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(54) Title: NOVEL ORGANIC ELECTROLUMINESCENT COMPOUNDS AND ORGANIC ELECTROLUMINESCENT DEVICE USING 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 compounds of the present invention, it is possible to manufacture an OLED device with a long operating lifespan and high luminous efficiency. In addition, the compounds can improve the power efficiency of the device and reduce overall power consumption.



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Description

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

Technical Field

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

Background Art

- [2] An electroluminescent (EL) device is a self-light-emitting device which has advantages over other types of display devices 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 for forming a light-emitting layer [Appl. Phys. Lett. 51, 913, 1987].
- [3] The most important factor determining luminous efficiency in an organic EL device is the light-emitting material. Until now, fluorescent materials have been widely used as a 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)picolinate iridium (Firpic) as red, green and blue materials, respectively.
- [4] Until now, 4,4'-N,N'-dicarbazol-biphenyl (CBP) was the most widely known host material for phosphorescent substances in conventional technologies. Further, an organic EL device using bathocuproine (BCP) and aluminum(III)bis(2-methyl-8-quinolate)(4-phenylphenolate) (BALq) for a hole blocking layer is also known, and Pioneer (Japan) et al. developed a high performance organic EL device employing a derivative of BALq as a host material.
- [5] Though these materials provide good light-emitting characteristics, they have the following disadvantages: (1) Due to their low glass transition temperature and poor thermal stability, degradation may occur during a high-temperature deposition process in a vacuum. (2) The power efficiency of an organic EL device is given by $[(\pi/\text{voltage}) \times \text{current efficiency}]$, and power efficiency is inversely proportional to voltage. An

organic EL device comprising phosphorescent host materials provides a higher current efficiency (cd/A) than one comprising fluorescent materials. However, it has a higher driving voltage, and thus, there is less advantages in terms of power efficiency (lm/W). (3) Further, the operating lifespan of the organic EL device is short, and luminous efficiency still needs improvement.

- [6] International Patent Publication No. WO 2011/081423 discloses a carbazole compound substituted with a triphenylene group and a fluorene group. However, the compound requires high driving voltage, and so is not suitable for commercialization.

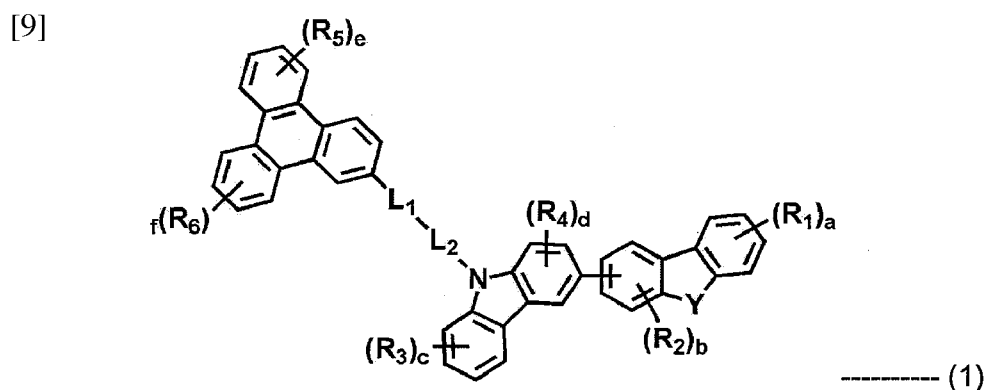
Disclosure of Invention

Technical Problem

- [7] The objective of the present invention is to provide an organic electroluminescent compound imparting high luminous efficiency and a long operating lifespan to a device, and having suitable color coordinate; and an organic electroluminescent device having high efficiency and a long lifespan, using said compound as a light-emitting material.

Solution to Problem

- [8] The present inventors found that the objective above is achievable by an organic electroluminescent compound represented by the following formula 1:



- [10] wherein
- [11] Li and L₂ each independently represent a single bond, a substituted or unsubstituted 3- to 30- membered heteroarylene group, or a substituted or unsubstituted (C6-C30)arylene group, provided that Li and L₂ are not simultaneously single bonds;
- [12] Y represents -O-, -S-, -CR₇R₈- or -NR₉-;
- [13] R₁ to R₉ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30- membered heteroaryl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted 5- to 7- membered heterocycloalkyl group, a (C6-C30)cycloalkyl group fused with one or more aromatic rings, -NRi₀Rn, -SRi₂, -OR₁₃ or -SiRi₄R₁₅R₁₆;

- [14] R_{10} to R_{16} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30- membered heteroaryl group;
- [15] a, c, e and f each independently represent an integer of 1 to 4; where a, c, e and f are integers of 2 or more, each of R_1 , R_3 , R_5 and R_6 is the same or different;
- [16] b and d each independently represent an integer of 1 to 3; where b and d are integers of 2 or more, each of R_2 and R_4 is the same or different; and
- [17] the heterocycloalkyl group, the heteroarylene group and the heteroaryl group contain at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

Advantageous Effects of Invention

- [18] The organic electroluminescent compounds according to the present invention have high luminous efficiency and good lifespan characteristics, and thus could provide an organic electroluminescent device having long operating lifespan.
- [19] In addition, the present invention makes it possible to manufacture a device free from crystallization since the organic electroluminescent compounds used in the present invention are highly efficient in transporting electrons. Further, the compounds have good layer foraiability and improve the current characteristics of the device. Therefore, it is possible to produce an organic electroluminescent device having lowered driving voltages and enhanced power efficiency.

Mode for the Invention

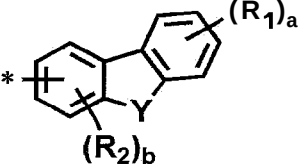
- [20] Hereinafter, the present invention will be described in detail. However, the following description is intended to explain the invention, and is not meant in any way to restrict the scope of the invention.
- [21] The present invention relates to an organic electroluminescent compound represented by formula 1, above, an organic electroluminescent material comprising the compound, and an organic electroluminescent device comprising the material.
- [22] Hereinafter, The organic electroluminescent compound represented by the above formula 1 will be described in detail.
- [23] Herein, "alkyl" includes methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, etc.; "alkenyl" includes vinyl, 1-propenyl, 2-propenyl, 1-butenyl, 2-butenyl, 3-butenyl, 2-methylbut-2-enyl, etc.; "alkynyl" includes ethynyl, 1-propynyl, 2-propynyl, 1-butylnyl, 2-butylnyl, 3-butylnyl, 1-methylpent-2-ynyl, etc.; "cycloalkyl" includes cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, etc.; "5- to 7-membered heterocycloalkyl" is a cycloalkyl having at least one heteroatom selected from B, N, O, S, P(=O), Si and P, preferably O, S and N, and 5 to 7 ring backbone atoms, and includes tetrahydrofuran, pyrrolidine, thiolan, tetrahydropyran, etc.; "aryl(ene)" is a monocyclic or fused ring derived from an (C6-C30)aromatic hydrocarbon, and includes phenyl,

biphenyl, terphenyl, naphthyl, binaphthyl, phenyl naphthyl, naphthyl phenyl, fluorenyl, phenyl fluorenyl, benzofluorenyl, dibenzofluorenyl, phenanthrenyl, phenyl phenanthrenyl, anthracenyl, indenyl, triphenylenyl, pyrenyl, tetracenyl, perylenyl, chrysenyl, naphthacenyl, fluoranthenyl, etc.; "3- to 30-membered heteroaryl(ene)" is an aryl group having at least one, preferably 1 to 4 heteroatom selected from the group consisting of B, N, O, S, P(=O), Si and P, and 3 to 30 ring backbone atoms; is 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 including 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 including benzofuranyl, benzothiophenyl, isobenzofuranyl, dibenzofuranyl, dibenzothiophenyl, benzoimidazolyl, benzothiazolyl, benzoisothiazolyl, benzoisoxazolyl, benzoxazolyl, isoindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolyl, quinazolinyl, quinoxalinyl, carbazolyl, phenoxazinyl, phenanthridinyl, benzodioxolyl, etc. Further, "Halogen" includes F, Cl, Br and I.

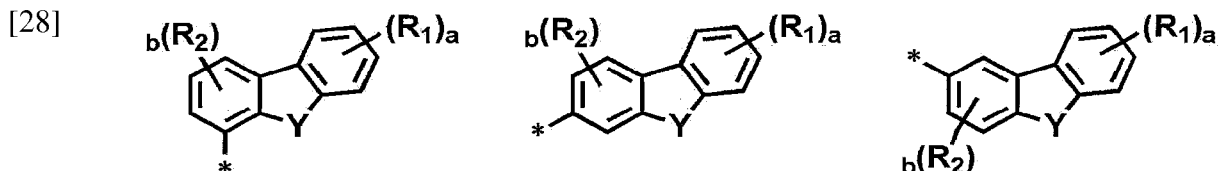
[24] Herein, "substituted" in the expression "substituted or unsubstituted" means that a hydrogen atom in a certain functional group is replaced with another atom or group, i.e., a substituent.

[25] The aryl(ene), heteroaryl(ene), alkyl, cycloalkyl, heterocycloalkyl, arylamino, trialkylsilyl and triarylsilyl groups in L_1 , L_2 , and R_1 to R_{16} of formula 1, can be further substituted with at least one selected from the group consisting of: deuterium; a halogen; a (Cl-C30)alkyl group substituted or unsubstituted with a halogen; a (C6-C30)aryl group substituted or unsubstituted with a halogen; a 3- to 30- membered heteroaryl group substituted or unsubstituted with a (Cl-C30)alkyl or a (C6-C30)aryl; a (C3-C30)cycloalkyl group; a (C6-C30)cycloalkyl group fused with one or more aromatic rings; a 5- to 7- membered heterocycloalkyl group; $R_a R_b R_c Si$ -; a carbazolyl group; $-NR_d R_e$; $-BR_f R_g$; $-PR_h R_i$; $-P(=O)R_j R_k$; a (C6-C30)aryl(Cl-C30)alkyl group; a (Cl-C30)alkyl(C6-C30)aryl group; a (Cl-C30)alkoxy group; and a (C6-C30)aryloxy group, wherein R_a to R_k each independently represent a (Cl-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30- membered heteroaryl group, preferably are at least one selected from the group consisting of deuterium, a halogen, a (Cl-C10)alkyl group, a (C6-C20)aryl group, a halo(C6-C20)aryl group, and a 5- to 20-membered heteroaryl group, more preferably are at least one selected from the group consisting of deuterium, a halogen, a (Cl-C6)alkyl group, a (C6-C12)aryl group, a halo(C6-C12)aryl group, and a 5- to 12-membered heteroaryl group.

[26]

[27] The moiety,  in formula 1, above, is selected from the

following structures, but not limited thereto:



[29] wherein R_i , R_2 , Y, a and b are as defined in formula 1.

[30] In formula 1, above, L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted 3- to 30- membered heteroarylene group, or a substituted or unsubstituted (C6-C30)arylene group, provided that L_1 and L_2 are not simultaneously single bonds, preferably a single bond, a substituted or unsubstituted 5- to 20- membered heteroarylene group, or a substituted or unsubstituted (C6-C20)arylene group, more preferably a single bond; a 5- to 12- membered heteroarylene group unsubstituted or substituted with a (C6-C12)aryl group or a halo(C6-C12)aryl group; or a (C6-C10)arylene group unsubstituted or substituted with a (C1-C6)alkyl group or a 5- to 12- membered heteroaryl group.

[31] Y represents -O-, -S-, $-CR_7R_8$ - or $-NR_9$ -;

[32] R_i to R_9 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30- membered heteroaryl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted 5- to 7- membered heterocycloalkyl group, a (C6-C30)cycloalkyl group fused with one or more aromatic rings, $-NR_{10}R_n$, $-SR_{12}$, $-OR_{13}$ or $-SiR_{14}R_{15}R_{16}$, preferably hydrogen, a halogen, a substituted or unsubstituted (C1-C10)alkyl group, a substituted or unsubstituted (C6-C20)aryl group, or a substituted or unsubstituted 5- to 20- membered heteroaryl group, more preferably hydrogen; a halogen; an unsubstituted (C1-C6)alkyl group; a (C6-C10)aryl group unsubstituted or substituted with deuterium; or an unsubstituted 5- to 10- membered heteroaryl group.

[33] R_{10} to R_{16} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30- membered heteroaryl group.

[34] According to one embodiment of the present invention in formula 1 above, L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted 5- to 20- membered heteroarylene group, or a substituted or unsubstituted (C6-C20)arylene group, provided that L_1 and L_2 are not simultaneously single bonds; Y represents -O-, -

S-, -CR₇R₈- or -NR₉-; R₁ to R₉ each independently represent hydrogen, a halogen, a substituted or unsubstituted (C6-C10)alkyl group, a substituted or unsubstituted (C6-C20)aryl group, or a substituted or unsubstituted 5- to 20- membered heteroaryl group.

[35] According to another embodiment of the present invention in formula 1 above, L₁ and L₂ each independently represent a single bond; a 5- to 12- membered heteroarylene group unsubstituted or substituted with a (C6-C12)aryl group or a halo(C6-C12)aryl group; or a (C6-C10)arylene group unsubstituted or substituted with a (C1-C6)alkyl group or a 5- to 12- membered heteroaryl group, provided that L₁ and L₂ are not simultaneously single bonds, Y represents -O-, -S-, -CR₇R₈- or -NR₉-, R₁ to R₉ each independently represent hydrogen; a halogen; an unsubstituted (C1-C6)alkyl group; a (C6-C10)aryl group unsubstituted or substituted with deuterium; or an unsubstituted 5- to 10- membered heteroaryl group.

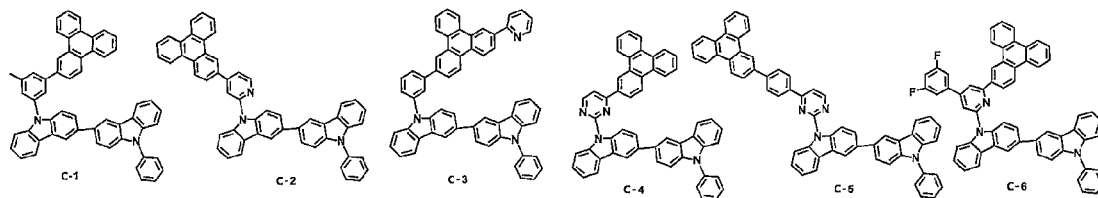
[36] Specifically, L₁ and L₂ can each independently represent a single bond, a substituted or unsubstituted 3- to 30- membered heteroarylene group, or a substituted or unsubstituted (C6-C30)arylene group, provided that L₁ and L₂ are not simultaneously single bonds; Y represents -O-, -S-, -CR₇R₈- or -NR₉-; R₁ to R₉ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30- membered heteroaryl group; and the heteroarylene and arylene groups in L₁ and L₂, and the alkyl, aryl and heteroaryl groups in R₁ to R₉ can be further substituted with at least one selected from the group consisting of deuterium; a halogen; a (C1-C30)alkyl group substituted or unsubstituted with a halogen; a (C6-C30)aryl group substituted or unsubstituted with a halogen; a 3- to 30- membered heteroaryl group substituted or unsubstituted with a (C1-C30)alkyl or a (C6-C30)aryl; a (C6-C30)aryl(C1-C30)alkyl group; and a (C1-C30)alkyl(C6-C30)aryl group.

[37] More specifically, L₁ can represent a single bond, a phenylene, a biphenylene, a terphenylene, a fluorenylene, a triphenylenylene, a pyrenylene, a perylenylene, a fluoranthylene, a thiophenylene, a pyrrolylene, a pyrazolylene, a thiazolylene, an oxazolylene, an oxadiazolylene, a triazinylene, a tetrazinylene, a triazolylene, a furazanylene, a pyridylene, a pyrimidylene, a quinolylene, an isoquinolylene, a quinoxalylene, a benzofuranylene, a benzothiophenylene, an indolylene, a benzoimidazolylene, a benzothiazolylene, a benzoisothiazolylene, a benzoisoxazolylene, a benzoxazolylene, a benzothiadiazolylene, a dibenzofuranylene or a dibenzothiophenylene, provided that L₁ and L₂ are not simultaneously single bonds; R₁ to R₉ each independently represent: hydrogen, deuterium, a fluorine, a methyl, an ethyl, an n-propyl, an i-propyl, an n-butyl, an i-butyl, a t-butyl, an n-pentyl, an i-pentyl, an n-hexyl, an n-heptyl, an n-octyl, a 2-ethylhexyl, an n-nonyl, a decyl, a dodecyl, a hexadecyl, a triflu-

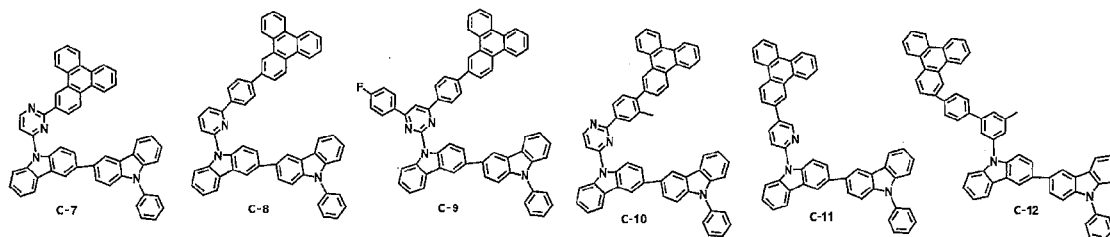
oromethyl, a perfluoroethyl, a trifluoroethyl, a perfluoropropyl, a perfluorobutyl, a phenyl, a biphenyl, a fluorenyl, a fluoranthenyl, a triphenylenyl, a pyrenyl, a chrysenyl, a naphthaceny, a perylenyl, a pyridyl, a pyrrolyl, a furanyl, a thiophenyl, an imidazolyl, a benzoimidazolyl, an indenyl, a pyrazinyl, a pyrimidyl, a pyridazinyl, a quinolyl, an isoquinolyl, a triazinyl, a benzofuranyl, a dibenzofuranyl, a benzothioiophenyl, a dibenzothiophenyl, a pyrazolyl, an indolyl, a carbazolyl, a thiazolyl, an oxazolyl, a benzothiazolyl, a benzoxazolyl, a phenanthrolinyl, or an N-carbazolyl; and the substituents in L₁, L₂ and ³/₄ to R₉ can be further substituted with at least one selected from the group consisting of: deuterium, a chlorine, a fluorine, a methyl, an ethyl, an n-propyl, an i-propyl, an n-butyl, an i-butyl, a t-butyl, an n-pentyl, an i-pentyl, an n-hexyl, an n-heptyl, an n-octyl, a 2-ethylhexyl, an n-nonyl, a decyl, a dodecyl, a hexadecyl, a trifluoromethyl, a perfluoroethyl, a trifluoroethyl, a perfluoropropyl, a perfluorobutyl, a phenyl, a biphenyl, a fluorenyl, a 9,9-dimethylfluorenyl, a fluoranthenyl, a triphenylenyl, a fluorophenyl, a difluorophenyl, a trifluorophenyl, a tetrafluorophenyl, a pentafluorophenyl, a pyridyl, a pyrimidyl, a benzothiophenyl, an imidazolyl, a benzothiazolyl, a benzoimidazolyl, a pyrenyl, a perylenyl, a quinolyl and an isoquinolyl.

[38] The representative organic electroluminescent compounds of the present invention include the following compounds, but not limited thereto:

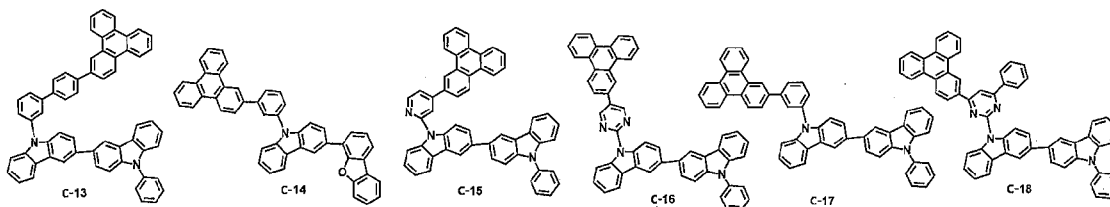
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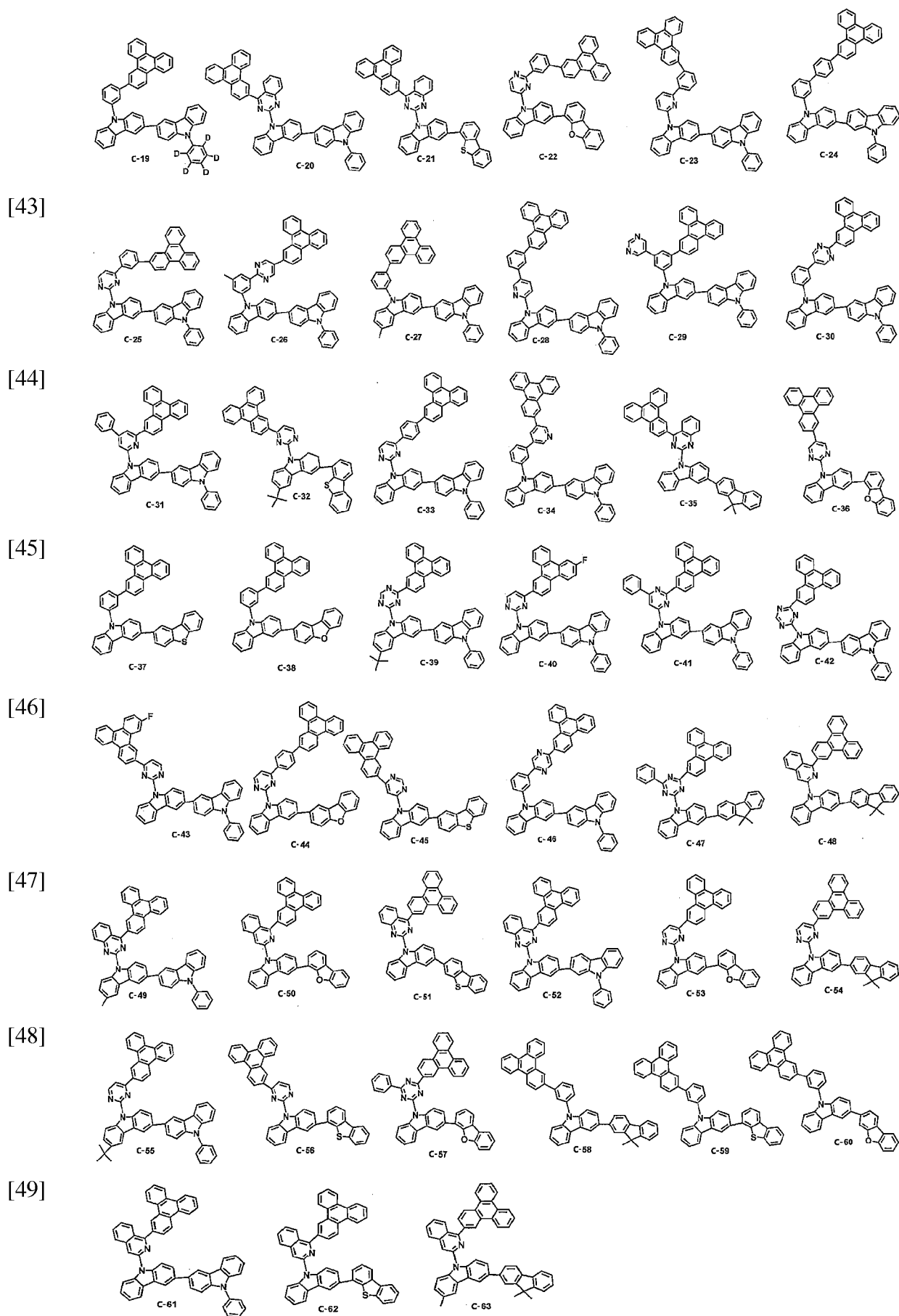
[40]



[41]



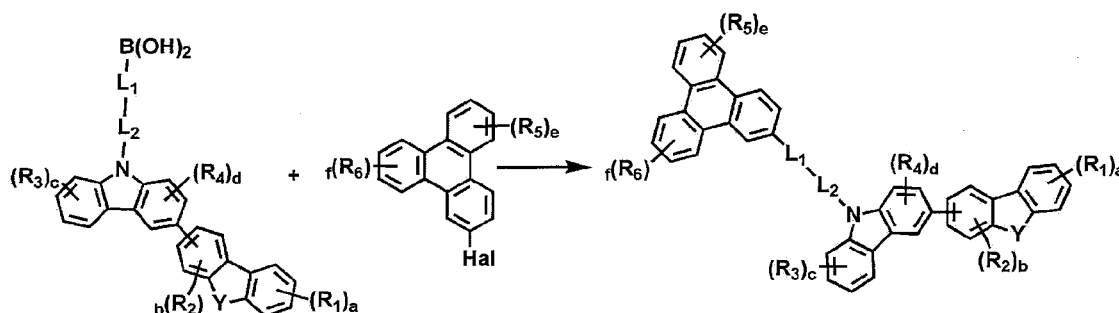
[42]



[50] The organic electroluminescent compounds of the present invention can be prepared according to the following reaction scheme.

[51] [Reaction Scheme 1]

[52]



[53] wherein L_1 , L_2 , R_1 to R_6 , Y , a , b , c , d , e and f are as defined in formula 1 above, and Hal represents a halogen.

[54] In addition, the present invention provides an organic electroluminescent material comprising the organic electroluminescent compound of formula 1, and an organic electroluminescent device comprising the material.

[55] The above material can be comprised of the organic electroluminescent compound according to the present invention alone, or can further include conventional materials generally used in organic electroluminescent materials.

[56] Said organic electroluminescent device comprises a first electrode, a second electrode, and at least one organic layer between said first and second electrodes. Said organic layer may comprise at least one organic electroluminescent compound of formula 1 according to the present invention, or an organic electroluminescent material comprising the compound.

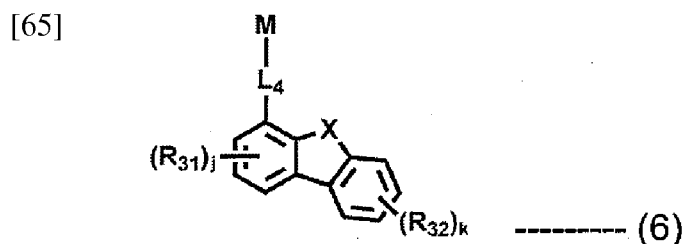
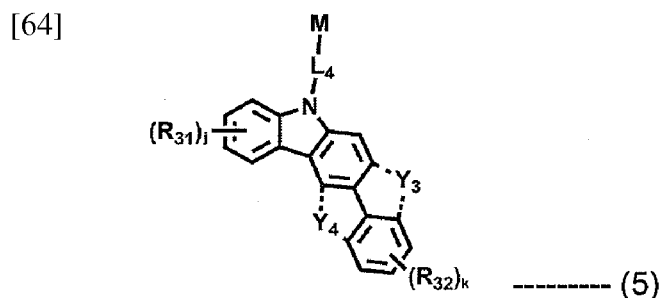
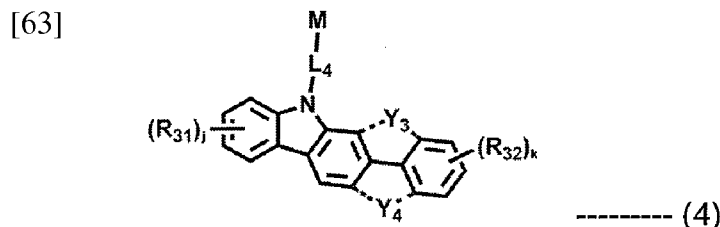
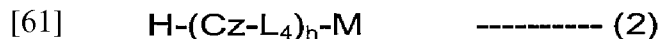
[57] One of the first and second electrodes is an anode, and the other is a cathode. The organic layer comprises a light-emitting layer, and at least one layer selected from the group consisting of a hole injection layer, a hole transport layer, an electron transport layer, an electron injection layer, an interlayer, a hole blocking layer and an electron blocking layer.

[58] The organic electroluminescent compound according to the present invention can be comprised of in the light-emitting layer. Where used in the light-emitting layer, the organic electroluminescent compound according to the present invention can be comprised as a host material.

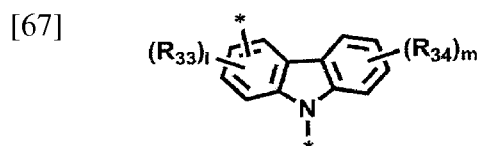
[59] The light-emitting layer can further comprise at least one dopant and, if needed, another compound as a second host material in addition to the organic electroluminescent compound according to the present invention, wherein the ratio of the organic electroluminescent compound according to the present invention (a first host material) to the second host material can be in the range of 1:99 to 99:1.

[60] The second host material can be from any of the known phosphorescent dopants.

Specifically, the phosphorescent dopant selected from the group consisting of the compounds of formula 2 to 6 below is preferable in view of luminous efficiency.



[66] wherein Cz represents the following structure;



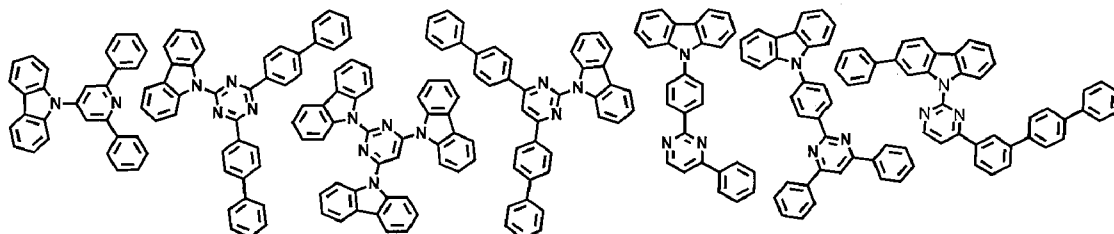
[68] X represents O or S;

[69] R₃₁ to R₃₄ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, or R₃₅R₃₆R₃₇Si-; R₃₅ to R₃₇ each independently represent a substituted or unsubstituted (C1-C30)alkyl group, or a substituted or unsubstituted (C6-C30)aryl group; L₄ represents a single bond, a substituted or unsubstituted (C6-C30)arylene group, or a substituted or unsubstituted 3- to 30-membered heteroarylene group; M represents a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30-membered heteroaryl group; Y₃ and Y₄ represent -O-, -S-, -N(R₄₁)- or -C(R₄₂)(R₄₃)-, provided that Y₃ and Y₄ do not simultaneously exist; R₄₁ to R₄₃ each independently represent a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsub-

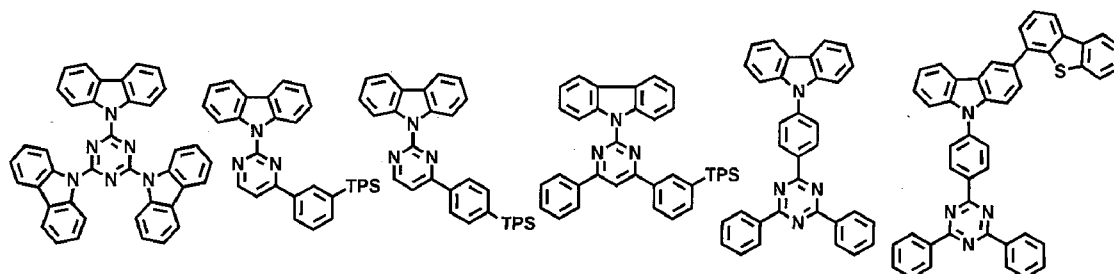
stituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30-membered heteroaryl group, and R_{42} and R_{43} are the same or different; h and i each independently represent an integer of 1 to 3; j, k, l and m each independently represent an integer of 0 to 4; and where h, i, j, k, l or m is an integer of 2 or more, each of (Cz-L₄), each of (Cz), each of R_{31} , each of R_{32} , each of R_{33} or each of R_{34} is the same or different.

[70] Specifically, preferable examples of the second host material are as follows:

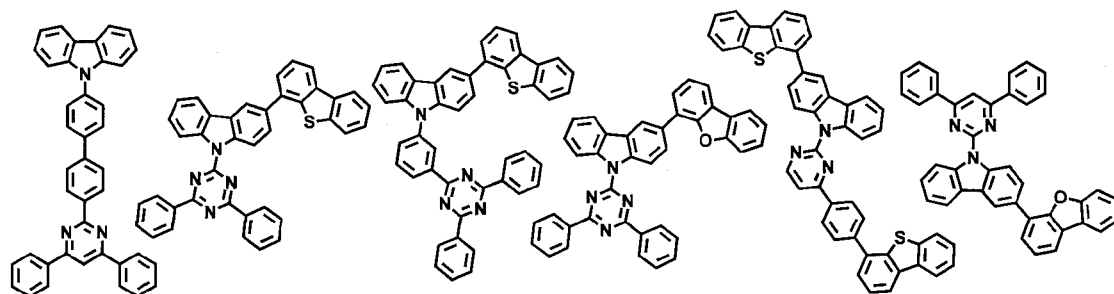
[71]



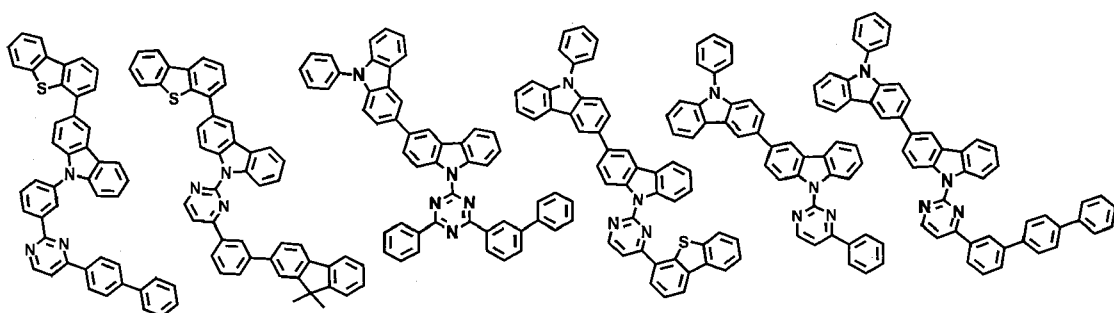
[72]



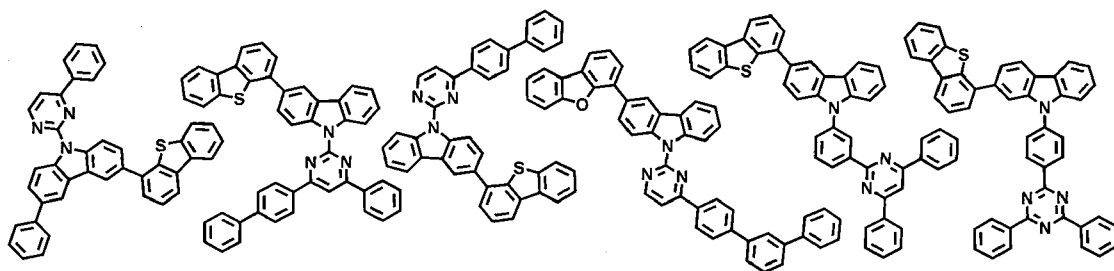
[73]



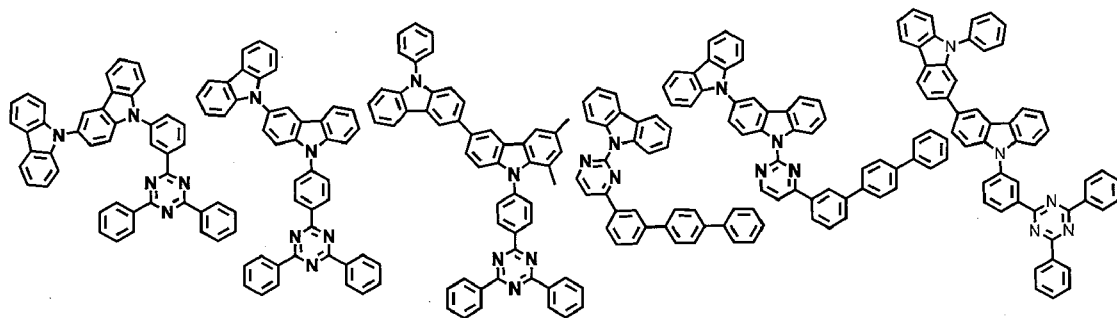
[74]



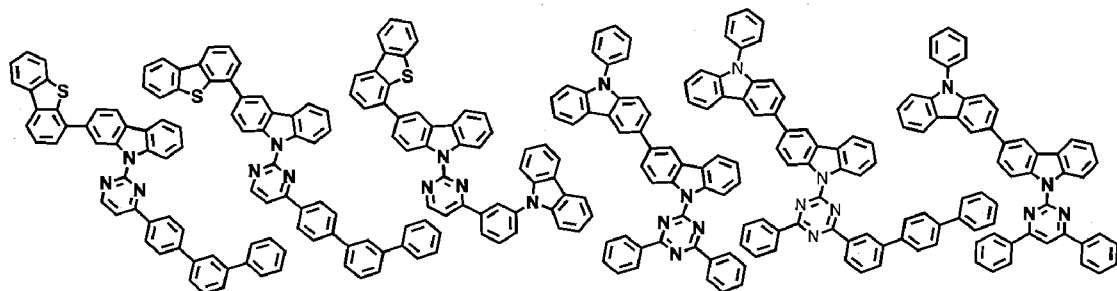
[75]



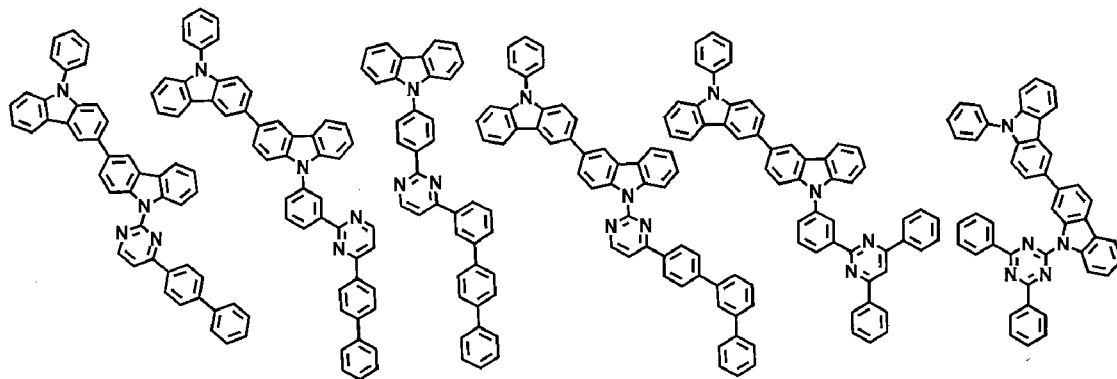
[76]



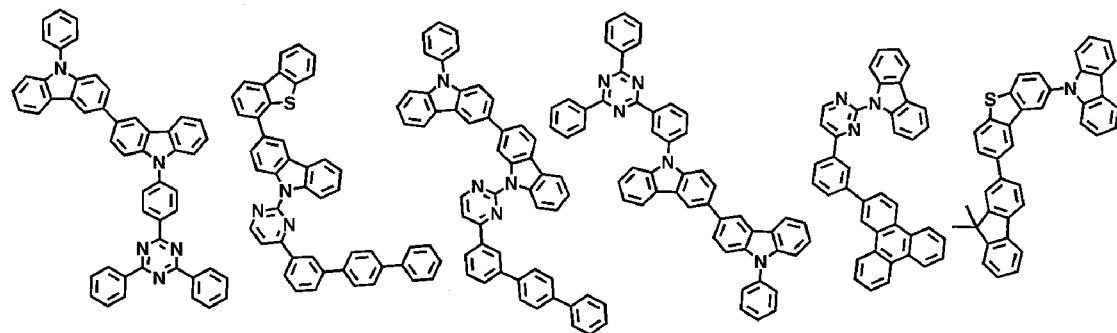
[77]



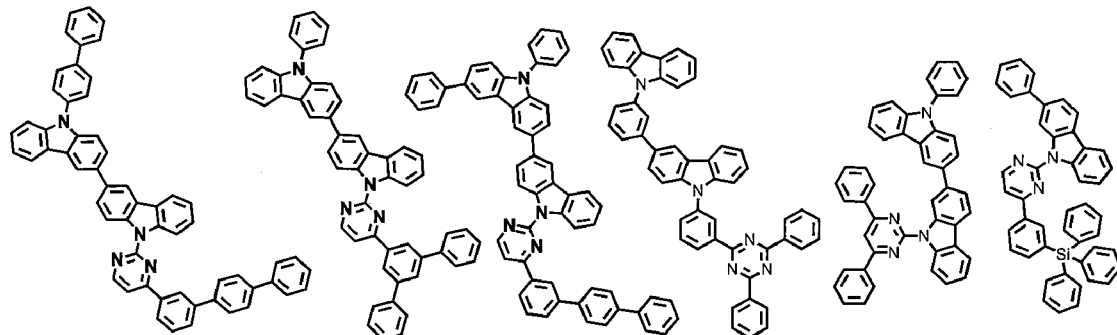
[78]



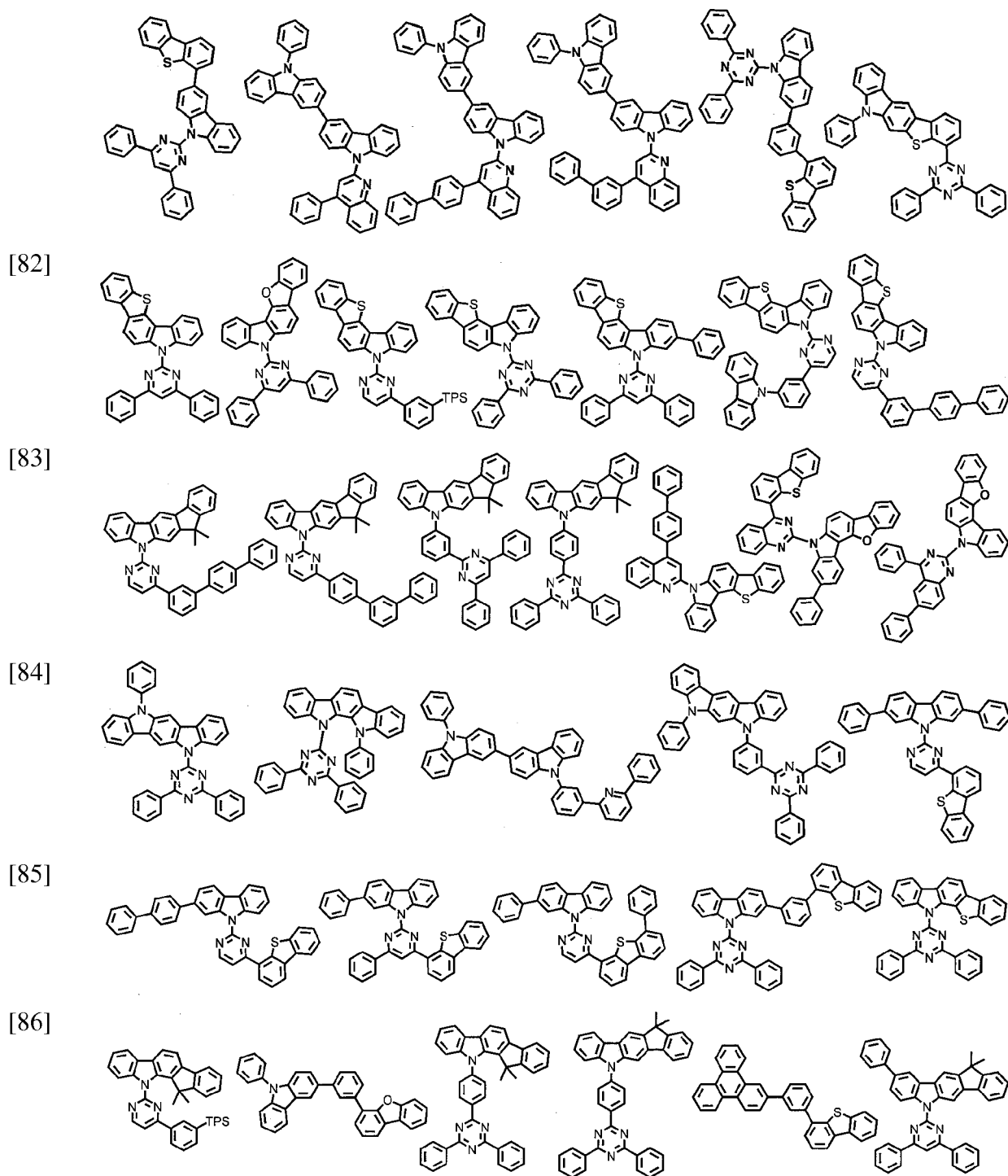
[79]



[80]



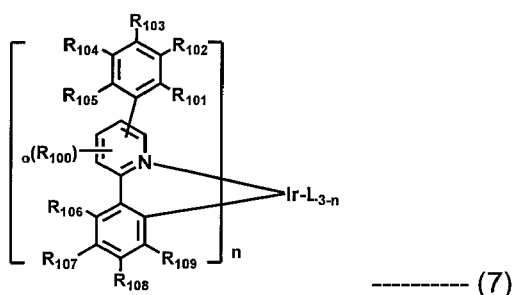
[81]



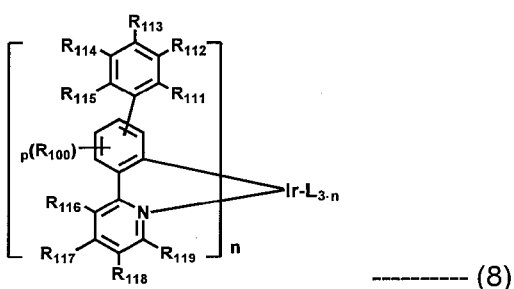
- [87] According to the present invention, the dopant used in the manufacture of the organic electroluminescent device is preferably one or more phosphorescent dopants. The phosphorescent dopant material applied to the electroluminescent device according to the present invention is not limited, but preferably may be selected from complex compounds of iridium, osmium, copper and platinum; more preferably ortho-metallated complex compounds of iridium, osmium, copper and platinum; and even more preferably ortho-metallated iridium complex compounds.

[88] According to the present invention, the dopant comprised in the organic electroluminescent device may be selected from compounds represented by the following formulas 7 to 9.

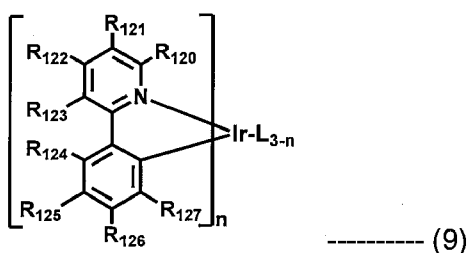
[89]



[90]

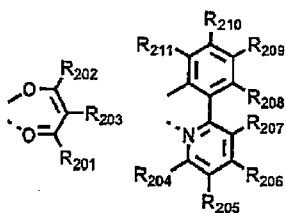


[91]



[92] wherein L is selected from the following structures:

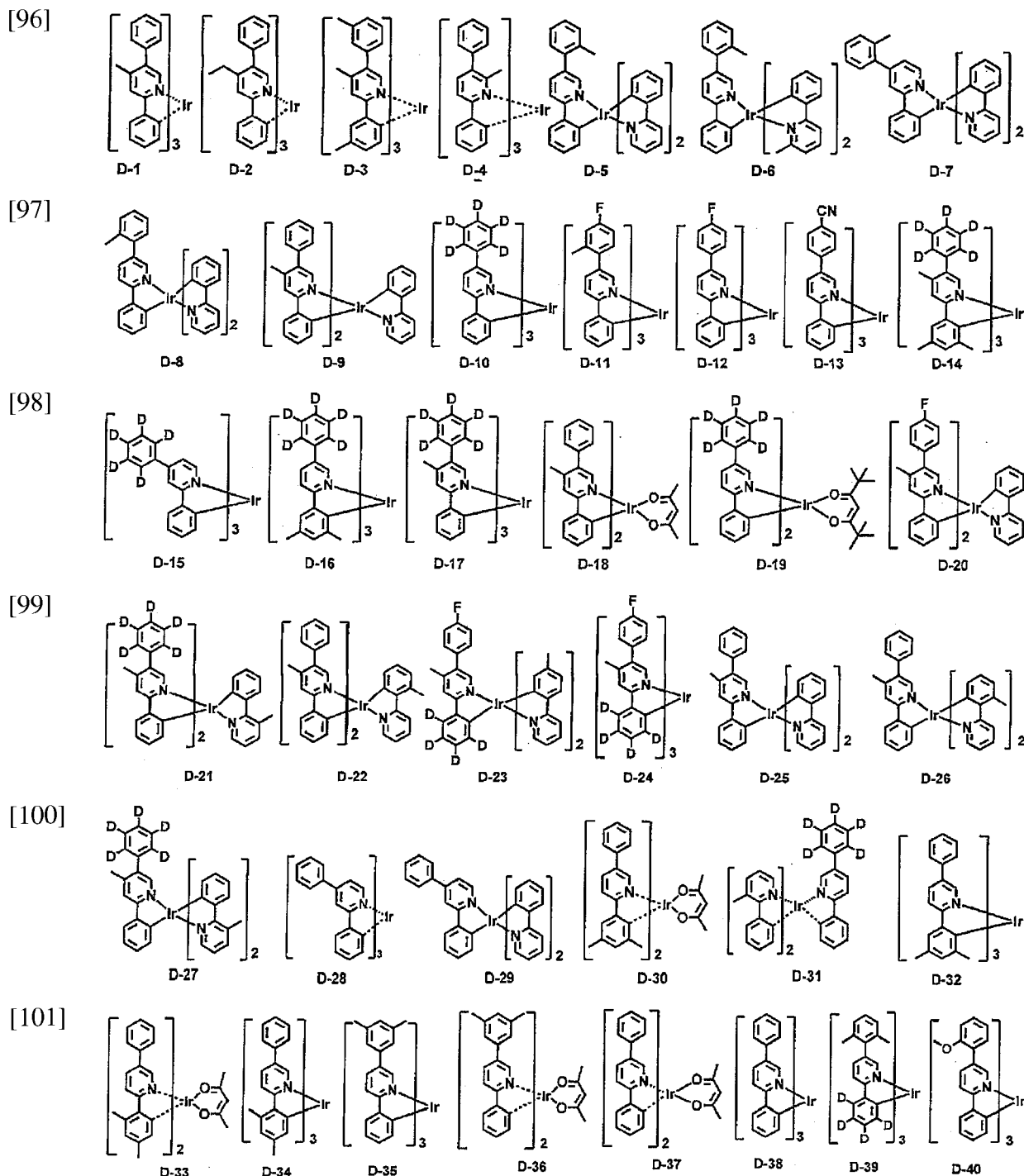
[93]



[94] R_{100} represents hydrogen, a substituted or unsubstituted (C1-C30)alkyl group, or a substituted or unsubstituted (C3-C30)cycloalkyl group; R_{101} to R_{109} , and R_{111} to R_{123} each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group unsubstituted or substituted with halogen(s), a substituted or unsubstituted (C3-C30)cycloalkyl group, a cyano group, or a substituted or unsubstituted (C1-C30)alkoxy group; R_{120} to R_{123} are linked to an adjacent substituent 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 group, or a substituted or unsubstituted (C6-C30)aryl group; where R_{124} to R_{127} are aryl groups, adjacent substituents may be linked to each other to form a fused ring, e.g. fluorene; R_{201} to R_{210} each inde-

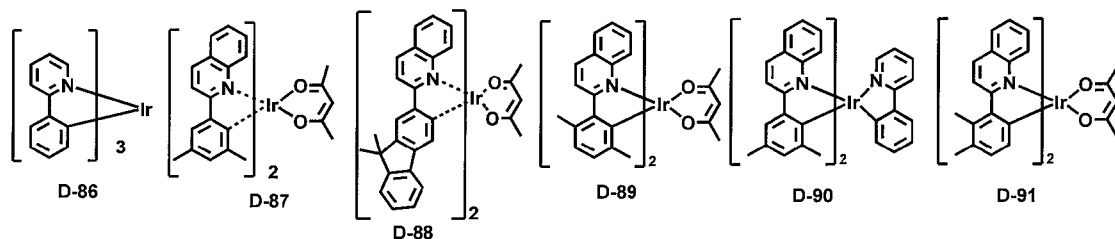
pendently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group unsubstituted or substituted with halogen(s), or a substituted or unsubstituted (C3-C30)cycloalkyl group; o and p each independently represent an integer of 1 to 3; where o or p is an integer of 2 or more, each of R₁₀₀ is the same or different; and n is an integer of 1 to 3.

[95] The phosphorescent dopant materials include the following:

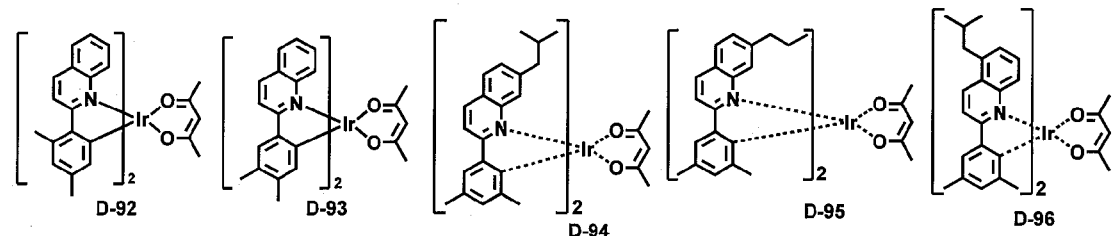


[102]

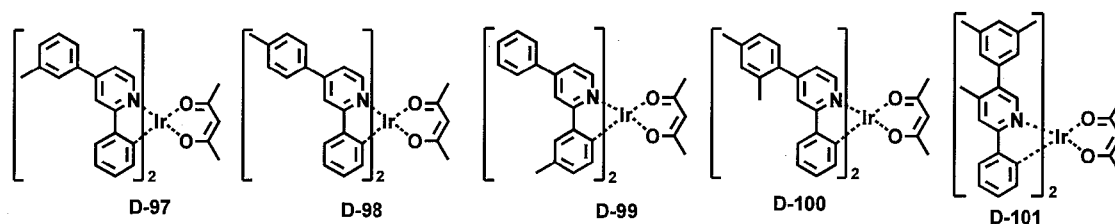




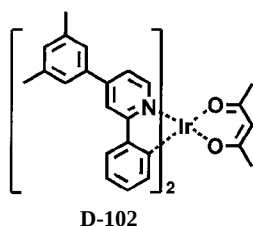
[110]



[111]



[112]

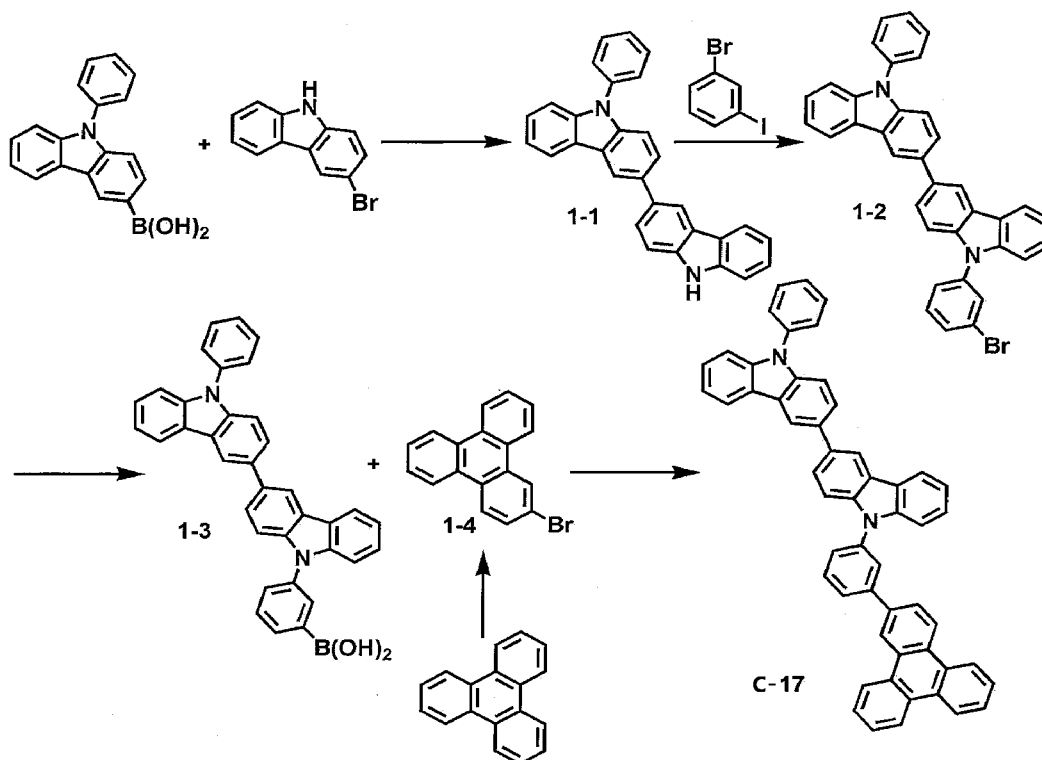


[113] The organic layer of the organic electroluminescent device according to the present invention may further comprise, in addition to the organic electroluminescent compounds represented by formula 1, at least one compound selected from the group consisting of arylamine-based compounds and styrylarylamine-based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

[114] In the organic electroluminescent device according to the present invention, the organic layer may further comprise 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 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.

[115] In addition, the organic electroluminescent device according to 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 organic electroluminescent compound according to the present invention. Also, if needed, a yellow or orange light-emitting layer can be comprised in the device.

- [116] According to the present invention, at least one layer (hereinafter, "a surface layer") of the organic electroluminescent device preferably selected from a chalcogenide layer, a metal halide layer and a metal oxide layer; may be placed on an inner surface(s) of one or both electrode(s). 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 placed on a cathode surface of an electroluminescent medium layer. Such a surface layer provides operation stability for the organic electroluminescent device. Preferably, said chalcogenide includes SiO_x ($1 \leq x < 2$), AlO_x ($1 \leq x < 1.5$), SiON, SiAlON, etc.; said metal halide includes LiF, MgF_2 , CaF_2 , a rare earth metal fluoride, etc.; and said metal oxide includes Cs_2O , Li_2O , MgO, SrO, BaO, CaO, etc.
- [117] Preferably, in the organic electroluminescent device according to the present invention, a mixed region of an electron transport compound and an 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. Further, 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.
- [118] As for the formation of the layers of the organic electroluminescent device according to 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, dipping, flow coating methods can be used.
- [119] When applying a wet film-forming method, a thin film can be formed by dissolving or diffusing materials forming each layer into any suitable solvent such as ethanol, chloroform, tetrahydrofuran, dioxane, etc. The solvent can be any solvent where the materials forming each layer can be dissolved or diffused.
- [120] Hereinafter, the organic electroluminescent compound, the preparation method of the compound, and the luminescent properties of the device comprising the compound of the present invention will be explained in detail with reference to the following examples:
- [121] Example 1: Preparation of compound C-17
- [122]



[123] Preparation of compound 1-1

[124] After mixing (9-phenyl-9H-carbazol-3-yl)boronic acid (36 g, 0.13 mol), 3-bromo-9H-carbazole (25.7 g, 0.11 mol), 2 M K_2CO_3 (125 mL), toluene (500 mL), and ethanol (125 mL), $Pd(PPh_3)_4$ (6.0 g, 0.005 mol) was added to the mixture. Then, the mixture was stirred for 5 hours at 120°C. After completing the reaction, the mixture was cooled to room temperature and distilled water (200 mL) was added, then the mixture was extracted with ethyl acetate (EA) (800 mL). Then, the organic layer was dried with anhydrous $MgSO_4$ to remove the remaining moisture, then distilled under reduced pressure, and then separated through a column to obtain compound 1-1 (38 g, 85 %).

[125] Preparation of compound 1-2

[126] After mixing compound 1-1 (25.4 g, 0.062 mol), 1,3-dibromobenzene (44 g, 0.186 mol), CuI (5.9 g, 0.031 mmol), ethylene diamine (EDA) (4.1 mL, 0.062 mol), K_3PO_4 (40.5 g, 0.186 mol) and toluene (500 mL), the mixture was stirred for 12 hours at 120°C. After completing the reaction, the mixture was washed with distilled water and then extracted with EA. The remaining product was dried with anhydrous $MgSO_4$ to remove the remaining moisture, then distilled under reduced pressure and then separated through a column to obtain compound 1-2 (21.5 g, 61 %).

[127] Preparation of compound 1-3

[128] After putting compound 1-2 (21.3 g, 37.80 mmol) in a flask, and vacuum drying the compound, nitrogen gas was filled and then tetrahydrofuran (THF) (400 mL) was

added. The mixture was cooled to -78°C , then n-buLi (19.6 mL, 49.14 mmol, 2.5 M in hexane) was slowly added to the mixture at the same temperature. After stirring the mixture for 1 hour, $\text{B}(\text{i-pro})_3$ (6.3 mL, 56.70 mmol) was added to the mixture at the same temperature, then the mixture was stirred for 12 hours. After completing the reaction, 1 M HCl (36 mL) was added to the mixture, and after 10 minutes, distilled water was added. Then the mixture was extracted with EA. The organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, then distilled under reduced pressure, and then separated through a column to obtain compound 1-3 (14.5 g, 72 %).

[129] Preparation of compound 1-4

[130] While slowly adding Br_2 (11.2 mL, 0.219 mol) and methylene chloride (MC) (2.1 L) to triphenylene (50 g, 0.219 mol), the mixture was stirred for 12 hours under room temperature. After completing the reaction, distilled water was added, then the mixture was extracted with EA. The organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, then distilled under reduced pressure, and then washed with hexane to obtain compound 1-4 (30 g, 44 %).

[131] Preparation of compound C-17

[132] After mixing compound 1-4 (5 g, 0.016 mol), compound 1-3 (10.3 g, 0.019 mol), $\text{Pd}(\text{PPh}_3)_4$ (940 mg, 0.81 mmol), 2 M K_2CO_3 (25 mL), ethanol (25 mL) and toluene (50 mL), the mixture was stirred for 5 hours at 120°C . After completing the reaction, distilled water was added, and then the mixture was extracted with EA. Then, the organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, then distilled under reduced pressure, and then separated through a column to obtain compound C-17 (2.9 g, 26 %).

[133] MS/EIMS found 710.86; calculated 710.27

[134]

[135] Device Example 1: Production of an PLED device using the compound according to the present invention

[136] An OLED device was produced using the compound according to the present invention. A transparent electrode indium tin oxide (ITO) thin film ($15 \Omega/\text{sq}$) 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. 4,4',4''-tris(N,N-(2-naphthyl)-phenylamino)triphenylamine 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 50 nm on the ITO substrate. Then, N,N'-di(4-biphenyl)-N,N'-di(4-biphenyl)-4,4'-diaminobiphenyl 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, compound **C-17** was introduced into one cell of the vacuum vapor depositing apparatus, as a host material, and 9-(4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-9H-carbazole was introduced into another cell as another host material, and the two materials were evaporated at the same rate, and was used as a host material. Compound **D-41** was introduced as a dopant, and the dopant was evaporated and 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-benzimidazole was introduced into a cell, and evaporated to form an electron transport layer having a thickness of 40 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.

[137] The produced OLED device showed a green emission having a luminance of 1260 cd/m² and a current density of 4.2 mA/cm² at a driving voltage of 5.1 V.

[138] Comparative Example 1: Production of an OLED device using conventional
 [139] electroluminescent compounds

[140] An OLED device was produced in the same manner as in Device Example 1, except that 4,4'-bis(carbazol-9-yl)biphenyl (CBP), instead of the compound according to the present invention, was introduced into the cell of the vacuum vapor depositing apparatus as a host material, and compound **D-41** was used as a dopant; and a hole blocking layer having a thickness of 10 nm was deposited between the light-emitting layer and the electron transport layer by using aluminum(III)bis(2-methyl-8-quinolinato)4-phenylphenolate (BALq).

[141] The produced OLED device showed a green emission having a luminance of 5015 cd/m² and a current density of 24.2 mA/cm² at a driving voltage of 7.9 V.

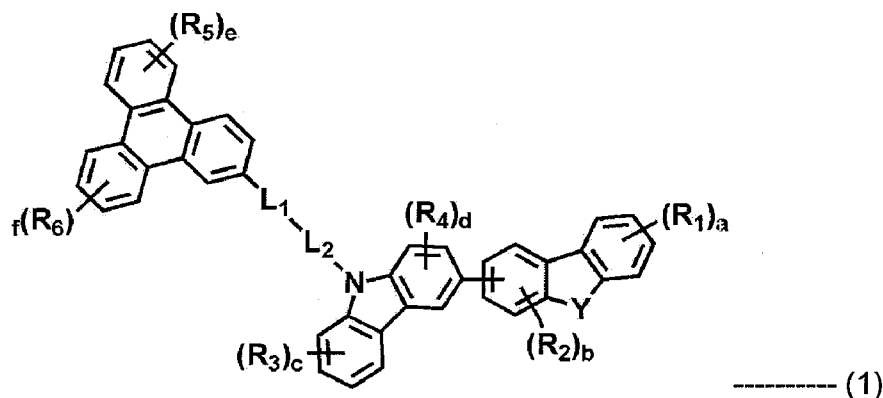
[142] It is verified that the organic electroluminescent compounds according to the present invention have superior luminous efficiency over conventional materials. In addition, the organic electroluminescent devices using the compounds according to the present invention as a light-emitting host material have not only superior luminous efficiency but also can improve the power efficiency of the device and reduce overall power con-

sumption by lowering driving voltages.

Claims

[Claim 1]

An organic electroluminescent compound represented by the following formula 1:



wherein

L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted 3- to 30- membered heteroarylene group, or a substituted or unsubstituted (C6-C30)arylene group, provided that L_1 and L_2 are not simultaneously single bonds;

Y represents -O-, -S-, $-CR_7R_8$ - or $-NR_9$ -;

R_i to R_9 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30- membered heteroaryl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted 5- to 7- membered heterocycloalkyl group, a (C6-C30)cycloalkyl group fused with one or more aromatic rings, $-NR_{10}R_n$, $-SR_{12}$, $-OR_{13}$ or $-SiR_{14}R_{15}R_{16}$;

R_{10} to R_{16} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30- membered heteroaryl group;

a, c, e and f each independently represent an integer of 1 to 4; where a, c, e and f are integers of 2 or more, each of R_i , R_3 , R_5 and R_6 is the same or different;

b and d each independently represent an integer of 1 to 3; where b and d are integers of 2 or more, each of R_2 and R_4 is the same or different; and

the heterocycloalkyl group, the heteroarylene group and the heteroaryl

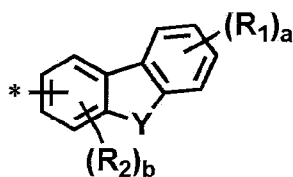
group contain at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

[Claim 2]

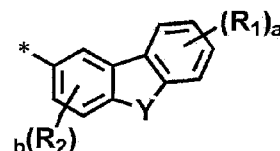
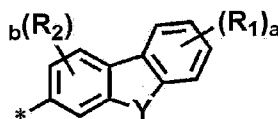
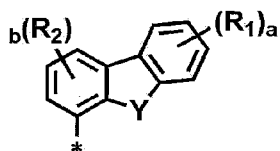
The organic electroluminescent compound according to claim 1, wherein the aryl(ene), heteroaryl(ene), alkyl, cycloalkyl and heterocycloalkyl groups in L_1 , L_2 , and $\frac{3}{4}$ to R_{16} can be further substituted with at least one selected from the group consisting of deuterium; a halogen; a (C1-C30)alkyl group substituted or unsubstituted with a halogen; a (C6-C30)aryl group substituted or unsubstituted with a halogen; a 3- to 30- membered heteroaryl group substituted or unsubstituted with a (C1-C30)alkyl or a (C6-C30)aryl; a (C3-C30)cycloalkyl group; a (C6-C30)cycloalkyl group fused with one or more aromatic rings; a 5- to 7- membered heterocycloalkyl group; $R_a R_b R_c Si-$; a carbazolyl group; $-NR_d R_e$; $-BR_f R_g$; $-PR^h$; $-P(=O)R_j R_k$; a (C6-C30)aryl(C1-C30)alkyl group; a (C1-C30)alkyl(C6-C30)aryl group; a (C1-C30)alkoxy group; and a (C6-C30)aryloxy group, wherein R_a to R_k each independently represent a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30- membered heteroaryl group.

[Claim 3]

The organic electroluminescent compound according to claim 1, wherein the moiety,



from the following structures:



wherein R_i , R_2 , Y, a and b are as defined in claim 1.

[Claim 4]

The organic electroluminescent compound according to claim 1, wherein

L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted 3- to 30- membered heteroarylene group, or a substituted or unsubstituted (C6-C30)arylene group, provided that L_1 and L_2 are not simultaneously single bonds;

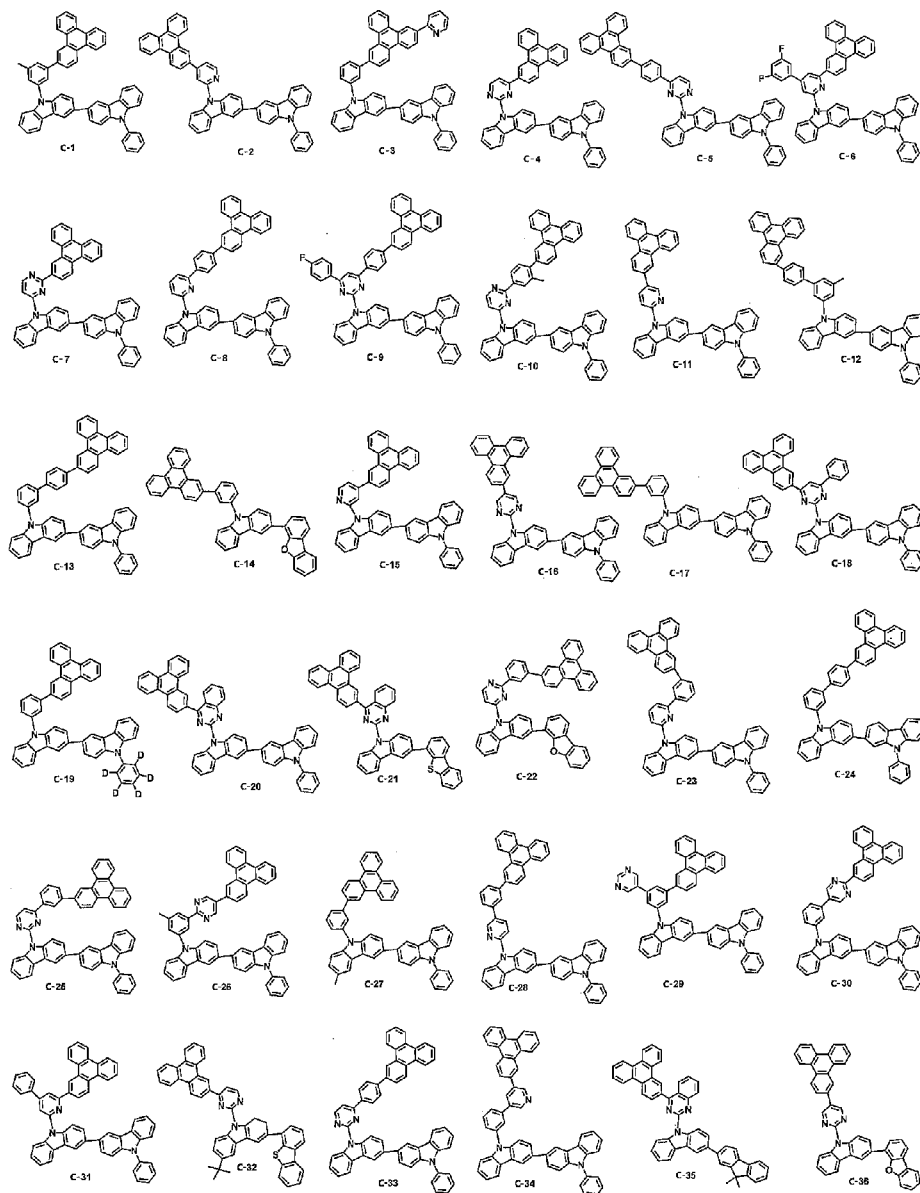
Y represents $-O-$, $-S-$, $-CR_7 R_8-$ or $-NR_9-$;

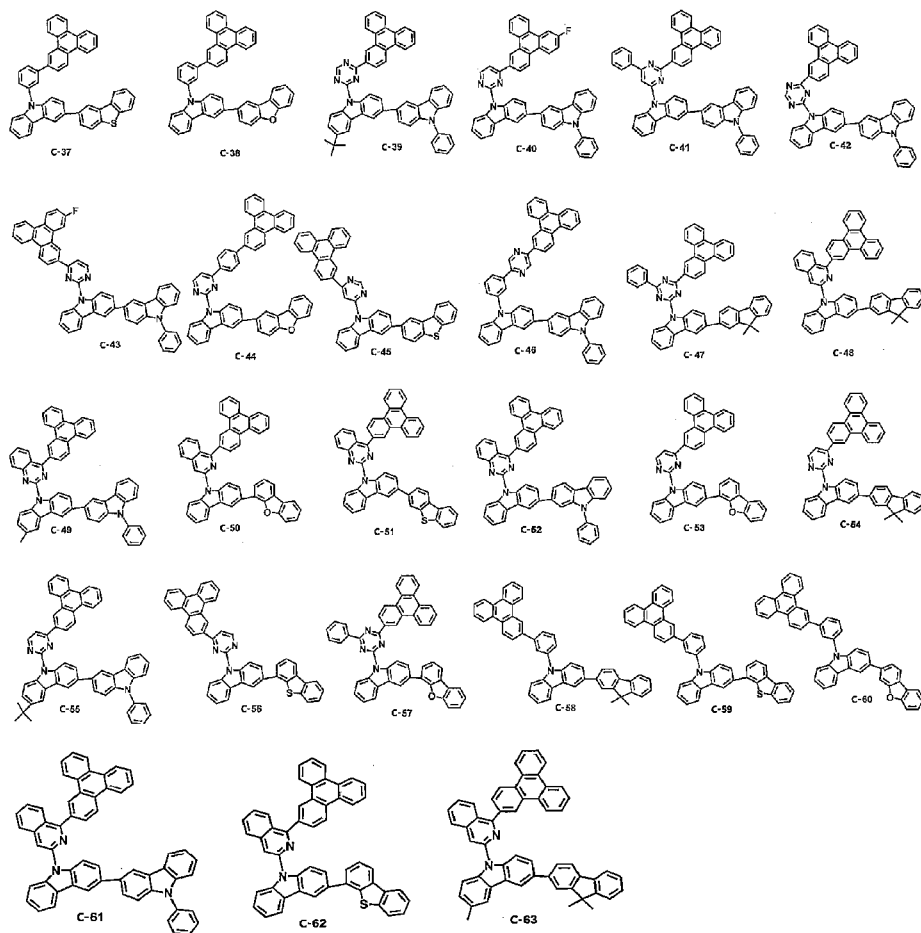
R_i to R_9 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to

30- membered heteroaryl group; and
 the heteroarylene and arylene groups in L_1 and L_2 , and the alkyl, aryl
 and heteroaryl groups in R_1 to R_9 can be further substituted with at least
 one selected from the group consisting of deuterium; a halogen; a
 (C1-C30)alkyl group substituted or unsubstituted with a halogen; a
 (C6-C30)aryl group substituted or unsubstituted with a halogen; a 3- to
 30- membered heteroaryl group substituted or unsubstituted with a
 (C1-C30)alkyl or a (C6-C30)aryl; a (C6-C30)aryl(C1-C30)alkyl group;
 and a (C1-C30)alkyl(C6-C30)aryl group.

[Claim 5]

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





[Claim 6]

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

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	FOR FURTHER ACTION see Form PCT/ISA/220 as well as, where applicable, item 5 below.	
International application No. PCT/KR2012/010412	International filing date (<i>day/month/year</i>) 04 December 2012	(Earliest) Priority Date (<i>day/month/year</i>) 05 December 2011

Applicant

ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.

This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This international search report consists of a total of **4** sheets.

☐

It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

a. With regard to the **language**, the international search was carried out on the basis of:

☒

The international application in the language in which it was filed.

☐

A translation of the international application into , which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).

b. ☐ This international search report has been established taking into account the **rectification of an obvious mistake** authorized by or notified to this Authority under Rule 91 (Rule **43.6bis(a)**).

c. ☒ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☐ **Certain claims were found unsearchable** (See Box No. II).

3. ☐ **Unity of invention is lacking** (See Box No. III).

4. With regard to the **title**,

☒

the text is approved as submitted by the applicant.

☐

the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☒

the text is approved as submitted by the applicant.

☐

the text has been established, according to Rule 38.2, by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the **drawings**,

a. the figure of the **drawings** to be published with the abstract is Figure No.

☐

as suggested by the applicant.

☐

as selected by this Authority, because the applicant failed to suggest a figure.

☐

as selected by this Authority, because this figure better characterizes the invention.

b. ☐ none of the figures is to be published with the abstract.

A. CLASSIFICATION OF SUBJECT MATTER

C09K 11/06 (2006.01) H01L 51/50 (2006.01) H01L 27/32 (2006.01) H05B 33/14 (2006.01) C07D 333/76 (2006.01)
 C07D 209/82 (2006.01) C07D 307/91 (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)

REGISTRY, CAPLUS: Substructure search based on present 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

* Special categories of cited documents:	
"A" 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

1 February 2013

Date of mailing of the international search report

01 February 2013

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INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2012/010412
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 201 1/139055 A2 (CHEIL INDUSTRIES INC.) 10 November 201 1 Abstract & compounds 41 & 44 on page 10	1-4, 6
P,X	WO 2012/161382 A1 (CHEIL INDUSTRIES INC.) 29 November 2012 Abstract & compounds associated with CAS Registry Numbers RN 1415 113-75-5; RN 14151 13-76-6; RN 14 15113-77-7; RN 1415113-78-8 & RN 14151 13-79-9	1-4, 6
P,X	KR 10-2012-01 16282 A (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 22 October 2012 Abstract & compound associated with CAS Registry Number RN 1404058-53-2	1-6
P,X	WO 2012/108388 A1 (IDEMITSU KOSAN CO. LTD.) 16 August 2012 Abstract & compound associated with CAS Registry Number RN 1392844-59-5	1-4, 6
P,X	WO 2012/077902 A2 (CHEIL INDUSTRIES INC.) 14 June 20 12 Abstract & compounds associated with CAS Registry Numbers RN 1361094-73-6 & RN 1361094-74-7	1-4, 6
P,X	WO 2012/023947 A1 (UNIVERSAL DISPLAY CORPORATION) 23 February 2012 Abstract & compounds associated with CAS Registry Numbers RN 1346668-75-4; RN 1360632-92-3; RN 1360632-93-4; RN 1360632-94-5; RN 1360632-95-6; RN 1360632-96-7; RN 1360632-97-8; RN 1360632-98-9; RN 1360632-99-0; RN 1360633-00-6; RN 1360633-01-7; RN 1360633-02-8; RN 1360633-03-9; RN 1360633-04-0; RN 1360633-05-1; RN 1360633-77-7; RN 1360633-78-8; RN 1360633-79-9; RN 1360633-80-2; RN 1360633-81-3; RN 1360633-82-4; RN 1360633-83-5; RN 1360633-84-6; RN 1360633-85-7; RN 1360633-86-8; RN 1360633-87-9; RN 1360633-88-0; RN 1360633-89-1; RN 1360633-90-4 & RN 1360633-91-5	1-4, 6

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR2012/010412

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	
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		WO 2012161382 A1	29 Nov 2012
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WO 2012/108388 A1	16 Aug 2012	TW 201235441 A	01 Sep 2012
		TW 201237140 A	16 Sep 2012
		US 2012223295 A1	06 Sep 2012
		WO 2012108388 A1	16 Aug 2012
		WO 2012108389 A1	16 Aug 2012
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		WO 2012077902 A2	14 Jun 20 12
WO 2012/023947 A1	23 Feb 2012	TW 201226407 A	01 Jul 2012
		WO 2012023947 A1	23 Feb 2012

End of Annex

Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001.

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