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

(57) Abstract: The present invention relates to a novel organic electroluminescent compound and an organic electroluminescent device comprising the same. The organic electroluminescent compound according to the present invention can be used in a light-emitting layer and has excellent luminescent efficiency; and an organic electroluminescent device comprising the organic electroluminescent compounds of the present invention has long lifespan, and improved current efficiency and power efficiency.



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Description

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

Technical Field

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

Background Art

- [2] An electroluminescent (EL) device is a self-light-emitting device with the advantage of providing 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 (see Appl. Phys. Lett. 51, 913, 1987).
- [3] The organic EL device generally comprises an anode, a cathode, and an organic layer formed between the two electrodes and emits light by the recombination of holes injected from an anode and electrons injected from a cathode. The organic layer of the organic EL device may be composed of a hole injection layer (HIL), a hole transport layer (HTL), a light-emitting layer (EML), an electron transport layer (ETL), an electron injection layer (EIL), etc.; the materials used in the organic layer can be classified into a hole injection material, a hole transport material, a light-emitting material, an electron transport material, an electron injection material, etc.
- [4] The most important factor determining luminescent efficiency in the organic EL device is the light-emitting material. The light-emitting material is required to have the following features: high quantum efficiency, high movement degree of an electron and a hole, formability of a uniform light-emitting material layer, and stability. The light-emitting material is classified into blue light-emitting materials, green light-emitting materials, or red light-emitting materials according to the light-emitting color, and further includes yellow light-emitting materials or orange light-emitting materials. The light-emitting materials are classified into fluorescent materials (singlet excited state) and phosphorescent materials (triplet excited state) according to the excited state. The fluorescent materials were initially used in an organic EL device. However, phosphorescent materials have efficiency for changing electricity into light (luminescent efficiency) by four (4) times over fluorescent materials, reduce consumption power, and increase lifespan. Thus, development of phosphorescent materials is widely being conducted.
- [5] Until now, 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)picolinateiridium (Firpic) as red, green and blue materials, respectively.

- [6] A mixed system of a dopant/host material can be used as a light-emitting material to improve color purity, luminescent efficiency, and stability. If the dopant/host material system is used, the selection of the host material is important since the host material greatly influences the efficiency and performance of a light-emitting device. In conventional technique, 4,4'-N,N'-dicarbazol-biphenyl (CBP) is the most widely known phosphorescent host material. Pioneer (Japan) et al., currently developed a high performance organic EL device by employing bathocuproine (BCP), aluminum(III) bis(2-methyl-8-quinolinato)(4-phenylphenolate) (BALq), etc., which were used in a hole blocking layer, as host materials.
- [7] Although these phosphorescent host materials provide good light-emitting characteristics, they have the following disadvantages: (1) Due to their low glass transition temperatures and poor thermal stability, their 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 the power efficiency is inversely proportional to voltage. An organic EL device comprising phosphorescent host materials provides higher current efficiency (cd/A) and has a higher driving voltage than one comprising fluorescent host materials. Thus, the organic EL device using conventional phosphorescent host materials has no advantage in terms of power efficiency (lm/W). (3) Furthermore, the operating lifespan and luminescent efficiency of the organic EL device are not satisfactory.
- [8] 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., have been used as hole injection and transport materials in the organic EL device. However, the organic EL device comprising the materials has low quantum efficiency and a short lifespan, because, when the organic EL device is driven at a high current, thermal stress is generated between an anode and a hole injection layer, thereby rapidly reducing the lifespan of the device. Furthermore, movement of holes is great in organic materials used in a hole injection layer, and thus the hole-electron charge balance is broken and quantum efficiency (cd/A) is reduced.
- [9] Thus, in order to embody excellent properties of the organic EL device, the materials constituting the organic layers in the device, in particular the host or dopant, should be suitably selected. Meanwhile, Korean Patent No. 10-1082144 discloses the organic

electroluminescent compound comprising a specific fused heterocyclic structure as a host. However, the organic EL devices comprising the compound recited in the above publication still do not satisfy power efficiency, luminescent efficiency, lifespan, etc. Thus, the present inventors have tried to find organic electroluminescent compounds that can provide the organic EL device with properties superior to the compounds recited in the above publication and have found the compound providing the device with high luminescent efficiency and excellent device properties.

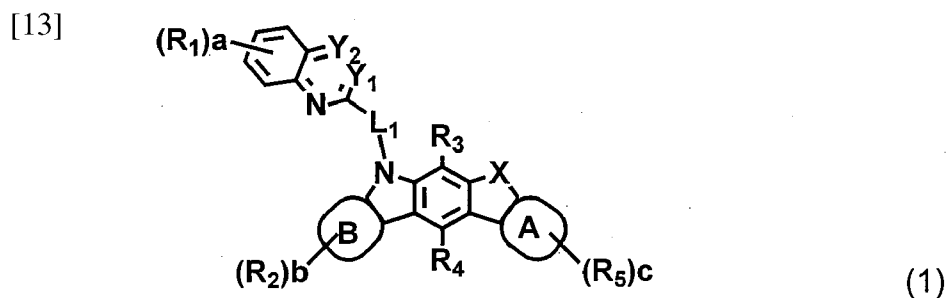
Disclosure of Invention

Technical Problem

- [10] The object of the present invention is to provide organic electroluminescent compounds having high luminescent efficiency and to provide an organic EL device comprising the organic electroluminescent compounds and having long driving lifespan, and improved power efficiency and current efficiency.

Solution to Problem

- [11] Korean Patent No. 10-1082144 discloses the compound wherein a HOMO site is directly connected to a LUMO site without a linker. The compound is in the form of an indolocarbazole structure, not a benzoindolocarbazole, and shows a remarkable characteristic difference as a red host. Generally, the structures of triazine, pyridine, pyrimidine, quinoline, etc., bonded to indolocarbazole are used as a phosphorescent green host, and the structures of quinazoline, quinoxaline, etc., bonded to benzoindolocarbazole have excellent features as only a phosphorescent red host. Thus, the red light-emitting device comprising the material of the present invention provides improved properties compared with the device comprising an indolocarbazole derivative having no phenyl as a linker which is supplied by other companies. Furthermore, the compound of the present invention, comprising benzoindolocarbazole bonded to quinoxaline, quinazoline, etc., by a linker, is structurally different from the compounds supplied by other companies in the chemical structure and has maximized efficiency properties.
- [12] The present inventors found that the above objective can be achieved by a compound represented by the following formula 1:



- [14] wherein
- [15] L_1 represents a substituted or unsubstituted (C6-C30)arylene group;
- [16] Ring A or B represents benzene or naphthalene, with the proviso that A and B do not simultaneously represent benzene;
- [17] X represents NR_6 , $-CR_7R_8$, O, or S;
- [18] Y_1 and Y_2 each independently represent $-CR_9-$ or $-N-$, and if Y_1 represents $-N-$, Y_2 represents $-CR_9-$, and if Y_1 represents $-CR_9-$, Y_2 represents $-N-$;
- [19] R_1 to R_5 each independently represent hydrogen, deuterium, a halogen, a cyano group, a carboxyl group, a nitro group, a hydroxyl group, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted (C3-C30)cycloalkenyl group, a substituted or unsubstituted 3- to 7-membered heterocycloalkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted (C6-C30)arylamine group, $-NR_{10}R_{11}$, or $-SiR_{12}R_{13}R_{14}$;
- [20] R_1 , R_2 and R_5 are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring;
- [21] R_6 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 substituted or unsubstituted (C6-C30)aryl(C1-C30)alkyl group, $-NR_{10}R_{11}$, $-SiR_{12}R_{13}R_{14}$, a cyano group, a nitro group, or a hydroxyl group;
- [22] R_{10} and R_{11} 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;
- [23] R_{12} to R_{14} 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 5- to 7-membered heterocycloalkyl group, or a substituted or unsubstituted (C3-C30)cycloalkyl group; or are linked to an adjacent substituent(s) to form a substituted or unsubstituted mono- or polycyclic (C3-C30) alicyclic or aromatic ring;
- [24] the carbon atom(s) of the alicyclic or aromatic ring may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;
- [25] the heteroaryl(ene) and heterocycloalkyl groups contain at least one hetero atom selected from B, N, O, S, $P(=O)$, Si, and P;
- [26] a represents an integer of 1 to 4; where a is an integer of 2 or more, each R_1 is the

same or different;

[27] if A or B represents benzene, b or c represents an integer of 1 to 4; and

[28] if A or B represents naphthalene, b or c represents an integer of 1 to 6; where b or c is an integer of 2 or more, each R₂ or each R₅ is the same or different.

Advantageous Effects of Invention

[29] The organic electroluminescent compounds according to the present invention have high luminescent efficiency and an excellent lifespan property, and thus the organic EL device comprising the compounds has long driving lifespan, and good current efficiency and power efficiency.

Mode for the Invention

[30] 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.

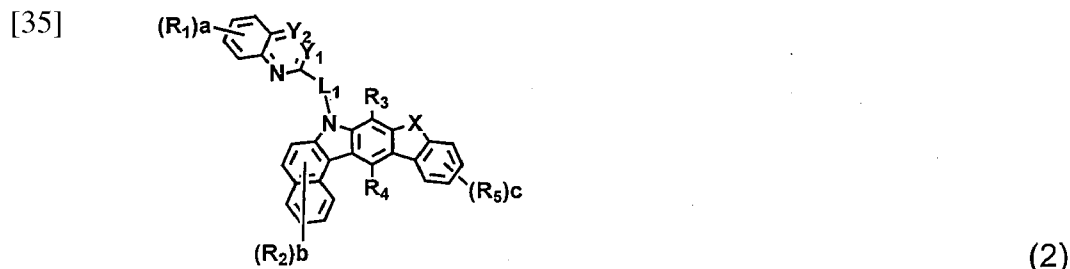
[31] The present invention relates to an organic electroluminescent compound represented by formula 1, an organic electroluminescent material comprising the organic electroluminescent compound, and an organic EL device comprising the material.

[32] Herein, “(C1-C30)alkyl(ene)” is meant to be a linear or branched alkyl(ene) having 1 to 30 carbon atoms, in which the number of carbon atoms is preferably 1 to 20, more preferably 1 to 10, and includes methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, etc. “(C2-C30)alkenyl” is meant to be a linear or branched alkenyl having 2 to 30 carbon atoms, in which the number of carbon atoms is preferably 2 to 20, more preferably 2 to 10, and includes vinyl, 1-propenyl, 2-propenyl, 1-butenyl, 2-butenyl, 3-butenyl, 2-methylbut-2-enyl, etc. “(C2-C30)alkynyl” is a linear or branched alkynyl having 2 to 30 carbon atoms, in which the number of carbon atoms is preferably 2 to 20, more preferably 2 to 10, and includes ethynyl, 1-propynyl, 2-propynyl, 1-butylnyl, 2-butylnyl, 3-butylnyl, 1-methylpent-2-ynyl, etc. “(C3-C30)cycloalkyl” is a mono- or polycyclic hydrocarbon having 3 to 30 carbon atoms, in which the number of carbon atoms is preferably 3 to 20, more preferably 3 to 7, and includes cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, etc. “3- 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 3 to 7, preferably 5 to 7 ring backbone atoms, and includes tetrahydrofuran, pyrrolidine, thiolan, tetrahydropyran, etc. “(C6-C30)aryl(ene)” is a monocyclic or fused ring derived from an aromatic hydrocarbon having 6 to 30 carbon atoms, in which the number of carbon atoms is preferably 6 to 20, more preferably 6 to 15, and includes phenyl, biphenyl, terphenyl, naphthyl, fluorenyl, 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; has preferably 3 to 20, more preferably 3 to 15 ring backbone atoms; 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. "Halogen" includes F, Cl, Br, and I.

- [33] 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. Substituents of the substituted (C1-C30)alkyl group, the substituted (C3-C30)cycloalkyl group, the substituted (C3-C30)cycloalkenyl group, the substituted 3- to 7-membered heterocycloalkyl group, the substituted (C6-C30)aryl(ene) group, the substituted 5- to 30-membered heteroaryl(ene) group, the substituted (C6-C30)aryl(C1-C30)alkyl group, and the substituted mono- or polycyclic (C3-C30) alicyclic or aromatic ring in L_1 , X, Y_1 , Y_2 , and R_1 to R_5 of the formulae are each independently at least one selected from the group consisting of deuterium; a halogen; a cyano group; a carboxyl group; a nitro group; a hydroxyl group; a (C1-C30)alkyl group; a halo(C1-C30)alkyl group; a (C2-C30)alkenyl group; a (C2-C30)alkynyl group; a (C1-C30)alkoxy group; a (C1-C30)alkylthio group; a (C3-C30)cycloalkyl group; a (C3-C30)cycloalkenyl group; a 3- to 7-membered heterocycloalkyl group; a (C6-C30)aryloxy group; a (C6-C30)arylthio group; a 3- to 30-membered heteroaryl group which is unsubstituted or substituted with a (C6-C30)aryl group; a (C6-C30)aryl group which is unsubstituted or substituted with a 3- to 30-membered heteroaryl group; a tri(C1-C30)alkylsilyl group; a tri(C6-C30)arylsilyl group; a di(C1-C30)alkyl(C6-C30)arylsilyl group; a (C1-C30)alkyldi(C6-C30)arylsilyl group; an amino group; a mono- or di(C1-C30)alkylamino group; a mono- or di(C6-C30)arylamino group; a (C1-C30)alkyl(C6-C30)arylamino group; a (C1-C30)alkylcarbonyl group; a (C1-C30)alkoxycarbonyl group; a (C6-C30)arylcarbonyl group; a di(C6-C30)arylboronyl group; a di(C1-C30)alkylboronyl group; a (C1-C30)alkyl(C6-C30)arylboronyl group; a (C6-C30)aryl(C1-C30)alkyl group; and a (C1-C30)alkyl(C6-C30)aryl group.

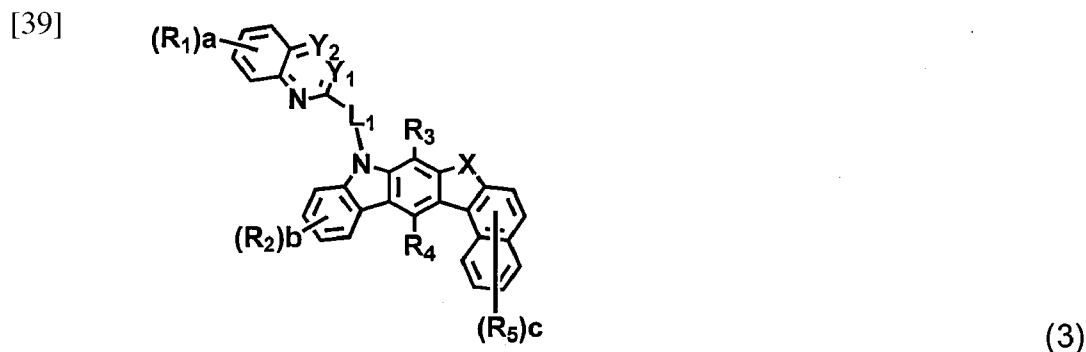
[34] The compound of formula 1 is represented by the following formula 2:



[36] wherein

[37] L_1 , X , Y_1 , Y_2 , R_1 to R_5 , a , b , and c are as defined in formula 1.

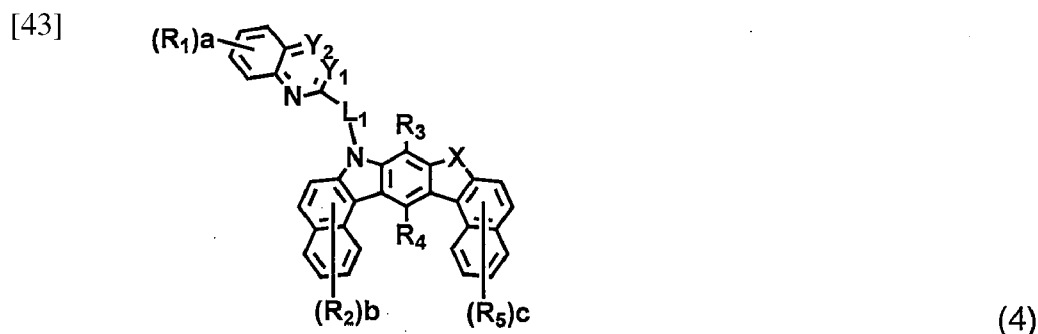
[38] The compound of formula 1 is represented by the following formula 3:



[40] wherein

[41] L_1 , X , Y_1 , Y_2 , R_1 to R_5 , a , b , and c are as defined in formula 1.

[42] The compound of formula 1 is represented by the following formula 4:



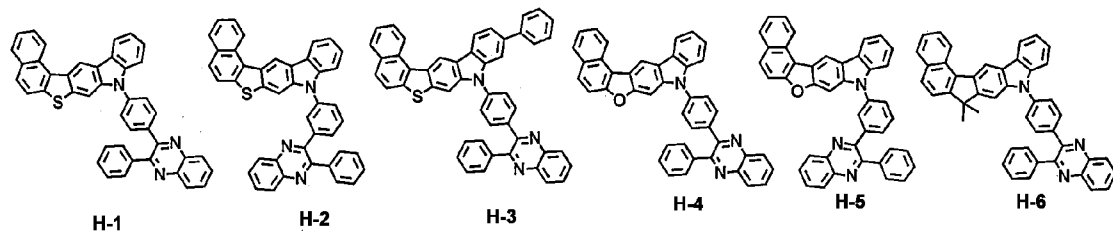
[44] wherein

[45] L_1 , X , Y_1 , Y_2 , R_1 to R_5 , a , b , and c are as defined in formula 1.

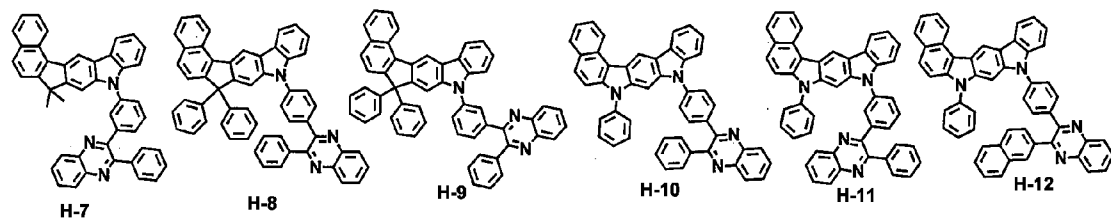
[46] In the compound of formula 1, 2, 3, and 4, preferably, L_1 represents a (C6-C20)arylene group which is unsubstituted or substituted by a (C1-C6)alkyl group; R_1 to R_5 each independently represent hydrogen, or a substituted or unsubstituted (C6-C20)aryl group; R_6 to R_9 each independently represent a substituted or unsubstituted (C1-C6)alkyl group, or a substituted or unsubstituted (C6-C20)aryl group.

[47] The compound of formula 1 can be selected from the group consisting of the following compounds:

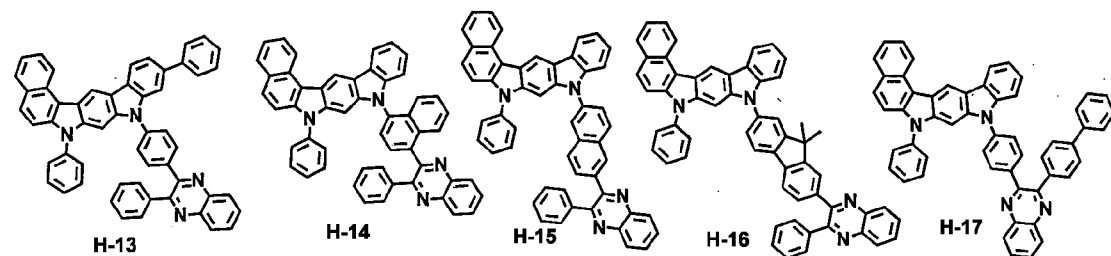
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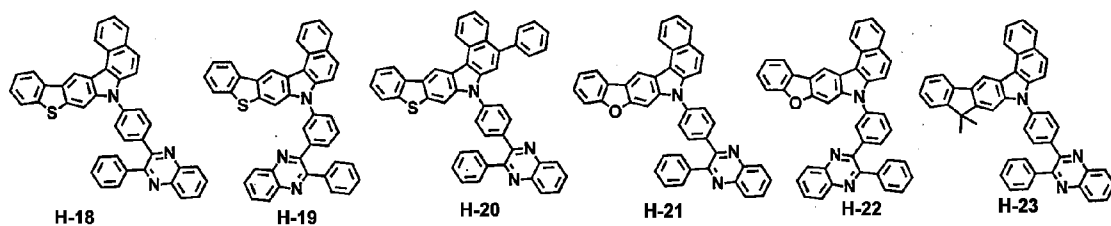
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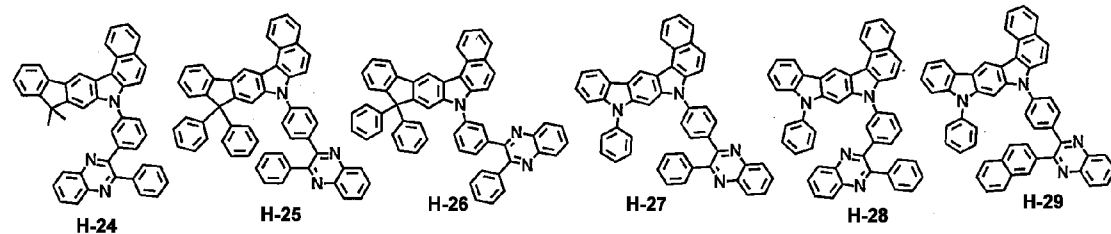
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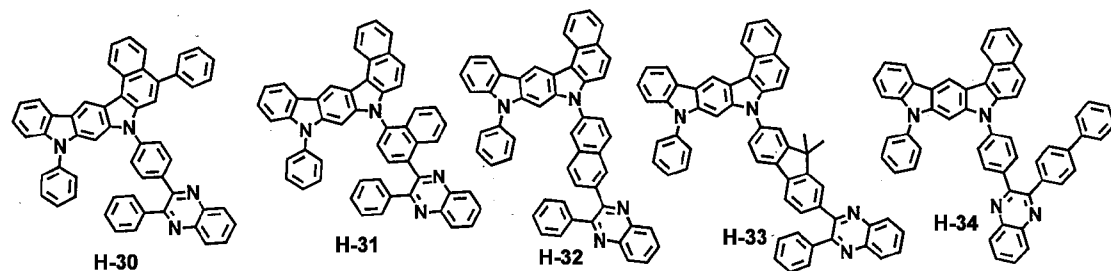
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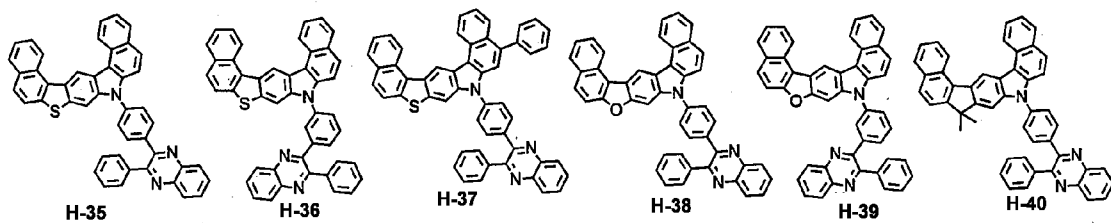
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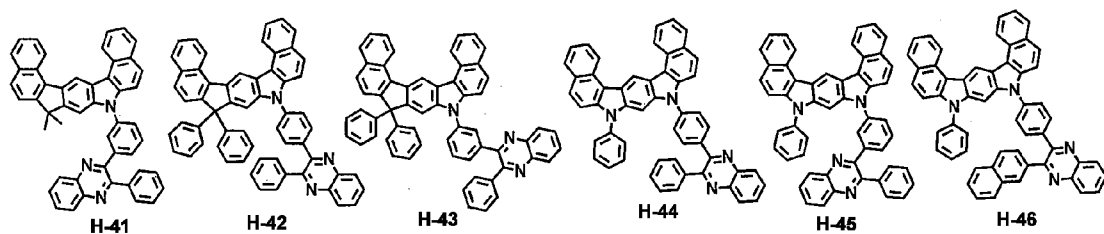
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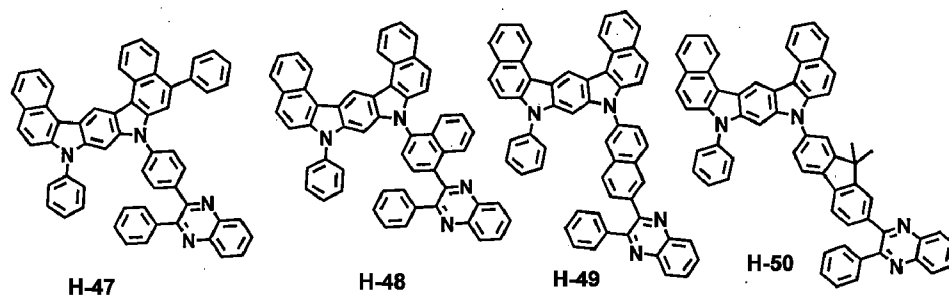
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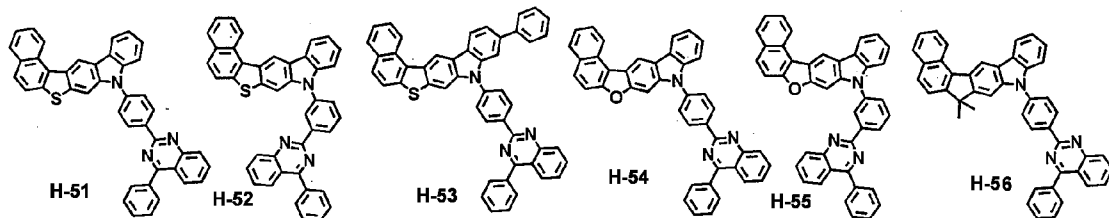
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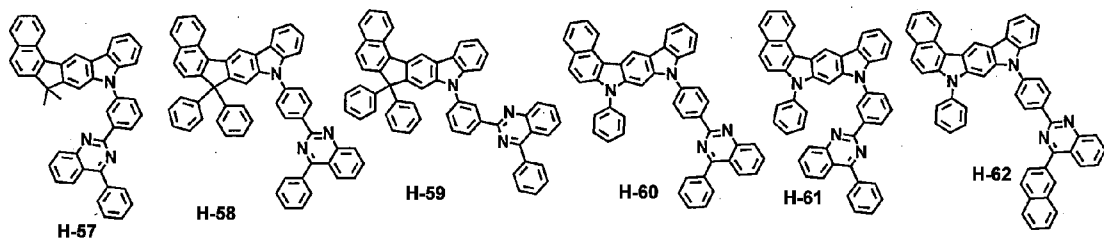
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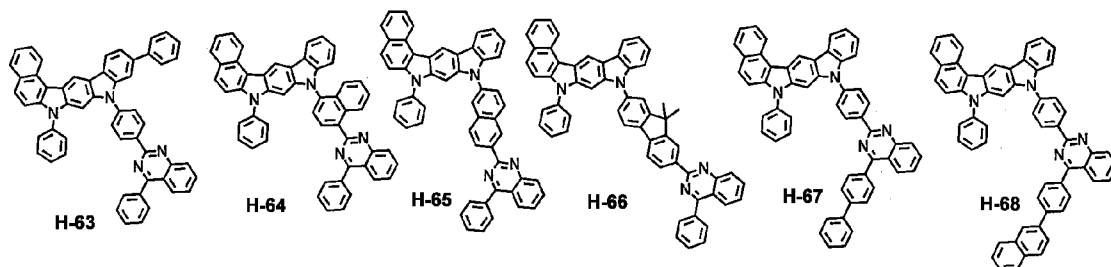
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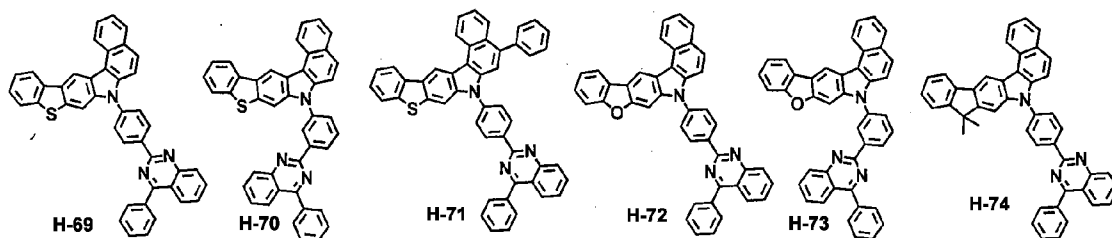
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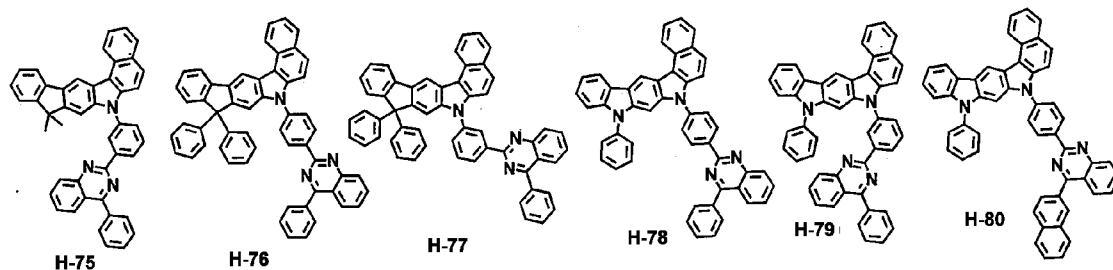
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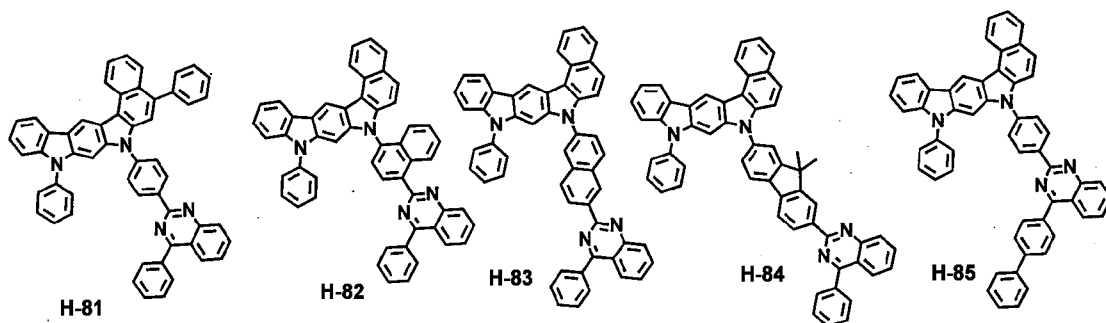
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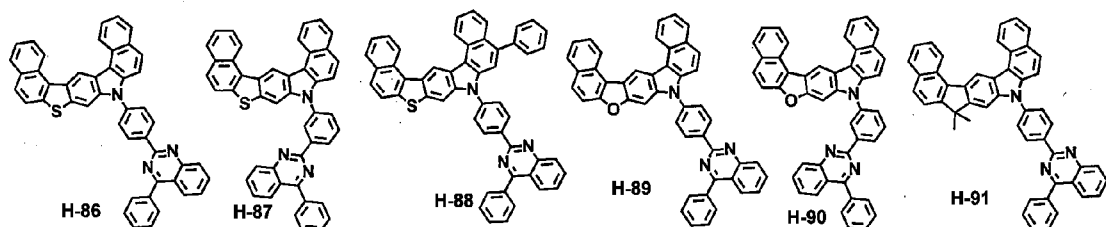
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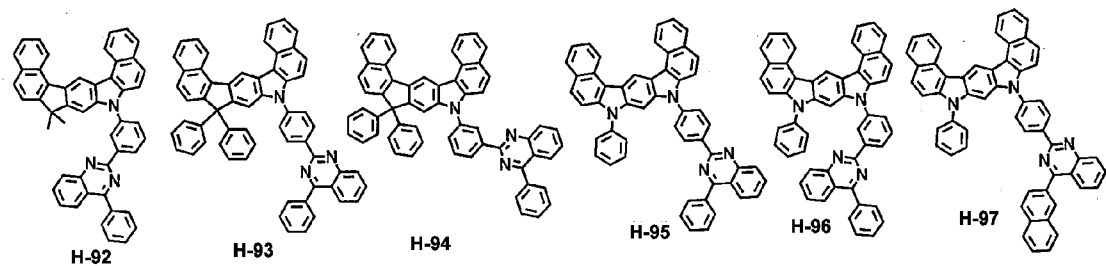
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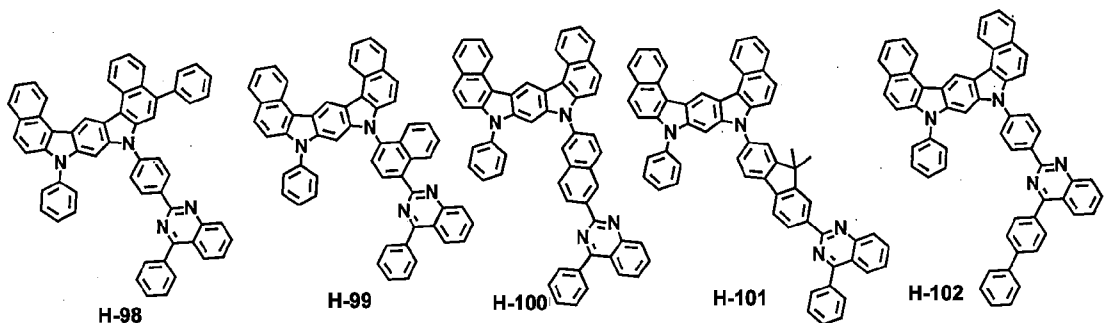
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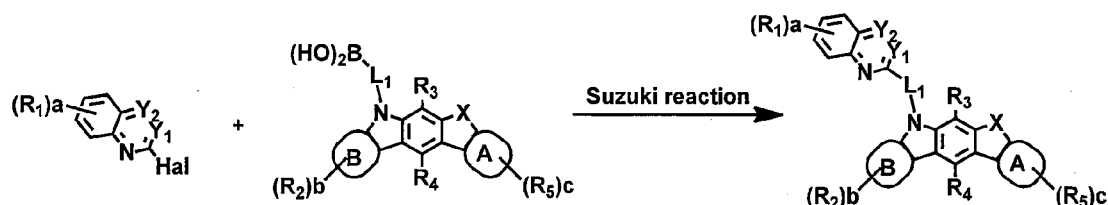
[65]



[66] The organic electroluminescent compounds according to the present invention can be prepared by known methods to one skilled in the art, and can be prepared, for example, according to the following reaction scheme 1:

[67] Reaction scheme 1

[68]



[69] The present invention further provides an organic electroluminescent material comprising the organic electroluminescent compound of formula 1, and an organic electroluminescent device comprising the material. The material can be comprised of the organic electroluminescent compound of the present invention alone, or can further include conventional materials generally included in organic electroluminescent materials.

[70] The organic electroluminescent device according to the present invention may comprise a first electrode, a second electrode, and at least one organic layer between the first and second electrodes, wherein the organic layer comprises at least one organic electroluminescent compound of formula 1.

[71] One of the first electrode and the second electrode can be an anode and the other can be a cathode. The organic layer may comprise a light-emitting layer, and may further comprise 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, and a hole blocking layer.

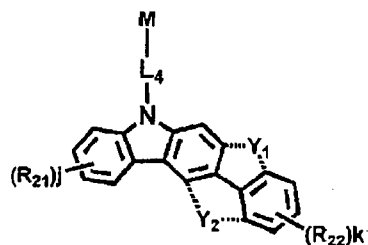
[72] The organic electroluminescent compound of formula 1 of the present invention can be included as a host material in the light-emitting layer. Preferably, the light-emitting layer may further include at least one dopant and, if necessary, may comprise other compounds, in addition to the organic electroluminescent compound of formula 1 of the present invention, as a second host material.

[73] The present invention further provides materials for preparing an organic EL device. The materials comprise the first and second host materials. The first host material includes the organic electroluminescent compound of the present invention. The first host material and the second host material may be in the range of 1:99 to 99:1 in a weight ratio.

[74] The second host material can be any of the known phosphorescent hosts and preferably is selected from the group consisting of the compounds of the following formulae 5 to 9:

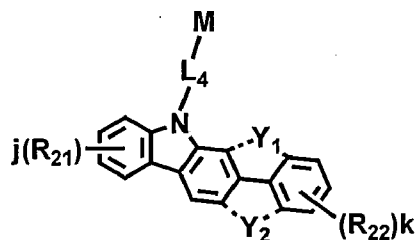


[77]



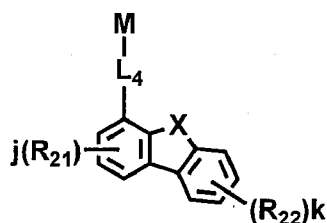
(7)

[78]



(8)

[79]



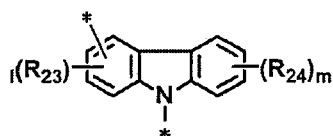
(9)

[80]

[81] wherein

[82] Cz represents the following structure:

[83]



[84] X represents O or S;

[85] R_{21} to R_{24} 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 5- or 30-membered heteroaryl group, or $R_{25}R_{26}R_{27}$ Si-; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C5-C30) alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen, and sulfur;

[86] R_{25} to R_{27} each independently represent a substituted or unsubstituted (C1-C30)alkyl group, or a substituted or unsubstituted (C6-C30)aryl group;

[87] L_4 represents a single bond, a substituted or unsubstituted (C6-C30)arylene group, or a substituted or unsubstituted 5- or 30-membered heteroarylene group;

[88] M represents a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 5- or 30-membered heteroaryl group;

[89] Y_1 and Y_2 each independently represent -O-, -S-, -N(R_{31})-, or -C(R_{32})(R_{33})-; and Y_1

and Y_2 are not simultaneously present;

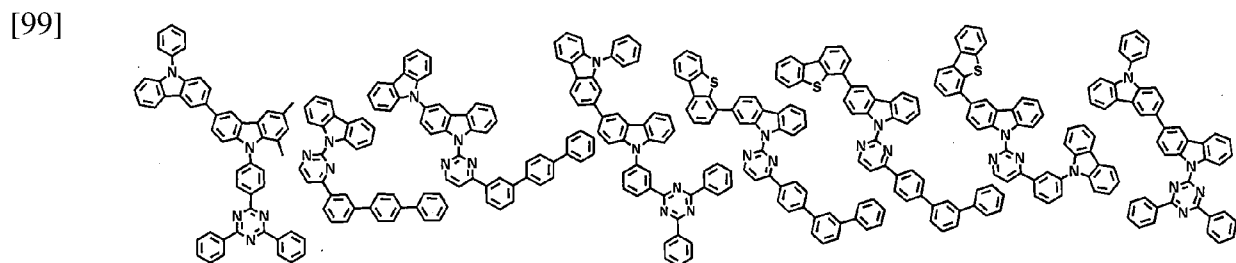
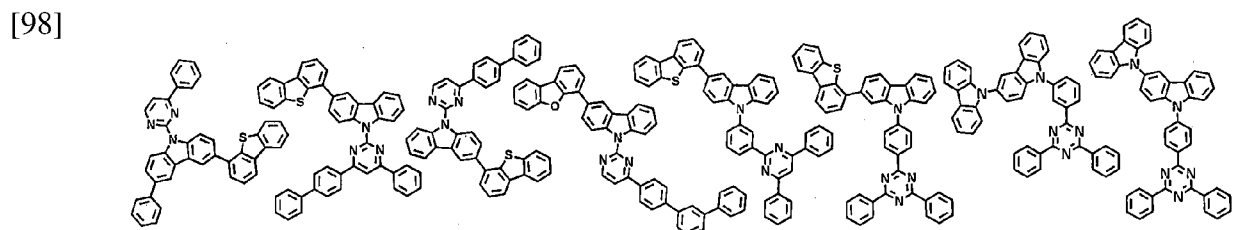
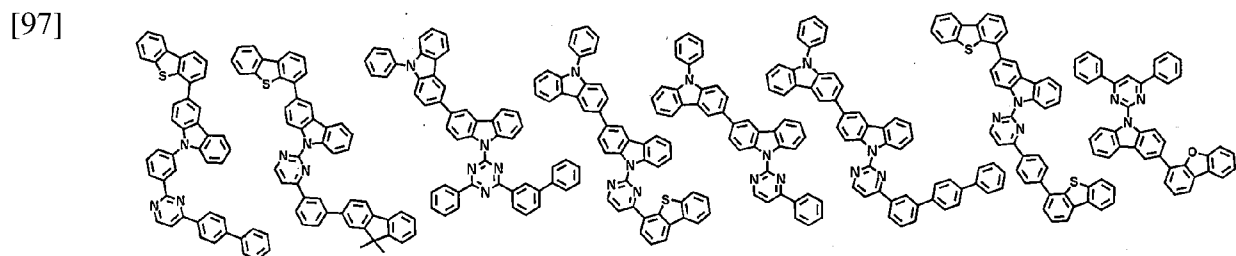
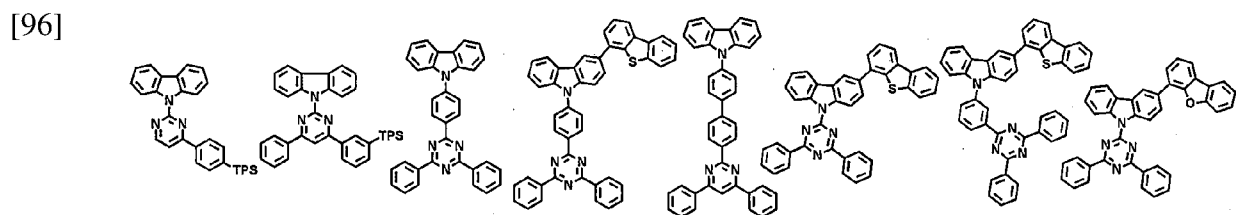
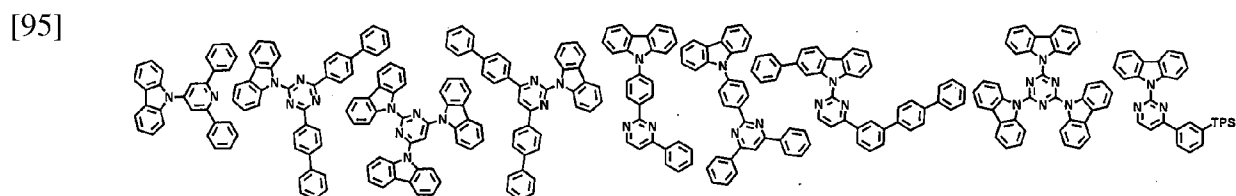
[90] R_{31} to R_{33} each independently represent a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 5- or 30-membered heteroaryl group; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C5-C30) alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur; and R_{32} and R_{33} may be the same or different;

[91] h and i each independently represent an integer of 1 to 3;

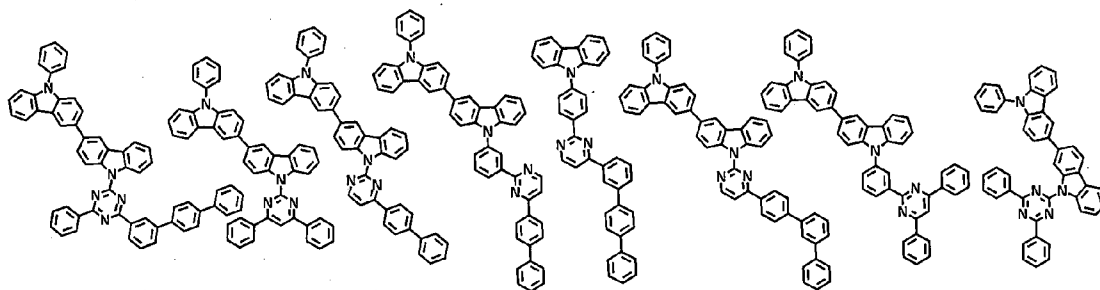
[92] j, k, l, and m each independently represent an integer of 0 to 4;

[93] where h, i, j, k, l, or m is an integer of 2 or more, each (Cz-L₄), each (Cz), each R_{21} , each R_{22} , each R_{23} , or each R_{24} is the same or different.

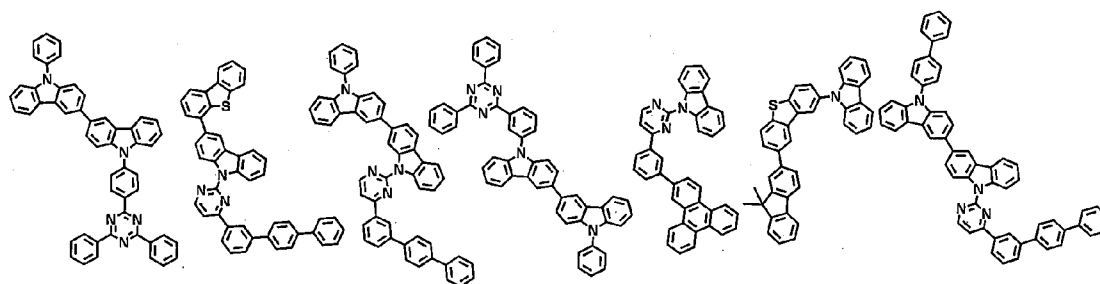
[94] Specifically, the second host material includes the following:



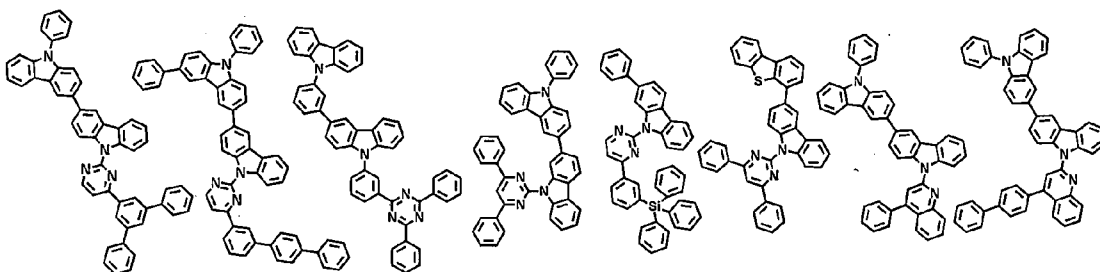
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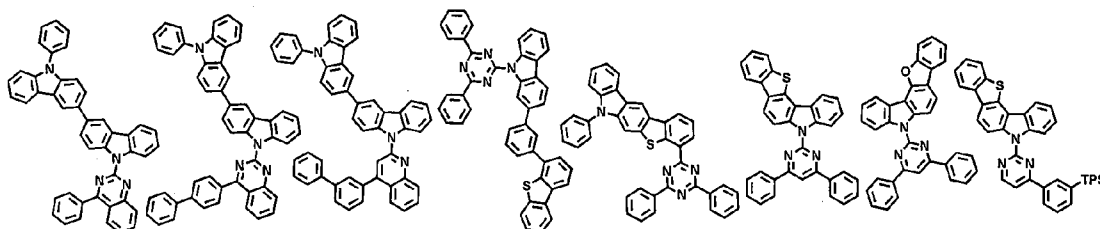
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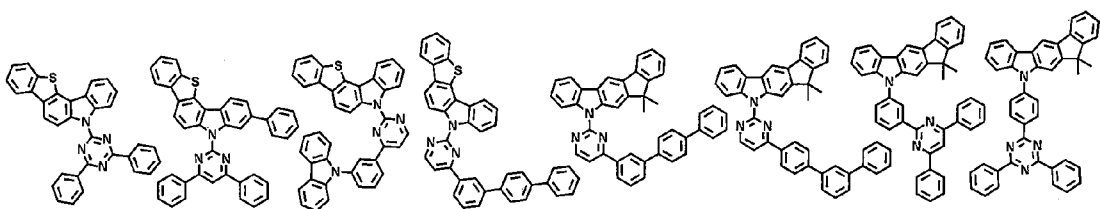
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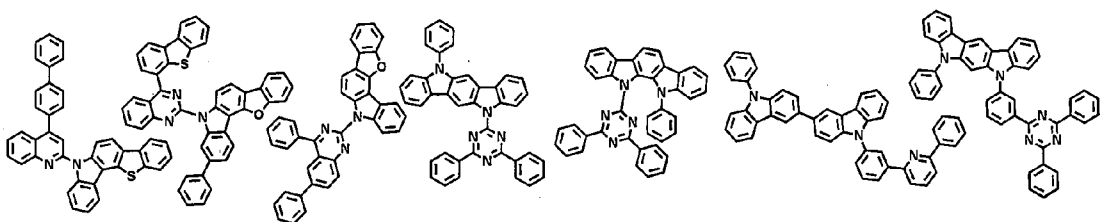
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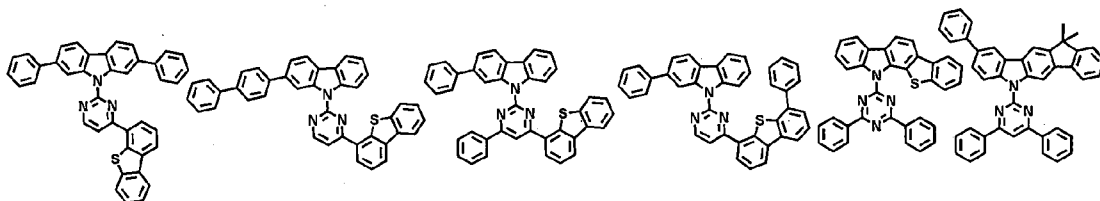
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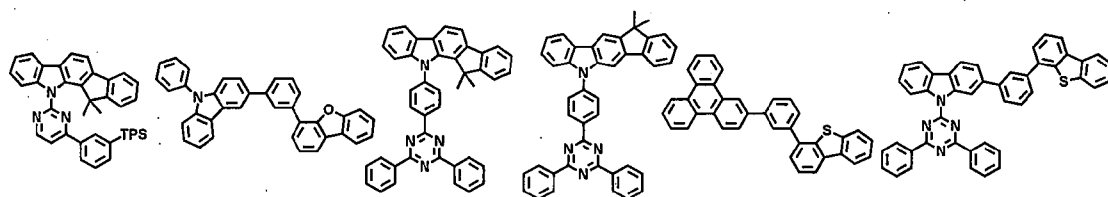
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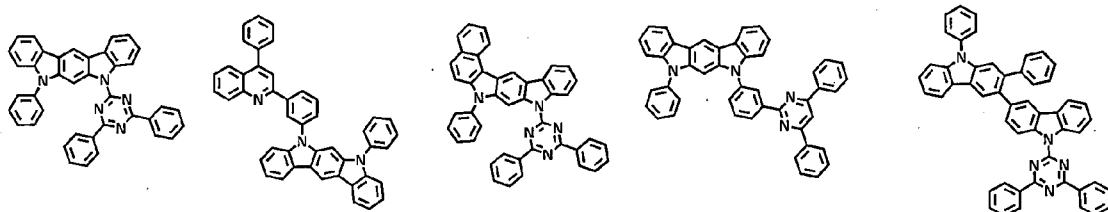
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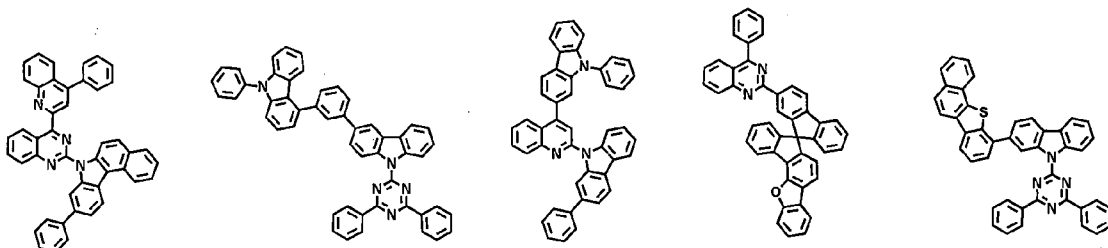
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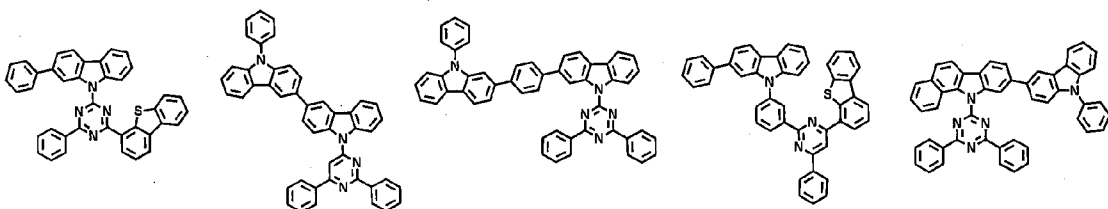
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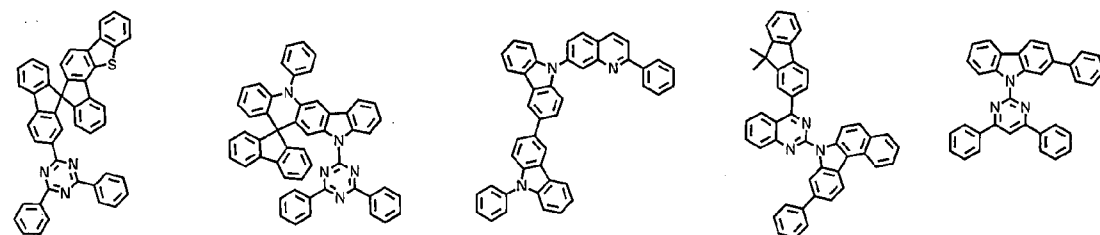
[109]



[110]



[111]



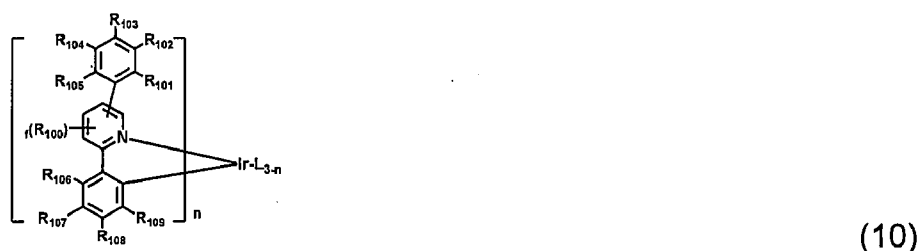
[112] wherein TPS represents triphenylsilyl.

[113] The dopants are preferably one or more phosphorescent dopants. The phosphorescent dopant material applied to the organic electroluminescent device of the present invention is not specifically limited, but preferably may be selected from complex compounds of iridium (Ir), osmium (Os), copper (Cu), and platinum (Pt), more preferably ortho-metallated complex compounds of iridium (Ir), osmium (Os), copper

(Cu), and platinum (Pt), and even more preferably ortho-metallated iridium complex compounds.

[114] The dopant included in the organic electroluminescent device of the present invention may be selected from the group consisting of the compounds represented by the following formulae 10 to 12:

[115]



[116]



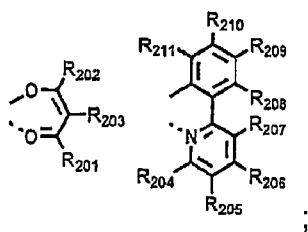
[117]



[118] wherein

[119] L is selected from the following structures:

[120]

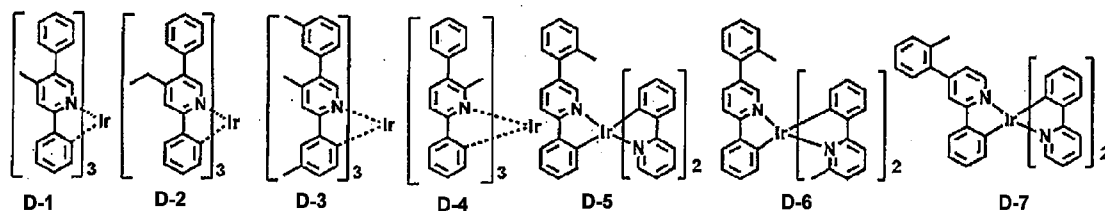


[121] R₁₀₀ represents hydrogen, a substituted or unsubstituted (C1-C30)alkyl group, or a substituted or unsubstituted (C3-C30)cycloalkyl group; R₁₀₁ to R₁₀₉ and R₁₁₁ to R₁₂₃ each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group which is unsubstituted or substituted with halogen(s), a substituted or unsubstituted (C3-C30)cycloalkyl group, a cyano group, or a substituted or unsubstituted (C1-C30)alkoxy group; R₁₂₀ to R₁₂₃ are linked to an adjacent substituent(s) to form a fused ring, for example, quinoline; R₁₂₄ to R₁₂₇ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, or a sub-

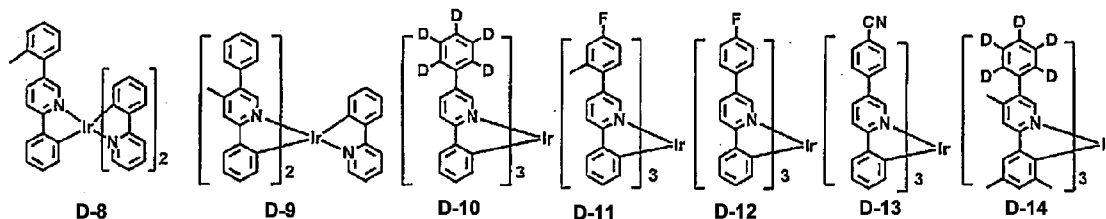
stituted or unsubstituted (C6-C30)aryl group; when R_{124} to R_{127} are aryl groups, they are linked to an adjacent substituent(s) to form a fused ring, for example, fluorene; R_{201} to R_{211} each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group which is unsubstituted or substituted with halogen(s), a substituted or unsubstituted (C3-C30)cycloalkyl group, or a (C6-C30)aryl group; f and g each independently represent an integer of 1 to 3; where f or g is an integer of 2 or more, each R_{100} may be the same or different; and n represents an integer of 1 to 3.

[122] The phosphorescent dopant material includes the following:

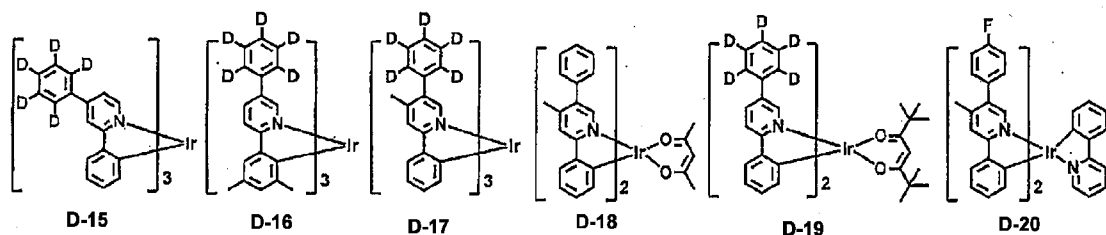
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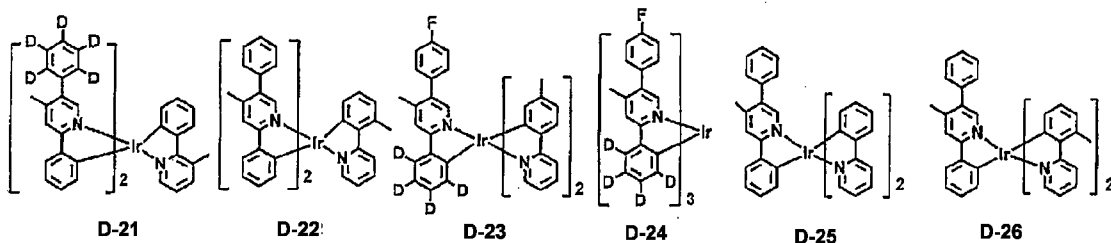
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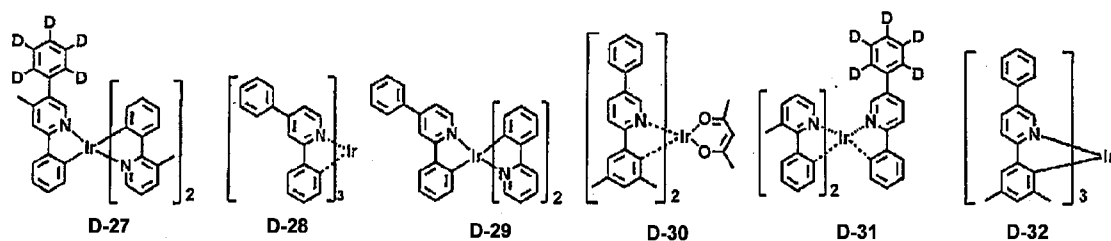
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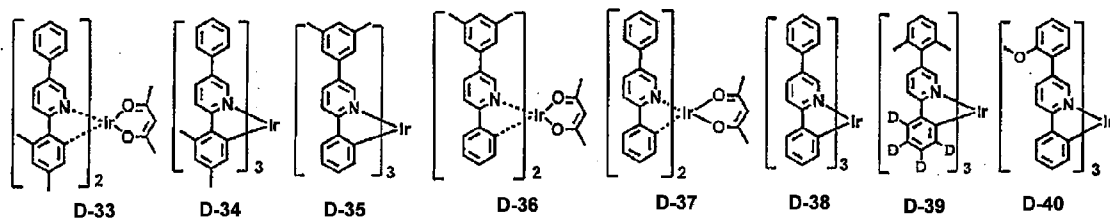
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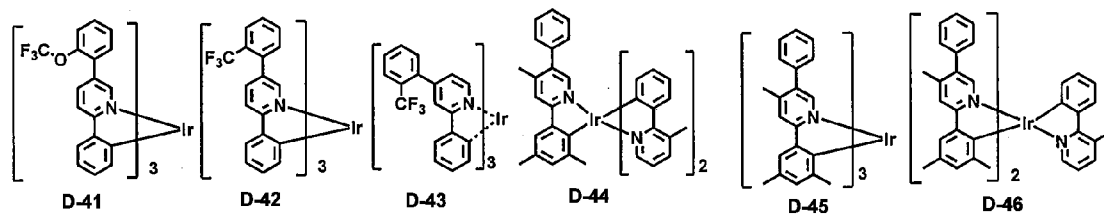
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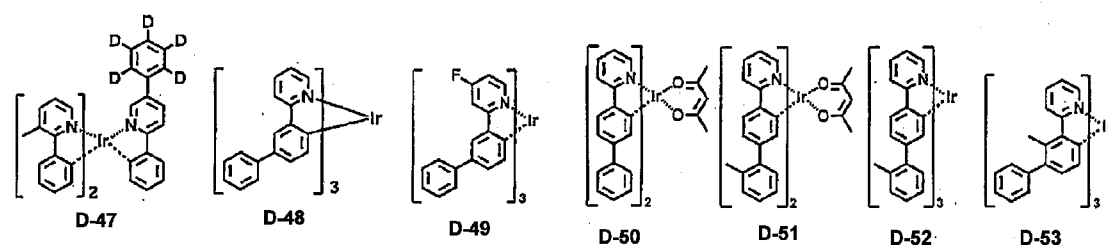
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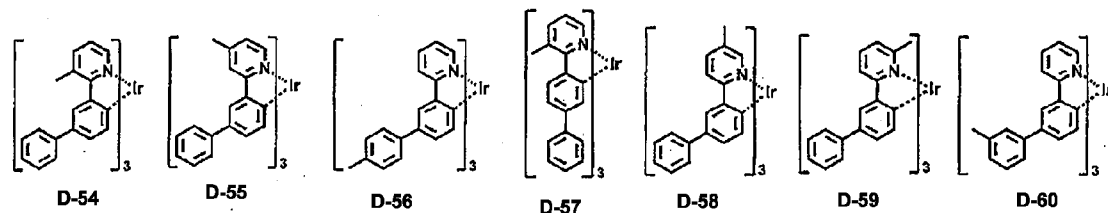
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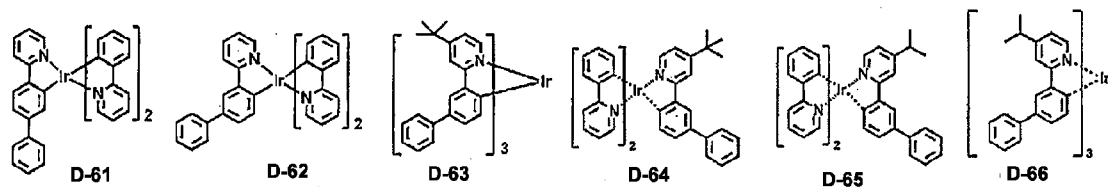
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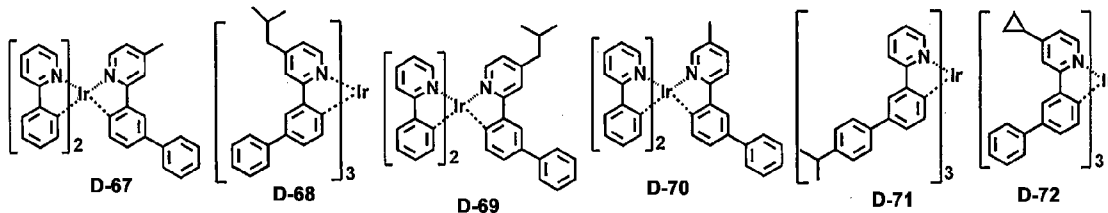
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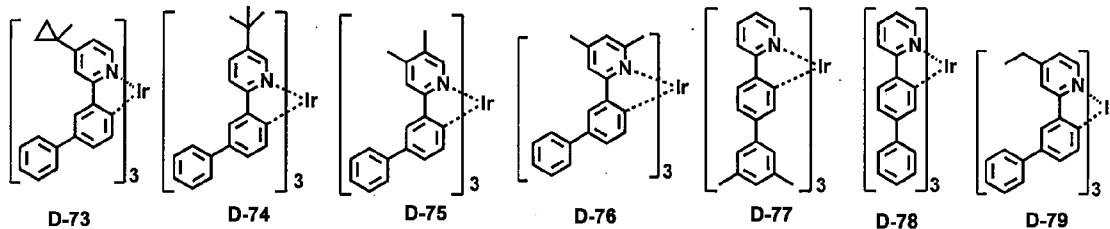
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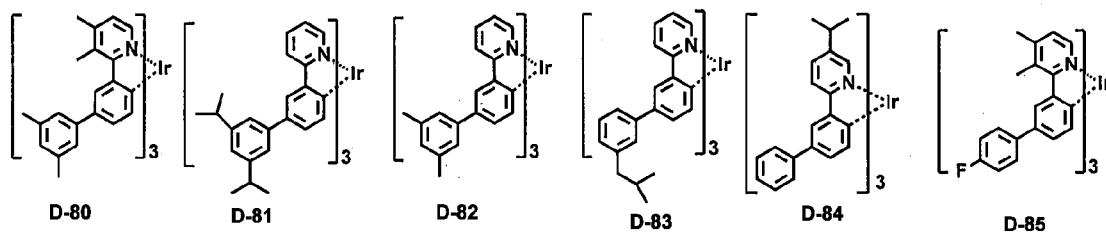
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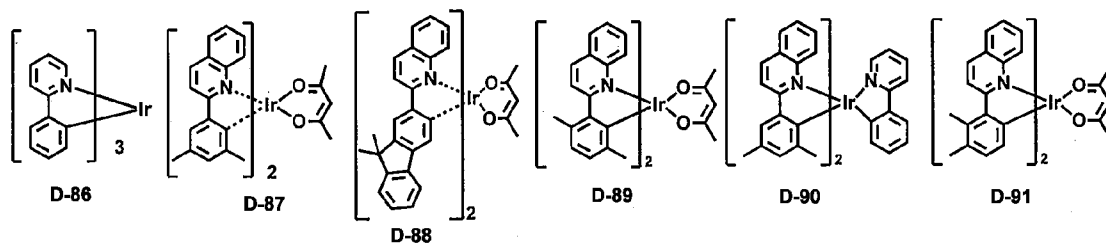
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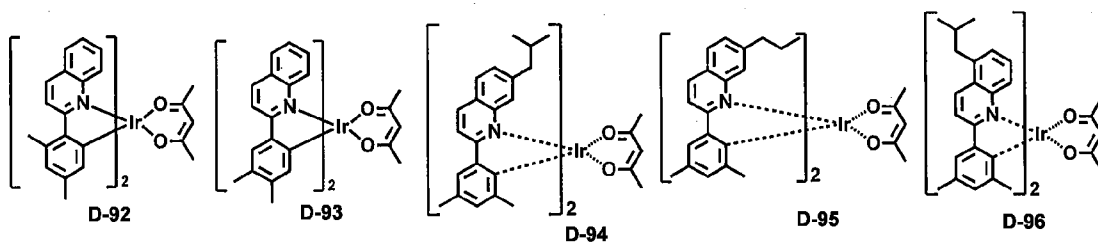
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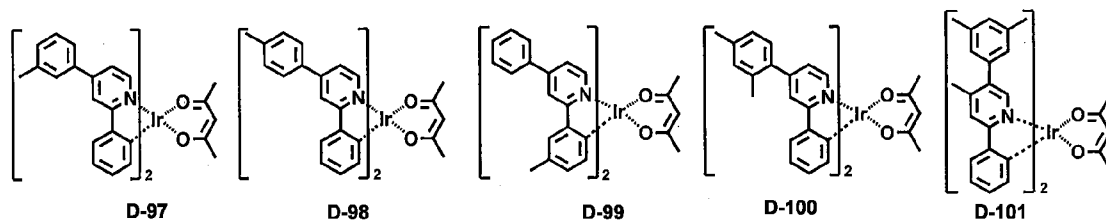
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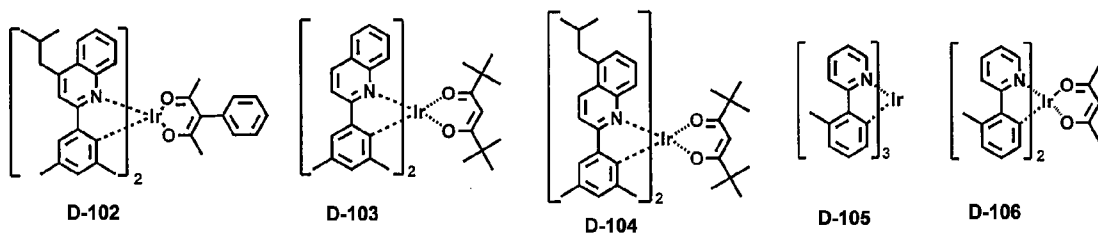
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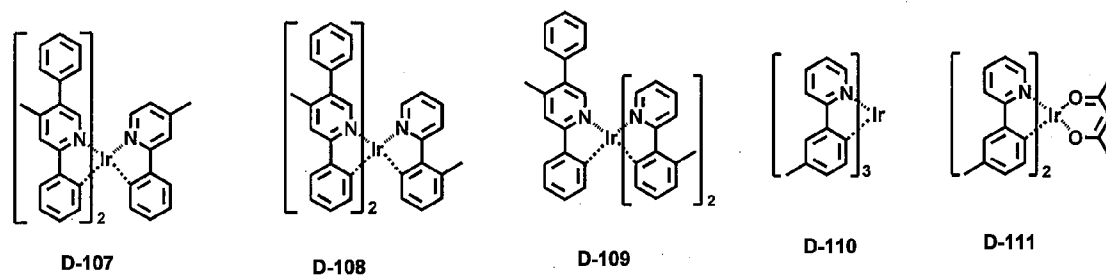
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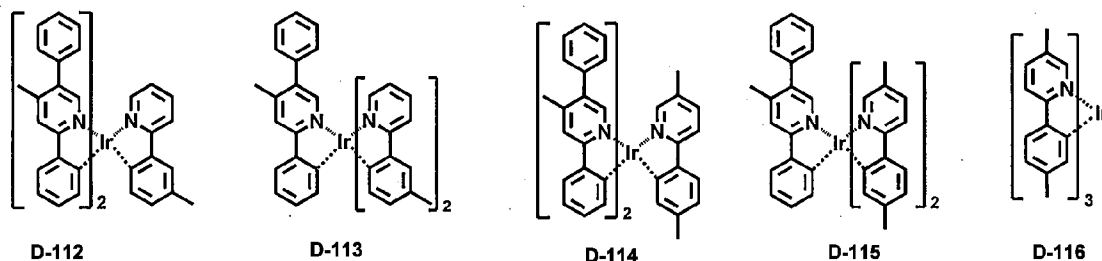
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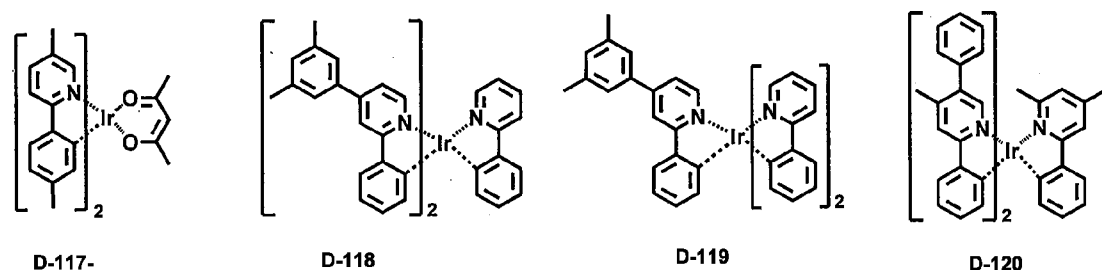
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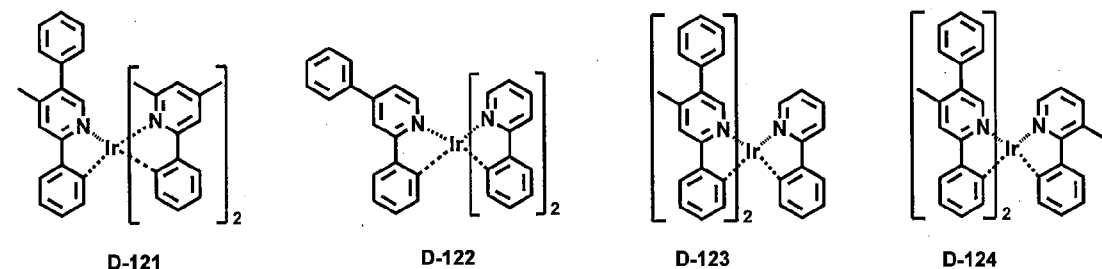
[141]



[142]



[143]



[144] The organic layer of the organic electroluminescent device of the present invention comprises the organic electroluminescent compounds of formula 1 and may further include at least one compound selected from the group consisting of arylamine-based compounds and styrylarylamine-based compounds.

[145] In the organic electroluminescent device of the present invention, the organic layer may further comprise, in addition to the organic electroluminescent compounds 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 d-transition elements of the Periodic Table, or at least one complex compound comprising the metal.

[146] 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, besides the organic electroluminescent compound of the present invention; and may further include a yellow or orange light-emitting layer, if necessary.

[147] Preferably, in the organic electroluminescent device of the present invention, at least one layer (hereinafter, "a surface layer") 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, it is preferred that a chalcogenide (including oxides) layer of silicon or aluminum is placed on an anode surface of a light-emitting medium layer, and a metal halide layer or metal oxide layer is placed on a cathode surface of an electroluminescent medium layer. The surface layer provides operating 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.

[148] Preferably, 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 a light-emitting 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 a light-emitting 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 organic electroluminescent device having two or more light-emitting layers and emitting white light.

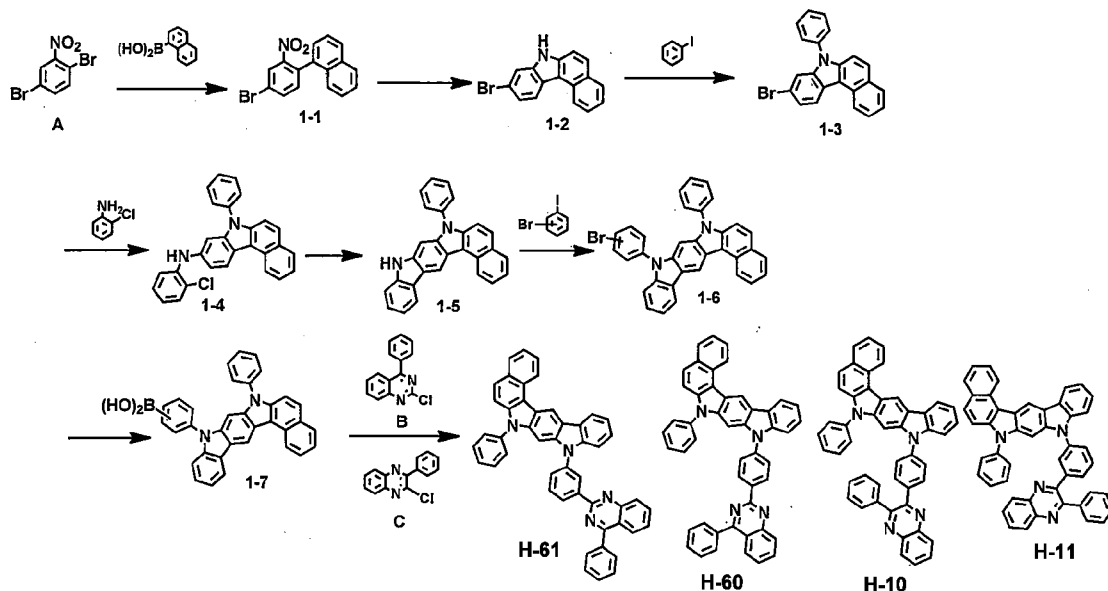
[149] In order to form each layer constituting the organic electroluminescent device of the present invention, dry film-forming methods, such as vacuum deposition, sputtering, plasma, ion plating methods, etc., or wet film-forming methods, such as spin coating, dip coating, flow coating methods, etc.,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

[150] When using a wet film-forming method, a thin film is formed by dissolving or dispersing the material constituting each layer in suitable solvents, such as ethanol, chloroform, tetrahydrofuran, dioxane, etc. The solvents are not specifically limited as long as the material constituting each layer is soluble or dispersible in the solvents, which do not cause any problems in forming a layer.

[151] Hereinafter, the organic electroluminescent compound of the present invention, the preparation method of the compound, and the luminescent properties of the device comprising the compound will be explained in detail with reference to the following examples:

[152] Example 1: Preparation of compounds H-61, H-60, H-10, and H-11

[153]

[154] 1) Preparation of compound 1-1

[155] Compound A (100.0 g, 356.0 mmol), 1-naphthyl boronic acid (51.0 g, 297.0 mmol), tetrakis(triphenylphosphine)palladium(O) ($\text{Pd}(\text{PPh}_3)_4$) (11.0 g, 9.0 mmol), 2M K_2CO_3 (500.0 mL), toluene (1000.0 mL), and ethanol (500.0 mL) in a flask were stirred under reflux for 5 hrs. After completing the reaction, the organic layer was extracted with ethyl acetate (EA) and dried by removing the remaining moisture with MgSO_4 . The residue was separated via column chromatography to obtain compound 1-1 (70.0 g, 72 %).

[156] 2) Preparation of compound 1-2

[157] Compound 1-1 (70.0 g, 213.0 mmol), triphenylphosphine (140.0 g, 533.0 mmol), and dichlorobenzene (1.0 L) were dissolved in a flask and the mixture was refluxed at 150 °C for 6 hrs. After completing the reaction, the mixture was distilled and triturated with methanol (MeOH) to obtain compound 1-2 (40.0 g, 64 %).

[158] 3) Preparation of compound 1-3

[159] Compound 1-2 (20.0 g, 67.53 mmol), iodobenzene (15.0 mL, 135.06 mmol), CuI (12.0 g, 33.77 mmol), Cs_2CO_3 (66.0 g, 202.59 mmol), ethylenediamine (EDA) (2.0 mL, 33.77 mmol), and toluene (400.0 mL) were dissolved in a flask and the mixture was refluxed at 120 °C for 5 hrs. After completing the reaction, the organic layer was extracted with EA and dried by removing the remaining moisture with MgSO_4 . The residue was separated via column chromatography to obtain compound 1-3 (15.0 g, 60 %).

[160] 4) Preparation of compound 1-4

[161] Compound 1-3 (26.0 g, 69.84 mmol), 2-chloroaniline (11.0 g, 104.77 mmol), palladium(II) acetate ($\text{Pd}(\text{OAc})_2$) (0.6 g, 2.79 mmol), sodium tert-butoxide (NaOtBu) (16.0 g, 174.6 mmol), tri-tert-butylphosphine ($\text{P}(\text{t-Bu})_3$) (3.0 mL, 5.58 mmol), and

toluene (350.0 mL) were dissolved in a flask and the mixture was refluxed at 120 °C for 5 hrs. After completing the reaction, the organic layer was extracted with EA and dried by removing the remaining moisture with MgSO₄. The residue was separated via column chromatography to obtain compound 1-4 (19.2 g, 66 %).

[162] 5) Preparation of compound 1-5

[163] Compound 1-4 (26.0 g, 69.84 mmol), Pd(OAc)₂ (0.6 g, 2.79 mmol), Cs₂CO₃ (45.0 g, 137.49 mmol), tricyclohexylphosphine tetrafluoroborate (PCy₃HBF₄) (1.7 g, 4.58 mmol), and toluene (300.0 mL) were dissolved in a flask and the mixture was refluxed at 120 °C for 5 hrs. After completing the reaction, the organic layer was extracted with EA and dried by removing the remaining moisture with MgSO₄. The residue was separated via column chromatography to obtain compound 1-5 (15.0 g, 88 %).

[164] 6) Preparation of compound 1-6

[165] Compound 1-5 (14.0 g, 36.60 mmol), 1-bromo-3-iodobenzene (15.0 g, 54.91 mmol), CuI (3.5 g, 18.30 mmol), K₃PO₄ (23.0 g, 109.8 mmol), EDA (3.0 mL, 36.60 mmol), and toluene (183.0 mL) were dissolved in a flask and the mixture was refluxed at 120 °C for 5 hrs. After completing the reaction, the organic layer was extracted with EA and dried by removing the remaining moisture with MgSO₄. The residue was separated via column chromatography to obtain compound 1-6 (12.4 g, 65 %).

[166] 7) Preparation of compound 1-7

[167] Compound 1-6 (12.4 g, 23.07 mmol) was dissolved in tetrahydrofuran (THF) (100.0 mL), and n-butyl lithium (n-BuLi) (14.0 mL, 34.61 mmol, 2.5 M in hexane) was slowly added thereto at -78 °C. After 1 hr, triisopropyl borate (8.0 mL, 34.61 mmol) was added to the mixture. After stirring the mixture at room temperature for 12 hrs, distilled water was added thereto. The organic layer was extracted with EA, dried over MgSO₄, and distilled under reduced pressure. Compound 1-7 (6.0 g, 52 %) was obtained by recrystallization with EA and hexane.

[168] 8) Preparation of compound H-61

[169] Compound 1-7 (7.4 g, 14.73 mmol), compound B (3.0 g, 12.27 mmol), Pd(PPh₃)₄ (1.4 g, 1.23 mmol), 2M K₂CO₃ (30.0 mL), toluene (60.0 mL), and ethanol (30.0 mL) in a flask were stirred under reflux for 5 hrs. The mixture was cooled to room temperature and distilled water was added thereto. The organic layer was extracted with EA and dried over MgSO₄. The residue was distilled under reduced pressure and recrystallized with EA and MeOH to obtain compound H-61 (2.6 g, 32 %).

[170] 9) Preparation of compound H-60

[171] Compound 1-7 (10.0 g, 17.11 mmol), compound B (5.3 g, 22.24 mmol), Pd(PPh₃)₄ (1.0 g, 0.86 mmol), 2M K₂CO₃ (50.0 mL), toluene (100.0 mL), and ethanol (50.0 mL) in a flask were stirred under reflux for 5 hrs. The mixture was cooled to room temperature and distilled water was added thereto. The organic layer was extracted with

EA and dried over MgSO_4 . The residue was distilled under reduced pressure and re-crystallized with EA and MeOH to obtain compound H-60 (4.1 g, 37 %).

[172] 10) Preparation of compound H-10

[173] Compound 1-7 (10.0 g, 17.11 mmol), compound C (3.5 g, 14.25 mmol), $\text{Pd}(\text{PPh}_3)_4$ (0.8 g, 0.712 mmol), 2M K_2CO_3 (35.0 mL), toluene (70.0 mL), and ethanol (35.0 mL) in a flask were stirred under reflux for 5 hrs. The mixture was cooled to room temperature and distilled water was added thereto. The organic layer was extracted with EA and dried over MgSO_4 . The residue was distilled under reduced pressure and re-crystallized with EA and MeOH to obtain compound H-10 (7.5 g, 79 %).

[174] 11) Preparation of compound H-11

[175] Compound 1-7 (13.0 g, 22.2 mmol), compound C (4.5 g, 18.6 mmol), $\text{Pd}(\text{PPh}_3)_4$ (1.0 g, 0.9 mmol), 2M K_2CO_3 (28.0 mL), toluene (60.0 mL), and ethanol (28.0 mL) in a flask were stirred under reflux for 5 hrs. The mixture was cooled to room temperature and distilled water was added thereto. The organic layer was extracted with EA and dried over MgSO_4 . The residue was distilled under reduced pressure and recrystallized with EA and MeOH to obtain compound H-11 (2.5 g, 21 %).

[176] The property data of the compounds prepared in Example 1 are provided in Table 1 below:

[177] Table 1

[178]

	Molecular weight (MW)	UV	PL (in Tol)	MP (°C)
H-61	662.78	353	501	272
H-60	662.78	418	466	217
H-10	662.78	402	477	295
H-11	662.78	344	513	263

[179] Device Example 1: Production of an OLED device by using the

[180] organic electroluminescent compound according to the present invention

[181] An organic light-emitting diode (OLED) device using the organic electroluminescent compound of the present invention was produced as follows: A transparent electrode indium tin oxide (ITO) thin film (15 Ω/sq) on a glass substrate for an OLED device (Samsung Corning, Republic of Korea) was subjected to an ultrasonic washing with trichloroethylene, acetone, ethanol, and distilled water, sequentially, and was then stored in isopropanol. Next, the ITO substrate was mounted on a substrate holder of a vacuum vapor depositing apparatus. $\text{N}^1, \text{N}^{1'} - ([1, 1' - \text{biphenyl}] - 4, 4' - \text{diyl}) \text{bis}(\text{N}^1 - (\text{naphthalene} - 1 - \text{yl}) - \text{N}^4, \text{N}^4 - \text{diphenylbenzene} - 1, 4 - \text{diamine})$ was introduced into a cell of the vacuum vapor depositing apparatus, and the pressure in the chamber of the apparatus was then controlled to 10^{-6} torr. Thereafter, an electric current was applied to

the cell to evaporate the introduced material, thereby forming a hole injection layer having a thickness of 60 nm on the ITO substrate. Next, N,N'-di(4-biphenyl)-N,N'-di(4-biphenyl)-4,4'-diaminobiphenyl was introduced into another cell of the vacuum vapor depositing apparatus, and an electric current was applied to the cell to evaporate the introduced material, thereby forming a hole transport layer having a thickness of 20 nm on the hole injection layer. Compound H-61 was introduced as a host into one cell of the vacuum vapor depositing apparatus, and compound D-87 as a dopant was introduced into another cell. The two materials were evaporated at different rates and the dopant was deposited in a doping amount of 4 wt%, based on the total weight of the host and dopant, to form a light-emitting layer having a thickness of 30 nm on the hole transport layer. 2-(4-(9,10-di(naphthalene-2-yl)anthracene-2-yl)phenyl)-1-phenyl-1H-benzo[d]imidazole was then introduced into one cell, and lithium quinolate was introduced into another cell. The two materials were evaporated at the same rates and were respectively deposited in a doping amount of 50 wt% to form an electron transport layer having a thickness of 30 nm on the light-emitting layer. Next, 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.

[182] The produced OLED device showed red emission having a current density of 8.1 mA/cm² and a luminance of 990 cd/m² at 3.5 V. Furthermore, the time taken to be reduced to 90 % of the luminescence at a luminance of 5,000nit was at least 130 hours.

[183]

[184] Device Example 2: Production of an OLED device by using the
[185] organic electroluminescent compound according to the present invention

[186] An OLED device was produced in the same manner as in Device Example 1, except that compound H-60 was used as a host in a light-emitting material.

[187] The produced OLED device showed red emission having a current density of 8.2 mA/cm² and a luminance of 1050 cd/m² at 3.4 V. Furthermore, the time taken to be reduced to 90 % of the luminescence at a luminance of 5,000nit was at least 130 hours.

[188]

[189] Device Example 3: Production of an OLED device by using the
[190] organic electroluminescent compound according to the present invention

[191] An OLED device was produced in the same manner as in Device Example 1, except that compound H-10 as a host and compound D-88 as a dopant were used in a light-emitting material.

[192] The produced OLED device showed red emission having a current density of 12.2 mA/cm² and a luminance of 920 cd/m² at 3.6 V. Furthermore, the time taken to be reduced to 90 % of the luminescence at a luminance of 5,000nit was at least 110 hours.

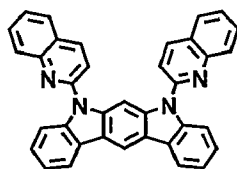
[193]

[194] Comparative Example 1: Production of an OLED device by
[195] using conventional light-emitting materials

[196] An OLED device was produced in the same manner as in Device Example 1, except that compound R-1 was used as a host in a light-emitting material.

[197] The produced OLED device showed red emission having a current density of 8.0 mA/cm² and a luminance of 1000 cd/m² at 4.7 V. Furthermore, the time taken to be reduced to 90 % of the luminescence at a luminance of 5,000nit was at least 10 hours.

[198]



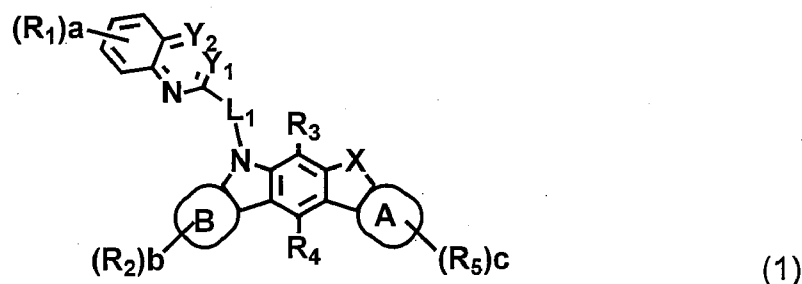
R-1

[199] The organic electroluminescent compound of the present invention has excellent luminescent efficiency and the organic electroluminescent device comprising the organic electroluminescent compound of the present invention provides long driving lifespan, and improved current efficiency and power efficiency.

Claims

[Claim 1]

An organic electroluminescent compound represented by the following formula 1:



wherein

L_1 represents a substituted or unsubstituted (C6-C30)arylene group;

Ring A or B represents benzene or naphthalene, with the proviso that A and B do not simultaneously represent benzene;

X represents NR_6 , $-CR_7R_8$, O, or S;

Y_1 and Y_2 each independently represent $-CR_9-$ or $-N-$, and if Y_1 represents $-N-$, Y_2 represents $-CR_9-$, and if Y_1 represents $-CR_9-$, Y_2 represents $-N-$;

R_1 to R_5 each independently represent hydrogen, deuterium, a halogen, a cyano group, a carboxyl group, a nitro group, a hydroxyl group, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted (C3-C30)cycloalkenyl group, a substituted or unsubstituted 3- to 7-membered heterocycloalkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted (C6-C30)arylamine group, $-NR_{10}R_{11}$, or $-SiR_{12}R_{13}R_{14}$;

R_1 , R_2 and R_5 are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring;

R_6 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 substituted or unsubstituted (C6-C30)aryl(C1-C30)alkyl group, $-NR_{10}R_{11}$, $-SiR_{12}R_{13}R_{14}$, a cyano group, a nitro group, or a hydroxyl group;

R_{10} and R_{11} 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;

R_{12} to R_{14} 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 5- to 7-membered heterocycloalkyl group, or a substituted or unsubstituted (C3-C30)cycloalkyl group; or are linked to an adjacent substituent(s) to form a substituted or unsubstituted mono- or polycyclic (C3-C30) alicyclic or aromatic ring;

the carbon atom(s) of the alicyclic or aromatic ring may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur; the heteroaryl(ene) and heterocycloalkyl groups contain at least one hetero atom selected from B, N, O, S, P(=O), Si, and P;

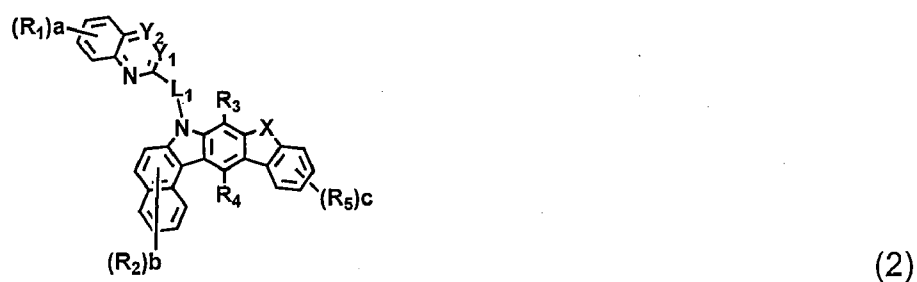
a represents an integer of 1 to 4; where a is an integer of 2 or more, each R_1 is the same or different;

if A or B represents benzene, b or c represents an integer of 1 to 4; and

if A or B represents naphthalene, b or c represents an integer of 1 to 6; where b or c is an integer of 2 or more, each R_2 or each R_5 is the same or different.

[Claim 2]

The organic electroluminescent compound according to claim 1, wherein the compound of formula 1 is represented by the following formula 2:

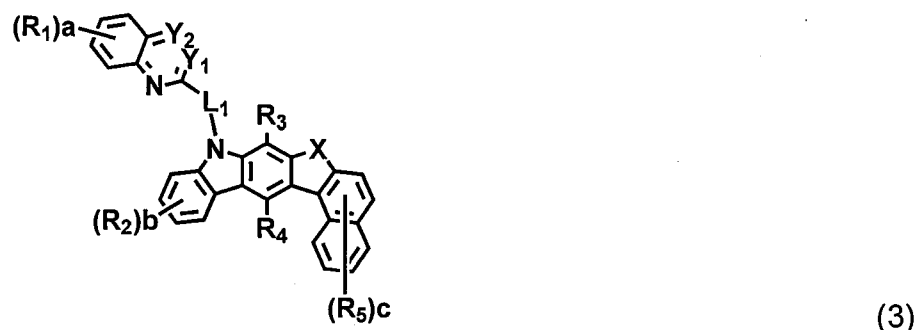


wherein

L_1 , X, Y_1 , Y_2 , R_1 to R_5 , a , b , and c are as defined in claim 1.

[Claim 3]

The organic electroluminescent compound according to claim 1, wherein the compound of formula 1 is represented by the following formula 3:

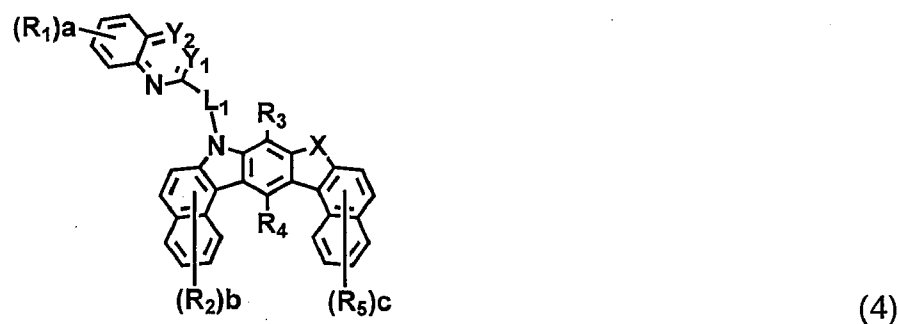


wherein

L_1 , X , Y_1 , Y_2 , R_1 to R_5 , a , b , and c are as defined in claim 1.

[Claim 4]

The organic electroluminescent compound according to claim 1, wherein the compound of formula 1 is represented by the following formula 4:



wherein

L_1 , X , Y_1 , Y_2 , R_1 to R_5 , a , b , and c are as defined in claim 1.

[Claim 5]

The organic electroluminescent compound according to any one of claims 1 to 4, wherein L_1 represents a (C6-C20)arylene group which is unsubstituted or substituted by a (C1-C6)alkyl group; R_1 to R_5 each independently represent hydrogen, or a substituted or unsubstituted (C6-C20)aryl group; R_6 to R_9 each independently represent a substituted or unsubstituted (C1-C6)alkyl group, or a substituted or unsubstituted (C6-C20)aryl group.

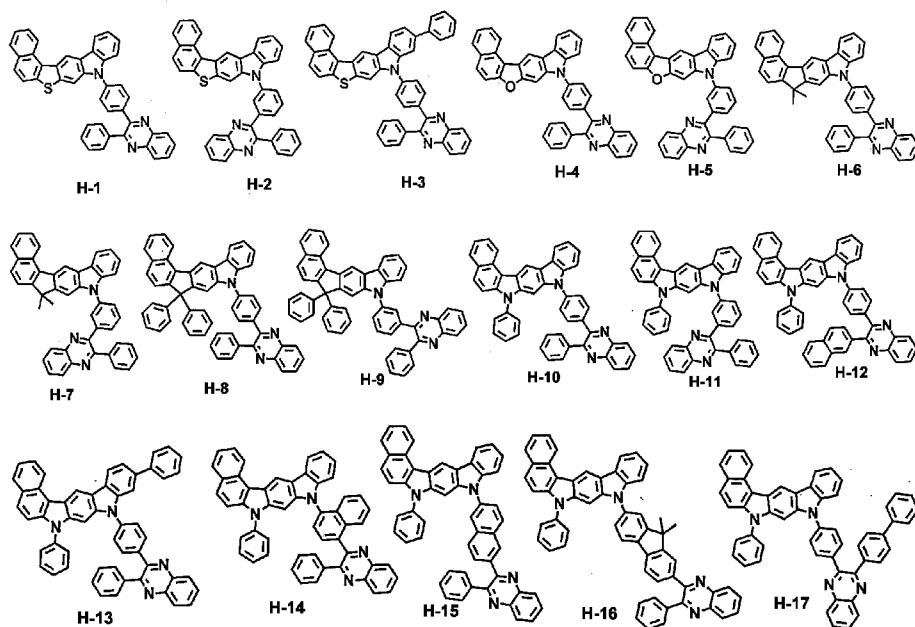
[Claim 6]

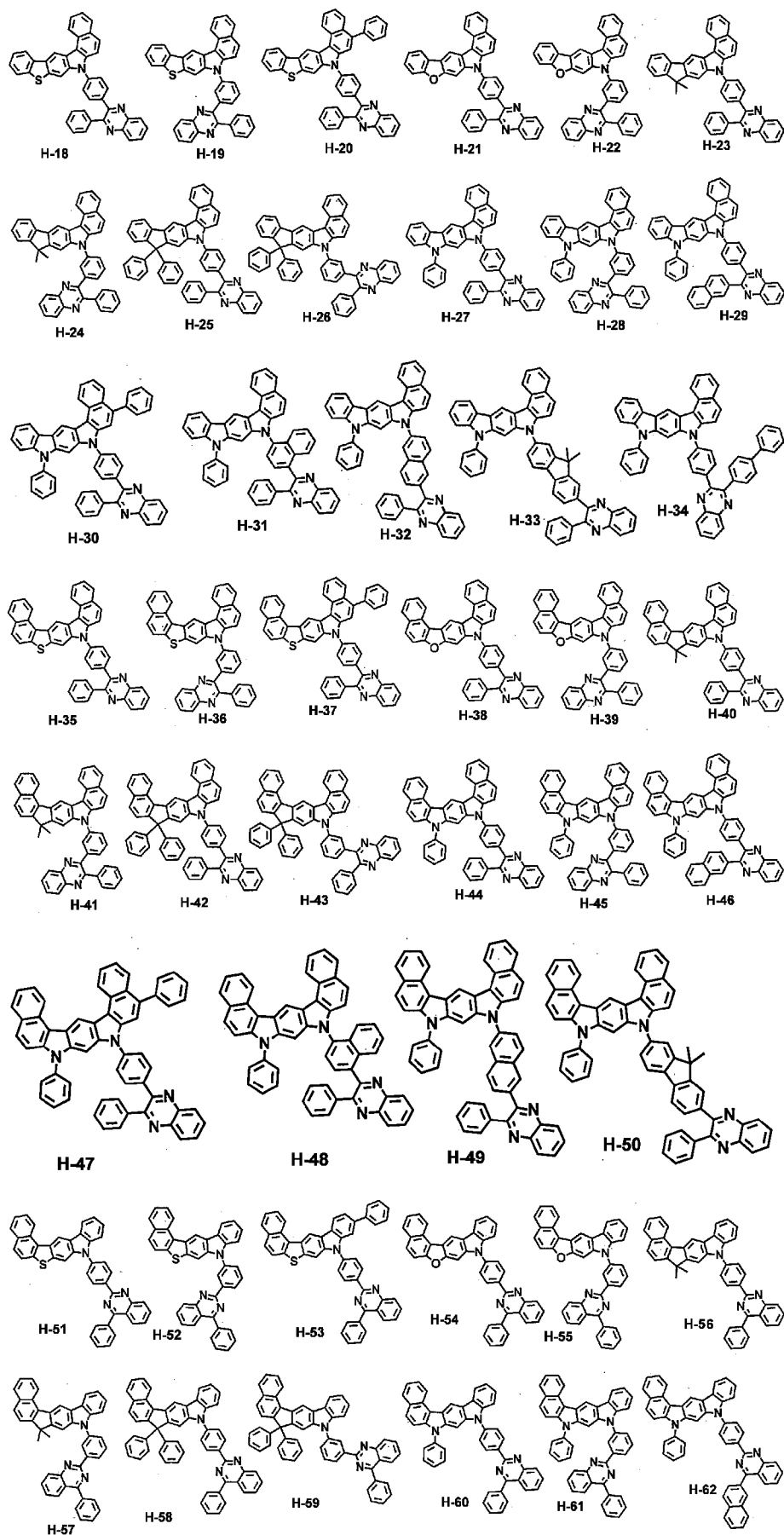
The organic electroluminescent compound according to claim 1, wherein the substituents of the substituted (C1-C30)alkyl group, the substituted (C3-C30)cycloalkyl group, the substituted (C3-C30)cycloalkenyl group, the substituted 3- to 7-membered heterocycloalkyl group, the substituted (C6-C30)aryl(ene) group, the substituted 3- to 30-membered heteroaryl(ene) group, the substituted (C6-C30)aryl(C1-C30)alkyl group, and the substituted mono- or polycyclic (C3-C30) alicyclic or aromatic ring in L_1 , X , Y_1 , Y_2 , and R_1 to R_5 are each independently at least one selected from the group

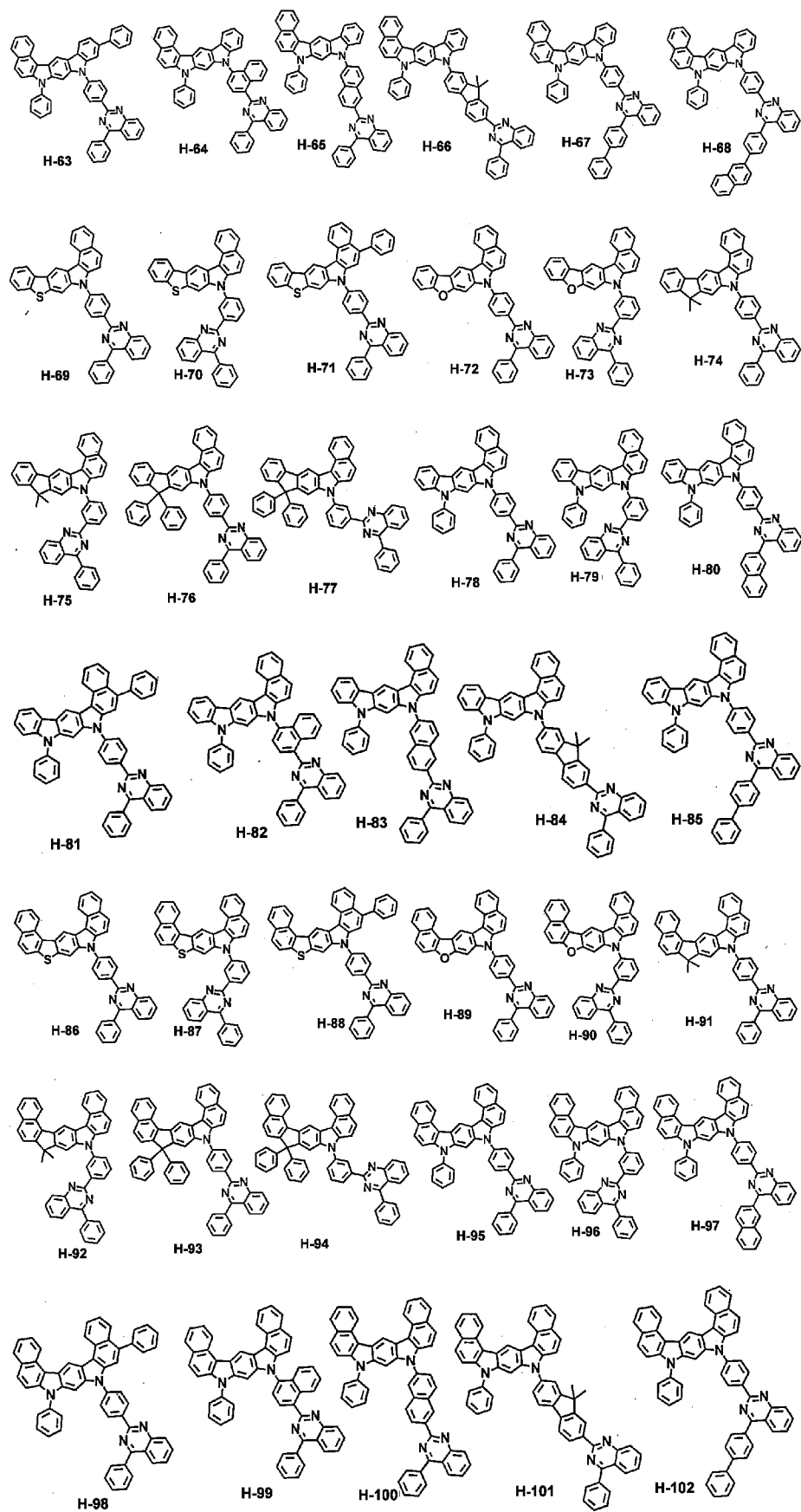
consisting of deuterium; a halogen; a cyano group; a carboxyl group; a nitro group; a hydroxyl group; a (C1-C30)alkyl group; a halo(C1-C30)alkyl group; a (C2-C30)alkenyl group; a (C2-C30)alkynyl group; a (C1-C30)alkoxy group; a (C1-C30)alkylthio group; a (C3-C30)cycloalkyl group; a (C3-C30)cycloalkenyl group; a 3- to 7-membered heterocycloalkyl group; a (C6-C30)aryloxy group; a (C6-C30)arylthio group; a 3- to 30-membered heteroaryl group which is unsubstituted or substituted with a (C6-C30)aryl group; a (C6-C30)aryl group which is unsubstituted or substituted with a 3- to 30-membered heteroaryl group; a tri(C1-C30)alkylsilyl group; a tri(C6-C30)arylsilyl group; a di(C1-C30)alkyl(C6-C30)arylsilyl group; a (C1-C30)alkyldi(C6-C30)arylsilyl group; an amino group; a mono- or di(C1-C30)alkylamino group; a mono- or di(C6-C30)arylamino group; a (C1-C30)alkyl(C6-C30)arylamino group; a (C1-C30)alkylcarbonyl group; a (C1-C30)alkoxycarbonyl group; a (C6-C30)arylcarbonyl group; a di(C6-C30)arylboronyl group; a di(C1-C30)alkylboronyl group; a (C1-C30)alkyl(C6-C30)arylboronyl group; a (C6-C30)aryl(C1-C30)alkyl group; and a (C1-C30)alkyl(C6-C30)aryl group.

[Claim 7]

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







[Claim 8]

An organic electroluminescent device comprising the compound

according to claim 1.

INTERNATIONAL SEARCH REPORT

 International application No.
PCT/KR2014/012895

A. CLASSIFICATION OF SUBJECT MATTER

C07D 487/04 (2006.01) C07D 403/10 (2006.01) C07D 491/048 (2006.01) C07D 495/04 (2006.01) C09K 11/06 (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)

Databases: Chemical Abstracts Registry / Chemical Abstracts Plus searched with structure based on claim 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 3 March 2015		Date of mailing of the international search report 03 March 2015	
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaustalia.gov.au		Authorised officer Andrew Bryce AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. +61 2 6283 3132	

INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2014/012895
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 2012/165832 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 06 December 2012 See abstract; Compounds C-79, C-80, C-98, C-100; Pages 9, 10; Claims 1, 5	1-8
A	WO 2012/050371 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 19 April 2012 See abstract; Compounds 19, 21, 27; Fig. 2, drawings page 2/12; Claim 1	1-8

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

INTERNATIONAL SEARCH REPORT		International application No.	
Information on patent family members		PCT/KR2014/012895	
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 2012/165832 A1	06 December 2012	CN 103717603 A	09 Apr 2014
		JP 2014522400 A	04 Sep 2014
		KR 20120132962 A	10 Dec 2012
		TW 201302974 A	16 Jan 2013
WO 2012/050371 A1	19 April 2012	CN 103249722 A	14 Aug 2013
		EP 2627640 A1	21 Aug 2013
		JP 2014501699 A	23 Jan 2014
		KR 20120038374 A	23 Apr 2012
		TW 201221619 A	01 Jun 2012
		US 2014061609 A1	06 Mar 2014
End of Annex			