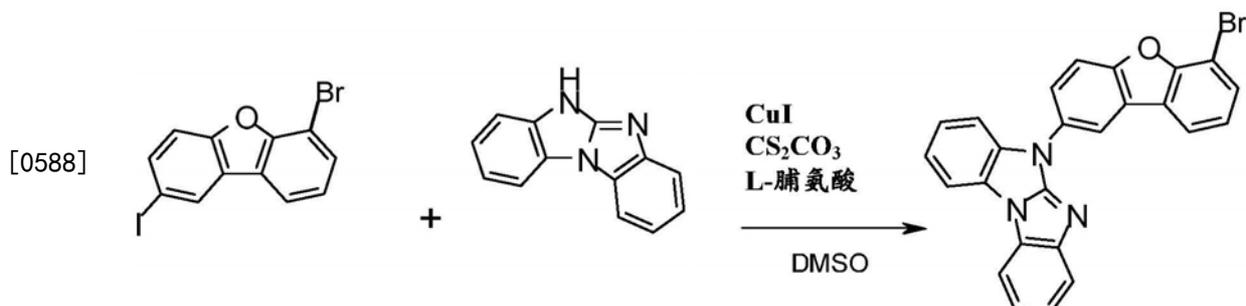
[0583] 实施例12



[0585] a) 4-溴二苯并呋喃根据US2011/0006670的实施例1制备并通过由甲醇结晶纯化。



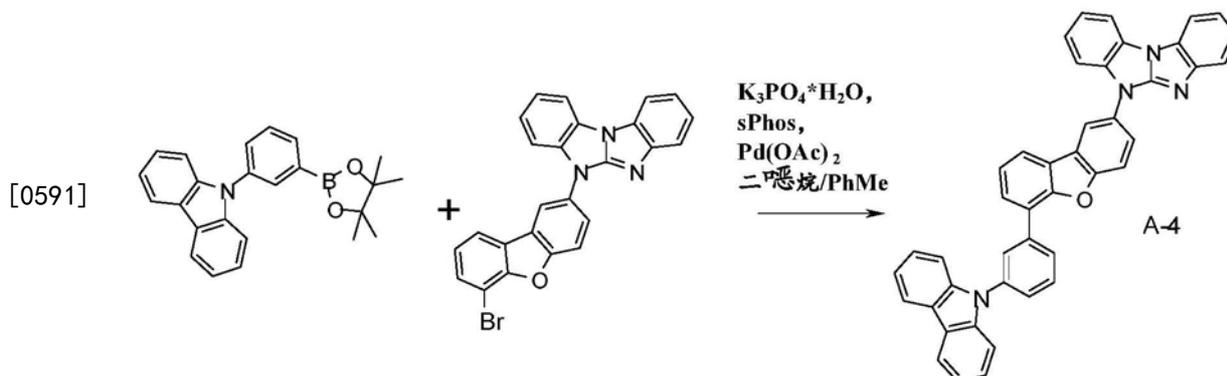
[0587] b) 6-溴-2-碘二苯并呋喃根据US2011/0006670的实施例1制备并通过由2-丙醇结晶纯化。



[0589] c) 在氮气下将1.00g (2.68mmol) 6-溴-2-碘二苯并呋喃、1.75g (5.36mmol) 碳酸铯、

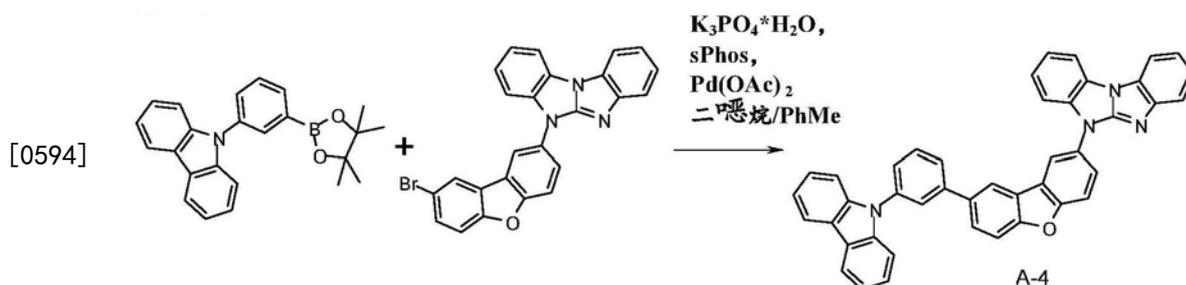
130mg (0.67mmol) 碘化铜 (I) 和150mg (1.34mmol) L-脯氨酸加入670mg (3.22mmol) 在20ml DMSO中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在100℃下搅拌18h并过滤。将THF和甲苯加入有机相并将有机相用水洗涤。将有机相用硫酸镁干燥并蒸除溶剂。产物可以不经进一步纯化用于步骤d) (产率=650mg (78%))。

[0590] ^1H NMR (400MHz, CDCl_3): δ 8.66 (d, $J=2.2\text{Hz}$, 1H), 8.13-8.19 (m, 2H), 7.96-8.07 (m, 3H), 7.66-7.78 (m, 3H), 7.25-7.45 (m, 5H) .



[0592] d) 实施例12的产物类似于实施例5中描述的制备。MS (APCI (pos), m/z): 615.5 (M^+). ^1H NMR (400MHz, THF- d_8): 8.68 (d, $J=2.1\text{Hz}$, 1H), 8.34 (t, $J=1.8\text{Hz}$, 1H), 8.21 (d, $J=7.7\text{Hz}$, 3H), 7.85-7.15 (m, 7H), 7.65-7.77 (m, 5H), 7.47-7.58 (m, 3H), 7.28-7.44 (m, 6H) .

[0593] 实施例13



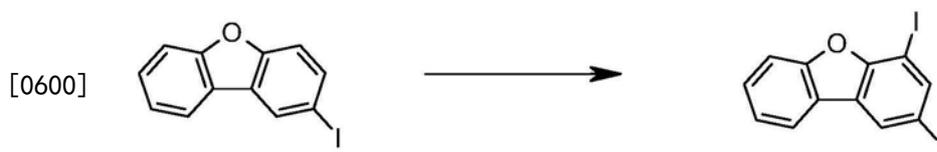
[0595] 9-[3-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基)苯基]咪唑的合成描述于 Chem.Mater.20 (2008) 1691-1693。实施例21的产物类似于实施例5中描述的制备。

[0596] 实施例14



[0598] 2-碘二苯并咪喃根据A.Krysk, Journal of Chemical Research, Miniprint 10 (1999) 2501制备并通过由甲醇结晶纯化。

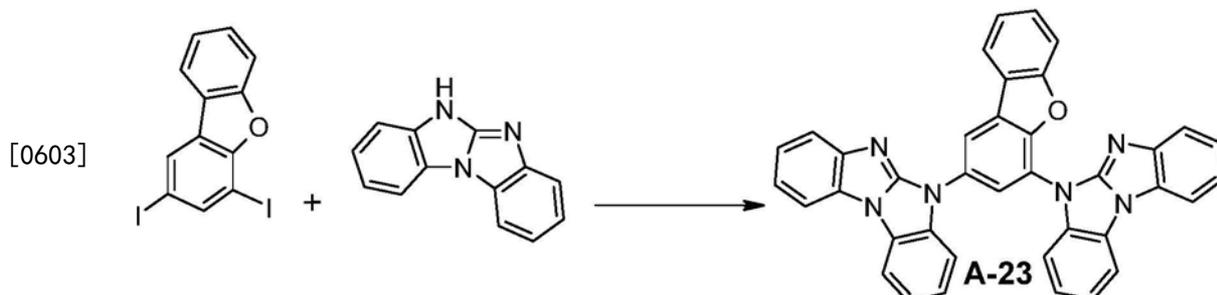
[0599] 实施例15



[0601] a) 2,4-二碘二苯并咪喃类似于实施例11中描述的制备,由2-碘二苯并咪喃

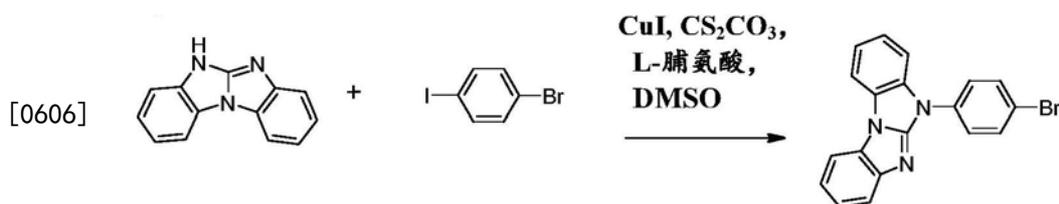
(实施例14) 开始并由2-丙醇结晶纯化(产率:80%)。

[0602] ^1H NMR (400MHz, CDCl_3): δ 8.17 (s, 1H), 8.07 (s, 1H), 7.84 (d, $J=7.6\text{Hz}$, 1H), 7.63 (d, $J=7.6\text{Hz}$, 1H), 7.51 (t, $J=7.6\text{Hz}$, 1H), 7.37 (t, $J=7.6\text{Hz}$, 1H) .



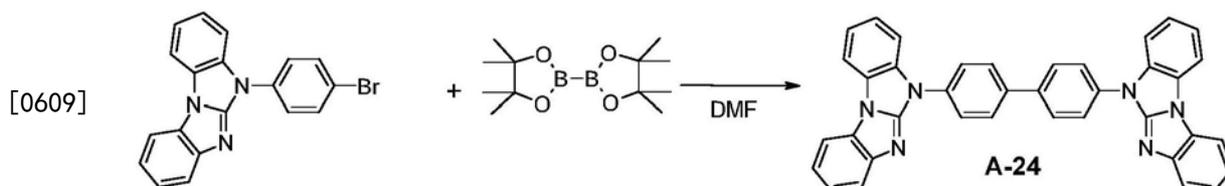
[0604] b) 实施例15的产物类似于实施例10中描述的程序制备。

[0605] 实施例16



[0607] a) 在氮气下将7.00g (24.7mmol) 1-溴-4-碘苯、10.5g (32.2mmol) 碳酸铯、2.36g (12.4mmol) 碘化铜(I) 和2.85g (24.7mmol) L-脯氨酸加入5.13g (24.7mmol) 在80ml DMSO中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在100°C下搅拌15h并在150°C下搅拌4h, 用二氯甲烷在Hyflo上过滤。将有机相用水洗涤。将有机相用硫酸镁干燥。将产物用乙醚和甲基乙基酮(MEK) 煎煮。产率:2.90g (77%)。

[0608] ^1H NMR (400MHz, DMF-d_7): δ 7.93-8.10 (m, 4H), 7.78-7.92 (m, 2H), 7.72-7.79 (m, 1H), 7.49-7.71 (m, 1H), 7.31-7.49 (m, 4H) .

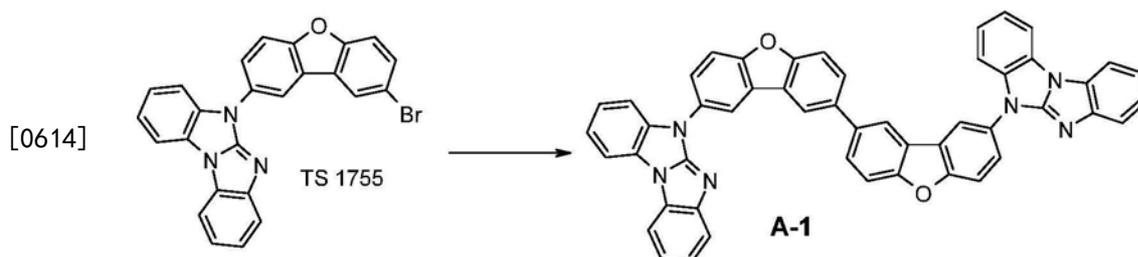


[0610] b) 实施例16b) 的产物类似于实施例4b) 中描述的程序制备。

[0611] ^1H NMR (400MHz, DMF-d_7): δ 8.19-8.33 (m, 10H), 7.83-7.87 (m, 2H), 7.73-7.77 (m, 2H), 7.35-7.54 (m, 4H) . 一个信号被DMF覆盖。

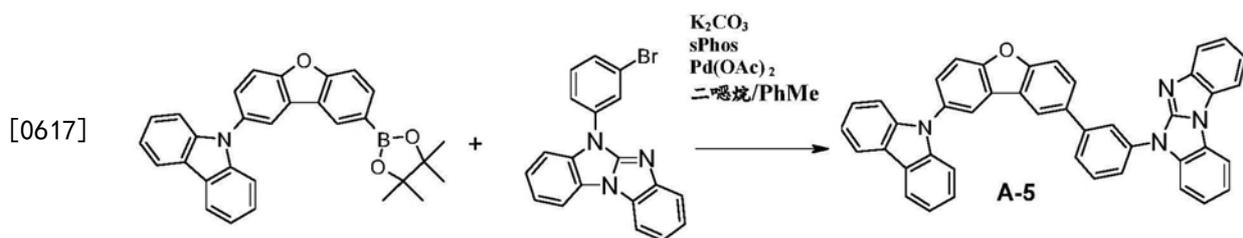
[0612] MS (APCI (pos), m/z): 565 (M^+) .

[0613] 实施例17



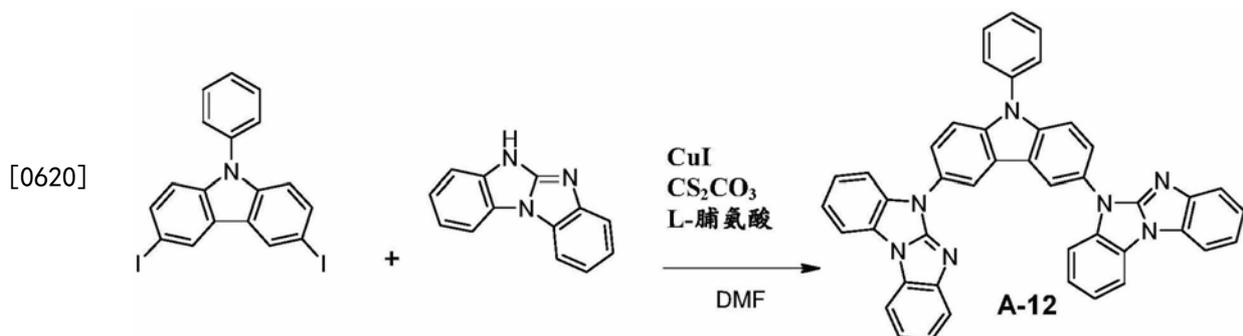
[0615] 实施例17的产物类似于实施例4b)中描述的制备。

[0616] 实施例18



[0618] 实施例18的产物类似于实施例5b)中描述的制备。MS (APCI (pos), m/z): 615 (M^{+1})。

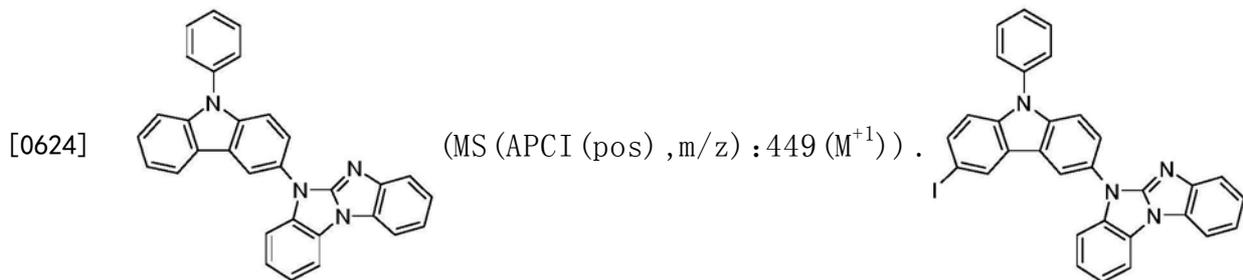
[0619] 实施例19



[0621] 在氮气下将2g (4.04mmol) 3,6-二碘-9-苯基咪唑、5.26g (16.2mmol) 碳酸铯、190mg (0.101mmol) 碘化铜(I) 和233mg (2.02mmol) L-脯氨酸加入1.84g (8.89mmol) 在40ml DMSO中的5H-苯并咪唑并[1,2-a]苯并咪唑。将反应混合物在150°C下搅拌10h,在Hyflo Super Cel (R) 介质(Fluka 56678, CAS[91053-39-3]) 上过滤并用二氯甲烷洗涤。将有机相用硫酸镁干燥并蒸除溶剂。用环己烷/甲苯(环己烷100%, 环己烷/甲苯10/1, 环己烷/甲苯4/1) 梯度柱层析得到产物(产率:70mg (3%))。MS (APCI (pos), m/z): 654 (M^{+1})。

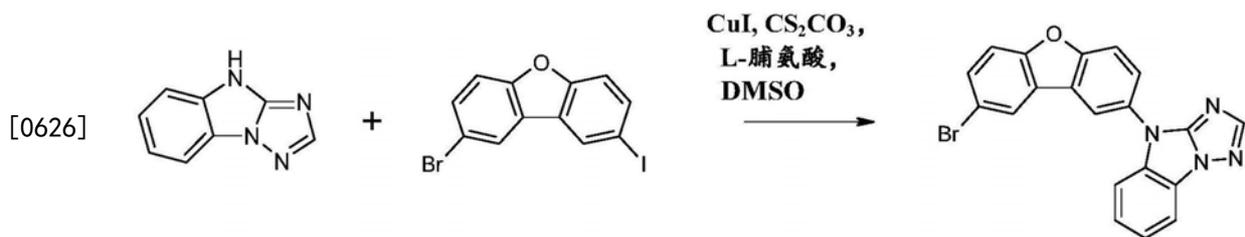
[0622] 1H NMR (400MHz, THF-d8): δ = 8.81 (d, J = 1.9Hz, 2H), 7.99-7.05 (m, 6H), 7.70-7.83 (m, 11H), 7.22-7.41 (m, 8H)。

[0623] 除了Cpd. A-12以外,通过HPLC-MS还检出如下化合物:

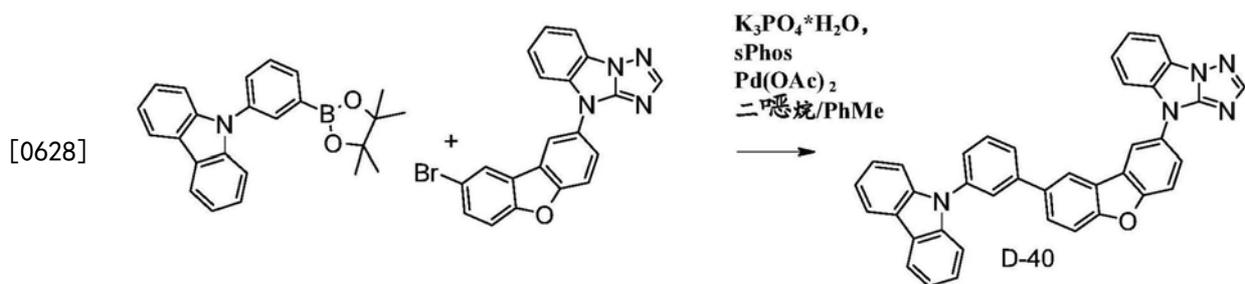


(MS (APCI (pos), m/z): 575 (M^{+1})) .

[0625] 实施例20

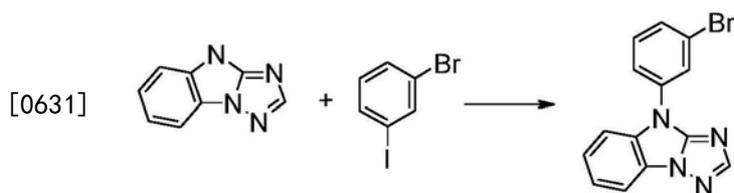


[0627] a) 实施例20a)的产物类似于实施例1c)中描述的程序制备。参考J.Heterocyclic Compounds (1989) 168和J.Org.Chem 42 (1977) 542关于4H-[1,2,4]三唑并[1,5-a]苯并咪唑的合成和用于其合成的原料。MS (MALDI-MS (m/z): 403 (M^{+1})).

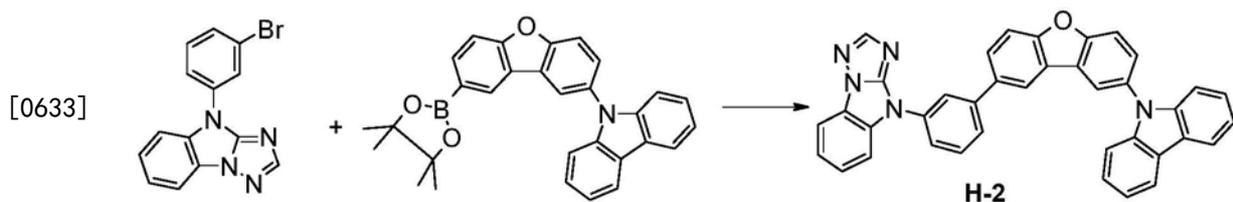


[0629] b) 实施例20b)的产物类似于实施例13)中描述的程序制备。

[0630] 实施例21



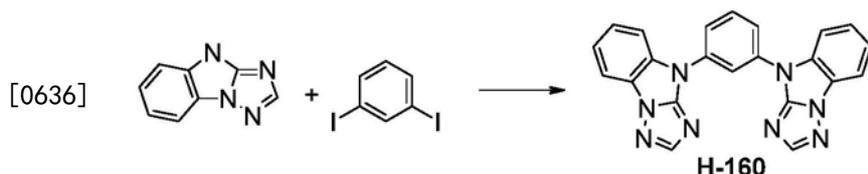
[0632] a) 在氩气下将5g (31.6mmol) 4H-[1,2,4]三唑并[1,5-a]苯并咪唑、20.6g (63.2mmol) 碳酸铯、1.5g (7.9mmol) 碘化铜(I)和910mg (7.9mmol) L-脯氨酸加入17.8g (8mL) (63.2mmol) 在60mL DMSO中的1-溴-3-碘苯。将反应混合物在85°C下搅拌15h。将反应混合物用二氯甲烷滤过硅胶。将有机相由水、NaCl溶液洗涤并使用硫酸钠干燥。将产物用乙醚煎煮(产率: 8.0g (80%))。¹H NMR (400MHz, CD₂Cl₂): δ8.02 (s, 1H), 7.98 (s, 1H), 7.90-7.88 (m, 1H), 7.71-7.68 (m, 1H), 7.60-7.58 (d, 1H), 7.52-7.48 (t, 1H), 7.46-7.40 (m, 2H)。¹³C NMR (500MHz, CD₂Cl₂): δ155.05 (d, 1C), 153.47 (s, 1C), 136.82 (s, 1C), 134.23 (s, 1C), 131.63 (d, 1C), 130.78 (d, 1C), 126.69 (d, 1C), 124.95 (s, 1C), 124.87 (d, 1C), 123.50 (s, 1C), 123.26 (d, 1C), 122.45 (d, 1C), 112.15 (d, 1C), 111.66 (d, 1C)。



[0634] b) 在氩气下将1.25g (4mmol) 4-(3-溴苯基)-[1,2,4]三唑并[1,5-a]苯并咪唑、2.4g (5.2mmol) 9-[8-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基)二苯并咪唑-2-基]咪唑和4.3g (20mmol) 在40mL甲苯中的磷酸钾加入在二噁烷/水40mL/10mL中的90mg (0.03mmol) 乙酸钡(II)、100mg (0.24mmol) 2-二环己基膦基-2',6'-二甲氧基联苯(sPhos)

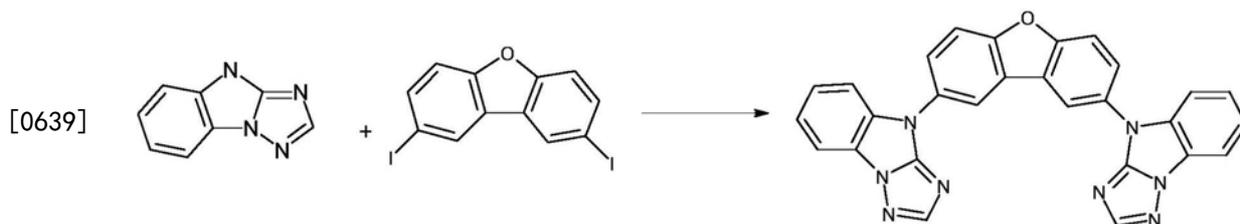
中。将反应混合物在85℃下搅拌15h并用二氯甲烷滤过C盐。将有机相由水、NaCl溶液洗涤并使用硫酸钠干燥(产率:1.75g (77%))。¹H NMR (400MHz, CD₂Cl₂): δ8.41 (d, 1H), 8.37 (d, 1H), 8.34 (d, 2H), 8.25 (t, 1H), 8.17 (s, 1H), 8.06-8.04 (m, 3H), 7.93-7.86 (m, 4H), 7.84 (d, 1H), 7.82 (d, 1H), 7.59-7.54 (m, 6H), 7.47-7.43 (m, 2H)。¹³C NMR (500MHz, CD₂Cl₂): δ157.22 (s, 1C), 155.97 (s, 1C), 155.07 (d, 1C), 153.80 (s, 1C), 143.30 (s, 1C), 141.80 (s, 2C), 136.12 (s, 1C), 135.79 (s, 1C), 134.68 (s, 1C), 133.14 (s, 1C), 130.81 (d, 1C), 127.78 (d, 1C), 127.31 (d, 1C), 126.79 (d, 1C), 126.36 (d, 2C), 125.87 (s, 1C), 124.91 (s, 1C), 124.89 (s, 1C), 124.76 (d, 1C), 123.51 (d, 2C), 122.92 (d, 1C), 122.88 (d, 1C), 120.61 (d, 2C), 120.31 (d, 1C), 120.26 (d, 2C), 122.65 (d, 1C), 120.03 (d, 1C), 113.40 (d, 1C), 112.66 (d, 1C), 112.21 (d, 1C), 111.58 (d, 1C), 110.02 (d, 2C)。

[0635] 实施例22



[0637] 将160mg (0.48mmol) 1,3-二碘苯和170mg (1.06mmol) 4H-[1,2,4]三唑并[1,5-a]苯并咪唑在10mL DMSO中在氩气下搅拌15分钟。加入625mg (1.9mmol) 碳酸铯、120mg (1.06mmol) L-脯氨酸和90mg (0.48mmol) 碘化铜(I)。将反应混合物在100℃下搅拌15h并用二氯甲烷滤过C盐。将有机相由水、NaCl溶液洗涤并使用硫酸钠干燥。将产物由异丙醇结晶(产率:130mg (69%))。¹H NMR (400MHz, CD₂Cl₂): δ8.35 (s, 1H), 8.06 (s, 2H), 7.94-7.84 (m, 7H), 7.50-7.42 (m, 4H)。¹³C NMR (500MHz, CD₂Cl₂): δ155.12 (d, 2C), 153.59 (s, 2C), 137.10 (s, 2C), 134.27 (s, 2C), 131.07 (d, 1C), 125.09 (s, 2C), 125.06 (d, 2C), 123.39 (d, 2C), 121.80 (d, 2C), 118.36 (d, 1C), 112.51 (d, 2C), 111.73 (d, 2C)。

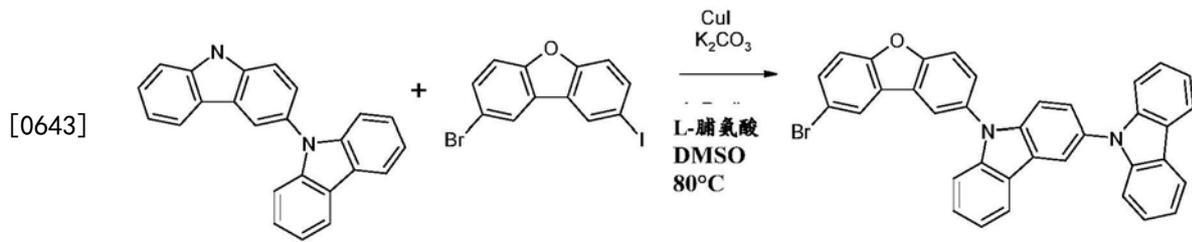
[0638] 实施例23



[0640] 将1g (2.4mmol) 2,8-二碘二苯并呋喃和840mg (5.3mmol) 4H-[1,2,4]三唑并[1,5-a]苯并咪唑在10mL DMSO中在氩气下搅拌15分钟。加入3.1g (9.6mmol) 碳酸铯、550mg (4.8mmol) L-脯氨酸和460mg (2.4mmol) 碘化铜(I)。将褐色反应混合物在100℃下搅拌15h。将水加入反应混合物中并过滤且用甲醇洗涤。将产物由甲苯结晶(产率:330mg (28%))。¹H NMR (400MHz, CD₂Cl₂): δ8.38 (s, 1H), 8.05 (s, 2H), 7.92-7.85 (m, 8H), 7.42-7.13 (dd, 2H)

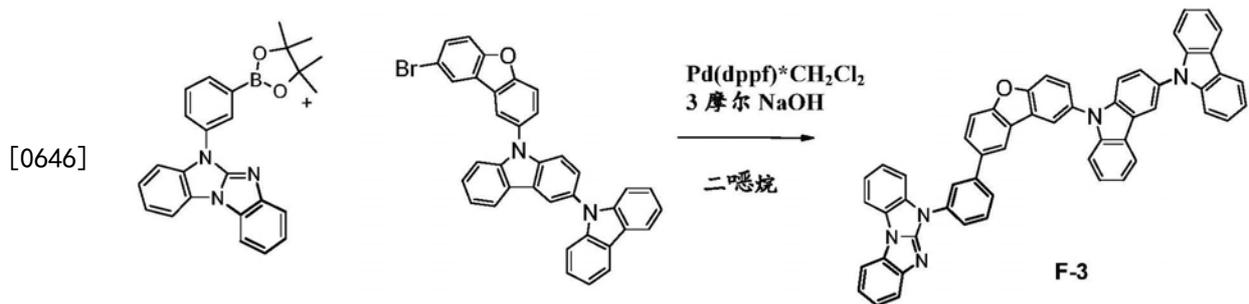
[0641] 实施例24

[0642] 9-(9H-咪唑-3-基)-9H-咪唑根据文献程序制备(J.Org.Chem, 2008, 73, 1809)。



[0644] a) 实施例24a) 的产物根据实施例10的程序制备。纯化:FC (SiO_2 , 环己烷/ CH_2Cl_2 4:1. 产率:85%。

[0645] ^1H NMR (400MHz, CD_2Cl_2): δ 8.34 (s, 1H), 8.18 (m, 5H), 7.87 (d, 1H), 7.77 (d, 1H), 7.67 (d, 1H), 7.59 (m, 3H), 7.51-7.39 (m, 6H), 7.31 (m, 3H) .



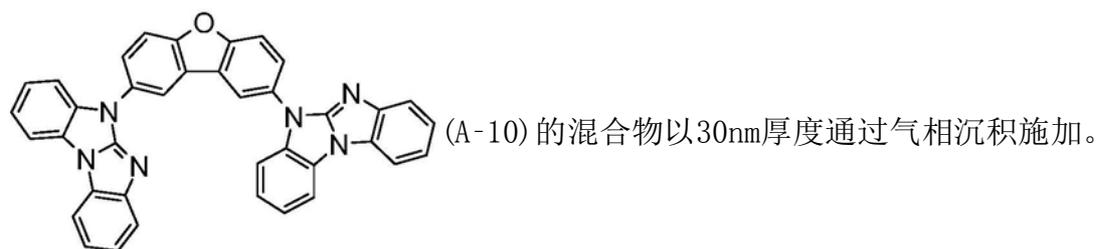
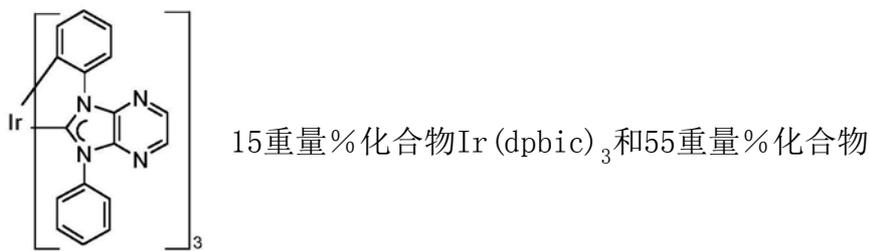
[0647] b) 将Pd (dppf) * CH_2Cl_2 (8mg, 0.01mmol) 加入5-[3-(4,4,5,5-四甲基-1,3,2-二氧杂戊硼烷-2-基) 苯基] 苯并咪唑并[1,2-a] 苯并咪唑 (133mg, 0.33mmol)、实施例24a) 的产物 (144mg, 0.25mmol) 在二噁烷 (5mL) 和NaOH (3M, 0.25mL) 中的经脱气 (Ar) 的混合物。将反应混合物在 80°C 下加热8h并滤过C盐和FC (SiO_2 , CH_2Cl_2) 得到产物 (产率:50mg, 26%)。

[0648] ^1H NMR (400MHz, CD_2Cl_2): δ 8.33 (2xs, 2+1H), 8.18 (m, 4H), 7.88 (m, 5H), 7.81-7.73 (m, 4H), 7.69-7.62 (m, 3H), 7.56 (d, 1H), 7.49 (d, 2H), 7.42-7.27 (m, 11H) .

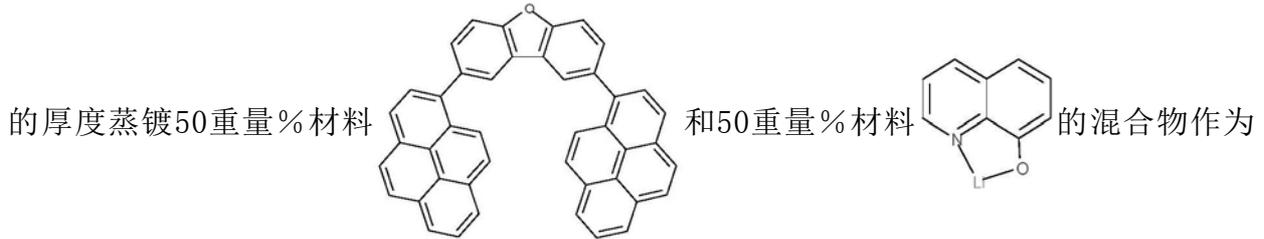
[0649] 应用实施例3

[0650] ITO基底的制备以及随后完整二极管的表征与应用实施例1相同,不同的是如下层的组成不同:

[0651] 作为空穴传输和激子阻断剂,将Ir (dpbic) $_3$ 以20nm的厚度施加于该基底上,其中最初10nm用MoO $_3$ (~10%) 掺杂以改善导电率。随后,将30重量%发光剂化合物



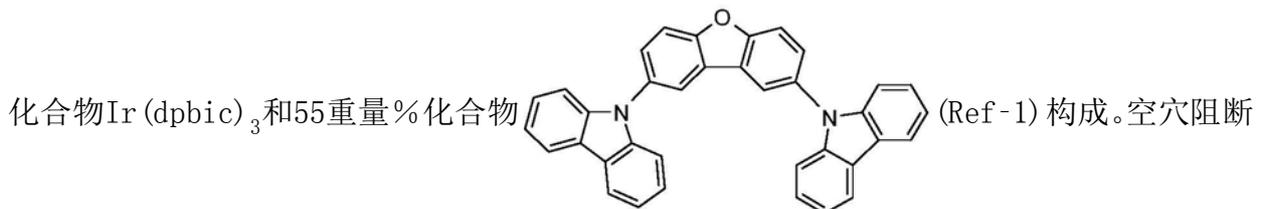
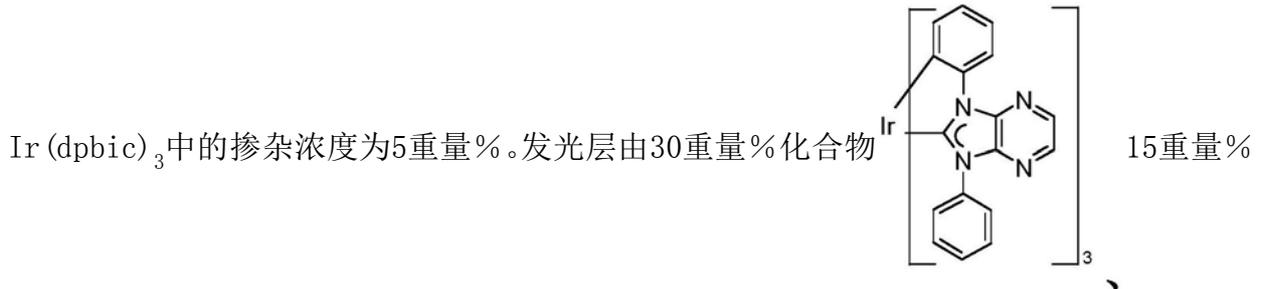
[0652] 然后通过气相沉积以5nm的厚度施加作为空穴阻断剂的化合物A-10。随后以20nm



电子传输层,最后沉积~2nm KF作为电子注入层并沉积100nm厚的Al电极完成器件。

[0653] 对比应用实施例1

[0654] OLED的生产和构造按照应用实施例3中进行,不同如下:在空穴传输层中,MoO₃在



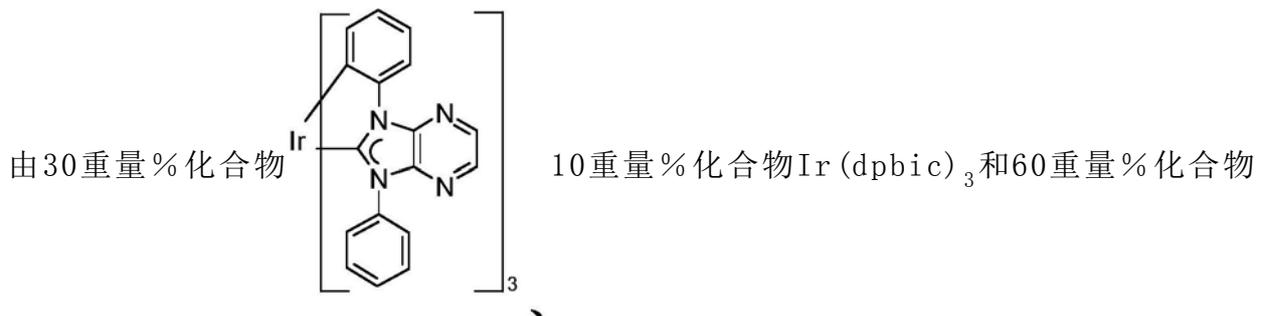
层由化合物Ref-1构成且电子传输层25nm厚。

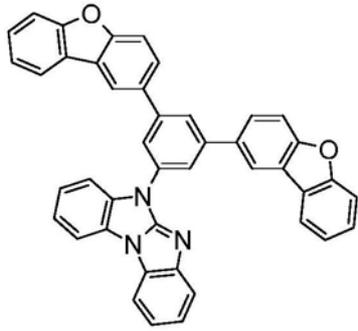
	主体材料	电压 @300nits[V]	EQE @300nits[%]	使用寿命 @4000nits*
[0655] 应用实施例 3	A-10	4.02	14.0%	100
对比应用实施例 1	Ref-1	4.00	10.8%	20

[0656] *将应用实施例3测量的使用寿命设定为100且对比应用实施例1的使用寿命的规定与应用实施例3的那些有关。

[0657] 应用实施例4

[0658] OLED的生产和构造按照应用实施例3中进行,不同如下:具有MoO₃和Ir (dpbic)₃的空穴传输层15nm厚且具有未掺杂的电子阻断剂Ir (dpbic)₃的空穴传输层仅5nm厚。发光层

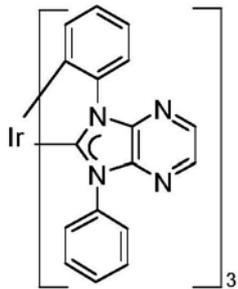




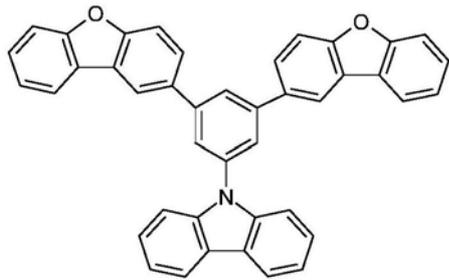
(B-5) 构成,厚度为40nm。空穴阻断层由材料B-5构成。

[0659] 对比应用实施例2

[0660] OLED的生产和构造按照应用实施例3中进行,不同如下:发光层由30重量%化合物



10重量%化合物Ir (dpbic)₃和60重量%化合物



(Ref-2) 构成,厚度为40nm。空穴阻断层由化合物Ref-2构

成。

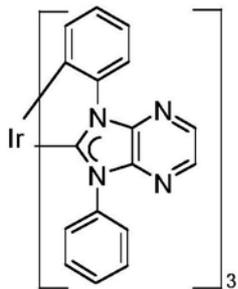
[0661]

	主体材料	电压@300nits [V]	使用寿命@4000nits*
应用实施例4	B-5	4.55	100
对比应用实施例2	Ref-2	4.34	70

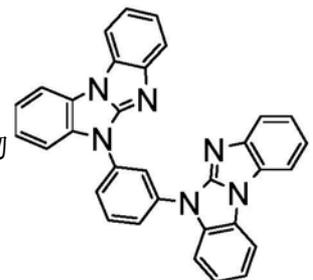
[0662] *将应用实施例4测量的使用寿命设定为100且对比应用实施例2的使用寿命的规定与应用实施例4的那些有关。

[0663] 应用实施例5

[0664] OLED的生产和构造按照应用实施例1中进行,不同如下:发光层由30重量%化合物



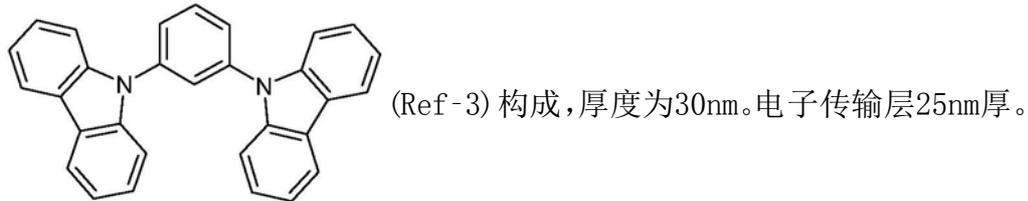
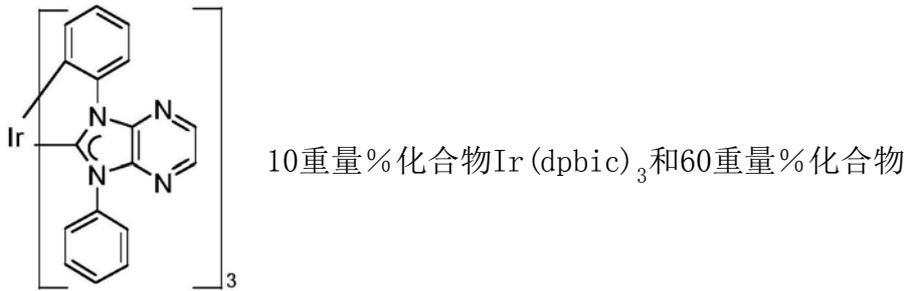
10重量%化合物Ir (dpbic)₃和60重量%化合物



(A-20) 构成,厚度为30nm。电子传输层25nm厚。

[0665] 对比应用实施例3

[0666] OLED的生产和构造按照应用实施例1中进行,不同如下:发光层由30重量%化合物



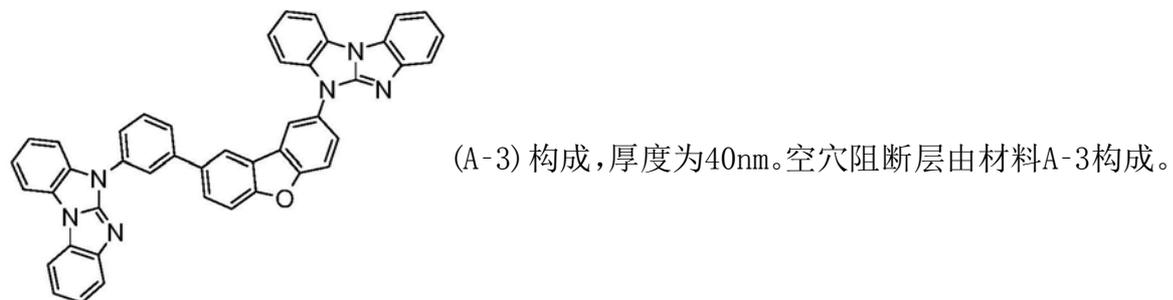
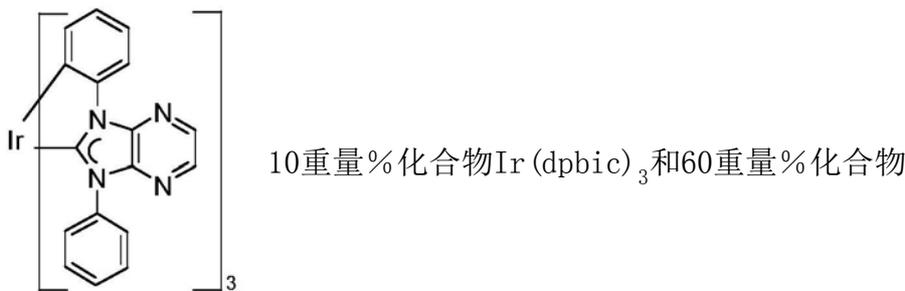
[0667]

	主体材料	电压@300nits [V]	EQE@300nits [%]
应用实施例5	A-20	3.43	11.3
对比应用实施例3	Ref-3	3.81	13.6

[0668] *将应用实施例5测量的使用寿命设定为100且对比应用实施例3的使用寿命的规定与应用实施例5的那些有关。

[0669] 应用实施例6

[0670] OLED的生产和构造按照应用实施例3中进行,不同如下:发光层由30重量%化合物



[0671]

	主体材料	电压@300nits [V]	EQE@300nits [%]	CIE
应用实施例6	A-20	4.56	12.8	0.18/0.36