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(54) Title: NOVEL ORGANIC ELECTROLUMINESCENT COMPOUNDS AND ORGANIC ELECTROLUMINESCENT DEVICE CONTAINING THE SAME

(57) Abstract: The present invention relates to a novel organic electroluminescent compound and an organic electroluminescent device containing the same. Using the organic electroluminescent compound according to the present invention, an organic electroluminescent device can have high luminescent efficiency, a long lifespan, and high current efficiency.



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Description

Title of Invention: NOVEL ORGANIC ELECTROLUMINESCENT COMPOUNDS AND ORGANIC ELECTROLUMINESCENT DEVICE CONTAINING THE SAME

Technical Field

- [1] The present invention relates to novel organic electroluminescent compounds and an organic electroluminescent device containing the same.

Background Art

- [2] An electroluminescent (EL) device is a self-light-emitting device which has advantages in that it provides a wider viewing angle, a greater contrast ratio, and a faster response time. An organic EL device was first developed by Eastman Kodak, by using small aromatic diamine molecules, and aluminum complexes as materials to form a light-emitting layer [Appl. Phys. Lett. 51, 913, 1987].
- [3] The most important factor determining luminous efficiency in the organic EL device is light-emitting materials. Until now, fluorescent materials have been widely used as the light-emitting material. However, in view of electroluminescent mechanisms, since phosphorescent materials theoretically enhance luminous efficiency by four (4) times compared to fluorescent materials, development of phosphorescent light-emitting materials are widely being researched. Iridium(III) complexes have been widely known as phosphorescent materials, including bis(2-(2'-benzothienyl)-pyridinato-N,C3')iridium(acetylacetonate) ((acac)Ir(btp)₂), tris(2-phenylpyridine)iridium (Ir(ppy)₃) and bis(4,6-difluorophenylpyridinato-N,C2)picolate iridium (Firpic) as red-, green- and blue-emitting materials, respectively.
- [4] The light-emitting material can be used as a combination of a host material and a dopant to improve color purity, luminous efficiency, and stability. The host materials greatly influence the efficiency and performance of the EL device when using a host material/dopant system as the light emitting material, and thus their selection is important. At present, 4,4'-N,N'-dicarbazol-biphenyl (CBP) is the most widely known host material for phosphorescent materials. Recently, Pioneer (Japan) et al., developed a high performance organic EL device using bathocuproine (BCP) and aluminum(III)bis(2-methyl-8-quinolate)(4-phenylphenolate) (BALq) etc., as host materials, which were known as hole blocking materials.
- [5] Although these phosphorescent host materials provide good light-emitting characteristics, they have the following disadvantages: (1) Due to their low glass transition temperature and poor thermal stability, their degradation may occur during a high-

temperature deposition process in a vacuum. (2) The power efficiency of the organic EL device is given by $[(\pi/\text{voltage}) \times \text{current efficiency}]$, and the power efficiency is inversely proportional to the voltage. Although the organic EL device comprising phosphorescent host materials provides higher current efficiency (cd/A) than one comprising fluorescent materials, a significantly high driving voltage is necessary. Thus, there is no merit in terms of power efficiency (lm/W). (3) Furthermore, the operational lifespan of the organic EL device is short, and luminous efficiency is still required in order to be improved.

- [6] Meanwhile, copper phthalocyanine (CuPc), 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB), N,N'-diphenyl-N,N'-bis(3-methylphenyl)-(1,1'-biphenyl)-4,4'-diamine (TPD), 4,4',4''-tris(3-methylphenylphenylamino)triphenylamine (MTDATA), etc., were used as a hole injection and transport material. However, the organic EL device using these materials is problematic in quantum efficiency and operational lifespan. It is because, when the organic EL device is driven under high current, thermal stress occurs between an anode and a hole injection layer. Thermal stress significantly reduces the operational lifespan of the device. Furthermore, since the organic material used in the hole injection layer has very high hole mobility, the hole-electron charge balance may be broken and quantum yield (cd/A) may decrease.
- [7] Korean Patent Application Laying-Open No. 20110129766 discloses compounds for an organic EL device, in which aryl or heteroaryl moiety is bonded to a carbon atom of a polycyclic compound having carbazole moiety fused with heteroaryl moiety such as benzofuran and benzothiophene. However, the above reference fails to specifically disclose compounds in which nitrogen-containing heteroaryl moiety, nitrogen-connected heteroaryl moiety (containing or not-containing nitrogen) or nitrogen-connected aryl moiety is bonded to a nitrogen atom of a carbazole moiety of a polycyclic backbone.

Disclosure of Invention

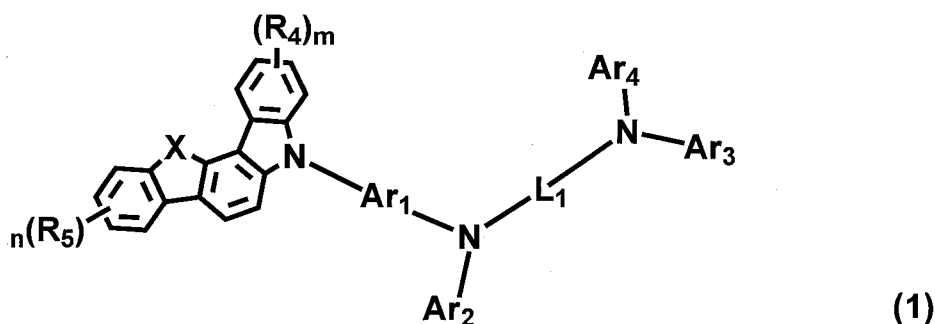
Technical Problem

- [8] The objective of the present invention is to provide an organic electroluminescent compound, which has higher luminous efficiency and a longer lifespan than the conventional materials, and an organic electroluminescent device, which has high efficiency and a long lifespan, using said compounds.

Solution to Problem

- [9] The present inventors found that the above objective can be achieved by a compound represented by the following formula 1:

[10]



- [11] wherein
- [12] X represents -O-, -S-, -CR₁R₂- or -NR₃-;
- [13] L₁ represents a substituted or unsubstituted 5- to 30-membered heteroarylene, or a substituted or unsubstituted (C6-C30)arylene;
- [14] Ar₁ to Ar₄ each independently represent a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (C6-C30)arylene, a substituted or unsubstituted 5- to 30-membered heteroaryl, or a substituted or unsubstituted 5- to 30-membered heteroarylene;
- [15] Ar₁ and Ar₂, Ar₃ and L₁, or Ar₃ and Ar₄ may be linked together with a nitrogen atom bonded thereto to form a nitrogen-containing 5- or 30-membered heteroaryl or a nitrogen-containing 5- or 30-membered heteroarylene;
- [16] R₁ to R₃ each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted 5- to 30-membered heteroaryl, or are linked to an adjacent substituent(s) to form a mono- or polycyclic, 3- to 30-membered alicyclic or aromatic ring;
- [17] R₄ and R₅ each independently represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C2-C30)alkenyl, a substituted or unsubstituted (C2-C30)alkynyl, a substituted or unsubstituted (C1-C30)alkoxy, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted 3- to 7-membered heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted 5- to 30-membered heteroaryl, -NR₁₁R₁₂, -SiR₁₃R₁₄R₁₅, -SR₁₆, -OR₁₇, -COR₁₈, or -B(OR₁₉)(OR₂₀);
- [18] R₁₁ to R₂₀ each independently represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C2-C30)alkenyl, a substituted or unsubstituted (C2-C30)alkynyl, a substituted or unsubstituted (C1-C30)alkoxy, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (C3-C30)cycloalkenyl, a substituted or unsubstituted 3- to 7-membered heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted 5- to 30-membered heteroaryl,

or are linked to an adjacent substituent(s) to form a mono- or polycyclic, 3- to 30-membered alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen, and sulfur;

[19] m and n each independently represent an integer of 1 to 4; where m or n is an integer of 2 or more, each of R₄ or each of R₅ may be the same or different; and

[20] the heteroaryl(ene) contains at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

Advantageous Effects of Invention

[21] The compounds according to the present invention show high luminous efficiency and a long lifespan. Using the compound according to the present invention, an electroluminescent device exhibits excellent luminous characteristics and current efficiency.

Mode for the Invention

[22] Hereinafter, the present invention will be described in detail. However, the following description is intended to explain the invention, and is not meant in any way to restrict the scope of the invention.

[23] The present invention provides the organic electroluminescent compound of formula 1 above, an organic electroluminescent material comprising the same, and an organic electroluminescent device comprising the material.

[24] Specifically, the compound of formula 1 of the present invention is as follows.

[25] In formula 1 above, preferably, X represents -O-, -S-, -CR₁R₂- or -NR₃-; L₁ represents a substituted or unsubstituted (C6-C30)arylene; Ar₁ to Ar₄ each independently represent a substituted or unsubstituted (C6-C15)aryl, a substituted or unsubstituted (C6-C15)arylene, a substituted or unsubstituted 5- to 15-membered heteroaryl, or a substituted or unsubstituted 5- to 15-membered heteroarylene; Ar₁ and Ar₂, Ar₃ and L₁, or Ar₃ and Ar₄ may be linked together with a nitrogen atom bonded thereto to form a nitrogen-containing 5- or 15-membered heteroaryl or a nitrogen-containing 5- or 15-membered heteroarylene; R₁ to R₃ each independently represent a substituted or unsubstituted (C1-C15)alkyl, a substituted or unsubstituted (C6-C15)aryl, or a substituted or unsubstituted 5- to 15-membered heteroaryl; R₄ and R₅ each independently represent hydrogen, a halogen, a substituted or unsubstituted (C1-C10)alkyl, a substituted or unsubstituted (C6-C15)aryl, a substituted or unsubstituted 5- to 15-membered heteroaryl, or -SiR₁₃R₁₄R₁₅; R₁₃ to R₁₅ each independently represent a substituted or unsubstituted (C1-C10)alkyl.

[26] In formula 1 above, more preferably, X represents -O-, -S-, -CR₁R₂- or -NR₃-; L₁ represents an unsubstituted (C6-C15)arylene; Ar₁ to Ar₄ each independently represent a substituted or unsubstituted (C6-C15)aryl, a substituted or unsubstituted

(C6-C15)arylene, a substituted or unsubstituted 5- to 15-membered heteroaryl, or a substituted or unsubstituted 5- to 15-membered heteroarylene; Ar₁ and Ar₂, Ar₃ and L₁, or Ar₃ and Ar₄ may be linked together with a nitrogen atom bonded thereto to form a nitrogen-containing 5- or 15-membered heteroaryl or a nitrogen-containing 5- or 15-membered heteroarylene; R₁ and R₂ each independently represent a substituted or unsubstituted (C1-C10)alkyl, or a substituted or unsubstituted (C6-C15)aryl; R₃ represents a substituted or unsubstituted (C6-C15)aryl; R₄ and R₅ each independently represent hydrogen, an unsubstituted (C1-C10)alkyl, or -SiR₁₃R₁₄R₁₅; R₁₃ to R₁₅ each independently represent a substituted or unsubstituted (C1-C10)alkyl.

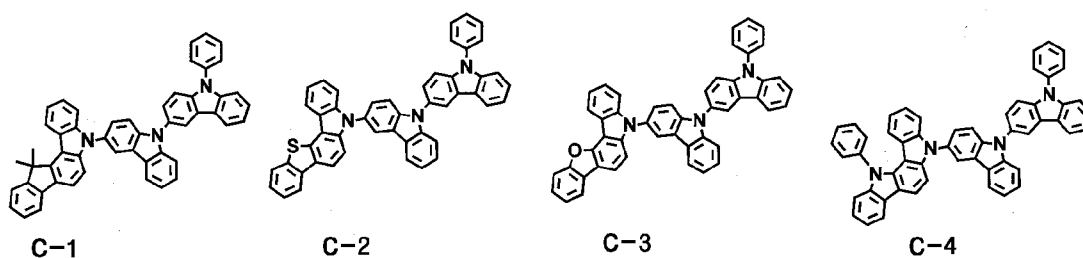
- [27] Herein, “(C1-C30)alkyl” indicates a linear or branched alkyl having 1 to 30, preferably 1 to 10 carbon atoms and includes methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, etc. “(C2-C30) alkenyl” indicates a linear or branched alkenyl having 2 to 30, preferably 2 to 20, more preferably 2 to 10 carbon atoms and includes vinyl, 1-propenyl, 2-propenyl, 1-butenyl, 2-butenyl, 3-butenyl, 2-methylbut-2-enyl, etc. “(C2-C30)alkynyl” indicates a linear or branched alkynyl having 2 to 30, preferably 2 to 20, more preferably 2 to 10 carbon atoms and includes ethynyl, 1-propynyl, 2-propynyl, 1-butyne, 2-butyne, 3-butyne, 1-methylpent-2-ynyl, etc. “(C3-C30)cycloalkyl” indicates a mono- or polycyclic hydrocarbon having 3 to 30, preferably 3 to 20, more preferably 3 to 7 carbon atoms and includes cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, etc. “3- to 7-membered heterocycloalkyl” indicates a cycloalkyl having 3 to 7 ring backbone atoms including at least one heteroatom selected from B, N, O, S, P(=O), Si and P, preferably O, S and N, and includes tetrahydrofuran, pyrrolidine, thiolan, tetrahydropyran, etc. Furthermore, “(C6-C30)aryl(ene)” indicates a monocyclic or fused ring derived from an aromatic hydrocarbon and having 6 to 30, preferably 6 to 15 ring backbone carbon atoms, and includes phenyl, biphenyl, terphenyl, naphthyl, fluorenyl, phenanthrenyl, anthracenyl, indenyl, triphenylenyl, pyrenyl, tetracenyl, perylenyl, chrysenyl, naphthacenyl, fluoranthenyl, etc. “5- to 30-membered heteroaryl(ene)” indicates an aryl group having 5 to 30, preferably 5 to 15 ring backbone atoms including at least one, preferably 1 to 4 heteroatom selected from the group consisting of B, N, O, S, P(=O), Si and P; may be a monocyclic ring, or a fused ring condensed with at least one benzene ring; may be partially saturated; may be one formed by linking at least one heteroaryl or aryl group to a heteroaryl group via a single bond(s); and includes a monocyclic ring-type heteroaryl such as furyl, thiophenyl, pyrrolyl, imidazolyl, pyrazolyl, thiazolyl, thiadiazolyl, isothiazolyl, isoxazolyl, oxazolyl, oxadiazolyl, triazinyl, tetrazinyl, triazolyl, tetrazolyl, furazanyl, pyridyl, pyrazinyl, pyrimidinyl, pyridazinyl, etc., and a fused ring-type heteroaryl such as benzofuranyl, benzothiophenyl, isobenzofuranyl, dibenzofuranyl, dibenzothiophenyl, benzoimidazolyl, benzothiazolyl, benzoisothiazolyl,

benzoxazolyl, benzothiazolyl, isindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolyl, quinazolinyl, quinoxalyl, carbazolyl, phenoxazinyl, phenanthridinyl, benzodioxolyl, etc. Furthermore, "halogen" includes F, Cl, Br and I.

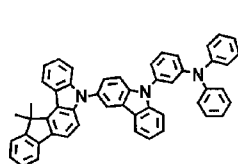
[28] Herein, "substituted" in the expression "substituted or unsubstituted" means that a hydrogen atom in a certain functional group is replaced with another atom or group, i.e., a substituent. The substituents of the substituted alkyl, the substituted alkenyl, the substituted alkynyl, the substituted alkoxy, the substituted cycloalkyl, the substituted cycloalkenyl, the substituted heterocycloalkyl, the substituted aryl(ene), and the substituted heteroaryl(ene) in formula 1 above, each independently, are preferably at least one selected from the group consisting of deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a (C1-C30)alkyl, a halo(C1-C30)alkyl, a (C2-C30)alkenyl, a (C2-C30)alkynyl, a (C1-C30)alkoxy, a (C1-C30)alkylthio, a (C3-C30)cycloalkyl, a (C3-C30)cycloalkenyl, a 3- to 7-membered heterocycloalkyl, a (C6-C30)aryloxy, a (C6-C30)arylthio, a 5- to 30-membered heteroaryl unsubstituted or substituted with a (C6-C30)aryl, a (C6-C30)aryl unsubstituted or substituted with a 5- to 30-membered heteroaryl, a tri(C1-C30)alkylsilyl, a tri(C6-C30)arylsilyl, a di(C1-C30)alkyl(C6-C30)arylsilyl, a (C1-C30)alkyldi(C6-C30)arylsilyl, an amino, a mono or di(C1-C30)alkylamino, a mono or di(C6-C30)arylamino, a (C1-C30)alkyl(C6-C30)arylamino, a (C6-C30)aryl(5- to 30-membered)heteroarylamino, a (C1-C30)alkylcarbonyl, a (C1-C30)alkoxycarbonyl, a (C6-C30)arylcarbonyl, a di(C6-C30)arylboronyl, a di(C1-C30)alkylboronyl, a (C1-C30)alkyl(C6-C30)arylboronyl, a (C6-C30)aryl(C1-C30)alkyl, and a (C1-C30)alkyl(C6-C30)aryl; more preferably, at least one selected from the group consisting of a (C1-C6)alkyl, an unsubstituted (C6-C30)aryl, an unsubstituted 5- to 30-membered heteroaryl, a tri(C1-C30)alkylsilyl, a (C6-C30)aryl(C1-C30)alkyl and a (C1-C30)alkyl(C6-C30)aryl; and even more preferably, at least one selected from the group consisting of a (C1-C30)alkyl, an unsubstituted (C6-C30)aryl, a (C6-C30)aryl(C1-C10)alkyl, and a (C1-C10)alkyl(C6-C30)aryl.

[29] Organic electroluminescent compounds of the present invention include the following, but are not limited thereto:

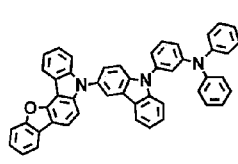
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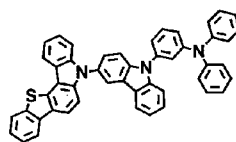
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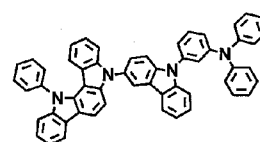
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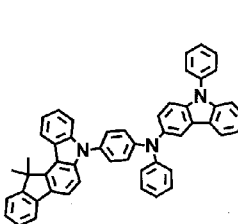
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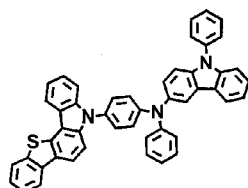
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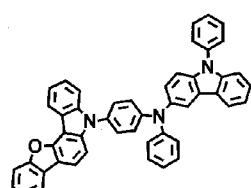
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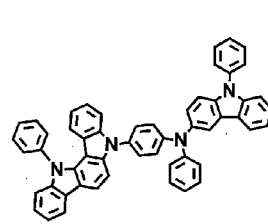
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C-10

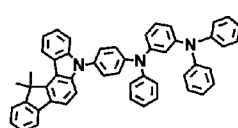


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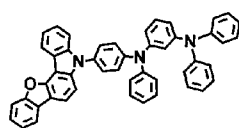


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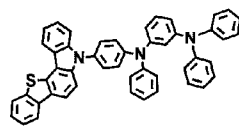
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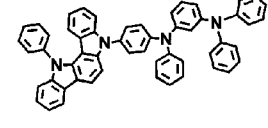
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C-14

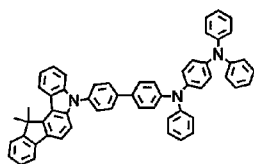


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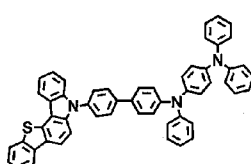


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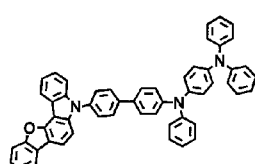
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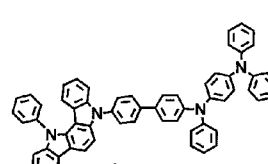
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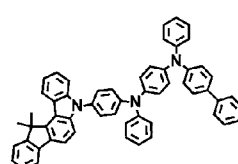


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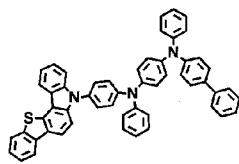


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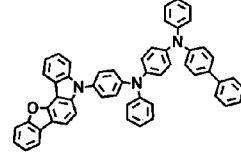
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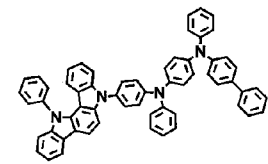
C-21



C-22

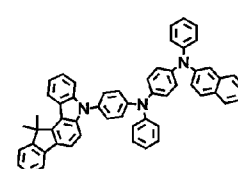


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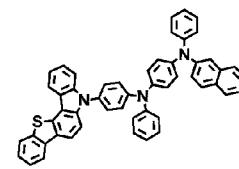


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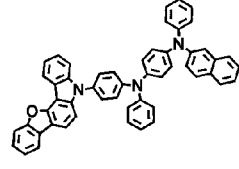
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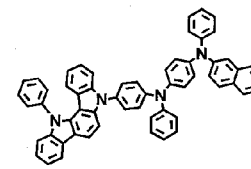
C-25



C-26

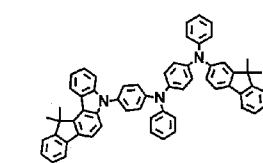


C-27

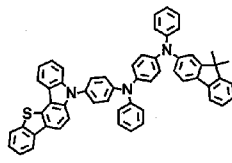


C-28

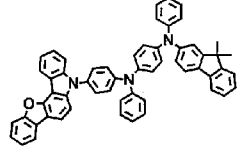
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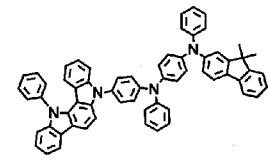
C-29



C-30

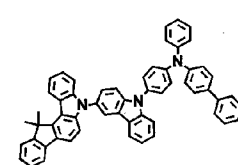


C-31

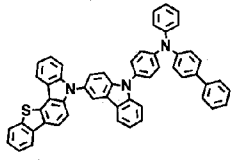


C-32

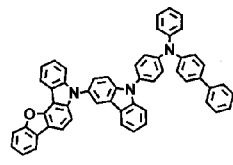
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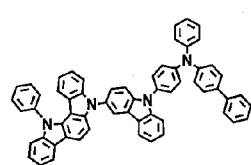
C-33



C-34

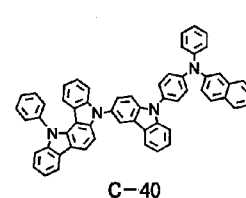
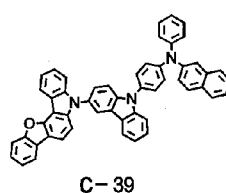
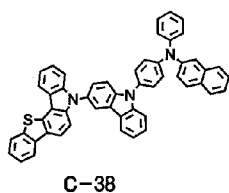
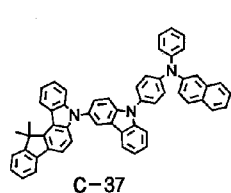


C-35

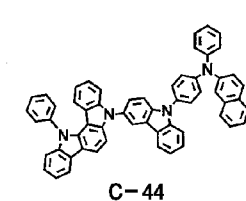
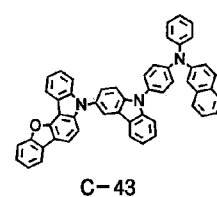
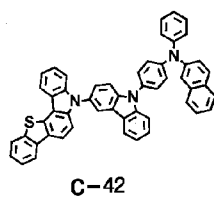
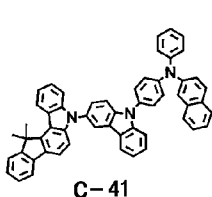


C-36

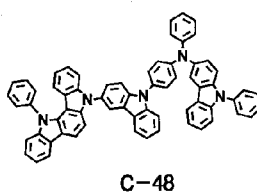
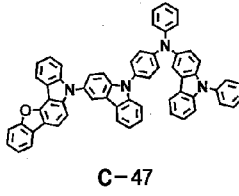
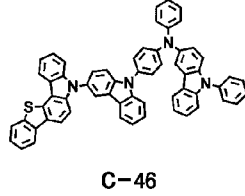
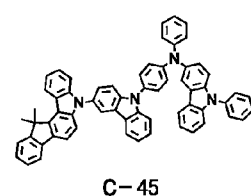
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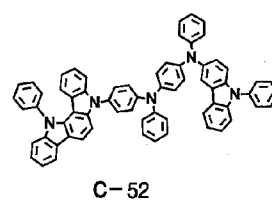
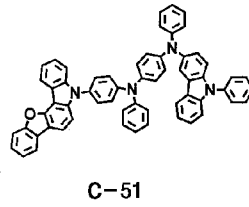
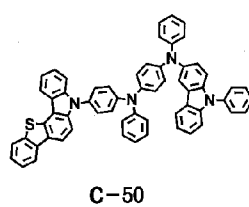
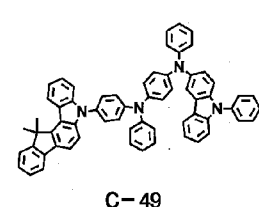
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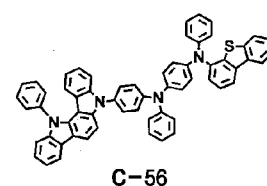
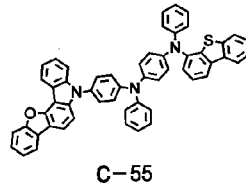
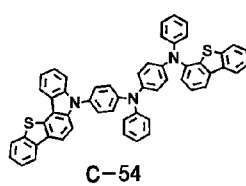
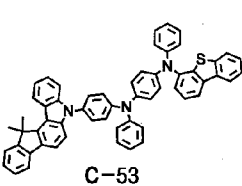
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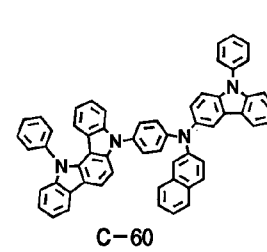
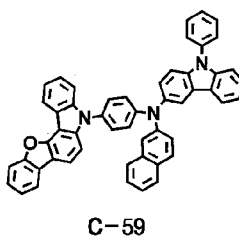
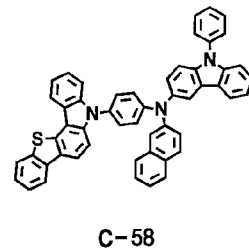
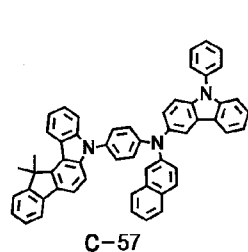
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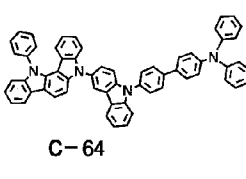
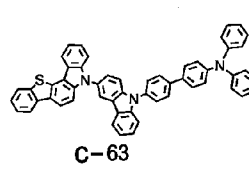
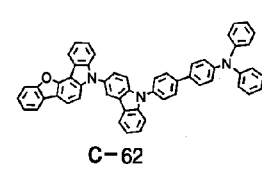
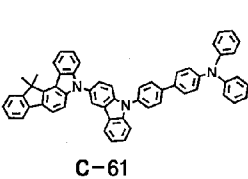
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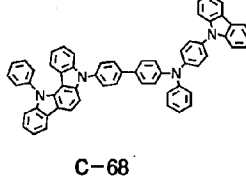
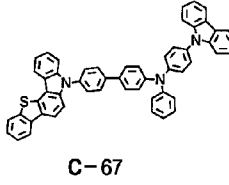
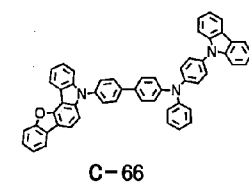
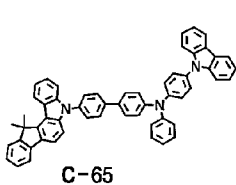
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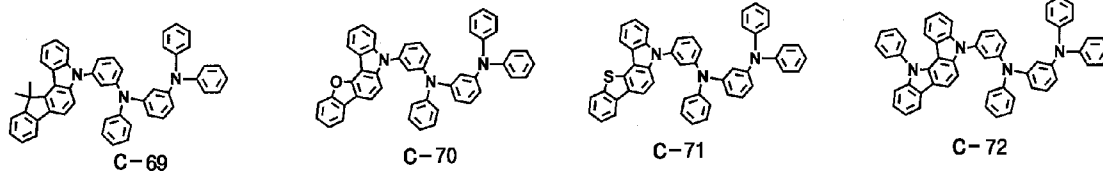
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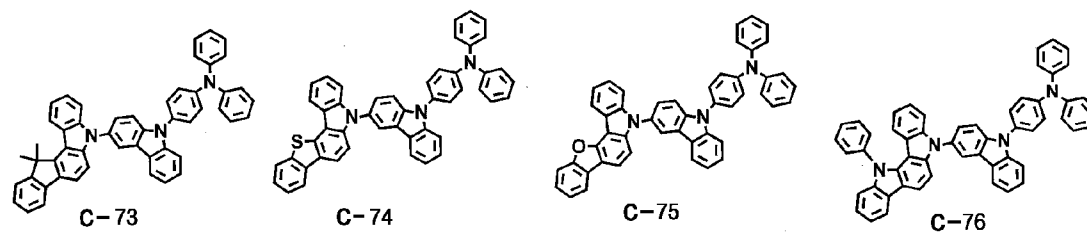
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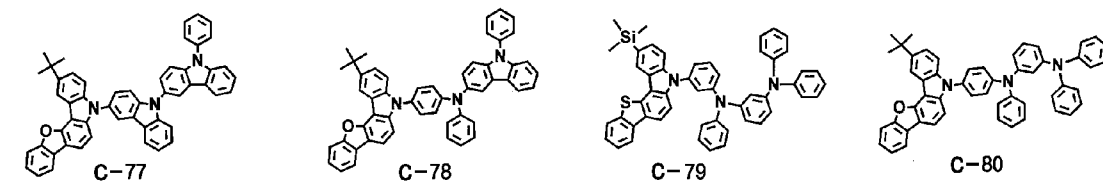
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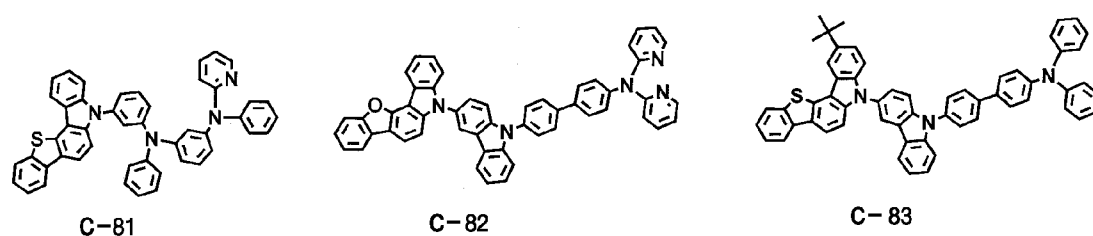
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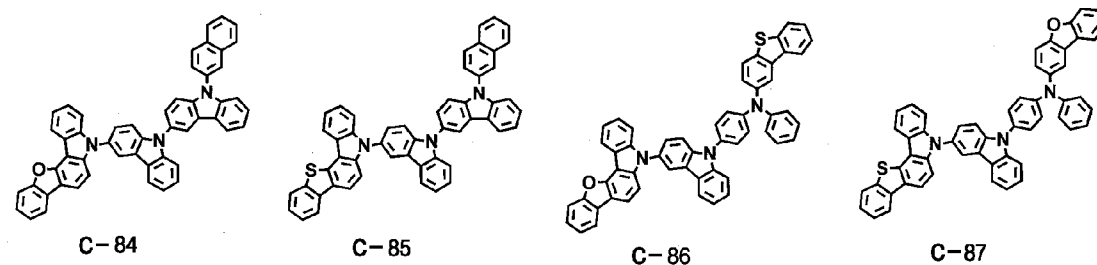
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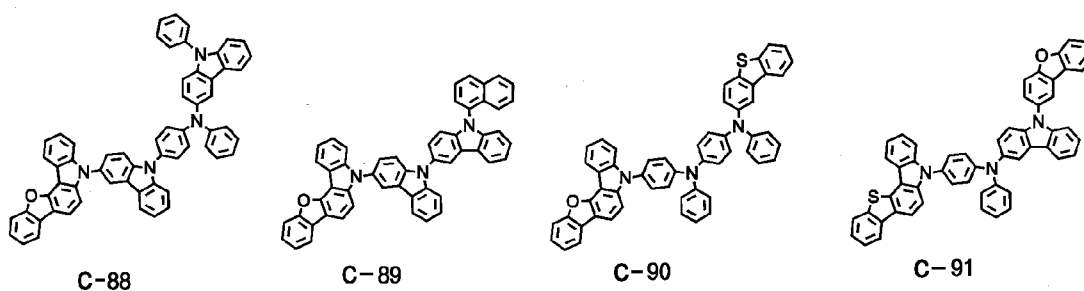
[49]



[50]



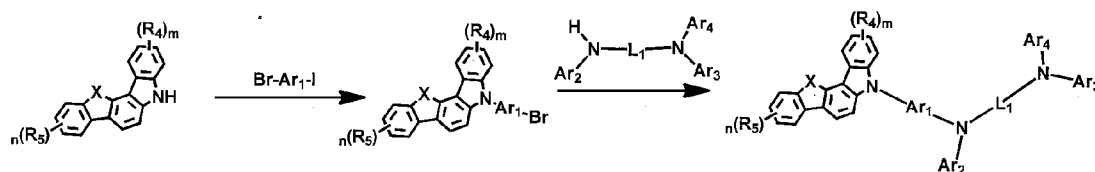
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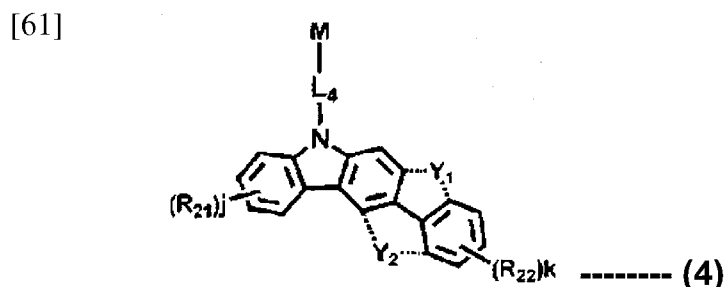
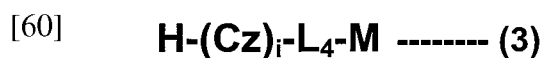
[52] The compounds of the present invention can be prepared by a synthetic method known to one skilled in the art. For example, they can be prepared according to the following reaction scheme 1.

[53] [Reaction Scheme 1]

[54]

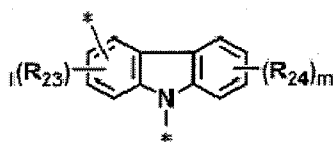


- [55] Furthermore, the present invention provides an organic electroluminescent material comprising the organic electroluminescent compound of formula 1, and an organic electroluminescent device comprising the material. The organic electroluminescent material may consist of the organic electroluminescent compound of formula 1. Otherwise, the organic electroluminescent material may further comprise a conventional compound(s), in addition to the organic electroluminescent compound of formula 1. The organic electroluminescent device may comprise a first electrode, a second electrode, and at least one organic layer disposed between the first and second electrodes. The organic layer may comprise at least one compound of formula 1.
- [56] One of the first and second electrodes may be an anode, and the other may be a cathode. The organic layer may comprise a light-emitting layer, and may further comprise at least one layer selected from a hole injection layer, a hole transport layer, an electron transport layer, an electron injection layer, an interlayer, a hole blocking layer, and an electron blocking layer.
- [57] The compound of formula 1 may be comprised in at least one of the light-emitting layer and the hole transport layer. When used in the hole transport layer, the compound of formula 1 may be comprised as a hole transport material. When used in the light-emitting layer, the compound of formula 1 may be comprised as a host material; preferably, the light-emitting layer may further comprise at least one dopant, and, if needed, a compound other than the compound of formula 1 may be comprised additionally as a second host material.
- [58] The second host material may be from any of the known phosphorescent host materials. Specifically, the phosphorescent host material selected from the compounds of formulae 2 to 4 below is preferable in view of luminous efficiency.



[62] Wherein, Cz represents the following structure;

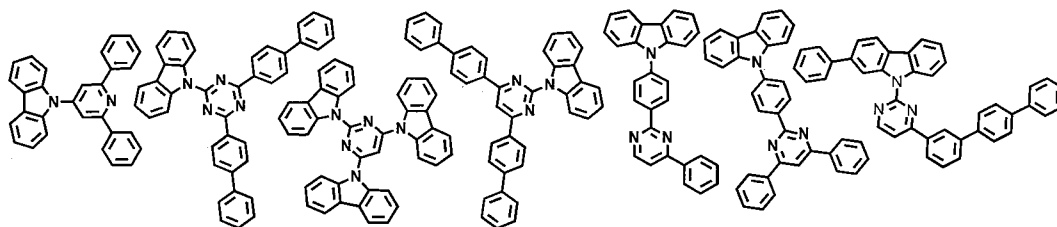
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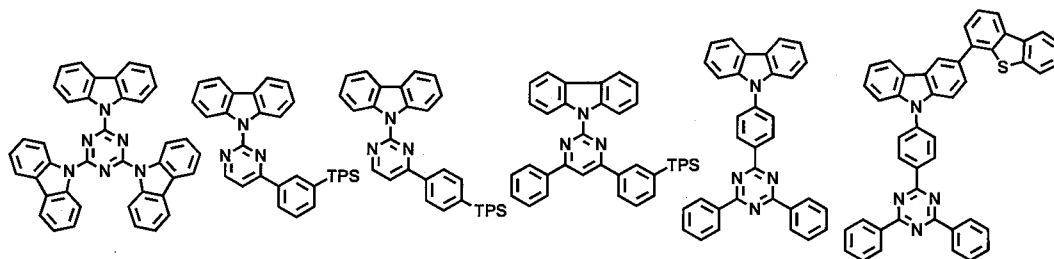
[64] R_{21} to R_{24} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted 5- to 30-membered heteroaryl, or $R_{25}R_{26}R_{27}Si-$; R_{25} to R_{27} each independently represent a substituted or unsubstituted (C1-C30)alkyl, or a substituted or unsubstituted (C6-C30)aryl; L_4 represents a single bond, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted 5- to 30-membered heteroarylene; M represents a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted 5- to 30-membered heteroaryl; Y_1 and Y_2 represent $-O-$, $-S-$, $-N(R_{31})-$ or $-C(R_{32})(R_{33})-$, provided that Y_1 and Y_2 do not simultaneously exist; R_{31} to R_{33} each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted 5- to 30-membered heteroaryl, and R_{32} and R_{33} may be the same or different; h and i each independently represent an integer of 1 to 3; j , k , l and m each independently represent an integer of 0 to 4, and where h , i , j , k , l or m is an integer of 2 or more, each of (Cz- L_4), each of (Cz), each of R_{21} , each of R_{22} , each of R_{23} or each of R_{24} may be the same or different.

[65] Specifically, the second host material includes the following:

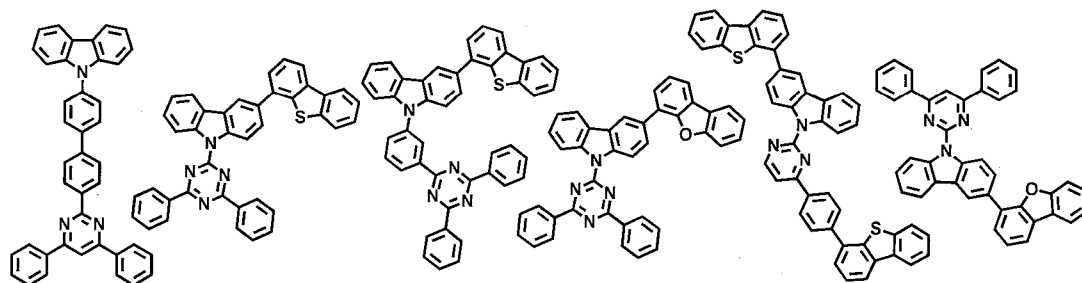
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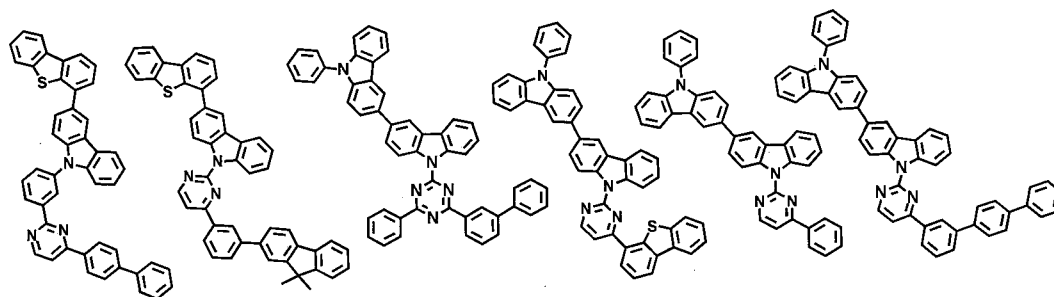
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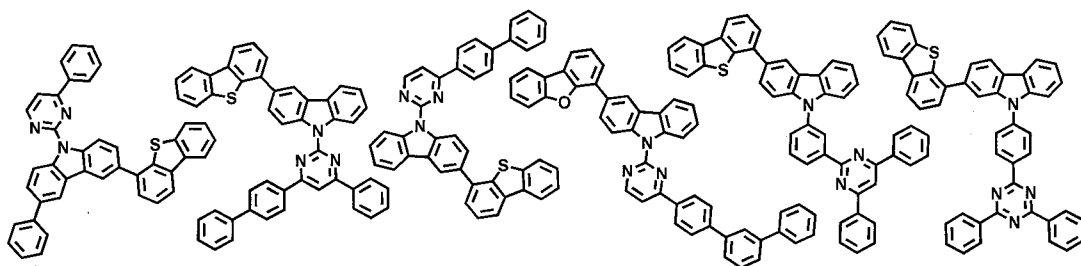
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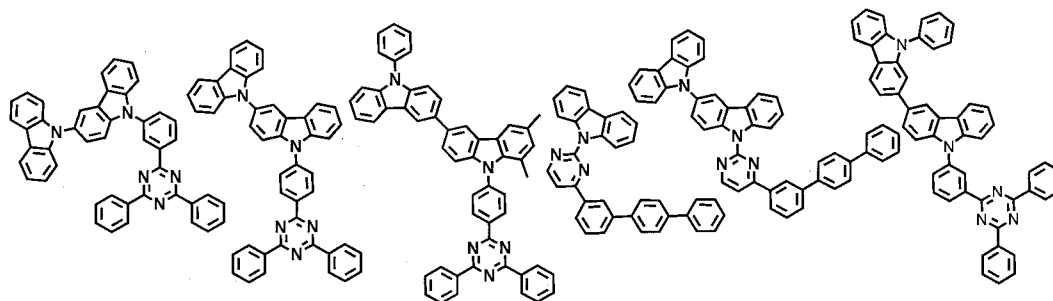
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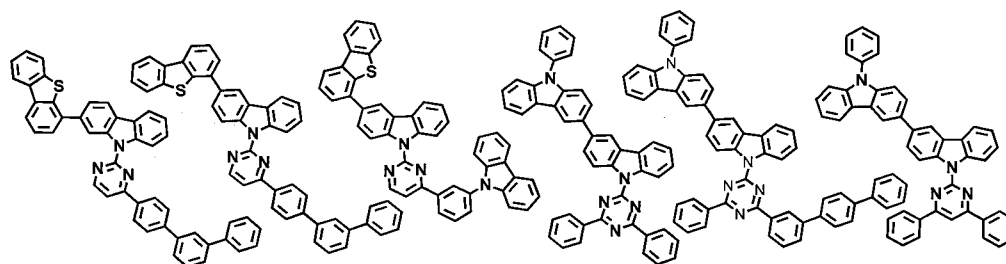
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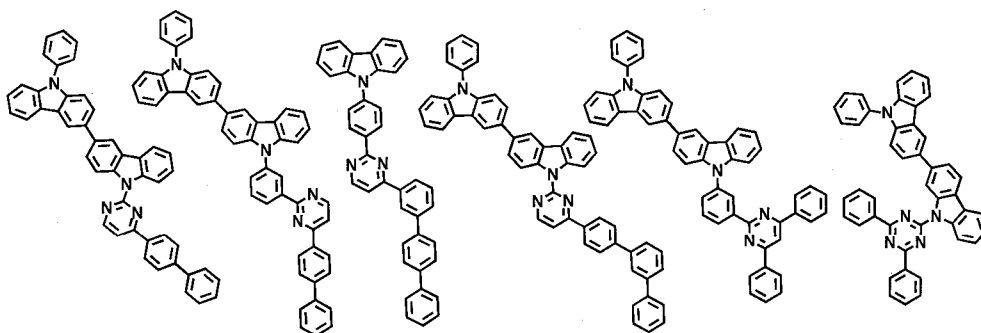
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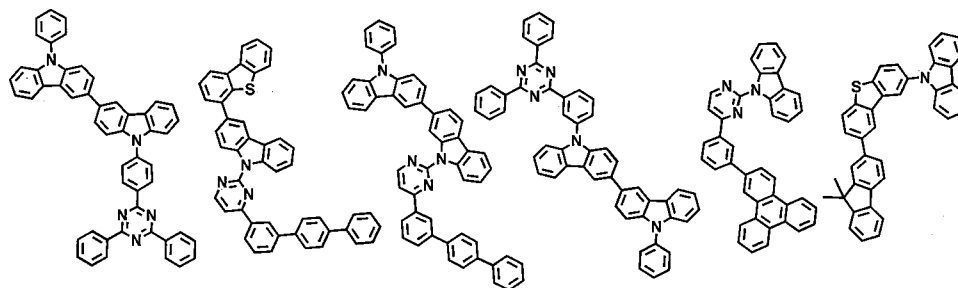
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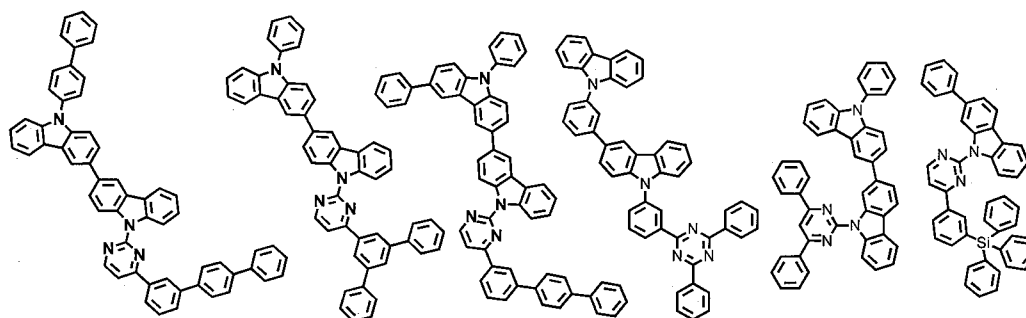
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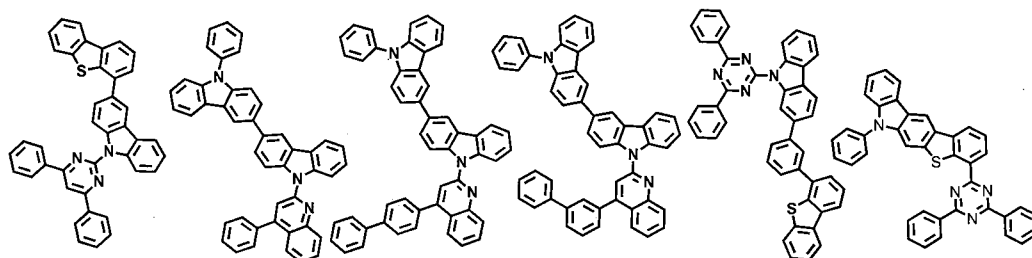
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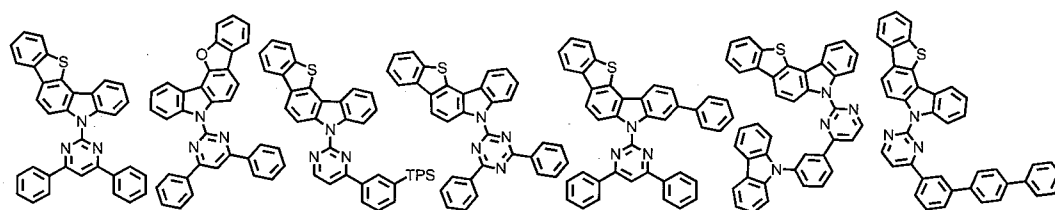
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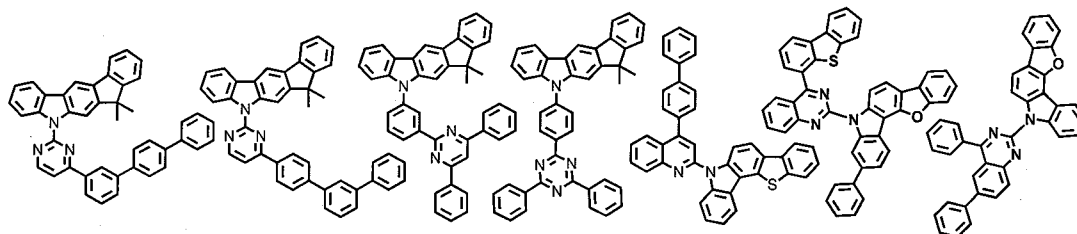
[76]



[77]



[78]

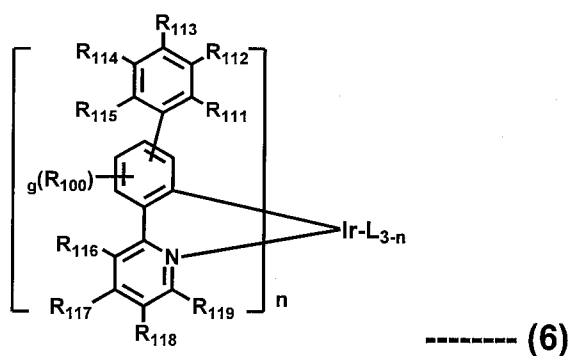
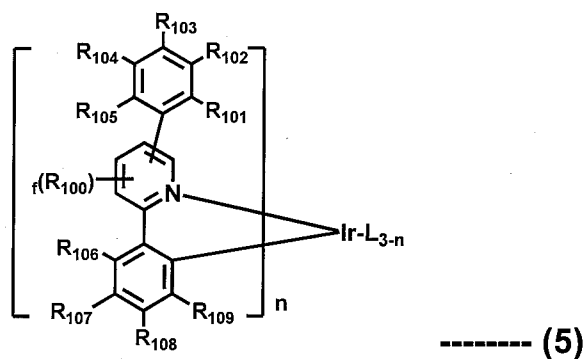


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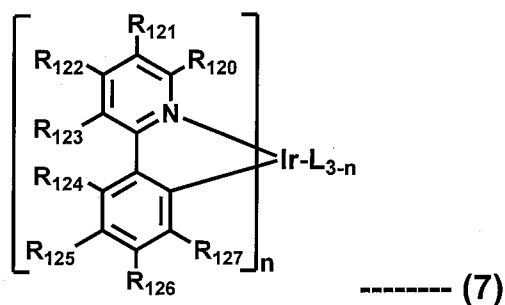
[80] The dopant for the organic electroluminescent device of the present invention is preferably at least one phosphorescent dopant. The phosphorescent dopant for the electroluminescent device of the present invention is not limited, but may be preferably selected from metallated complex compounds of iridium (Ir), osmium (Os), copper (Cu) and platinum (Pt), more preferably selected from ortho-metallated complex compounds of iridium (Ir), osmium (Os), copper (Cu) and platinum (Pt), and even more preferably ortho-metallated iridium complex compounds.

[81] The phosphorescent dopants may be preferably selected from compounds represented by the following formulae 5 to 7.

[82]

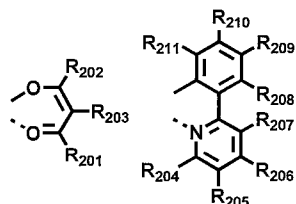


[83]



[84] wherein L is selected from the following:

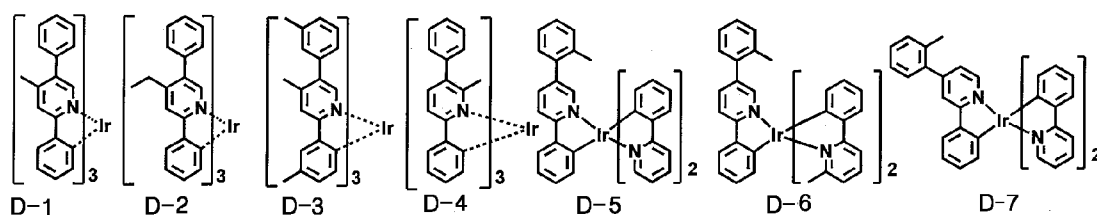
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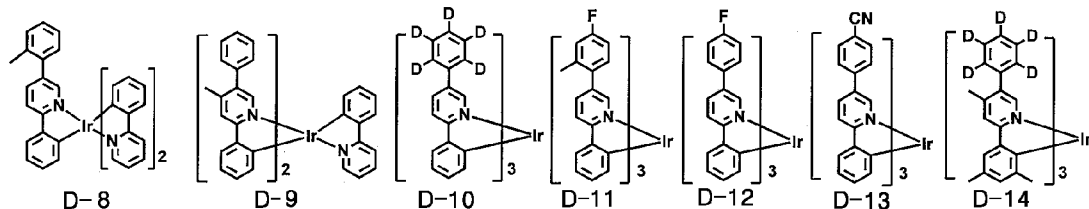
- [86] R_{100} represents hydrogen, or a substituted or unsubstituted (C1-C30)alkyl; R_{101} to R_{109} , and R_{111} to R_{123} each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl unsubstituted or substituted with halogen(s), a cyano, or a substituted or unsubstituted (C1-C30)alkoxy; R_{120} to R_{123} may be linked with an adjacent substituent(s) to form a fused ring, e.g. quinoline; R_{124} to R_{127} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, or a substituted or unsubstituted (C6-C30)aryl; where R_{124} to R_{127} are aryl, they may be linked with an adjacent substituent(s) to form a fused ring, e.g. fluorene; R_{201} to R_{211} each independently represent hydrogen, deuterium, a halogen, or a (C1-C30)alkyl unsubstituted or substituted with halogen(s); f and g each independently represent an integer of 1 to 3, where f or g is an integer of 2 or more, each of R_{100} may be the same or different, and n is an integer of 1 to 3.

- [87] Specifically, the phosphorescent dopant materials include the following:

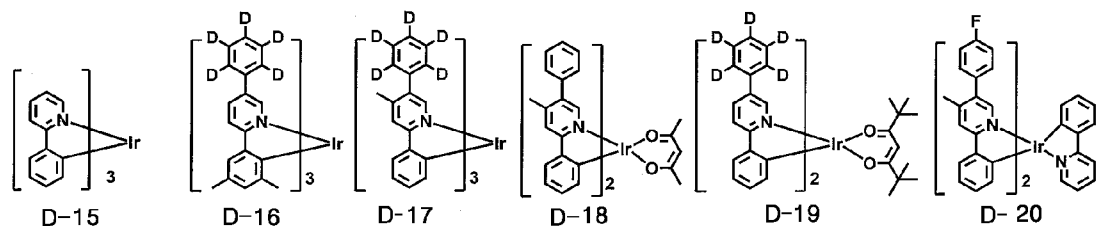
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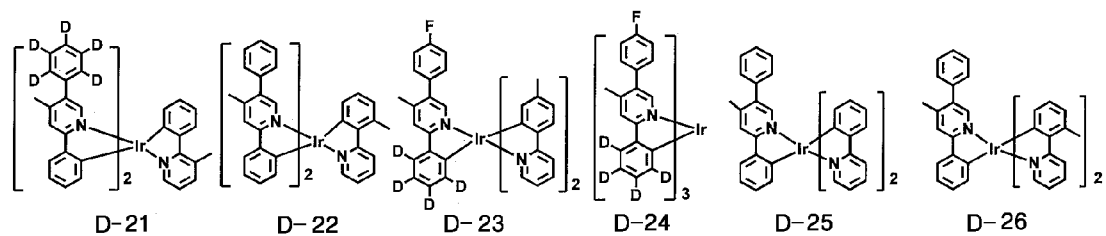
[89]



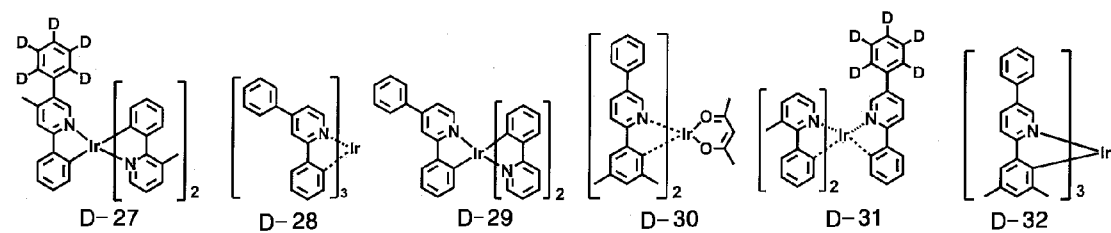
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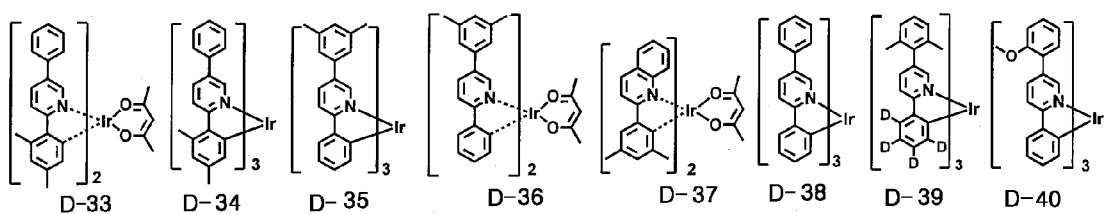
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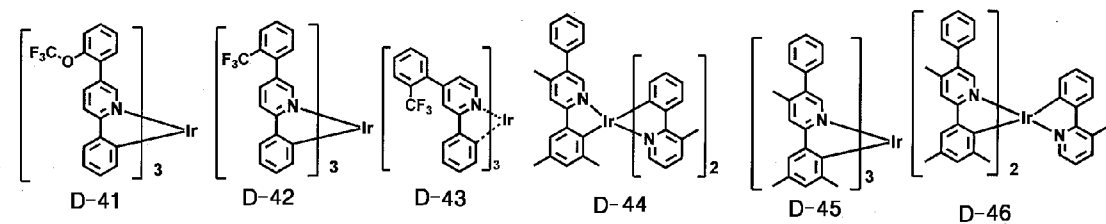
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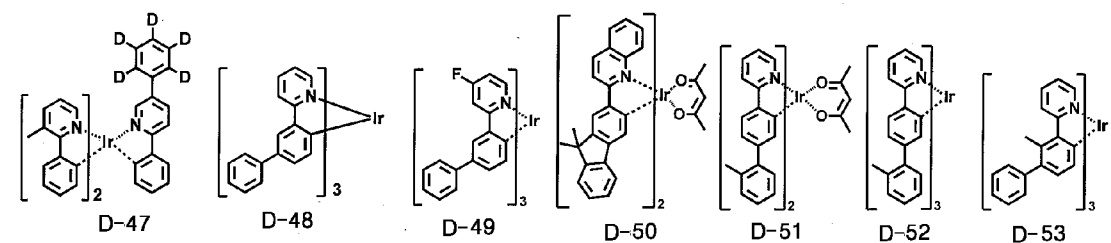
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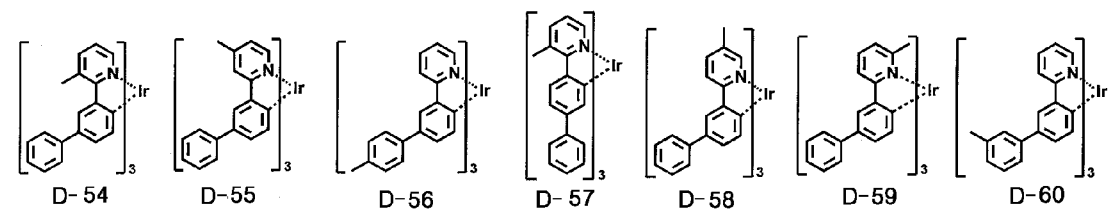
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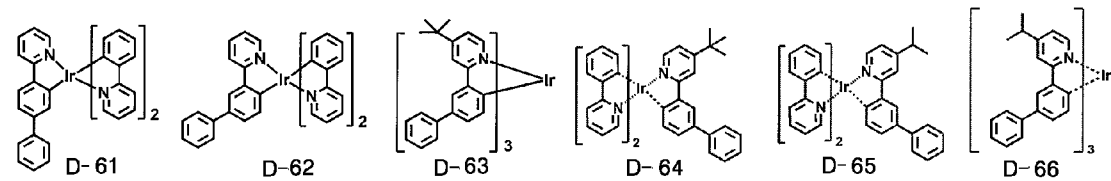
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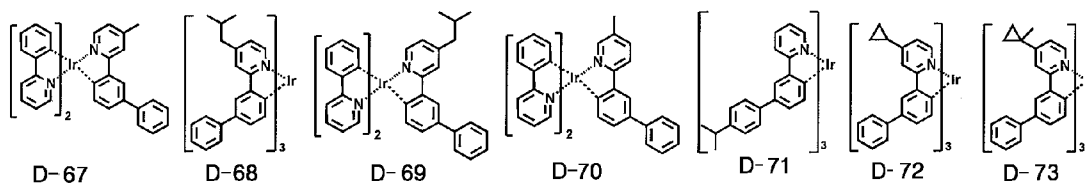
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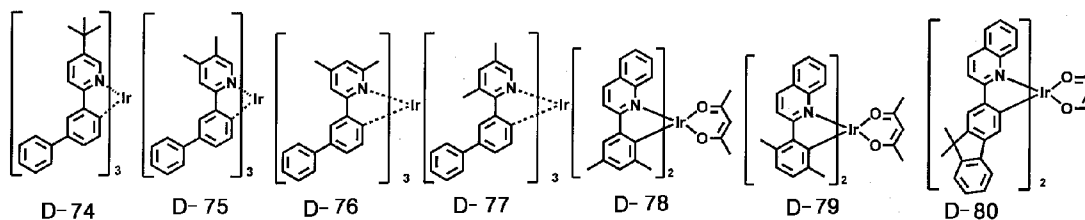
[97]



[98]



[99]



[100] Furthermore, the present invention provides a material for preparing an organic electroluminescent device. The material comprises the organic electroluminescent compound of the present invention as a host material or a hole transport material.

When the compound of the present invention is comprised as a host material, the material for preparing an organic electroluminescent device may further comprise a second host material, in which the ratio of the first host material to the second host material may be in the range of 1:99 to 99:1.

[101] In addition, the organic electroluminescent device of the present invention comprises a first electrode, a second electrode, and at least one organic layer disposed between the first and second electrodes. The organic layer comprises the material for preparing the organic electroluminescent device of the present invention.

[102] The organic layer of the organic electroluminescent device of the present invention may further comprise, in addition to the compound of formula 1, at least one compound selected from the group consisting of arylamine-based compounds and styrylarylamine-based compounds.

[103] In the organic electroluminescent device of the present invention, the organic layer may further comprise, in addition to the compound of formula 1, at least one metal selected from the group consisting of metals of Group 1, metals of Group 2, transition metals of the 4th period, transition metals of the 5th period, lanthanides and organic metals of the d-transition elements of the Periodic Table, or at least one complex compound comprising said metal. The organic layer may comprise a light-emitting layer and a charge generating layer.

[104] In addition, the organic electroluminescent device of the present invention may emit white light by further comprising at least one light-emitting layer, which comprises a blue electroluminescent compound, a red electroluminescent compound or a green electroluminescent compound known in the field, besides the compound of the present invention. Also, if needed, a yellow or orange light-emitting layer can be comprised in the device.

[105] According to the present invention, at least one layer (hereinafter, "a surface layer") may be placed on an inner surface(s) of one or both electrode(s), selected from a chalcogenide layer, a metal halide layer and a metal oxide layer. Specifically, a chalcogenide (includes oxides) layer of silicon or aluminum is preferably placed on an anode surface of an electroluminescent medium layer, and a metal halide layer or a metal oxide layer is preferably placed on a cathode surface of an electroluminescent medium layer. Such a surface layer provides operation stability for the organic electroluminescent device. Preferably, the chalcogenide includes SiO_x ($1 \leq x \leq 2$), AlO_x ($1 \leq x \leq 1.5$), SiON , SiAlON , etc.; the metal halide includes LiF , MgF_2 , CaF_2 , a rare earth metal fluoride, etc.; and the metal oxide includes Cs_2O , Li_2O , MgO , SrO , BaO , CaO , etc.

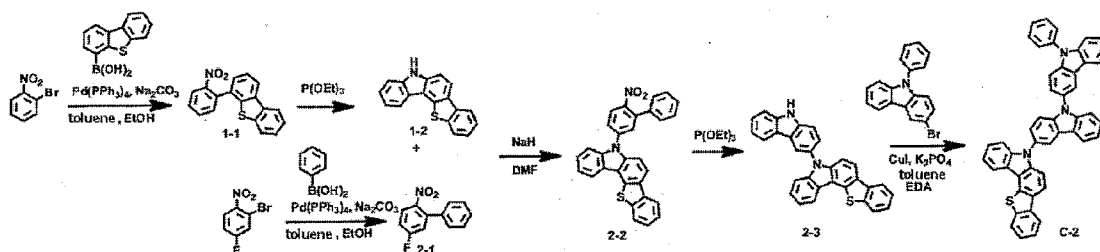
[106] In the organic electroluminescent device of the present invention, a mixed region of an electron transport compound and a reductive dopant, or a mixed region of a hole transport compound and an oxidative dopant may be placed on at least one surface of a pair of electrodes. In this case, the electron transport compound is reduced to an anion, and thus it becomes easier to inject and transport electrons from the mixed region to an electroluminescent medium. Furthermore, the hole transport compound is oxidized to a cation, and thus it becomes easier to inject and transport holes from the mixed region to the electroluminescent medium. Preferably, the oxidative dopant includes various Lewis acids and acceptor compounds, and the reductive dopant includes alkali metals, alkali metal compounds, alkaline earth metals, rare-earth metals, and mixtures thereof. A reductive dopant layer may be employed as a charge generating layer to prepare an electroluminescent device having two or more electroluminescent layers and emitting white light.

[107] In order to form each layer of the organic electroluminescent device of the present invention, dry film-forming methods such as vacuum evaporation, sputtering, plasma and ion plating methods, or wet film-forming methods such as spin coating, dip coating, and flow coating methods can be used.

[108] Hereinafter, the compound, the preparation method of the compound, and the luminescent properties of the device will be explained in detail with reference to the following examples.

[109] **Example 1: Preparation of compound C-2**

[110]



[111] Preparation of compound 1-1

[112] After adding distilled water(H₂O) (200 ml) to a mixture of 2-bromonitrobenzene (30g, 148mmol), 4-dibenzothiophene boric acid (41g, 178mmol), tetrakis(triphenylphosphine)palladium(0) [Pd(PPh₃)₄] (6.5g, 5.9mmol), Na₂CO₃ (39g, 370mmol), toluene (600ml), and ethanol (EtOH) (200ml), the mixture was stirred at 120°C for 2 hours. After completing the reaction, the mixture was washed with distilled water, extracted with ethyl acetate (EA), and the organic layer was dried with MgSO₄. Then, the solvent was removed with a rotary evaporator, and the remaining product was purified with a column chromatography to obtain compound 1-1 (39g, 86%).

[113] Preparation of compound 1-2

[114] A mixture of 2-(dibenzothiophen-4-yl)-nitrobenzene (39g, 128mmol), triethyl phosphite [P(OEt)₃] (300mL) and 1,2-dichlorobenzene (300mL) was stirred in a round bottom flask (RBF) at 150°C for 8 hours. After removing the solvent by distillation under vacuum, the obtained crude product was purified with a column chromatography to obtain compound 1-2 (21g, 61%).

[115] Preparation of compound 2-1

[116] After adding H₂O (90 ml) to a mixture of 4-bromo-2-fluoronitrobenzene (15g, 68 mmol), phenyl boric acid (9.1g, 75mmol), Pd(PPh₃)₄ (3.5g, 4.6mmol), Na₂CO₃ (18g, 170mmol), toluene (270mL), and EtOH (90mL), the mixture was stirred at 120°C for 2 hours. After completing the reaction, the mixture was washed with distilled water, extracted with EA, and the organic layer was dried with MgSO₄. Then, the solvent was removed with a rotary evaporator, and the remaining product was purified with a column chromatography to obtain compound 2-1 (9.2 g, 62%).

[117] Preparation of compound 2-2

[118] After dissolving 5H-[1]benzothieno[3,2-c]carbazole (8.8g, 40.5mmol) in dimethyl-formamide (DMF) (180 mL), NaH (1.9g, 60% in mineral oil, 48.6mmol) was added slowly to the solution. After stirring for 15 minutes, 4-fluoro-2-phenylnitrobenzene (8.8g, 40.5mmol) in DMF (20mL) was added, slowly and dropwise, to the solution. The reaction solution was stirred at room temperature for 4 hours, and then was quenched by adding methanol. After treating the resultant with EA/H₂O, the crude product was purified with a column chromatography to obtain compound 2-2 (19g, 100%).

[119] Preparation of compound 2-3

[120] A mixture of 2-phenyl-4-(benzothieno[3,2-c]carbazolyl)-nitrobenzene (19g, 40.3mmol), P(OEt)₃ (80mL), and 1,2-dichlorobenzene (120mL) was stirred in a RBF at 150°C for 8 hours. After removing the solvent by distillation under vacuum, the obtained crude product was purified with a column chromatography to obtain

compound 2-3 (12 g, 68%).

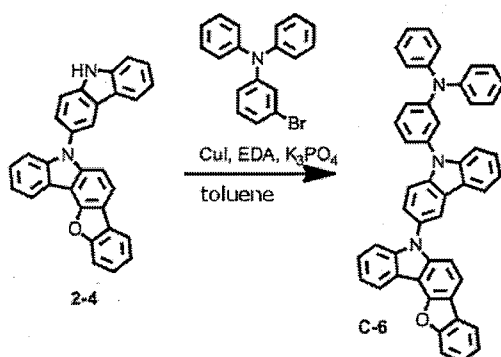
[121] Preparation of compound C-2

[122] A mixture of 3-(benzothieno[3,2-c]carbazolyl)-carbazole (10g, 22.8mmol), 9-phenyl-3-bromocarbazole (8g, 25mmol), CuI (2g, 11.4 mmol), ethylene diamine (1.5mL, 22.8mmol), potassium phosphate (12g, 57mmol), and toluene (200mL) was refluxed in a RBF for 8 hours. After cooling to room temperature, the reaction mixture was filtered and washed with methylene chloride (MC). The filtrate was distilled under reduced pressure and was purified with a column chromatography to obtain compound C-2 (11 g, 71%).

[123] UV=308nm, PL=383nm, mp=241°C, MS/EIMS calculated: 679.21, found: 679.83

[124] Example 2: Preparation of compound C-6

[125]

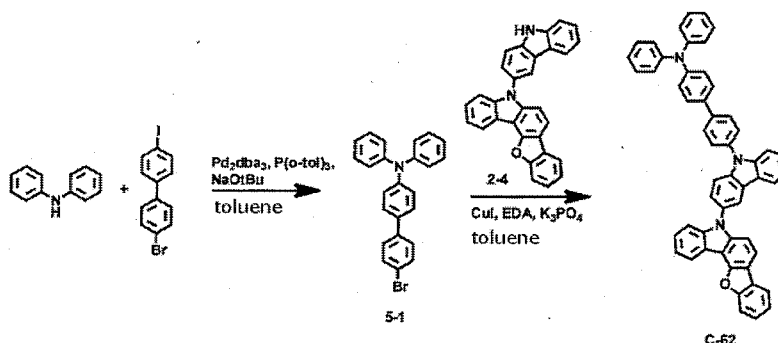


[126] Compound 2-4 was prepared in the same manner as compound 2-3 of Example 1. A mixture of compound 2-3 (5.9g, 14mmol), 3-bromo-N,N-diphenylaniline (5g, 15 mmol), CuI (1.3g, 6.9mmol), K₃PO₄ (8.9g, 42mmol), ethylene diamine (1.9ml, 28mmol), and toluene (70mL) was stirred in a 250mL sized RBF at 120°C overnight. After treating the reaction mixture with EA/H₂O, it was dried with MgSO₄ and distilled under reduced pressure. The crude product was purified with a column chromatography using MC/hexane(Hx) to obtain compound C-6 (white solids, 6.9 g, 74%).

[127] UV=314nm, PL= 381nm, mp= 236°C, MS/EIMS calculated: 665.25; found: 665.78

[128] Example 3: Preparation of compound C-62

[129]



[130] Preparation of compound 5-1

[131] A mixture of diphenylamine (20g, 118mmol), 4-bromo-4'-iodophenyl (63.6g, 177mmol), tris(dibenzylideneacetone)dipalladium(0) [$\text{Pd}_2(\text{dba})_3$] (3.2g, 3.5mmol), tri(o-tolyl)phosphine [$\text{P}(\text{o-tol})_3$] (2.9g, 9.4mmol), sodium tert-butoxide (NaO^tBu) (22.7g, 236mmol), and toluene (1000mL) was stirred in a 2L sized RBF at 120°C for 1 hour. After treating the reaction mixture with EA/ H_2O , it was dried with MgSO_4 , and distilled under reduced pressure. The crude product was purified with a column chromatography using MC:Hx to obtain compound 5-1 (13.4g, 28%).

[132] Preparation of compound C-62

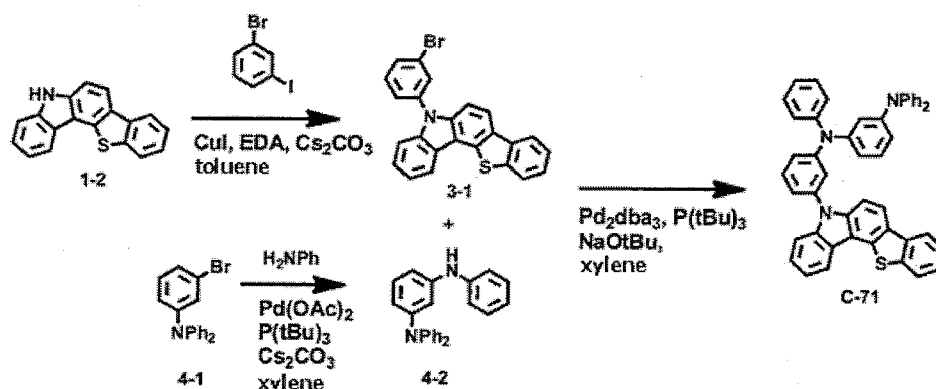
[133] A mixture of compound 2-4 (6.3g, 15mmol), compound 5-1 (6.6g, 16.5mmol), CuI (1.4g, 7.5mmol), K_3PO_4 (9.5g, 45mmol), ethylene diamine (2mL, 30mmol), and toluene (75mL) was stirred in a 250 mL sized RBF at 120°C for 8 hours. The reaction mixture was treated with EA/ H_2O , dried with MgSO_4 , and distilled under reduced pressure. The crude product was purified with a column chromatography using MC:Hx to obtain compound C-62 (white solids, 7.6 g, 68%).

[134] UV=344nm, PL=397nm, mp= 173°C MS/EIMS calculated: 741.28; found: 741.88

[135]

[136] Example 4: Preparation of compound C-71

[137]



[138] Preparation of compound 3-1

[139] A mixture of compound 1-2 (20g, 73mmol), 3-bromoiodobenzene (41g, 146mmol), CuI (7g, 36.5mmol), ethylene diamine (9mL, 146mmol), cesium carbonate (45g, 146mmol), and toluene (400mL) was refluxed in a RBF for 3 hours. The reaction mixture was cooled to room temperature. Then, it was filtered and washed with MC. The filtrate was distilled under reduced pressure and purified with a column chromatography to obtain compound 3-1 (23 g, 54%).

[140] Preparation of compound 4-2

[141] A mixture of compound 4-1 (25g, 77mmol), aniline (14g, 154mmol), palladium(II) acetate [$\text{Pd}(\text{OAc})_2$] (0.35g, 1.54mmol), tri-tert-butylphosphine [$\text{P}(\text{tBu})_3$] (3mL, 7.7 mmol), cesium carbonate (50g, 154mmol), and xylene (400mL) was refluxed in a RBF for 8 hours. The reaction mixture was cooled to room temperature. Then, it was filtered

and washed with MC. The filtrate was distilled under reduced pressure and purified with a column chromatography to obtain compound 4-2 (20 g, 77%).

[142] Preparation of compound C-71

[143] A mixture of compound 3-1 (10g, 23.3mmol), compound 4-2 (7.9g, 23.3mmol), Pd₂(dba)₃ (0.43g, 0.47mmol), P(tBu)₃ (1mL, 2.4mmol), sodium tert-butoxide (3.5g, 35mmol), and xylene (120mL) was refluxed in a RBF for 8 hours. The reaction mixture was cooled to room temperature. Then, it was filtered and washed with MC. The filtrate was distilled under reduced pressure and purified with a column chromatography to obtain compound C-71 (10.2 g, 64%).

[144] UV=310nm, PL=383nm, mp=205°C MS/EIMS calculated: 683.24; found: 683.86

[145]

[146] Device Example 1: an OLED device using the compound of the present invention

[147] An OLED device was produced using the material according to the present invention. A transparent electrode indium tin oxide (ITO) thin film (15 Ω/sq) on a glass substrate for an organic light-emitting diode (OLED) device (Samsung Corning, Republic of Korea) was subjected to an ultrasonic washing with trichloroethylene, acetone, ethanol and distilled water, sequentially, and then was stored in isopropanol. Then, the ITO substrate was mounted on a substrate holder of a vacuum vapor depositing apparatus. N¹,N^{1'}-([1,1'-biphenyl]-4,4'-diyl)bis(N¹-(naphthalen-1-yl)-N⁴,N^{4'}-diphenylbenzen-1,4-diamine) was introduced into a cell of said vacuum vapor depositing apparatus, and then the pressure in the chamber of said apparatus was controlled to 10⁻⁶ torr. Thereafter, an electric current was applied to the cell to evaporate the above introduced material, thereby forming a hole injection layer having a thickness of 60 nm on the ITO substrate. Then, organic electroluminescent compound **C-2** was introduced into another cell of said vacuum vapor depositing apparatus, and was evaporated by applying an electric current to the cell, thereby forming a hole transport layer having a thickness of 20 nm on the hole injection layer. Thereafter,

5-(4-([1,1':4',1''-terphenyl]-3-yl)pyrimidin-2-yl)-5H-benzo[4,5]thieno[3,2-c]carbazole was introduced into one cell of the vacuum vapor depositing apparatus, as a host material, and compound **D-1** was introduced into another cell as a dopant. The two materials were evaporated at different rates and were deposited in a doping amount of 15 wt% based on the total amount of the host and dopant to form a light-emitting layer having a thickness of 30 nm on the hole transport layer. Then, 2-(4-(9,10-di(naphthalen-2-yl)anthracen-2-yl)phenyl)-1-phenyl-1H-benzo[d]imidazole was introduced into one cell and lithium quinolate was introduced into another cell. The two materials were evaporated at the same rate and were deposited in a doping amount of 50 wt% each to form an electron transport layer having a thickness of 30 nm

on the light-emitting layer. Then, after depositing lithium quinolate as an electron injection layer having a thickness of 2 nm on the electron transport layer, an Al cathode having a thickness of 150 nm was deposited by another vacuum vapor deposition apparatus on the electron injection layer. Thus, an OLED device was produced. All the materials used for producing the OLED device were purified by vacuum sublimation at 10^{-6} torr prior to use. The produced OLED device showed a green emission having a luminance of 5350 cd/m² and a current density of 13.1 mA/cm².

[148]

[149] Device Example 2: an OLED device using the organic electroluminescent compound of the present invention

[150] An OLED device was produced in the same manner as in Device Example 1, except that compound **C-71** was used to form a hole transport layer having a thickness of 20 nm, and both 9-(4-([1,1':3',1''-terphenyl]-4-yl)pyrimidin-2-yl)-3-(dibenzo[b,d]thiophen-4-yl)-9H-carbazole in one cell of the vacuum vapor depositing apparatus, as a host material, and compound **D-25** in another cell as a dopant were evaporated at different rates and were deposited in a doping amount of 15 wt% based on the total amount of the host and dopant to form a light-emitting layer having a thickness of 30 nm on the hole transport layer. The produced OLED device showed a green emission having a luminance of 11000 cd/m² and a current density of 24.1 mA/cm².

[151]

[152] Device Example 3: an OLED device using the organic electroluminescent compound of the present invention

[153] An OLED device was produced in the same manner as in Device Example 1, except that compound **C-6** was used to form a hole transport layer having a thickness of 20 nm, and both 9-(4-([1,1'-biphenyl]-4-yl)quinazolin-2-yl)-9'-phenyl-9H,9'H-3,3'-bicarbazole in one cell of the vacuum vapor depositing apparatus as a host material, and compound **D-78** in another cell as a dopant were evaporated at different rates and were deposited in a doping amount of 3 wt% based on the total amount of the host and dopant to form a light-emitting layer having a thickness of 30 nm on the hole transport layer. The produced OLED device showed a red emission having a luminance of 7000 cd/m² and a current density of 50.4 mA/cm².

[154]

[155] Device Example 4: an OLED device using the organic electroluminescent compound of the present invention

[156] An OLED device was produced in the same manner as in Device Example 1, except

that compound **C-62** was used to form a hole transport layer having a thickness of 20 nm. The produced OLED device showed a green emission having a luminance of 3000 cd/m² and a current density of 7.1 mA/cm².

[157]

[158] Comparative Device Example 1: an OLED device using conventional materials

[159] An OLED device was produced in the same manner as in Device Example 1, except that N,N'-di(4-biphenyl)-N,N'-di(4-biphenyl)-4,4'-diaminobiphenyl was used to form a hole transport layer having a thickness of 20 nm; 4,4'-N,N'-dicarbazole-biphenyl as a host material and Ir(ppy)₃ (**D-15**) as a dopant were used to form a light-emitting layer having a thickness of 30 nm on the hole transport layer; and aluminum(III) bis(2-methyl-8-quinolinato)-4-phenylphenolate was used to form a hole blocking layer having a thickness of 10 nm. The produced OLED device showed a green emission having a luminance of 2750 cd/m² and a current density of 8.30 mA/cm².

[160]

[161] Comparative Device Example 2: an OLED device using conventional materials

[162] An OLED device was produced in the same manner as in Device Example 1, except that N,N'-di(4-biphenyl)-N,N'-di(4-biphenyl)-4,4'-diaminobiphenyl was used to form a hole transport layer having a thickness of 20 nm; both CBP as a host material and **D-80** as a dopant were used to form a light-emitting layer having a thickness of 30 nm on the hole transport layer; and aluminum(III) bis(2-methyl-8-quinolinato)-4-phenylphenolate was used to form a hole blocking layer having a thickness of 10 nm. The produced OLED device showed a red emission having a luminance of 1700 cd/m² and a current density of 32.1 mA/cm².

[163]

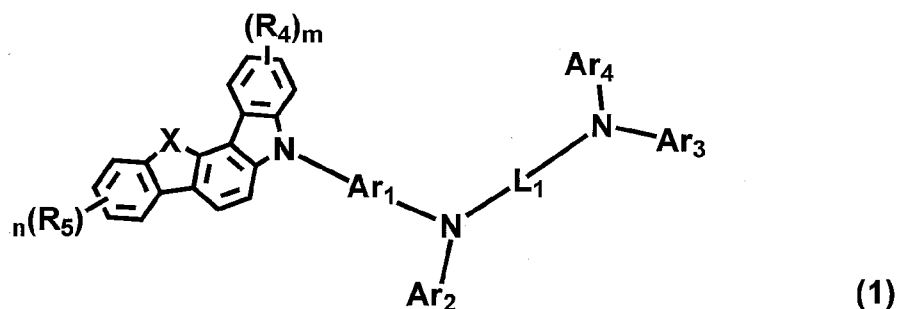
[164] As shown above, the organic electroluminescent compounds of the present invention have superior characteristics over conventional materials, and so the devices using the organic electroluminescent compounds of the present invention exhibit superior luminous characteristics and current efficiency.

[165]

Claims

[Claim 1]

An organic electroluminescent compound represented by the following formula 1:



wherein

X represents -O-, -S-, -CR₁R₂- or -NR₃-;

L₁ represents a substituted or unsubstituted 5- to 30-membered heteroarylene, or a substituted or unsubstituted (C6-C30)arylene;

Ar₁ to Ar₄ each independently represent a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (C6-C30)arylene, a substituted or unsubstituted 5- to 30-membered heteroaryl, or a substituted or unsubstituted 5- to 30-membered heteroarylene;

Ar₁ and Ar₂, Ar₃ and L₁, or Ar₃ and Ar₄ may be linked together with a nitrogen atom bonded thereto to form a nitrogen-containing 5- or 30-membered heteroaryl or a nitrogen-containing 5- or 30-membered heteroarylene;

R₁ to R₃ each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted 5- to 30-membered heteroaryl, or are linked to an adjacent substituent(s) to form a mono- or polycyclic, 3- to 30-membered alicyclic or aromatic ring;

R₄ and R₅ each independently represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C2-C30)alkenyl, a substituted or unsubstituted (C2-C30)alkynyl, a substituted or unsubstituted (C1-C30)alkoxy, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted 3- to 7-membered heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted 5- to 30-membered heteroaryl, -NR₁₁R₁₂, -SiR₁₃R₁₄R₁₅, -SR₁₆, -OR₁₇, -COR₁₈, or -B(OR₁₉)(OR₂₀);

R₁₁ to R₂₀ each independently represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C2-C30)alkenyl, a substituted or unsubstituted (C2-C30)alkynyl, a substituted or unsubstituted (C1-C30)alkoxy, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (C3-C30)cycloalkenyl, a substituted or unsubstituted 3- to 7-membered heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted 5- to 30-membered heteroaryl, or are linked to an adjacent substituent(s) to form a mono- or polycyclic, 3- to 30-membered alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen, and sulfur;

m and n each independently represent an integer of 1 to 4; where m or n is an integer of 2 or more, each of R₄ or each of R₅ may be the same or different; and

the heteroaryl(ene) contains at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

[Claim 2]

The organic electroluminescent compound according to claim 1, wherein X represents -O-, -S-, -CR₁R₂- or -NR₃-;

L₁ represents a substituted or unsubstituted (C6-C30)arylene;

Ar₁ to Ar₄ each independently represent a substituted or unsubstituted (C6-C15)aryl, a substituted or unsubstituted (C6-C15)arylene, a substituted or unsubstituted 5- to 15-membered heteroaryl, or a substituted or unsubstituted 5- to 15-membered heteroarylene;

Ar₁ and Ar₂, Ar₃ and L₁, or Ar₃ and Ar₄ may be linked together with a nitrogen atom bonded thereto to form a nitrogen-containing 5- or 15-membered heteroaryl or a nitrogen-containing 5- or 15-membered heteroarylene;

R₁ to R₃ each independently represent a substituted or unsubstituted (C1-C15)alkyl, a substituted or unsubstituted (C6-C15)aryl, or a substituted or unsubstituted 5- to 15-membered heteroaryl;

R₄ and R₅ each independently represent hydrogen, a halogen, a substituted or unsubstituted (C1-C10)alkyl, a substituted or unsubstituted (C6-C15)aryl, a substituted or unsubstituted 5- to 15-membered heteroaryl, or -SiR₁₃R₁₄R₁₅;

R₁₃ to R₁₅ each independently represent a substituted or unsubstituted (C1-C10)alkyl.

[Claim 3]

The organic electroluminescent compound according to claim 1,
 wherein X represents -O-, -S-, -CR₁R₂- or -NR₃-;
 L₁ represents an unsubstituted (C6-C15)arylene;
 Ar₁ to Ar₄ each independently represent a substituted or unsubstituted (C6-C15)aryl, a substituted or unsubstituted (C6-C15)arylene, a substituted or unsubstituted 5- to 15-membered heteroaryl, or a substituted or unsubstituted 5- to 15-membered heteroarylene;
 Ar₁ and Ar₂, Ar₃ and L₁, or Ar₃ and Ar₄ may be linked together with a nitrogen atom bonded thereto to form a nitrogen-containing 5- or 15-membered heteroaryl or a nitrogen-containing 5- or 15-membered heteroarylene;
 R₁ and R₂ each independently represent a substituted or unsubstituted (C1-C10)alkyl, or a substituted or unsubstituted (C6-C15)aryl;
 R₃ represents a substituted or unsubstituted (C6-C15)aryl;
 R₄ and R₅ each independently represent hydrogen, an unsubstituted (C1-C10)alkyl, or -SiR₁₃R₁₄R₁₅;
 R₁₃ to R₁₅ each independently represent a substituted or unsubstituted (C1-C10)alkyl.

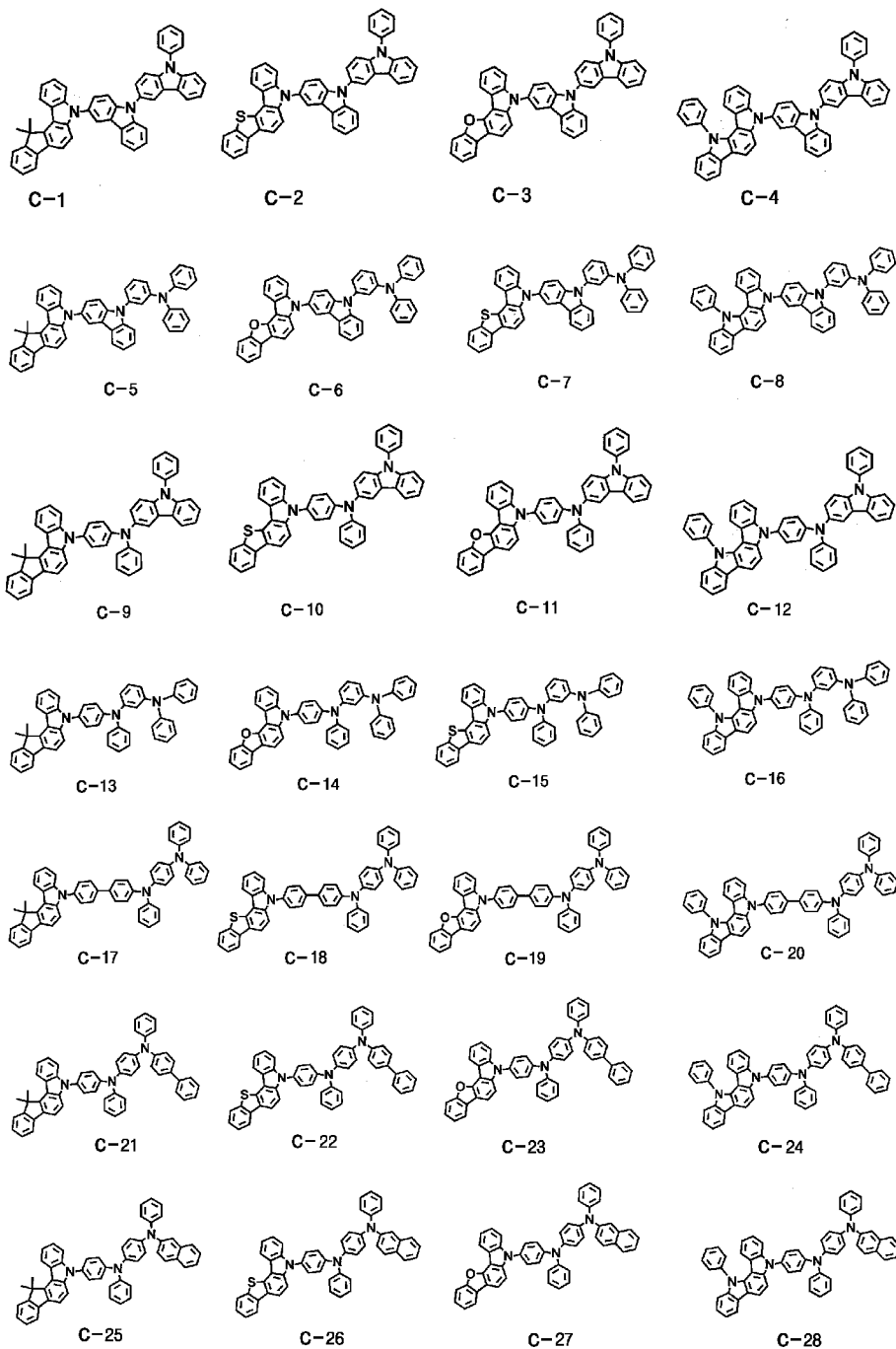
[Claim 4]

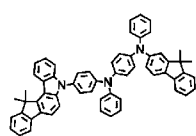
The organic electroluminescent compound according to claim 1,
 wherein the substituents of the substituted alkyl, the substituted alkenyl, the substituted alkynyl, the substituted alkoxy, the substituted cycloalkyl, the substituted cycloalkenyl, the substituted heterocycloalkyl, the substituted aryl(ene), and the substituted heteroaryl(ene) in L₁, Ar₁ to Ar₄, R₁ to R₅, and R₁₁ to R₂₀ each independently are at least one selected from the group consisting of deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a (C1-C30)alkyl, a halo(C1-C30)alkyl, a (C2-C30)alkenyl, a (C2-C30)alkynyl, a (C1-C30)alkoxy, a (C1-C30)alkylthio, a (C3-C30)cycloalkyl, a (C3-C30)cycloalkenyl, a 3- to 7-membered heterocycloalkyl, a (C6-C30)aryloxy, a (C6-C30)arylthio, a 5- to 30-membered heteroaryl unsubstituted or substituted with a (C6-C30)aryl, a (C6-C30)aryl unsubstituted or substituted with a 5- to 30-membered heteroaryl, a tri(C1-C30)alkylsilyl, a tri(C6-C30)arylsilyl, a di(C1-C30)alkyl(C6-C30)arylsilyl, a (C1-C30)alkyldi(C6-C30)arylsilyl, an amino, a mono or di(C1-C30)alkylamino, a mono or di(C6-C30)arylamino, a (C1-C30)alkyl(C6-C30)arylamino, a (C6-C30)aryl(5- to 30-membered)heteroarylamino, a (C1-C30)alkylcarbonyl, a (C1-C30)alkoxycarbonyl, a (C6-C30)arylcarbonyl, a

di(C6-C30)arylboronyl, a di(C1-C30)alkylboronyl, a (C1-C30)alkyl(C6-C30)arylboronyl, a (C6-C30)aryl(C1-C30)alkyl, and a (C1-C30)alkyl(C6-C30)aryl.

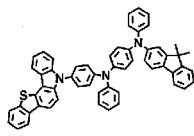
[Claim 5]

The organic electroluminescent compound according to claim 1, wherein the compound represented by formula 1 is selected from the group consisting of:

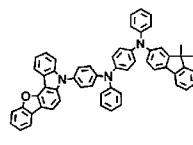




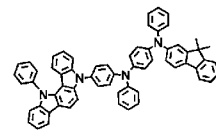
C-29



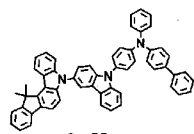
C-30



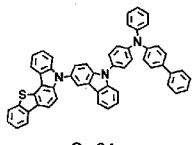
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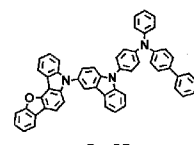
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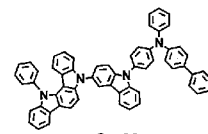
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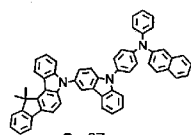
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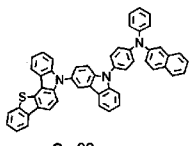
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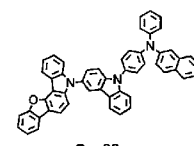
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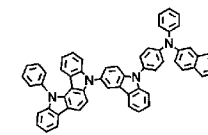
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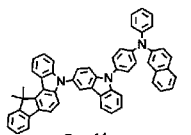
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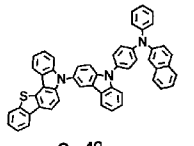
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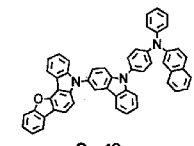
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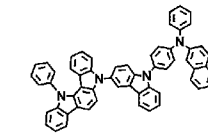
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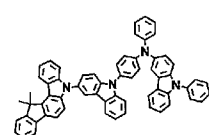
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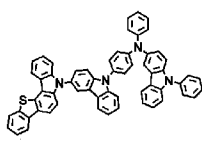
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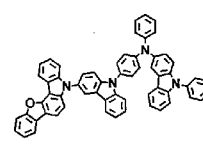
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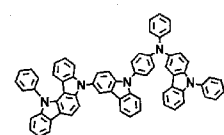
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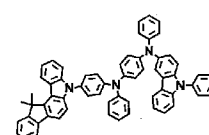
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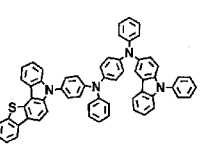
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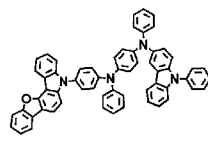
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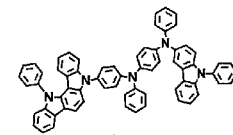
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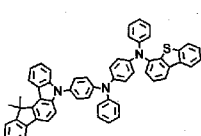
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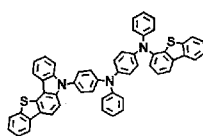
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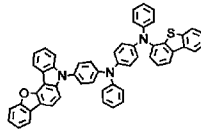
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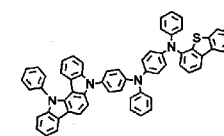
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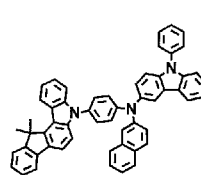
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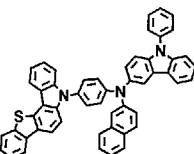
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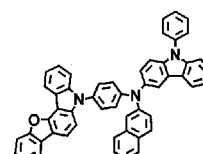
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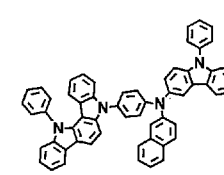
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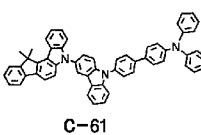
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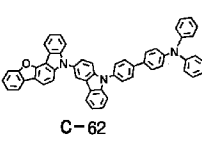
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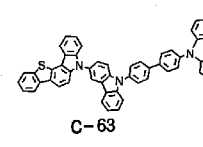
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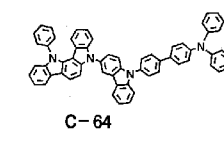
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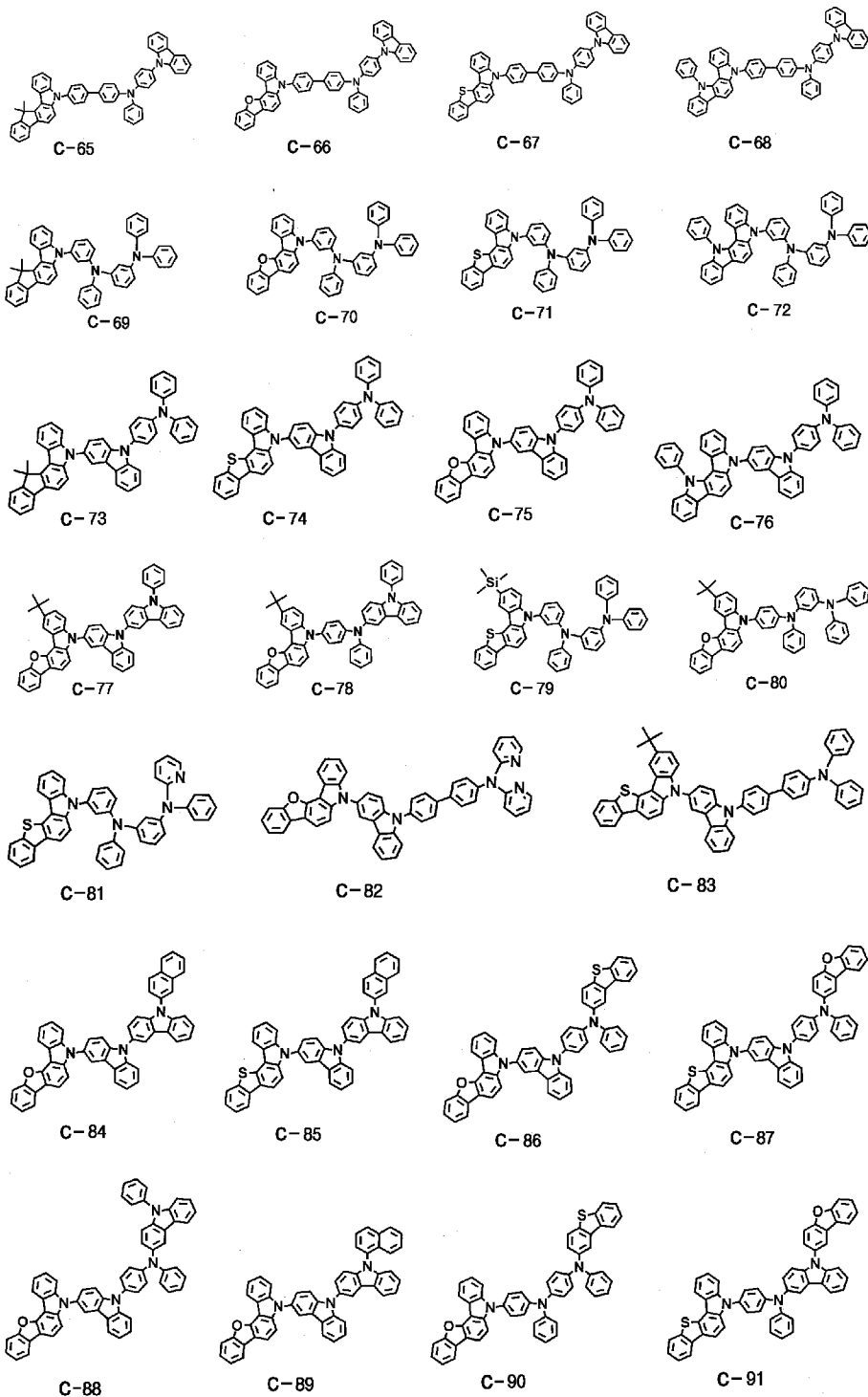
C-62



C-63



C-64



[Claim 6]

An organic electroluminescent device comprising the compound according to claim 1.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2013/008868

A. CLASSIFICATION OF SUBJECT MATTER

C07D 209/82 (2006.01) C07D 487/02 (2006.01) C07D 491/02 (2006.01) C07D 495/02 (2006.01) C09K 11/06 (2006.01)
H01L 27/32 (2006.01) H01L 51/54 (2006.01)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

STN REGISTRY, CAPLUS: Structure search based on compounds of formula (1).

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
	Documents are listed in the continuation of Box C	



Further documents are listed in the continuation of Box C



See patent family annex

* "A"	Special categories of cited documents: document defining the general state of the art which is not considered to be of particular relevance	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"E"	earlier application or patent but published on or after the international filing date	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"O"	document referring to an oral disclosure, use, exhibition or other means	"&"	document member of the same patent family
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search
22 November 2013

Date of mailing of the international search report
22 November 2013

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INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2013/008868
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2012/026780 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 01 March 2012 Abstract; compounds 32-33, 47-48, 63-71, 110-114, 116-117, 119, 136, and 145-146; paragraphs [80]-[83]; examples.	1-6
P,X	WO 2013/109045 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 25 July 2013 Abstract; compounds C-118, C-165, and C-202; paragraphs [86]-[90]; Device Examples.	1-3, 5-6

Form PCT/ISA/210 (fifth sheet) (July 2009)

INTERNATIONAL SEARCH REPORT Information on patent family members		International application No. PCT/KR2013/008868	
This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.			
Patent Document/s Cited in Search Report		Patent Family Member/s	
Publication Number	Publication Date	Publication Number	Publication Date
WO 2012/026780 A1	01 Mar 2012	JP 2013537556 A	03 Oct 2013
		KR 20120042633 A	03 May 2012
		TW 201213504 A	01 Apr 2012
		WO 2012026780 A1	01 Mar 2012
WO 2013/109045 A1	25 Jul 2013	WO 2013109045 A1	25 Jul 2013
End of Annex			
Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001. Form PCT/ISA/210 (Family Annex)(July 2009)			