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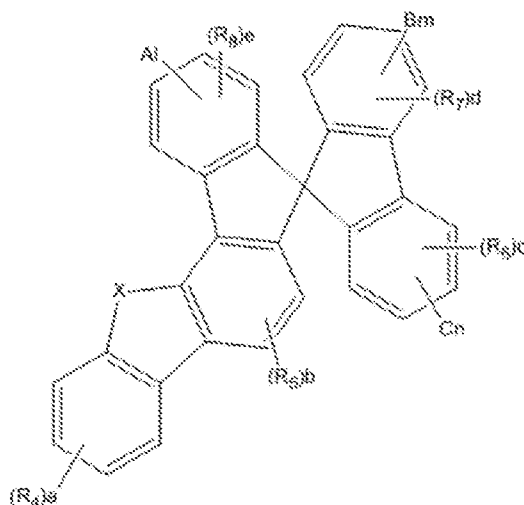
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[Continued on next page]

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



(1)

(57) Abstract: The present invention relates to organic electroluminescent compounds of formula 1 (see below) wherein X, A, B, C and R₄, R₅, R₆, R₇, a, b, c, d, e, l, m, and n are as defined in the description. The invention also relates to organic electroluminescent devices using compounds of formula 1. An organic electroluminescent compound according to the present invention and its use in an organic electroluminescent device can have high luminescent efficiency and high current efficiency.

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Description

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

Technical Field

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

Background Art

- [2] An electroluminescent device (EL device) is a self-light-emitting device which has advantages in that it provides a wider viewing angle, a greater contrast ratio, and a faster response time. An organic EL device was first developed by Eastman Kodak, by using small aromatic diamine molecules, and aluminum complexes as materials 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)picolate iridium (Firpic) as red, green and blue materials, respectively.
- [4] At present, 4,4'-N,N'-dicarbazol-biphenyl (CBP) is the most widely known phosphorescent host materials. Recently, Pioneer (Japan) et al. developed a high performance organic EL device using bathocuproine (BCP) and aluminum(III)bis(2-methyl-8-quinolinate)(4-phenylphenolate) (BALq) etc. as host materials, which were known as hole blocking layer materials.
- [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, 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 the voltage. Although an organic EL device comprising phosphorescent host materials provides higher current efficiency (cd/A) than one comprising fluorescent materials, a

significantly high driving voltage is necessary. Thus, there is no merit in terms of power efficiency (lm/W). (3) Further, the operational lifespan of an organic EL device is short and luminous efficiency is still required to be improved.

[6] Meanwhile, copper phthalocyanine (CuPc), 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB), N,N'-diphenyl-N,N'-bis(3-methylphenyl)-(1,1'-biphenyl)-4,4'-diamine (TPD), 4,4',4''-tris(3-methylphenylphenylamino)triphenylamine (MTDATA), etc. were used as a hole injection and transport material.

[7] However, an organic EL device using these materials is problematic in quantum efficiency and operational lifespan. It is because, when an organic EL device is driven under high current, thermal stress occurs between an anode and the hole injection layer. Thermal stress significantly reduces the operational lifespan of the device. Further, since the organic material used in the hole injection layer has very high hole mobility, the hole-electron charge balance may be broken and quantum yield (cd/A) may decrease.

[8] International Patent Publication No. WO 2012/034627 A1 discloses a compound in which a diarylamine group or a heteroaryl group is bonded directly or via an aryl group to a benzene ring of a spiro[fluoren-9,9'-fluorene] backbone unsubstituted or substituted with a halogen, an alkyl, or an aryl as a compound for an organic EL device.

[9] US Patent No. 7,714,145 B2 discloses a compound in which a substituent such as a diarylamine group is bonded via a linker such as an aryl group to a 2,2'-disubstituted 9,9'-spirobifluorene-based triaryldiamine as a compound for an organic EL device.

[10] However, the above references do not disclose a compound in which one or more diarylamine groups are bonded directly or via an aryl group to a benzene ring of a spiro[fluoren-9,9'-fluorene] backbone fused with an aryl group or a heteroaryl group, or the benzene ring is substituted with an indole group, nor an organic EL device using the compound for a hole transport layer.

Disclosure of Invention

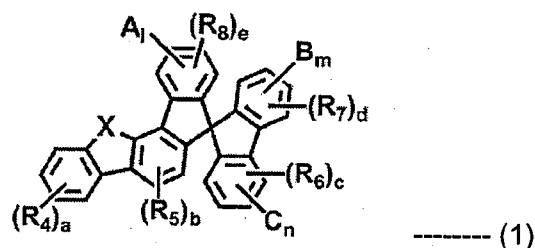
Technical Problem

[11] The objective of the present invention is to provide an organic electroluminescent compound having excellent current efficiency and luminous efficiency.

Solution to Problem

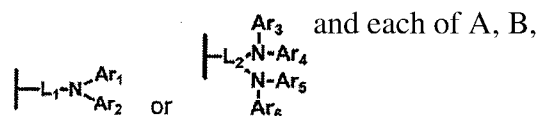
[12] The present inventors found that the above objective can be achieved by an organic electroluminescent compound represented by the following formula 1:

[13]



[14] wherein

[15] A, B, and C, each independently, represent



and C are the same or different;

[16] L₁ represents a single bond, a substituted or unsubstituted (C1-C30)alkylene, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (5- to 30-membered)heteroarylene;[17] L₂ represents a tertiary residue derived from a substituted or unsubstituted (C1-C30)acyclic hydrocarbon, a substituted or unsubstituted (C6-C30) aromatic hydrocarbon ring, or a substituted or unsubstituted (5- to 30-membered) aromatic heterocyclic ring;[18] Ar₁ to Ar₆, each independently, represent a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl; or Ar₁ and Ar₂, Ar₃ and Ar₄, or Ar₅ and Ar₆ may be linked to each other to form a substituted or unsubstituted (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;[19] X represents -O-, -S-, -C(R₁)(R₂)- or -N(R₃)-;[20] R₁ to R₃, each independently, represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (C3-C30)cycloalkenyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl; or R₁ and R₂ may be linked to each other to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;[21] R₄ to R₈, each independently, represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (5- to 30-membered)heteroaryl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted

(C6-C30)aryl(C1-C30)alkyl, -N(R₁₀)(R₁₁), -Si(R₁₂)(R₁₃)(R₁₄), -S(R₁₅), -O(R₁₆), a cyano, a nitro, or a hydroxyl; or may be linked to an adjacent substituent(s) to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

[22] R₁₀ to R₁₆, each independently, represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (5- to 30-membered)heteroaryl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, or a substituted or unsubstituted (C3-C30)cycloalkyl; or may be linked to an adjacent substituent(s) to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

[23] a, c, d, and e, each independently, represent an integer of 1 to 4; where a, c, d, or e is an integer of 2 or more, each of the substituents are the same or different;

[24] b represents 1 or 2;

[25] l, m, and n, each independently, represent an integer of 0 to 2;

[26] l+m+n is 1 or more; and

[27] the heterocycloalkyl and the heteroaryl(ene), each independently, contain at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

Advantageous Effects of Invention

[28] By using the organic electroluminescent compound according to the present invention, it is possible to manufacture an organic electroluminescent device having excellent current efficiency and luminous efficiency.

Mode for the Invention

[29] 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.

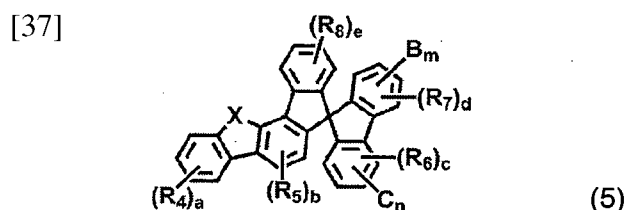
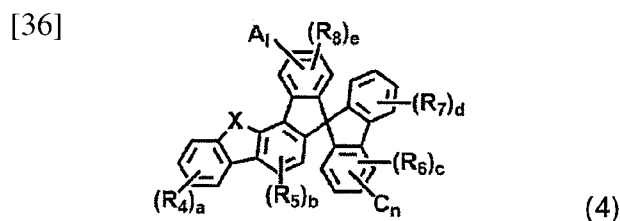
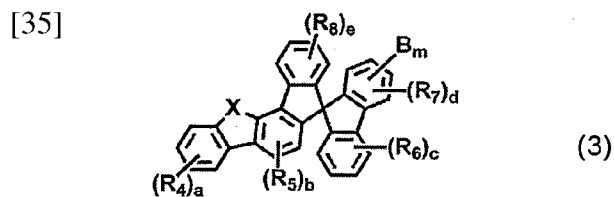
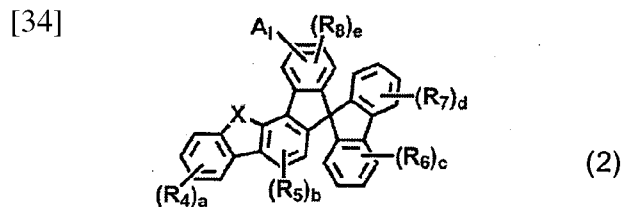
[30] The present invention relates to an organic electroluminescent compound of formula 1, an organic electroluminescent material comprising the compound, and an organic electroluminescent device comprising the material.

[31] The organic electroluminescent compound represented by the above formula 1 will be described in detail.

[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 10, more preferably 1 to 6, 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 meant to be 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-butyne, 2-butyne, 3-butyne, 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 3 to 7 ring backbone atoms including at least one heteroatom selected from B, N, O, S, P(=O), Si and P, preferably O, S and N, and includes tetrahydrofuran, pyrrolidine, thiolan, tetrahydropyran, etc.; “(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, binaphthyl, phenylnaphthyl, naphthylphenyl, fluorenyl, phenylfluorenyl, benzofluorenyl, dibenzofluorenyl, phenanthrenyl, phenylphenanthrenyl, anthracenyl, indenyl, triphenylenyl, pyrenyl, tetracenyl, perylenyl, chrysenyl, naphthacenyl, fluoranthenyl, etc.; “(5- to 30-membered)heteroaryl(ene)” is an aryl having 5 to 30 ring backbone atoms including at least one, preferably 1 to 4 heteroatom selected from the group consisting of B, N, O, S, P(=O), Si and P; 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, furazanil, 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, cinnolinyl, quinazolinyl, quinoxalinyl, carbazolyl, phenoxazinyl, phenanthridinyl, benzodioxolyl, etc. Further, “halogen” includes F, Cl, Br and I.

[33] The compound represented by formula 1 can be represented by one selected from formulae 2 to 5:



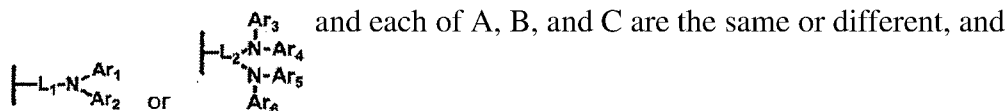
[38] wherein

[39] A, B, C, X, R₄ to R₈, a, b, c, d, and e are as defined in formula 1.

[40] Herein, "substituted" in the expression "substituted or unsubstituted" means that a hydrogen atom in a certain functional group is replaced with another atom or group, i.e., a substituent. The substituents of the substituted (C1-C30)alkyl(ene), the substituted (C3-C30)cycloalkyl, the substituted (C3-C30)cycloalkenyl, the substituted (3- to 7- membered)heterocycloalkyl, the substituted (C6-C30)aryl(ene), the substituted (5- to 30- membered)heteroaryl(ene), and the substituted (C6-C30)aryl(C1-C30)alkyl in R₁ to R₈, R₁₀ to R₁₆, L₁, L₂, and Ar₁ to Ar₆ in formulae 1 to 5 each independently are at least one selected from the group consisting of deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a (C1-C30)alkyl, a halo(C1-C30)alkyl, a (C2-C30)alkenyl, a (C2-C30)alkynyl, a (C1-C30)alkoxy, a (C1-C30)alkylthio, a (C3-C30)cycloalkyl, a (C3-C30)cycloalkenyl, a (3- to 7- membered)heterocycloalkyl, a (C6-C30)aryloxy, a (C6-C30)arylthio, a (5- to 30- membered)heteroaryl unsubstituted or substituted with a (C6-C30)aryl, a (C6-C30)aryl unsubstituted or substituted with a (5- to 30- membered)heteroaryl, a tri(C1-C30)alkylsilyl, a tri(C6-C30)arylsilyl, a di(C1-C30)alkyl(C6-C30)arylsilyl, a (C1-C30)alkyldi(C6-C30)arylsilyl, an amino, a mono- or di-(C1-C30)alkylamino, a mono- or di-(C6-C30)arylamino, a (C1-C30)alkyl(C6-C30)arylamino, a (C1-C30)alkylcarbonyl, a

(C1-C30)alkoxycarbonyl, a (C6-C30)arylcarbonyl, a di(C6-C30)arylboronyl, a di(C1-C30)alkylboronyl, a (C1-C30)alkyl(C6-C30)arylboronyl, a (C6-C30)aryl(C1-C30)alkyl, and a (C1-C30)alkyl(C6-C30)aryl, and preferably each independently are at least one selected from the group consisting of deuterium, a halogen, a (C1-C6)alkyl, a (C6-C15)aryl, a (5- to 15- membered)heteroaryl, and a di(C6-C15)arylamino.

[41] In formula 1 above, A, B, and C, each independently, represent



[42] L_1 represents a single bond, a substituted or unsubstituted (C1-C30)alkylene, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (5- to 30-membered)heteroarylene, preferably represents a single bond, a substituted or unsubstituted (C6-C20)arylene, or an unsubstituted (5- to 20-membered)heteroarylene, and more preferably represents a single bond, a (C6-C15)arylene unsubstituted or substituted with a (C1-C6)alkyl(s), or an unsubstituted (5- to 15-membered)heteroarylene.

[43] L_2 represents a tertiary residue derived from a substituted or unsubstituted (C1-C30)acyclic hydrocarbon, a substituted or unsubstituted (C6-C30) aromatic hydrocarbon ring, or a substituted or unsubstituted (5- to 30-membered) aromatic heterocyclic ring, preferably represents a tertiary residue derived from an unsubstituted (C6-C20) aromatic hydrocarbon ring, and more preferably represents a tertiary residue derived from an unsubstituted (C6-C15) aromatic hydrocarbon ring.

[44] Ar_1 to Ar_6 , each independently, represent a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl; or Ar_1 and Ar_2 , Ar_3 and Ar_4 , or Ar_5 and Ar_6 may be linked to each other to form a substituted or unsubstituted (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur; preferably, each independently, represent a substituted or unsubstituted (C6-C20)aryl, or a substituted or unsubstituted (5- to 20-membered)heteroaryl; or Ar_1 and Ar_2 , Ar_3 and Ar_4 , or Ar_5 and Ar_6 may be linked to each other to form a substituted or unsubstituted (3- to 20-membered) polycyclic aromatic ring whose carbon atom(s) may be replaced with nitrogen; and more preferably, each independently, represent a (C6-C20)aryl unsubstituted or substituted with deuterium, a halogen(s), a (C1-C6)alkyl(s), a (C6-C15)aryl(s), a di(C6-C15)arylamino(s), or a (5- to 15-membered)heteroaryl(s); or a (5- to 15-membered)heteroaryl unsubstituted or substituted with a (C6-C15)aryl(s); or Ar_1 and Ar_2 , Ar_3 and Ar_4 , or Ar_5 and Ar_6 may be linked to each other to form an indole ring unsubstituted or substituted with a

(C6-C15)aryl(s); or an indolocarbazole ring substituted with a (C6-C15)aryl(s) unsubstituted or substituted with a (C1-C4)alkyl(s), a phenyl(s) or a naphthyl(s), or a (5- to 15-membered)heteroaryl(s) unsubstituted or substituted with a phenyl(s).

[45] X represents -O-, -S-, -C(R₁)(R₂)- or -N(R₃)-.

[46] R₁ to R₃, each independently, represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (C3-C30)cycloalkenyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl; or R₁ and R₂ may be linked to each other to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur; preferably, each independently, represent an unsubstituted (C1-C10)alkyl or an unsubstituted (C6-C20)aryl; or R₁ and R₂ may be linked to each other to form a (3- to 20-membered) monocyclic alicyclic ring; and more preferably, each independently, represent an unsubstituted (C1-C6)alkyl or an unsubstituted (C6-C15)aryl; or R₁ and R₂ may be linked to each other to form a cyclopentane ring or a cyclohexane ring.

[47] R₄ to R₈, each independently, represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (5- to 30-membered)heteroaryl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl(C1-C30)alkyl, -N(R₁₀)(R₁₁), -Si(R₁₂)(R₁₃)(R₁₄), -S(R₁₅), -O(R₁₆), a cyano, a nitro, or a hydroxyl; or may be linked to an adjacent substituent(s) to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur; preferably, each independently, represent hydrogen or an unsubstituted (C6-C20)aryl; or may be linked to an adjacent substituent(s) to form a (6- to 20-membered) monocyclic aromatic ring; and more preferably, each independently, represent hydrogen or an unsubstituted (C6-C15)aryl; or may be linked to an adjacent substituent(s) to form a benzene ring.

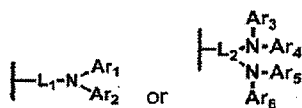
[48] a, c, d, and e, each independently, represent an integer of 1 to 4; where a, c, d, or e is an integer of 2 or more, each of the substituents are the same or different. Preferably, a represents 1 or 4, c and d each independently, represent an integer of 2 to 4, and e represents 3 or 4.

[49] b represents 1 or 2, and preferably 2.

[50] l, m, and n, each independently, represent an integer of 0 to 2. l+m+n is 1 or more,

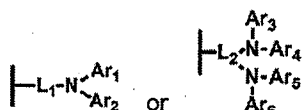
and preferably 1 or 2.

- [51] According to one embodiment of the present invention, in formula 1 above, A, B, and C, each independently, represent



are the same; L_1 represents a single bond, a substituted or unsubstituted (C6-C20)arylene, or an unsubstituted (5- to 20-membered)heteroarylene; L_2 represents a tertiary residue derived from an unsubstituted (C6-C20) aromatic hydrocarbon ring; Ar_1 to Ar_6 , each independently, represent a substituted or unsubstituted (C6-C20)aryl, or a substituted or unsubstituted (5- to 20-membered)heteroaryl; or Ar_1 and Ar_2 , Ar_3 and Ar_4 , or Ar_5 and Ar_6 may be linked to each other to form a substituted or unsubstituted (3- to 20-membered) polycyclic aromatic ring whose carbon atom(s) may be replaced with nitrogen; X represents -O-, -S-, -C(R_1)(R_2)- or -N(R_3)-; R_1 to R_3 , each independently, represent an unsubstituted (C1-C10)alkyl or an unsubstituted (C6-C20)aryl; or R_1 and R_2 may be linked to each other to form a (3- to 20-membered) monocyclic alicyclic ring; R_4 to R_8 , each independently, represent hydrogen or an unsubstituted (C6-C20)aryl, or may be linked to an adjacent substituent(s) to form a (6- to 20-membered) monocyclic aromatic ring; a represents 1 or 4; c and d, each independently, represent an integer of 2 to 4; e represents 3 or 4; where a, c, d or e is an integer of 2 or more, each of the substituents are the same or different; b represents 2; l, m, and n, each independently, represent an integer of 0 to 2; and $l+m+n$ is 1 or 2.

- [52] According to another embodiment of the present invention, in formula 1 above, A, B, and C, each independently, represent

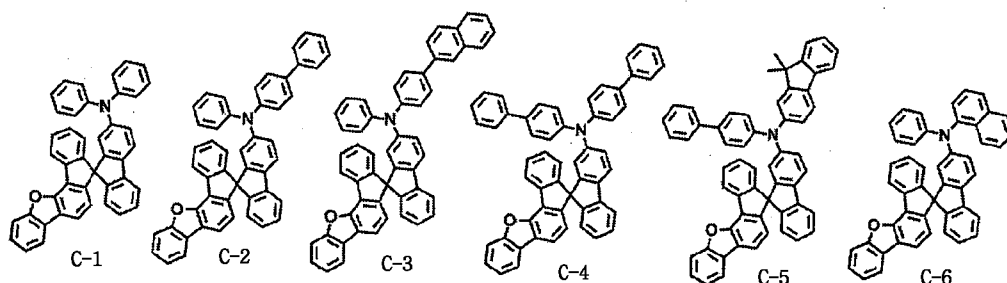


are the same; L_1 represents a single bond, a (C6-C15)arylene unsubstituted or substituted with a (C1-C6)alkyl(s), or an unsubstituted (5- to 15-membered)heteroarylene; L_2 represents a tertiary residue derived from an unsubstituted (C6-C15) aromatic hydrocarbon ring; Ar_1 to Ar_6 , each independently, represent a (C6-C20)aryl unsubstituted or substituted with deuterium, a halogen(s), a (C1-C6)alkyl(s), a (C6-C15)aryl(s), a di(C6-C15)arylamino(s), or a (5- to 15-membered)heteroaryl(s); or a (5- to 15-membered)heteroaryl unsubstituted or substituted with a (C6-C15)aryl(s); or Ar_1 and Ar_2 , Ar_3 and Ar_4 , or Ar_5 and Ar_6 may be linked to each other to form an indole ring unsubstituted or substituted with a (C6-C15)aryl(s); or an indolocarbazole ring substituted with a (C6-C15)aryl(s) unsubstituted or substituted with a (C1-C4)alkyl(s), a phenyl(s) or a naphthyl(s), or a (5- to 15-membered)heteroaryl(s) unsubstituted or substituted with a phenyl(s); X represents -O-, -S-, -C(R_1)(R_2)- or -N(R_3)-; R_1 to R_3 , each independently, represent an unsubstituted (C1-C6)alkyl or an unsubstituted

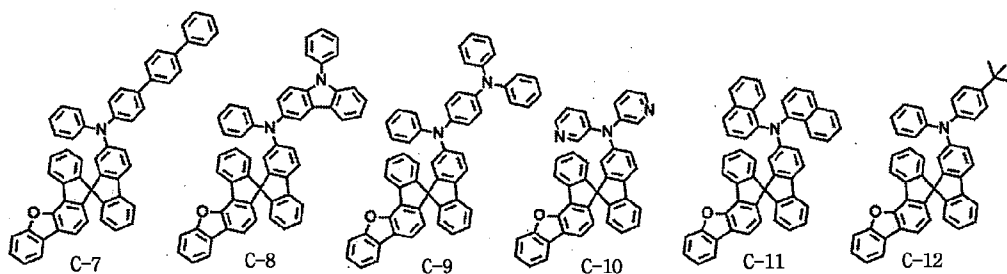
(C6-C15)aryl; or R₁ and R₂ may be linked to each other to form a cyclopentane ring or a cyclohexane ring; R₄ to R₈, each independently, represent hydrogen or an unsubstituted (C6-C15)aryl, or may be linked to an adjacent substituent(s) to form a benzene ring; a represents 1 or 4; c and d, each independently, represent an integer of 2 to 4; e represents 3 or 4; where a, c, d, or e is an integer of 2 or more, each of the substituents are the same or different; b represents 2; l, m, and n, each independently, represent an integer of 0 to 2; and l+m+n is 1 or 2.

[53] The specific compounds of the present invention include the following compounds, but are not limited thereto:

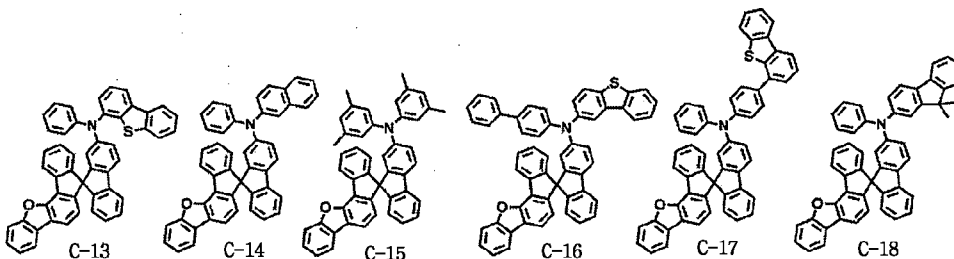
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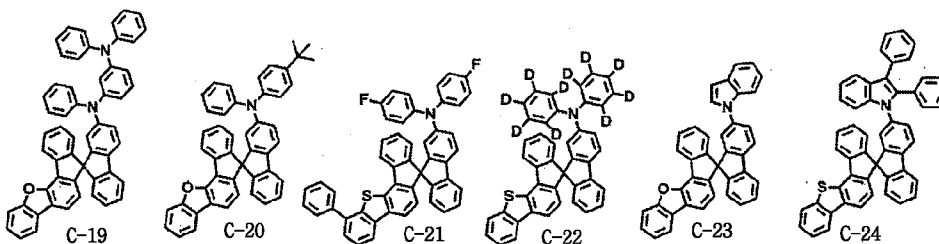
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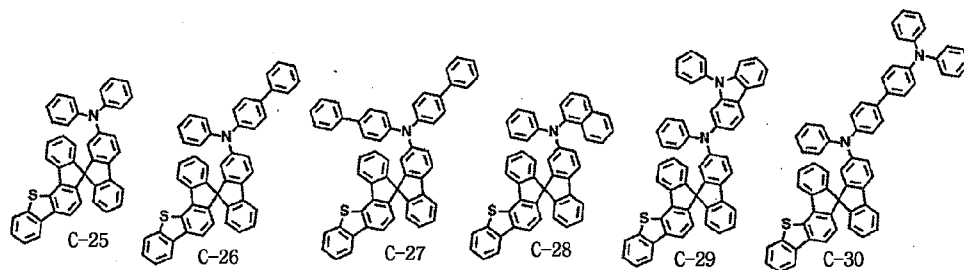
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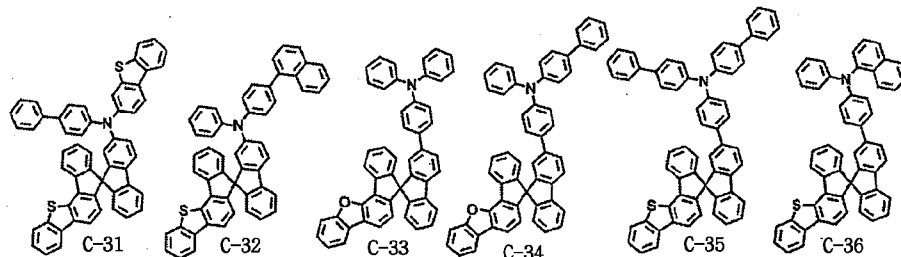
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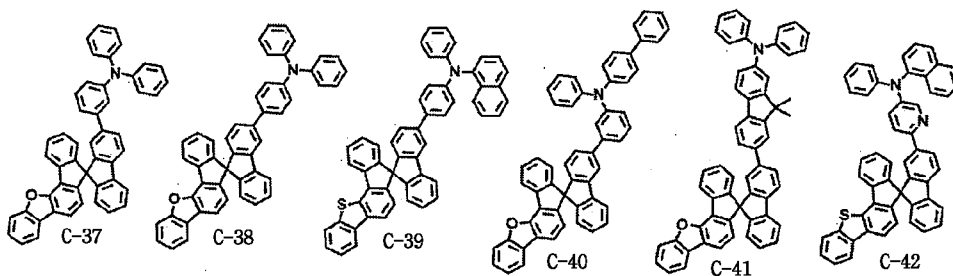
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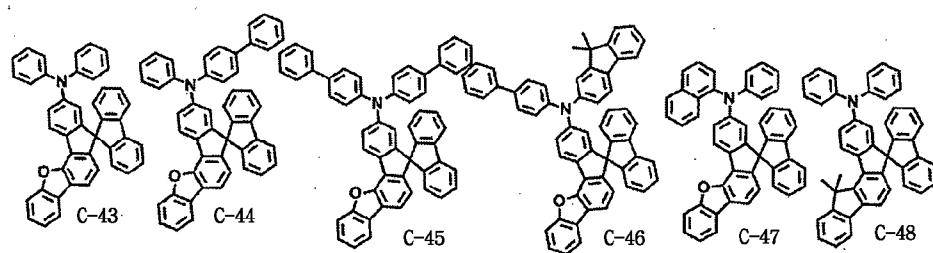
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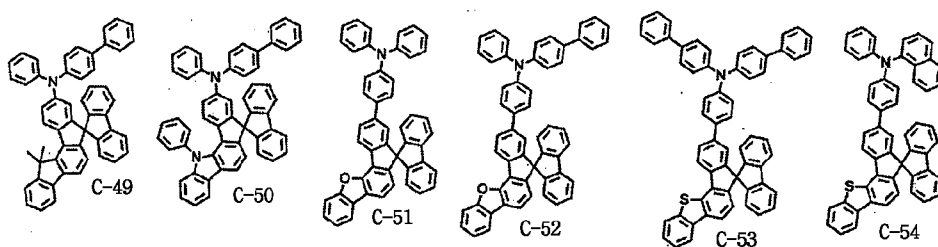
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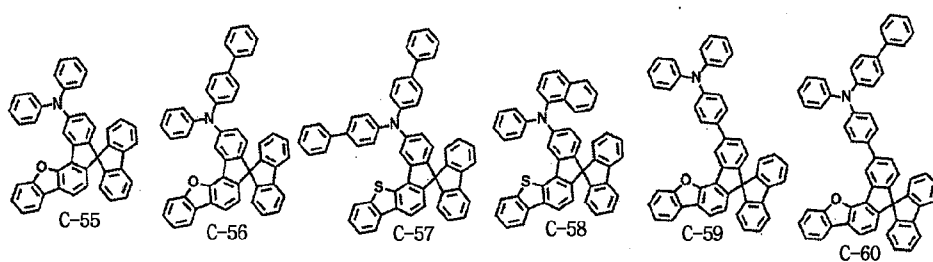
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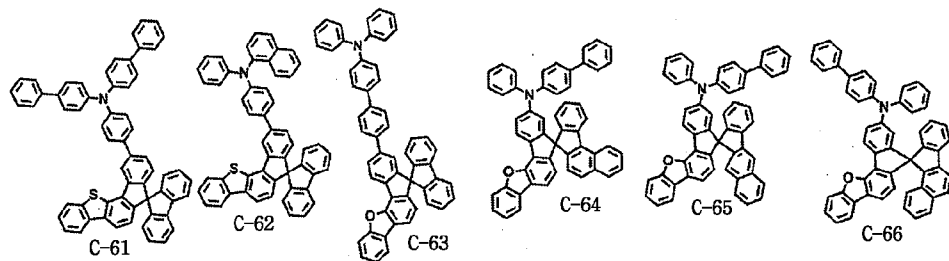
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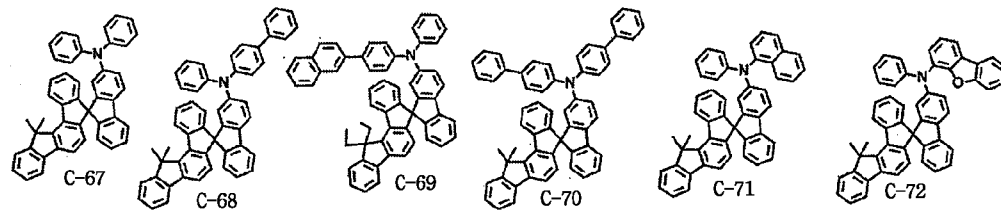
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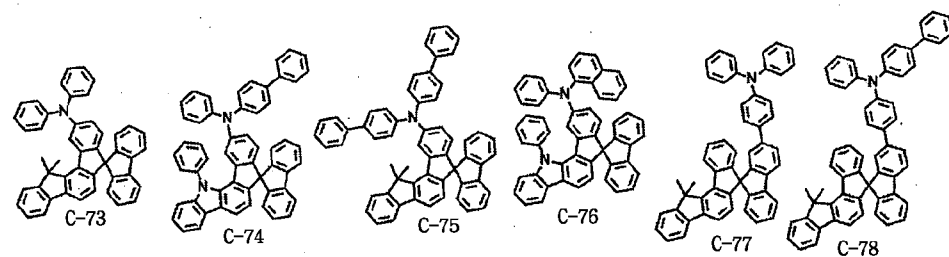
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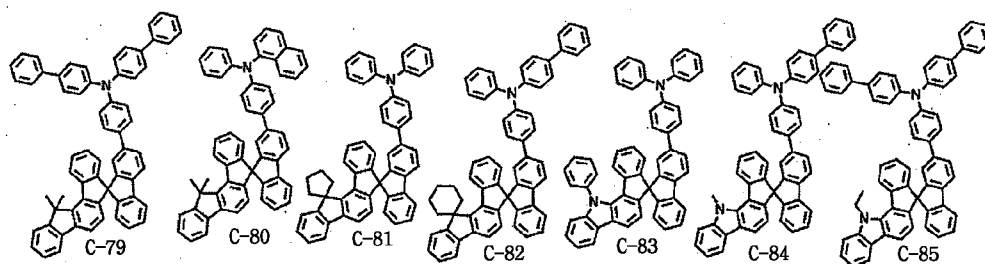
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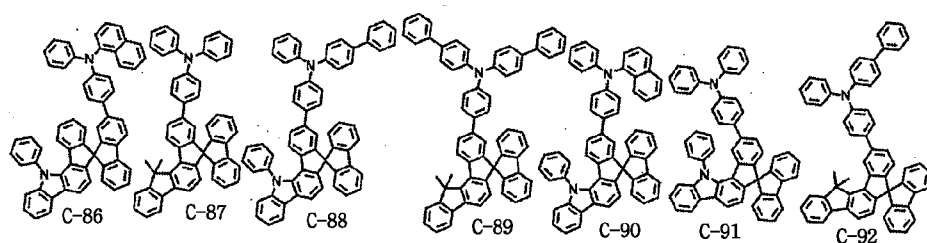
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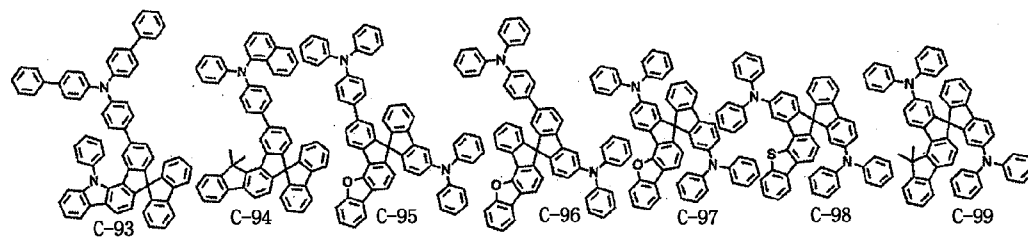
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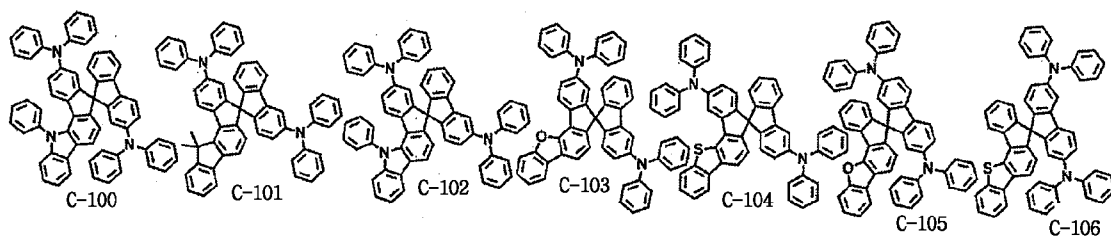
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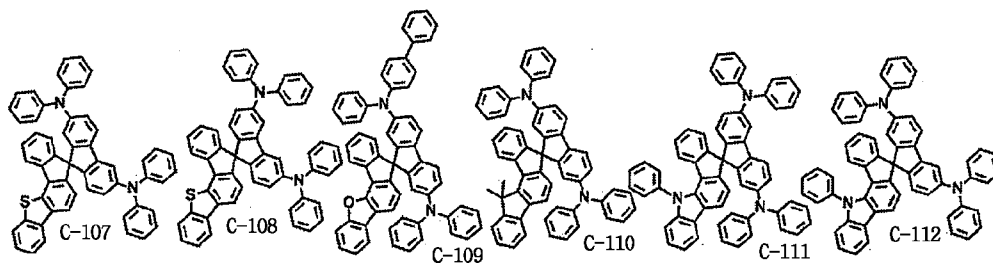
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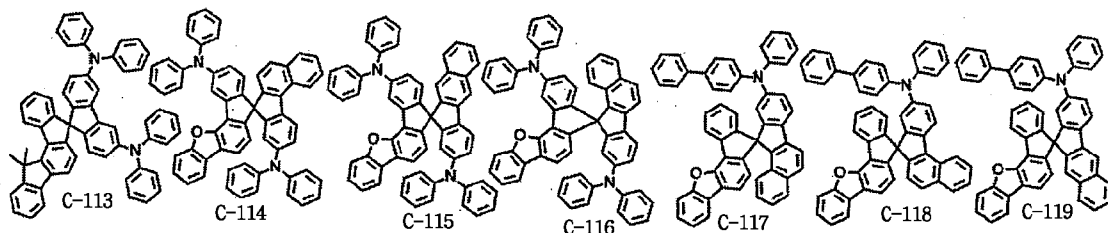
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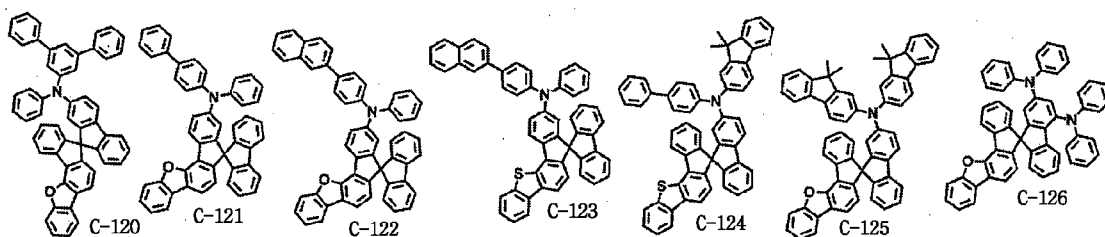
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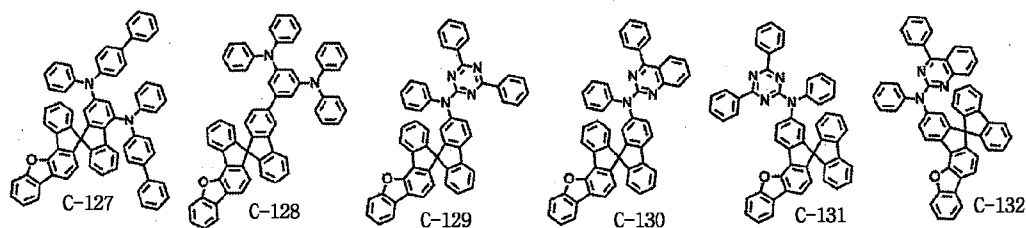
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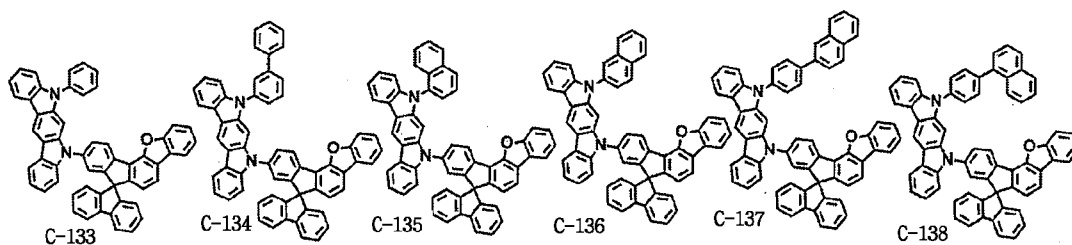
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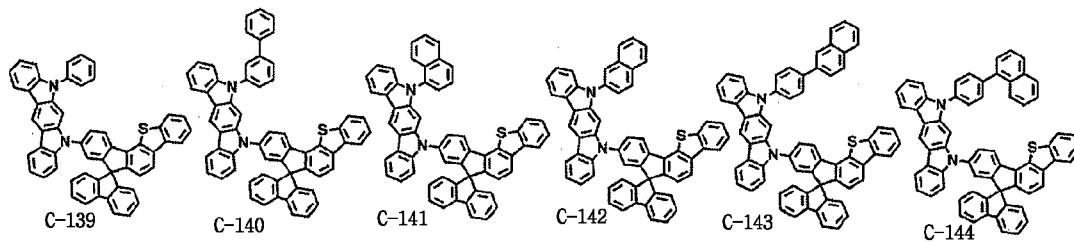
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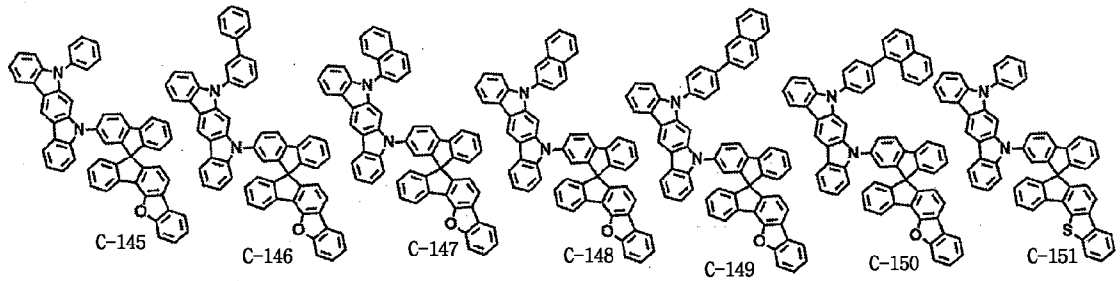
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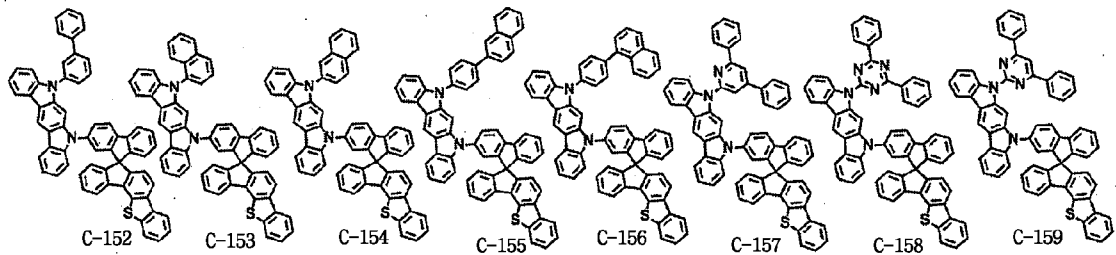
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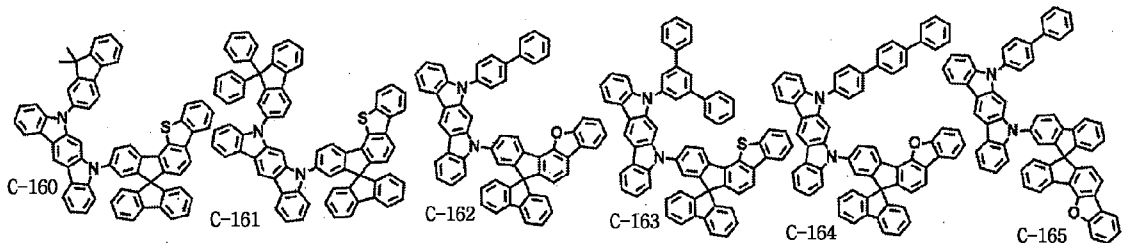
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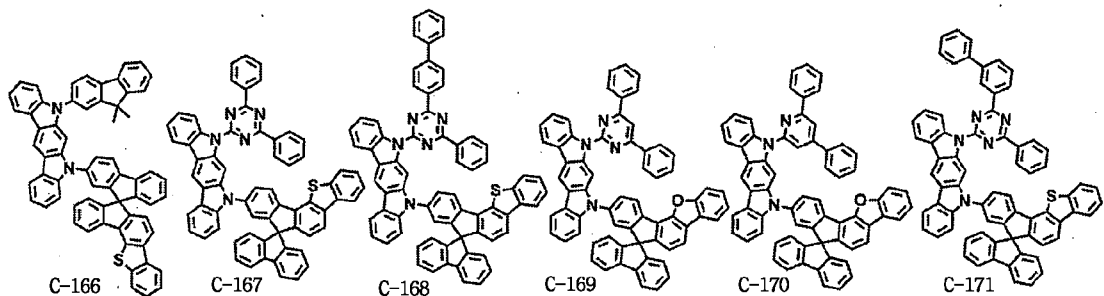
[78]



[79]



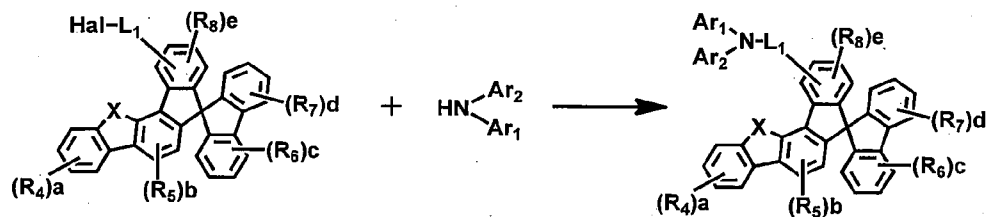
[80]



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

[82] [Reaction Scheme 1]

[83]



[84] wherein L₁, Ar₁, Ar₂, R₄ to R₈, and a to e are as defined in formula 1 above, and Hal represents a halogen.

[85] The present invention provides an organic electroluminescent material comprising the organic electroluminescent compound of formula 1, and an organic electrolu-

minescent device comprising the material.

[86] 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.

[87] The organic electroluminescent device comprises a first electrode; a second electrode; and at least one organic layer between the first and second electrodes. The organic layer may comprise at least one organic electroluminescent compound of formula 1.

[88] 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 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, a hole blocking layer, and an electron blocking layer.

[89] The organic electroluminescent compound according to the present invention can be comprised in at least one of the light-emitting layer and the hole transport layer. Where used in the hole transport layer, the organic electroluminescent compound represented by formula 1 can be comprised as a hole transport material. Where used in the light-emitting layer, the organic electroluminescent compound represented by formula 1 can be comprised as a host material.

[90] The organic electroluminescent device comprising the organic electroluminescent compound of the present invention can further comprise one or more compounds other than the organic electroluminescent compound according to the present invention as host materials, and can further comprise one or more dopants.

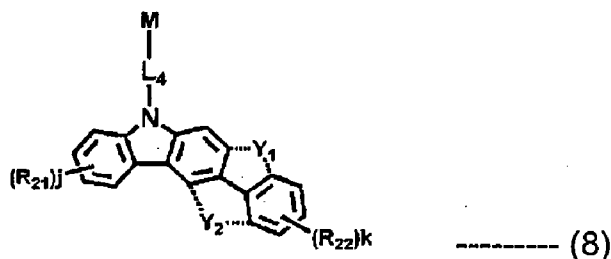
[91] Where the organic electroluminescent compound according to the present invention is comprised as a host material (first host material), the other compound may be comprised as a second host material. Herein, the ratio of the first host material to the second host material is in the range of 1:99 to 99:1.

[92] The host material other than the organic electroluminescent compound according to the present invention can be from any of the known fluorescent or phosphorescent hosts. Specifically, the phosphorescent host selected from the group consisting of the compounds of formulae 6 to 8 below is preferable in view of luminous efficiency.

[93] $H-(Cz-L_4)_n-M$ ----- (6)

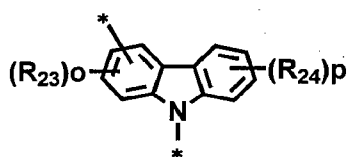
[94] $H-(Cz)_i-L_4-M$ ----- (7)

[95]



[96] wherein Cz represents the following structure;

[97]

[98] R₂₁ to R₂₄ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (3- to 30-membered)heteroaryl, or -SiR₂₅R₂₆R₂₇;[99] R₂₅ to R₂₇ each independently represent a substituted or unsubstituted (C1-C30)alkyl, or a substituted or unsubstituted (C6-C30)aryl;[100] L₄ represents a single bond, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (5- to 30-membered)heteroarylene;

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

[102] Y₁ and Y₂ each independently represent -O-, -S-, -N(R₃₁)- or -C(R₃₂)(R₃₃)-, provided that Y₁ and Y₂ do not simultaneously exist;[103] R₃₁ to R₃₃ each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl, and R₃₂ and R₃₃ may be the same or different;

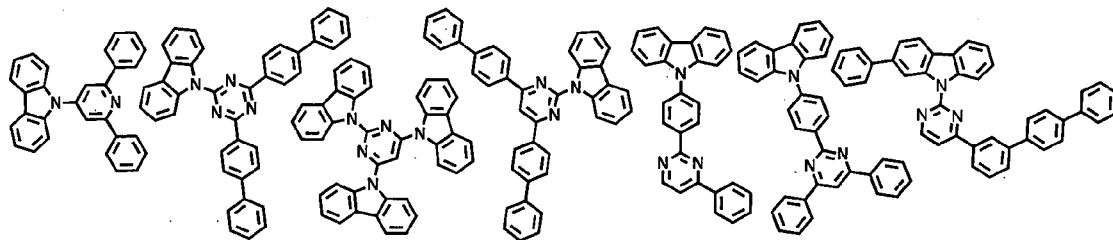
[104] h and i each independently represent an integer of 1 to 3;

[105] j, k, o and p each independently represent an integer of 0 to 4; and

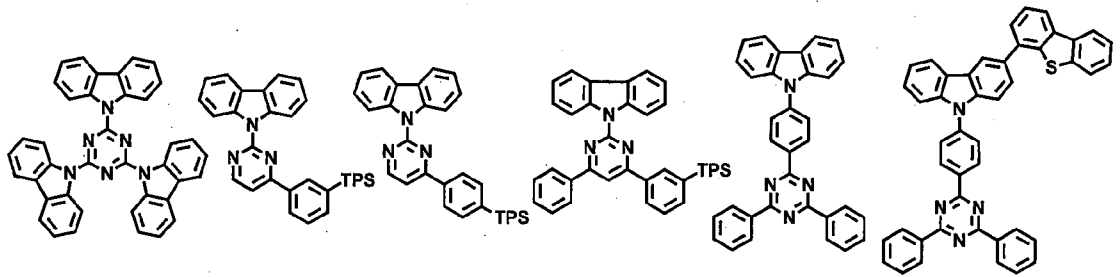
[106] where h, i, j, k, o or p is an integer of 2 or more, each of (Cz-L₄), each of (Cz), each of R₂₁, each of R₂₂, each of R₂₃ or each of R₂₄ may be the same or different.

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

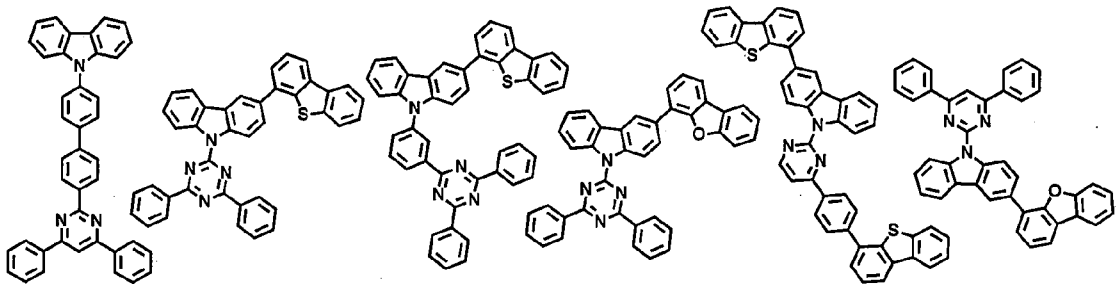
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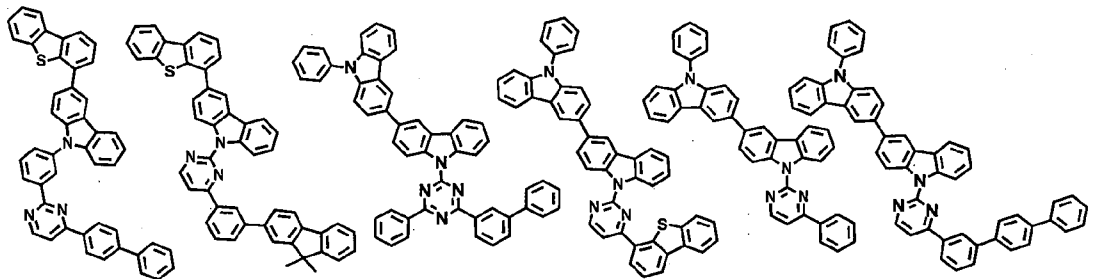
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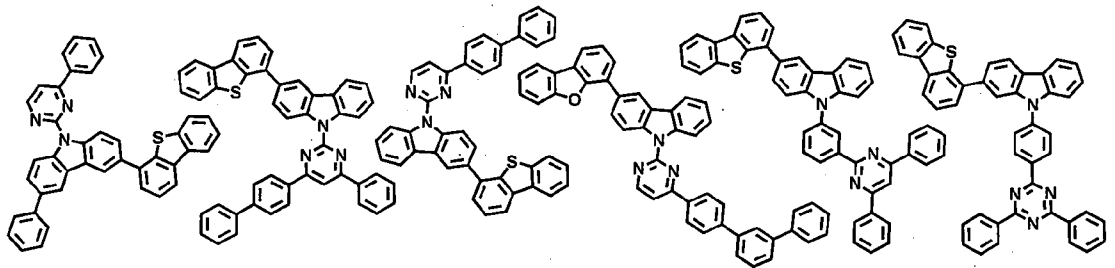
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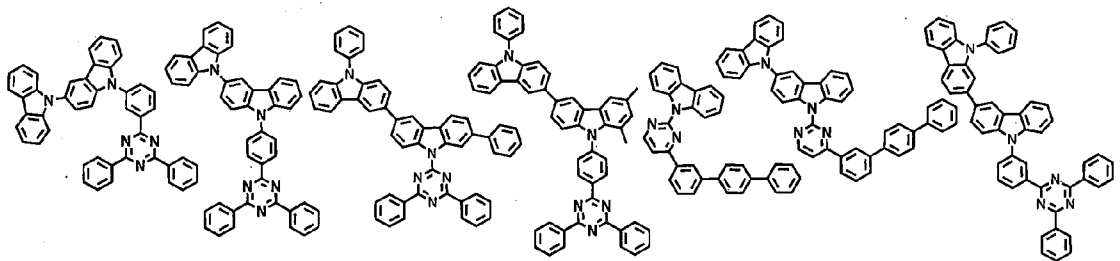
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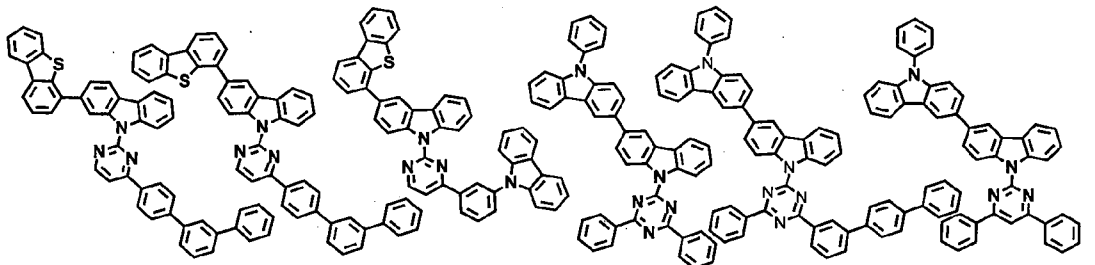
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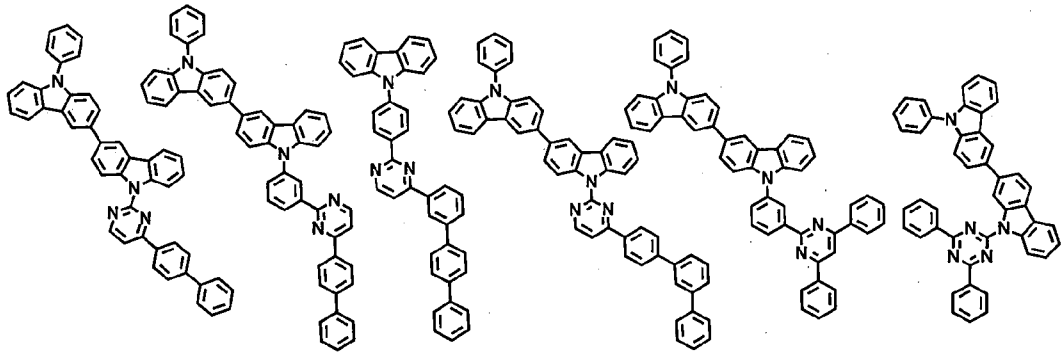
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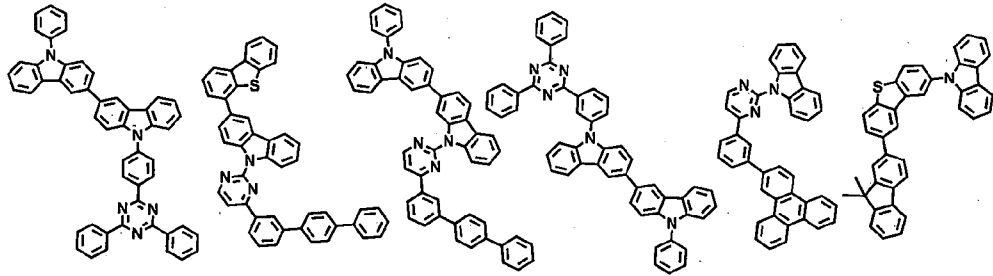
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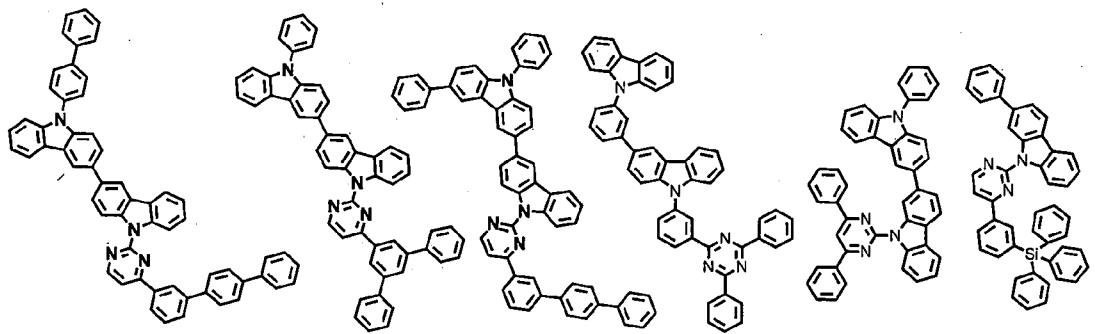
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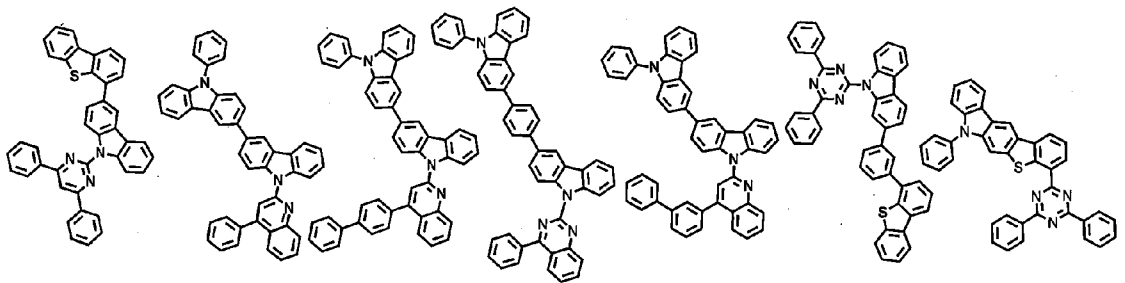
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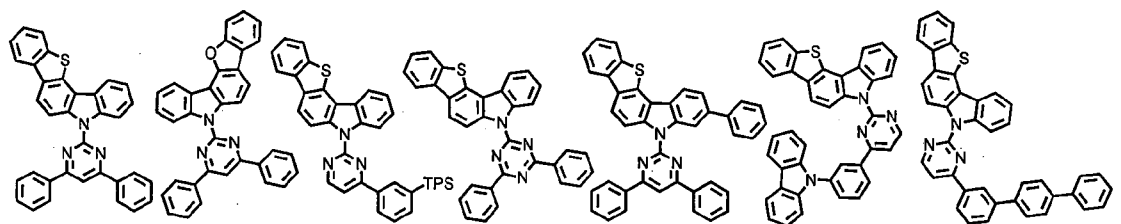
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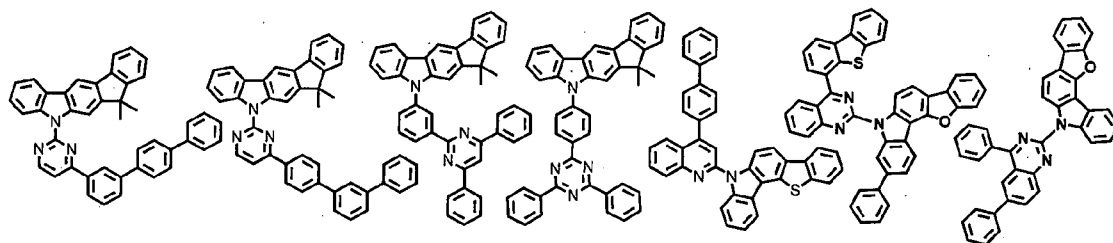
[118]



[119]



[120]

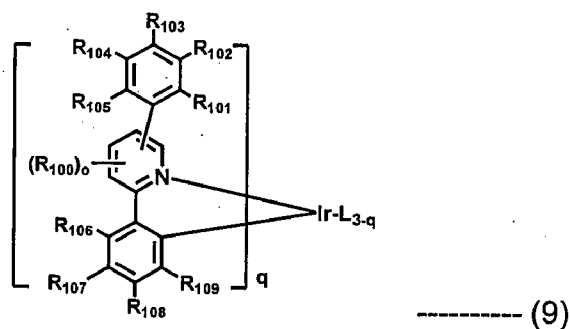


[121] [wherein TPS represents triphenylsilyl]

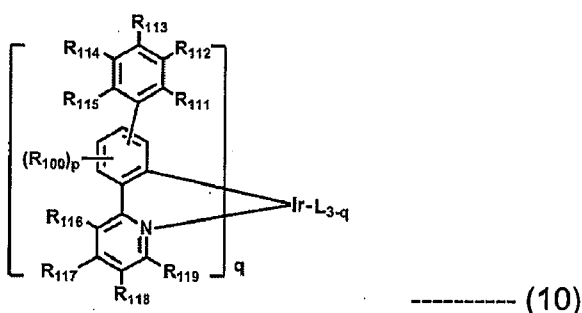
[122] The dopant comprised in the organic electroluminescent device according to the present invention can be a fluorescent or phosphorescent dopant, and preferably is at least one phosphorescent dopant. The dopant materials applied to the organic electroluminescent device according to the present invention are not limited, but may be preferably selected from metallated complex compounds of iridium, osmium, copper and platinum, more preferably selected from ortho-metallated complex compounds of iridium, osmium, copper and platinum, and even more preferably ortho-metallated iridium complex compounds.

[123] The phosphorescent dopants may be preferably selected from compounds represented by the following formulae 9 to 11.

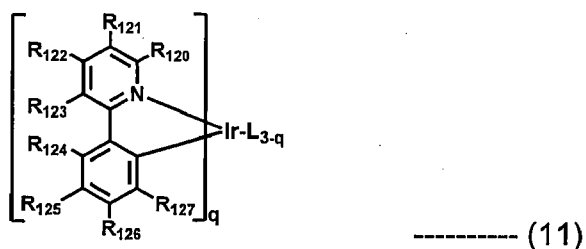
[124]



[125]

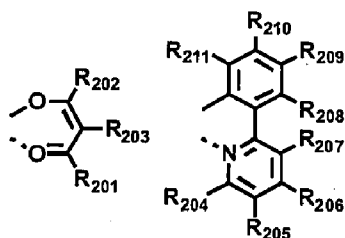


[126]



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

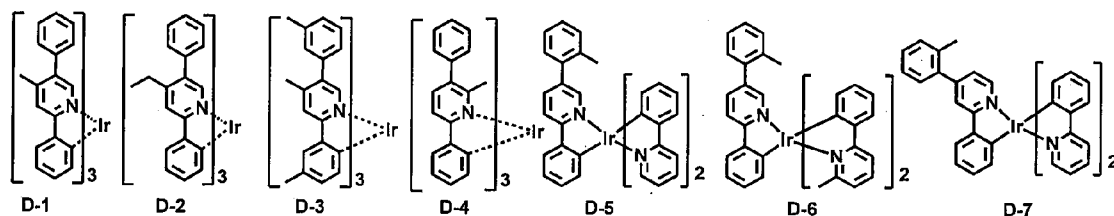
[128]

[129] R₁₀₀ represents hydrogen, or a substituted or unsubstituted (C1-C30)alkyl;[130] R₁₀₁ to R₁₀₉, and R₁₁₁ to R₁₂₃ each independently represent hydrogen; deuterium; a halogen; a (C1-C30)alkyl unsubstituted or substituted with a halogen(s); a cyano; a substituted or unsubstituted (C1-C30)alkoxy; or a substituted or unsubstituted (C3-C30)cycloalkyl; and adjacent substituents of R₁₂₀ to R₁₂₃ may be linked to each other to form a fused ring, e.g. quinoline;[131] R₁₂₄ to R₁₂₇ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, or a substituted or unsubstituted (C6-C30)aryl; where R₁₂₄ to R₁₂₇ are aryl groups, adjacent substituents may be linked to each other to form a fused ring, e.g. fluorene;[132] R₂₀₁ to R₂₁₁ each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl unsubstituted or substituted with a halogen(s), or a substituted or unsubstituted (C6-C30)aryl;[133] 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 may be the same or different; and

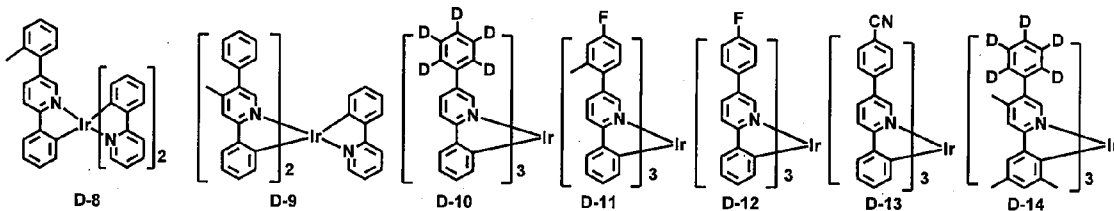
[134] q represents an integer of 1 to 3.

[135] Specifically, the phosphorescent dopant materials include the following:

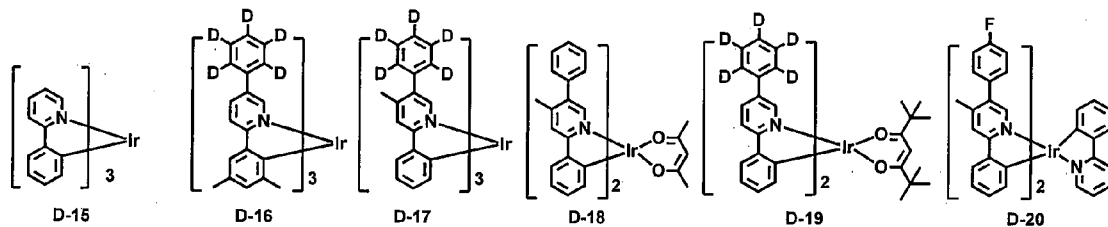
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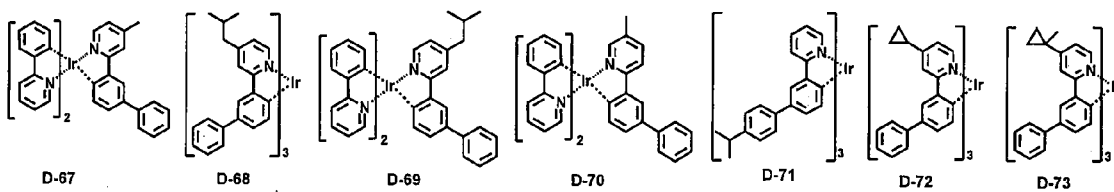
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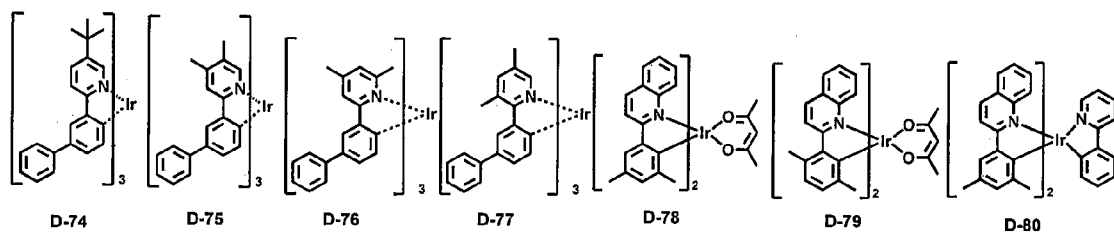
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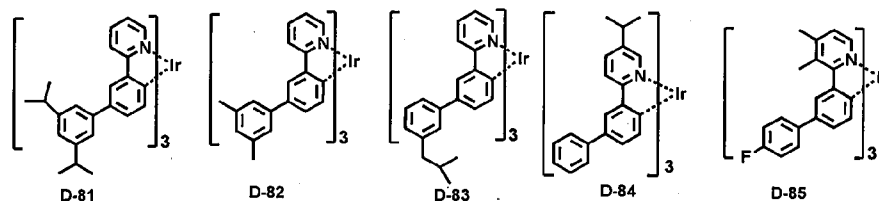
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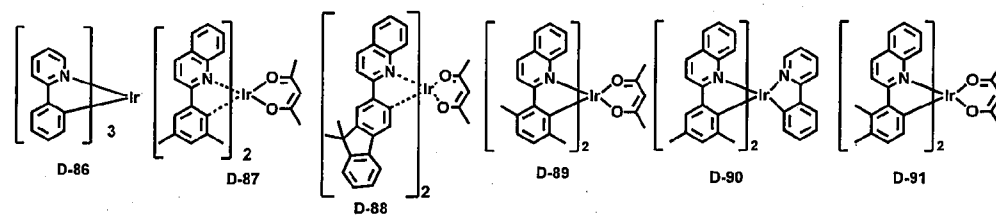
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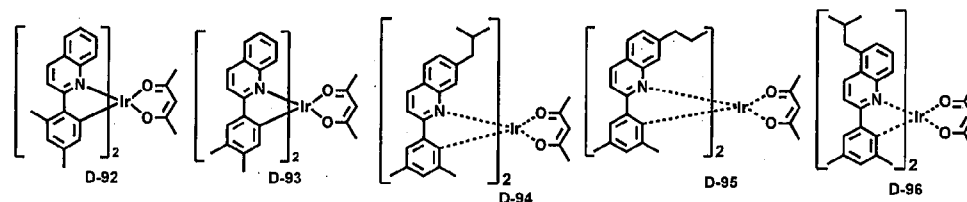
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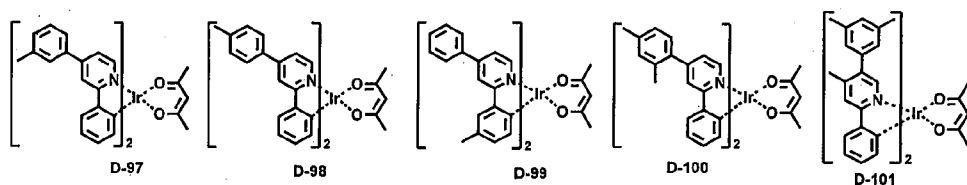
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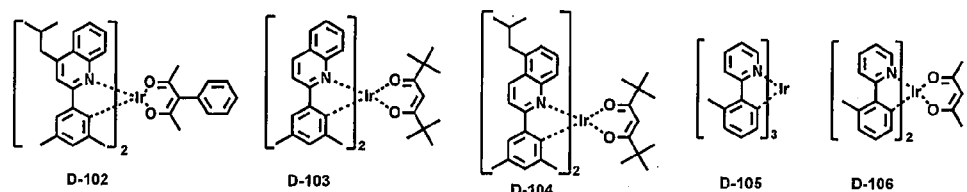
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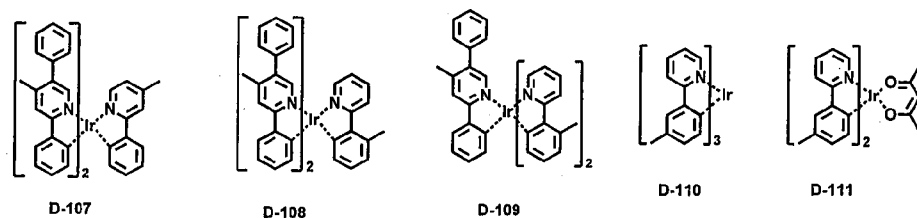
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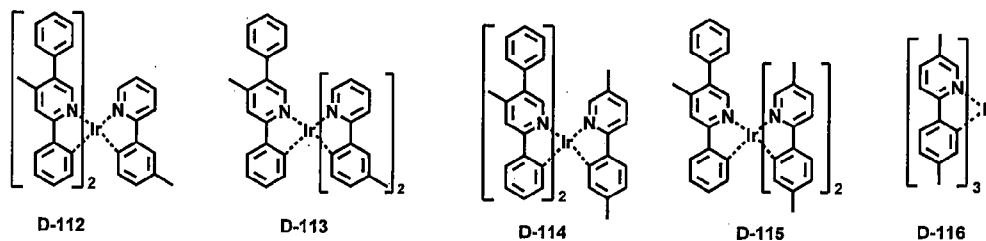
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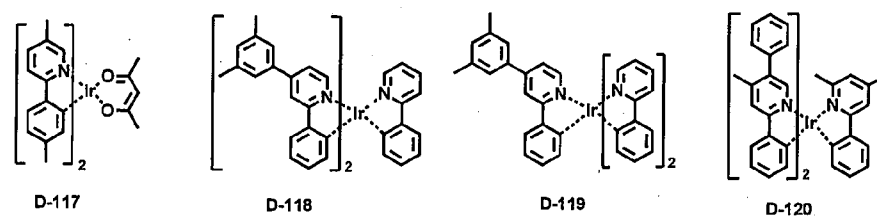
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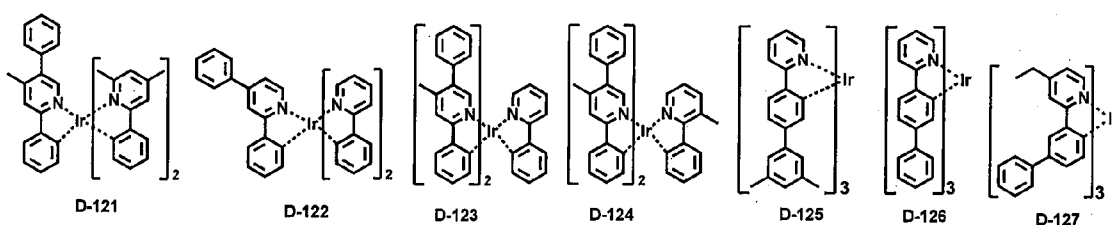
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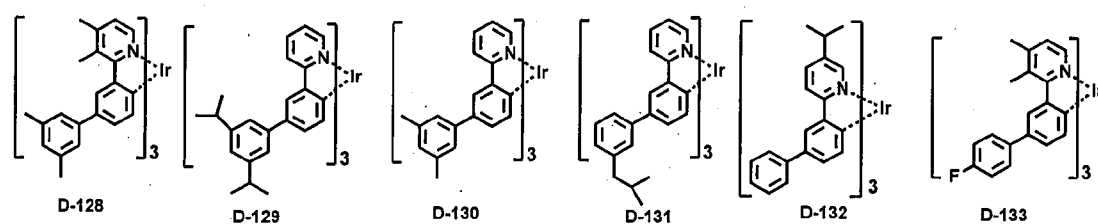
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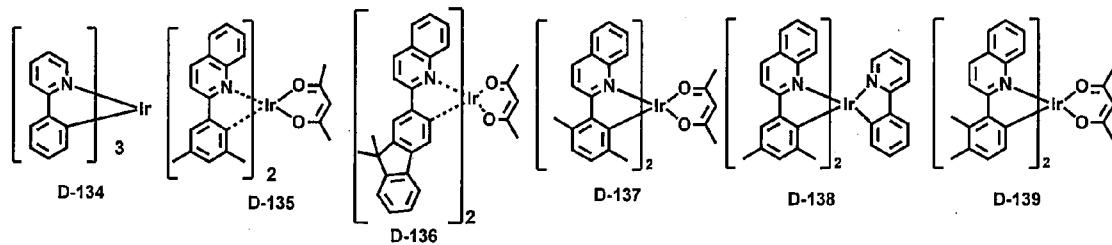
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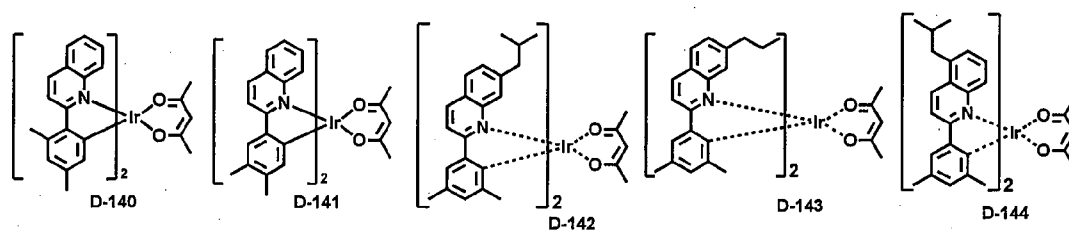
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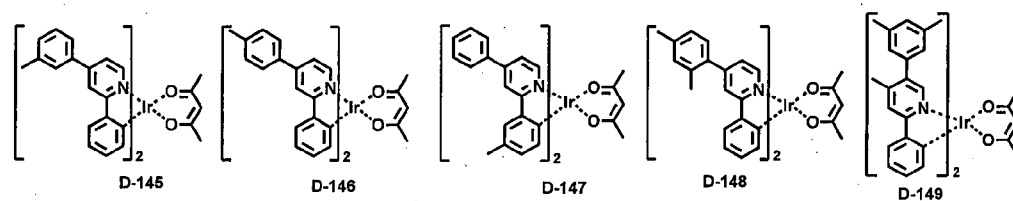
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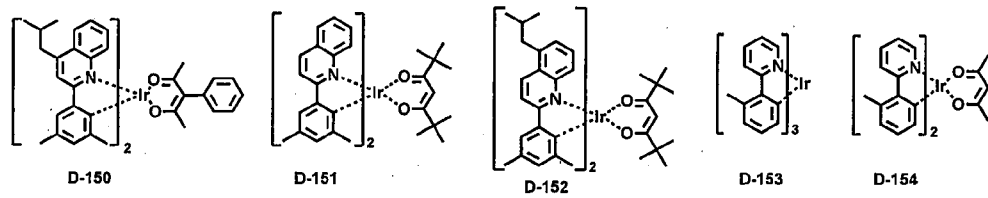
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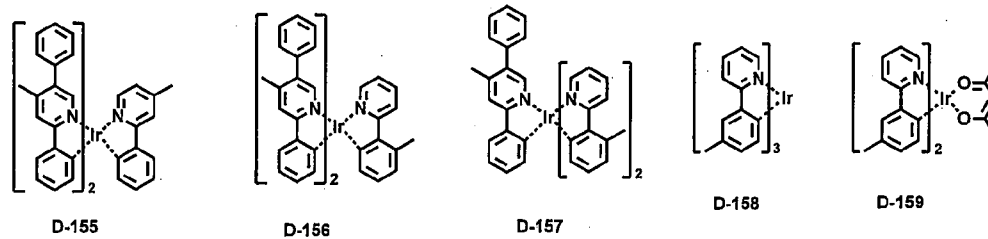
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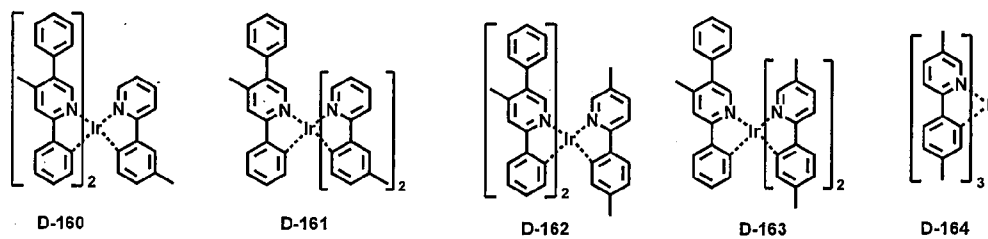
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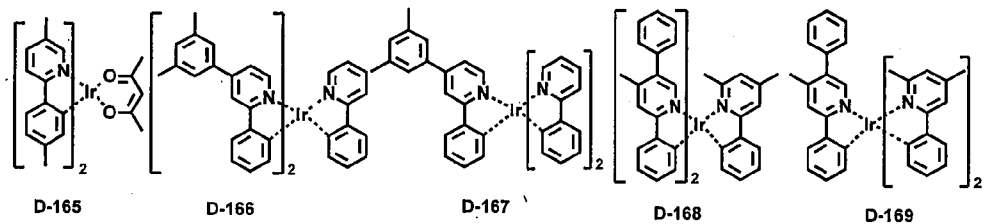
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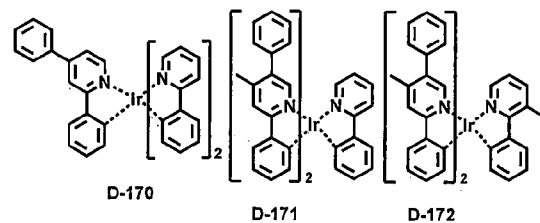
[163]



[164]



[165]



[166] In another embodiment of the present invention, a composition for an organic electroluminescent device is provided. The composition comprises the compound according to the present invention as a host material or a hole transport material.

[167] In addition, the organic electroluminescent device according to the present invention comprises a first electrode; a second electrode; and at least one organic layer between the first and second electrodes. The organic layer comprises a light-emitting layer, and the light-emitting layer may comprise the composition for the organic electroluminescent device according to the present invention.

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

- [169] 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 further comprise a light-emitting layer and a charge generating layer.
- [170] 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 compound according to the present invention. Also, if needed, a yellow or orange light-emitting layer can be comprised in the device.
- [171] According to the present invention, at least one layer (hereinafter, "a surface layer") is preferably placed on an inner surface(s) of one or both electrode(s); selected from a chalcogenide layer, a metal halide layer and a metal oxide layer. Specifically, a chalcogenide(includes oxides) layer of silicon or aluminum is preferably placed on an anode surface of an electroluminescent medium layer, and a metal halide layer or a metal oxide layer is preferably placed on a cathode surface of an electroluminescent medium layer. Such a surface layer provides operation stability for the organic electroluminescent device. Preferably, said chalcogenide includes SiO_x ($1 \leq X \leq 2$), AlO_x ($1 \leq X \leq 1.5$), 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.
- [172] 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 is preferably 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.
- [173] In order to form each layer 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, dip coating, and flow coating methods can be used.

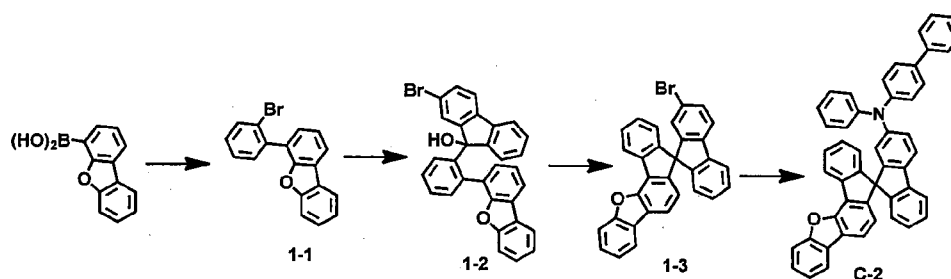
[174] When using a wet film-forming method, a thin film can be formed by dissolving or diffusing materials forming each layer into any suitable solvent such as ethanol, chloroform, tetrahydrofuran, dioxane, etc. The solvent can be any solvent where the materials forming each layer can be dissolved or diffused, and where there are no problems in film-formation capability.

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

[176]

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

[178]



[179] **Preparation of compound 1-1**

[180] After mixing 2-bromoiodobenzene (60 g, 212 mmol), 4-dibenzofuranboronic acid (30 g, 142 mmol), tetrakis(triphenylphosphine)palladium (3 g, 2.8 mmol), sodium carbonate (37 g, 354 mmol), toluene 1000 mL, and ethanol 200 mL in a reaction container, distilled water 200 mL was added to the mixture, and the mixture was stirred at 120°C for 6 hours. After the reaction, the mixture was washed with distilled water, and an organic layer was extracted with ethyl acetate. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 1-1 (32 g, 68%).

[181] **Preparation of compound 1-3**

[182] After mixing compound 1-1 (30 g, 93 mmol) and tetrahydrofuran 300 mL in a reaction container, the container was cooled to -78°C under nitrogen atmosphere. N-butyl lithium 48 mL (2.5 M, 120 mmol) was then slowly added dropwise to the mixture. After stirring the mixture for 2 hours at -78°C, 2-bromofluorenone dissolved in tetrahydrofuran 400 mL was slowly added dropwise to the mixture. After adding, the reaction temperature was slowly heated to room temperature, and the mixture was additionally stirred for 30 minutes. Ammonium chloride aqueous solution was then

added to the reaction solution to complete the reaction, and the mixture was extracted with ethylacetate. The obtained organic layer was then dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. Acetic acid 900 mL and HCl 0.5 mL were added to the produced compound 1-2, and the mixture was stirred at 120°C overnight. The solvent was then removed using a rotary evaporator, and the remaining substance was purified with column chromatography to obtain compound 1-3 (28 g, 75%).

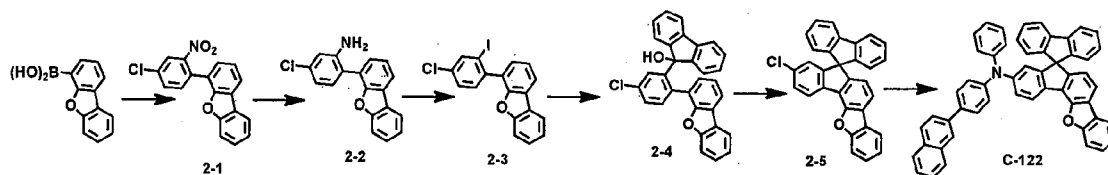
[183] Preparation of compound C-2

[184] After mixing compound 1-3 (10 g, 23.3 mmol), N-phenylbiphenyl-4-amine (6 g, 24.5 mmol), palladium (II) acetate (0.2 g, 0.98 mmol), tri-t-butyl phosphine 1 mL (50%, 2.45 mmol), sodium tert-butoxide (3.5 g, 37 mmol), and o-xylene 120 mL in a reaction container, the mixture was stirred for 8 hours under reflux. The reaction mixture was then cooled to room temperature, and the produced solid was filtered, and washed with methylene chloride (MC). The filtrate was then distilled under reduced pressure, and purified with column chromatography to obtain compound C-2 (13 g, 82%).

[185]

[186] Example 2: Preparation of compound C-122

[187]



[188] Preparation of compound 2-1

[189] After mixing 1-bromo-4-chloro-2-nitrobenzene (38 g, 161.82 mmol), 4-dibenzofuranboronic acid (37.7 g, 178 mmol), tetrakis(triphenylphosphine)palladium (5.6 g, 4.85 mmol), sodium carbonate (43 g, 404.55 mmol), toluene 800 mL, and ethanol 200 mL in a reaction container, distilled water 200 mL was added to the mixture, and the mixture was stirred at 120°C for 2 hours. After the reaction, the mixture was washed with distilled water, and an organic layer was extracted with ethyl acetate. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 2-1 (51 g, 97%).

[190] Preparation of compound 2-2

[191] After mixing compound 2-1 (51 g, 157.54 mmol), tin chloride (107 g, 472.62 mmol), and ethylacetate 1.6 L in a reaction container, the mixture was stirred for 4 hours under reflux. After the reaction, the mixture was washed with distilled water, and an organic layer was extracted with ethyl acetate. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain

compound 2-2 (31 g, 70%).

[192] Preparation of compound 2-3

[193] After dissolving compound 2-2 (28 g, 95.2 mmol) and para toluene sulfonic acid (56 g, 285.88 mmol) in acetonitrile in a reaction container, sodium nitrite (14 g, 190.68 mmol) and potassium iodide (39.2 g, 238.00 mmol) dissolved in water of 0°C were added to the mixture. The mixture was then stirred for 6 hours, washed with distilled water, and an organic layer was extracted with ethyl acetate. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 2-3 (30.4 g, 79%).

[194] Preparation of compound 2-5

[195] After mixing compound 2-3 (30.4 g, 75.12 mmol) and tetrahydrofuran 260 mL in a reaction container, the container was cooled to -78°C under nitrogen atmosphere. N-butyl lithium 40 mL (2.5 M, 97.68 mmol) was then slowly added dropwise to the mixture. After stirring the mixture for 2 hours at -78°C, fluorenone dissolved in tetrahydrofuran 260 mL was slowly added dropwise to the mixture. After adding, the reaction temperature was slowly heated to room temperature, and the mixture was additionally stirred for 30 minutes. Ammonium chloride aqueous solution was then added to the reaction solution to complete the reaction, and the mixture was extracted with ethylacetate. The obtained organic layer was then dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. Acetic acid 720 mL and HCl 72 mL were added to the produced compound 2-4, and the mixture was stirred at 120°C overnight. The solvent was then removed using a rotary evaporator, and the remaining substance was purified with column chromatography to obtain compound 2-5 (18 g, 54%).

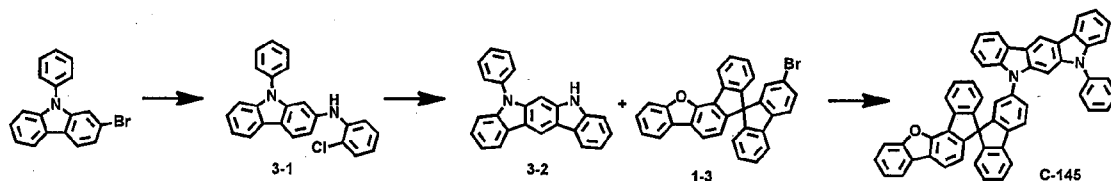
[196] Preparation of compound C-122

[197] After mixing compound 2-5 (4.5 g, 10.21 mmol), 4-(naphthalen-2-yl)-N-phenylaniline (3.3 g, 11.23 mmol), palladium (II) acetate (0.12 g, 0.51 mmol), 2-dicyclohexylphosphino-2',6'-dimethoxybiphenyl (0.4 g, 1.02 mmol), sodium tert-butoxide (2.0 g, 20.42 mmol), and o-xylene 51 mL in a reaction container, the mixture was stirred for 8 hours under reflux. The reaction mixture was then cooled to room temperature, the produced solid was filtered, and washed with methylene chloride (MC). The filtrate was then distilled under reduced pressure, and purified with column chromatography to obtain compound C-122 (4.1 g, 58%).

[198]

[199] Example 3: Preparation of compound C-145

[200]

[201] Preparation of compound 3-1

[202] After mixing 2-bromo-9-phenylcarbazole (43 g, 133 mmol), 2-chloroaniline (20.7 mL, 200 mmol), palladium acetate (1.2 g, 5.33 mmol), tri-*t*-butyl phosphine 5.2 mL (50%, 10.6 mmol), sodium *tert*-butoxide (32 g, 333 mmol), and toluene 380 mL in a reaction container, the mixture was stirred for 3 hours under reflux. After the reaction, the mixture was washed with distilled water, and an organic layer was extracted with methylene chloride. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 3-1 (47 g, 96%).

[203] Preparation of compound 3-2

[204] After mixing compound 3-1 (47 g, 127 mmol), palladium acetate (1.4 g, 6.38 mmol), tricyclohexylphosphonium tetrafluoroborate (4.7 g, 12.7 mmol), cesium carbonate (124 g, 383 mmol), and dimethylacetamide 600 mL in a reaction container, the mixture was stirred for 3 hours under reflux. After the reaction, the mixture was washed with distilled water, and an organic layer was extracted with methylene chloride. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 3-2 (25 g, 59%).

[205] Preparation of compound C-145

[206] After mixing compound 1-3 (7.3 g, 15.0 mmol), compound 3-2 (5 g, 15.0 mmol), copper iodide (1.4 g, 7.52 mmol), ethylenediamine (2 mL, 30.0 mmol), cesium carbonate (12.2 g, 37.6 mmol), and *ortho*-xylene 75 mL in a reaction container, the mixture was stirred for 3 hours under reflux. After the reaction, the mixture was washed with distilled water, and an organic layer was extracted with methylene chloride. The extracted organic layer was dried using magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound C-145 (8.1 g, 73%).

[207] Compounds C-1 to C-171 were prepared by the same method as in Examples 1 to 3. The detailed data of the representative compounds are shown in Table 1 below.

[208]

[209] [Table 1]

[210]

Compound	Yield (%)	UV Spectrum	PL Spectrum	MP(°C)	MS/EIMS	
		(in toluene, nm)	(in toluene, nm)		found	calculated
C-2	82	344	399	269	650.2	649.2
C-3	45	396	411	270	699.8	699.3
C-4	82	344	399	292	725.9	725.3
C-5	72	334	407	208	765.9	265.3
C-26	46	388	399	252	665.8	665.2
C-27	68	334	399	303	742	741.3
C-44	62	398	409	270	649.8	649.2
C-45	29	402	413	232	725.9	725.3
C-105	66	382	421	152	740.3	740.01
C-120	75	376	404	163	725.9	725.3
C-121	80	378	402	269	699.8	699.3
C-122	58	397	413	271	699.8	699.3
C-123	71	388	409	225	715.9	715.2
C-124	16	496	407	193	782	781.3
C-125	81	332	410	302	860	805.3
C-145	73	360	390	215	736.9	736.25

[211]

Device Example 1: Production of an OLED device using the organic

[212]

electroluminescent compound according to the present invention

[213]

An OLED device was produced using the light emitting material according to the present invention. A transparent electrode indium tin oxide (ITO) thin film (15 Ω/sq) on a glass substrate for an organic light-emitting diode (OLED) device (Geomatec, Japan) was subjected to an ultrasonic washing with acetone and isopropanol alcohol, sequentially, and then was stored in isopropanol alcohol. Then, the ITO substrate was mounted on a substrate holder of a vacuum vapor depositing apparatus. N¹,N^{1'}-([1,1'-biphenyl]-4,4'-diyl)bis(N¹-(naphthalen-1-yl)-N⁴,N^{4'}-diphenylbenzen-1,4-diamine) was introduced into a cell of said vacuum vapor depositing apparatus, and then the pressure in the chamber of said apparatus was controlled to 10⁻⁶ torr. Thereafter, an electric current was applied to the cell to evaporate the above introduced material, thereby forming a hole injection layer having a thickness of 60 nm on the ITO substrate. Then, compound **C-2** according to the present invention 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, 9-(3-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl)-9'-phenyl-9H,9'H-3,3'-bicarbazole was introduced into one cell of the vacuum vapor depositing apparatus, as a host material, and compound **D-1** was introduced into another cell as a dopant. The two materials were evaporated at different rates and were deposited in a doping amount of 15 wt% based on the total amount of the host and dopant to form a light-emitting layer having a

thickness of 30 nm on the hole transport layer. Then, 2-(4-(9,10-di(naphthalene-2-yl)anthracen-2-yl)phenyl)-1-phenyl-1H-benzimidazole was introduced into one cell and lithium quinolate was introduced into another cell. The two materials were evaporated at the same rate and were deposited in a doping amount of 50 wt% each to form an electron transport layer having a thickness of 30 nm on the light-emitting layer. Then, after depositing lithium quinolate as an electron injection layer having a thickness of 2 nm on the electron transport layer, an Al cathode having a thickness of 150 nm was deposited by another vacuum vapor deposition apparatus on the electron injection layer. Thus, an OLED device was produced. All the materials used for producing the OLED device were purified by vacuum sublimation at 10^{-6} torr prior to use.

[214] The produced OLED device showed a green emission having a luminance of 1100 cd/m² and a current density of 2.4 mA/cm².

[215]

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

[217] electroluminescent compound according to the present invention

[218] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **C-4** to form a hole transport layer in a thickness of 20 nm.

[219] The produced OLED device showed a green emission having a luminance of 2200 cd/m² and a current density of 4.9 mA/cm².

[220]

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

[222] electroluminescent compound according to the present invention

[223] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **C-27** to form a hole transport layer in a thickness of 20 nm.

[224] The produced OLED device showed a green emission having a luminance of 1500 cd/m² and a current density of 3.3 mA/cm².

[225]

[226] Device Example 4: Production of an OLED device using the organic electroluminescent compound according to the present invention

[227] electroluminescent compound according to the present invention

[228] An OLED device was produced in the same manner as in Device Example 1, except for forming a hole transport layer having a thickness of 20 nm by using compound **C-44**; introducing

9-phenyl-3-(4-(9-(4-phenylquinazolin-2-yl)-9H-carbazol-3-yl)phenyl)-9H-carbazole into one cell of the vacuum vapor depositing apparatus as a host, introducing compound **D-37** into another cell as a dopant, and evaporating the two materials at different rates and depositing them in a doping amount of 3 wt% based on the total amount of the host and dopant to form a light-emitting layer having a thickness of 30

nm on the hole transport layer.

[229] The produced OLED device showed a red emission having a luminance of 1800 cd/m² and a current density of 13.2 mA/cm².

[230]

[231] Device Example 5: Production of an OLED device using the organic electroluminescent compound according to the present invention

[232] An OLED device was produced in the same manner as in Device Example 4, except for evaporating compound **C-125** to form a hole transport layer in a thickness of 20 nm.

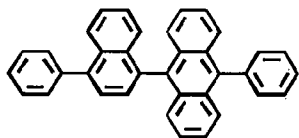
[234] The produced OLED device showed a red emission having a luminance of 1100 cd/m² and a current density of 8.3 mA/cm².

[235]

[236] Device Example 6: Production of an OLED device using the organic electroluminescent compound according to the present invention

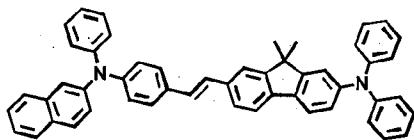
[237] An OLED device was produced in the same manner as in Device Example 1, except for using compound **C-3** for the hole transport layer; compound **H-1** as below for the host; and compound **H-2** as below for the dopant.

[239]



----- (H-1)

[240]



----- (H-2)

[241] The produced OLED device showed a blue emission having a luminance of 1200 cd/m² and a current density of 28.6 mA/cm².

[242]

[243] Device Example 7: Production of an OLED device using the organic electroluminescent compound according to the present invention

[244] An OLED device was produced in the same manner as in Device Example 6, except for evaporating compound **C-5** to form a hole transport layer in a thickness of 20 nm.

[246] The produced OLED device showed a blue emission having a luminance of 800 cd/m² and a current density of 17.8 mA/cm².

[247]

[248] Device Example 8: Production of an OLED device using the organic electroluminescent compound according to the present invention

[249] An OLED device was produced in the same manner as in Device Example 6, except

[250]

for evaporating compound **C-123** to form a hole transport layer in a thickness of 20 nm.

[251] The produced OLED device showed a blue emission having a luminance of 900 cd/m² and a current density of 20.5 mA/cm².

[252]

[253] Device Example 9: Production of an OLED device using the organic

[254] electroluminescent compound according to the present invention

[255] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **C-145** to form a hole transport layer in a thickness of 20 nm.

[256] The produced OLED device showed a green emission having a luminance of 1600 cd/m² and a current density of 3.2 mA/cm².

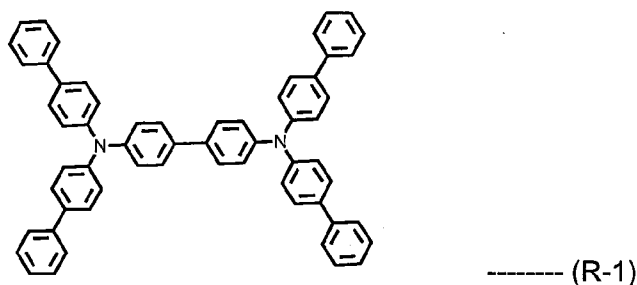
[257]

[258] Comparative Example 1: Production of an OLED device using a conventional

[259] organic electroluminescent compound

[260] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **R-1** as below to form a hole transport layer in a thickness of 20 nm.

[261]



[262] The produced OLED device showed a green emission having a luminance of 11400 cd/m² and a current density of 30.7 mA/cm².

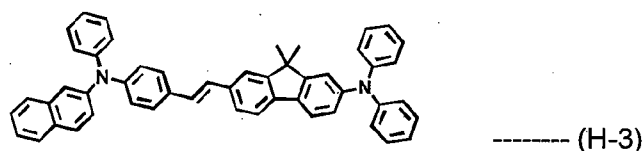
[263]

[264] Comparative Example 2: Production of an OLED device using a conventional

[265] organic electroluminescent compound

[266] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **R-1** to form a hole transport layer in a thickness of 20 nm; using compound **H-1** for the host, and using compound **H-3** as below for the dopant of the light-emitting material to form a light-emitting layer having a thickness of 30 nm on the hole transport layer.

[267]



[268] The produced OLED device showed a blue emission having a luminance of 3500 cd/m² and a current density of 100 mA/cm².

[269]

[270] Comparative Example 3: Production of an OLED device using a conventional organic electroluminescent compound

[271] organic electroluminescent compound

[272] An OLED device was produced in the same manner as in Device Example 4, except for evaporating compound **R-1** to form a hole transport layer in a thickness of 20 nm.

[273] The produced OLED device showed a red emission having a luminance of 4800 cd/m² and a current density of 57.8 mA/cm².

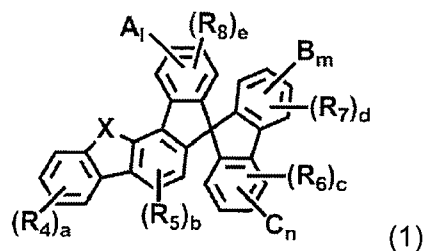
[274]

[275] It is verified that the luminous characteristics of the organic electroluminescent compound according to the present invention is superior to the conventional compounds. In addition, an organic electroluminescent device using the organic electroluminescent compound according to the present invention has excellent luminous characteristics, especially luminescent efficiency and current efficiency.

Claims

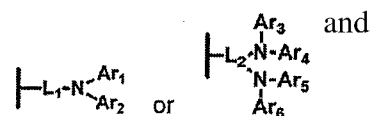
[Claim 1]

An organic electroluminescent compound represented by the following formula 1:



wherein

A, B, and C, each independently, represent



each of A, B, and C are the same or different;

L₁ represents a single bond, a substituted or unsubstituted (C1-C30)alkylene, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (5- to 30-membered)heteroarylene;

L₂ represents a tertiary residue derived from a substituted or unsubstituted (C1-C30) acyclic hydrocarbon, a substituted or unsubstituted (C6-C30) aromatic hydrocarbon ring, or a substituted or unsubstituted (5- to 30-membered) aromatic heterocyclic ring;

Ar₁ to Ar₆, each independently, represent a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl; or Ar₁ and Ar₂, Ar₃ and Ar₄, or Ar₅ and Ar₆ may be linked to each other to form a substituted or unsubstituted (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

X represents -O-, -S-, -C(R₁)(R₂)- or -N(R₃)-;

R₁ to R₃, each independently, represent hydrogen, deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (C3-C30)cycloalkenyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl; or R₁ and R₂ may be linked to each other to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic

ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

R₄ to R₈, each independently, represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (5- to 30-membered)heteroaryl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl(C1-C30)alkyl, -N(R₁₀)(R₁₁), -Si(R₁₂)(R₁₃)(R₁₄), -S(R₁₅), -O(R₁₆), a cyano, a nitro, or a hydroxyl; or may be linked to an adjacent substituent(s) to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

R₁₀ to R₁₆, each independently, represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (5- to 30-membered)heteroaryl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, or a substituted or unsubstituted (C3-C30)cycloalkyl; or may be linked to an adjacent substituent(s) to form a (3- to 30-membered) mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

a, c, d, and e, each independently, represent an integer of 1 to 4; where a, c, d, or e is an integer of 2 or more, each of the substituents are the same or different;

b represents 1 or 2;

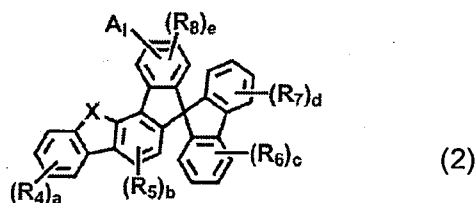
l, m, and n, each independently, represent an integer of 0 to 2;

l+m+n is 1 or more; and

the heterocycloalkyl and the heteroaryl(ene), each independently, 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 compound is represented by the following formula 2:

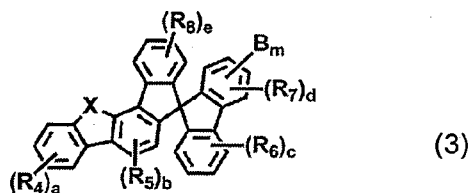


wherein

A, X, R₄ to R₈, a, b, c, d, and e are as defined in claim 1; and l is 1 or 2.

[Claim 3]

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

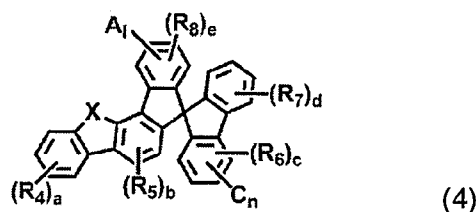


wherein

B, X, R₄ to R₈, a, b, c, d, and e are as defined in claim 1; and m is 1 or 2.

[Claim 4]

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

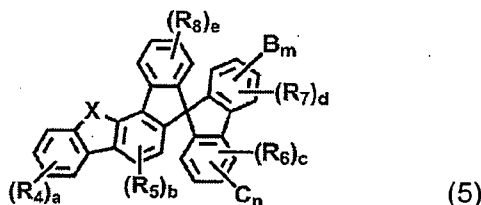


wherein

A, C, X, R₄ to R₈, a, b, c, d, and e are as defined in claim 1; and l and n, each independently, are 1.

[Claim 5]

The organic electroluminescent compound according to claim 1, wherein the compound is represented by the following formula 5:



wherein

B, C, X, R₄ to R₈, a, b, c, d, and e are as defined in claim 1; and m and n, each independently, are 1.

[Claim 6]

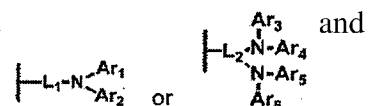
The organic electroluminescent compound according to claim 1, wherein the substituents of the substituted (C1-C30)alkyl(ene), the substituted (C3-C30)cycloalkyl, the substituted (C3-C30)cycloalkenyl, the substituted (3- to 7-membered)heterocycloalkyl, the substituted (C6-C30)aryl(ene), the substituted (5- to 30-membered)heteroaryl(ene), and the substituted (C6-C30)aryl(C1-C30)alkyl in R₁ to R₈, R₁₀ to R₁₆,

L_1 , L_2 , and Ar_1 to Ar_6 , each independently, are at least one selected from the group consisting of deuterium, a halogen, a cyano, a carboxyl, a nitro, a hydroxyl, a (C1-C30)alkyl, a halo(C1-C30)alkyl, a (C2-C30)alkenyl, a (C2-C30)alkynyl, a (C1-C30)alkoxy, a (C1-C30)alkylthio, a (C3-C30)cycloalkyl, a (C3-C30)cycloalkenyl, a (3- to 7-membered)heterocycloalkyl, a (C6-C30)aryloxy, a (C6-C30)arylthio, a (5- to 30-membered)heteroaryl unsubstituted or substituted with a (C6-C30)aryl(s), a (C6-C30)aryl unsubstituted or substituted with a (5- to 30-membered)heteroaryl(s), a tri(C1-C30)alkylsilyl, a tri(C6-C30)arylsilyl, a di(C1-C30)alkyl(C6-C30)arylsilyl, a (C1-C30)alkyldi(C6-C30)arylsilyl, an amino, a mono- or di-(C1-C30)alkylamino, a mono- or di-(C6-C30)arylamino, a (C1-C30)alkyl(C6-C30)arylamino, a (C1-C30)alkylcarbonyl, a (C1-C30)alkoxycarbonyl, a (C6-C30)arylcarbonyl, a di(C6-C30)arylboronyl, a di(C1-C30)alkylboronyl, a (C1-C30)alkyl(C6-C30)arylboronyl, a (C6-C30)aryl(C1-C30)alkyl, and a (C1-C30)alkyl(C6-C30)aryl.

[Claim 7]

The organic electroluminescent compound according to claim 1, wherein

A, B, and C, each independently, represent



A, B, and C are the same;

L_1 represents a single bond, a substituted or unsubstituted (C6-C20)arylene, or an unsubstituted (5- to 20-membered)heteroarylene;

L_2 represents a tertiary residue derived from an unsubstituted (C6-C20) aromatic hydrocarbon ring;

Ar_1 to Ar_6 , each independently, represent a substituted or unsubstituted (C6-C20)aryl, or a substituted or unsubstituted (5- to 20-membered)heteroaryl; or Ar_1 and Ar_2 , Ar_3 and Ar_4 , or Ar_5 and Ar_6 may be linked to each other to form a substituted or unsubstituted (3- to 20-membered) polycyclic aromatic ring whose carbon atom(s) may be replaced with nitrogen;

X represents -O-, -S-, -C(R_1)(R_2)- or -N(R_3)-;

R_1 to R_3 , each independently, represent an unsubstituted (C1-C10)alkyl or an unsubstituted (C6-C20)aryl; or R_1 and R_2 may be linked to each

other to form a (3- to 20-membered) monocyclic alicyclic ring;

R₄ to R₈, each independently, represent hydrogen or an unsubstituted (C6-C20)aryl; or may be linked to an adjacent substituent(s) to form a (6- to 20-membered) monocyclic aromatic ring;

a represents 1 or 4; c and d, each independently, represent an integer of 2 to 4; e represents 3 or 4; where a, c, d or e is an integer of 2 or more, each of the substituents are the same or different;

b represents 2;

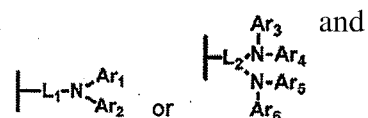
l, m, and n, each independently, represent an integer of 0 to 2; and

l+m+n is 1 or 2.

[Claim 8]

The organic electroluminescent compound according to claim 1, wherein

A, B, and C, each independently, represent



A, B, and C are the same;

L₁ represents a single bond, a (C6-C15)arylene unsubstituted or substituted with a (C1-C6)alkyl(s), or an unsubstituted (5- to 15-membered)heteroarylene;

L₂ represents a tertiary residue derived from an unsubstituted (C6-C15) aromatic hydrocarbon ring;

Ar₁ to Ar₆, each independently, represent a (C6-C20)aryl unsubstituted or substituted with deuterium, a halogen(s), a (C1-C6)alkyl(s), a (C6-C15)aryl(s), a di(C6-C15)arylamino(s), or a (5- to 15-membered)heteroaryl(s); or a (5- to 15-membered)heteroaryl unsubstituted or substituted with a (C6-C15)aryl(s); or Ar₁ and Ar₂, Ar₃ and Ar₄, or Ar₅ and Ar₆ may be linked to each other to form an indole ring unsubstituted or substituted with a (C6-C15)aryl(s); or an indolo-carbazole ring substituted with a (C6-C15)aryl(s) unsubstituted or substituted with a (C1-C4)alkyl(s), a phenyl(s) or a naphthyl(s), or a (5- to 15-membered)heteroaryl(s) unsubstituted or substituted with a phenyl(s);

X represents -O-, -S-, -C(R₁)(R₂)- or -N(R₃)-;

R₁ to R₃, each independently, represent an unsubstituted (C1-C6)alkyl or an unsubstituted (C6-C15)aryl; or R₁ and R₂ may be linked to each other to form a cyclopentane ring or a cyclohexane ring;

R₄ to R₈, each independently, represent hydrogen or an unsubstituted (C6-C15)aryl; or may be linked to an adjacent substituent(s) to form a

benzene ring;

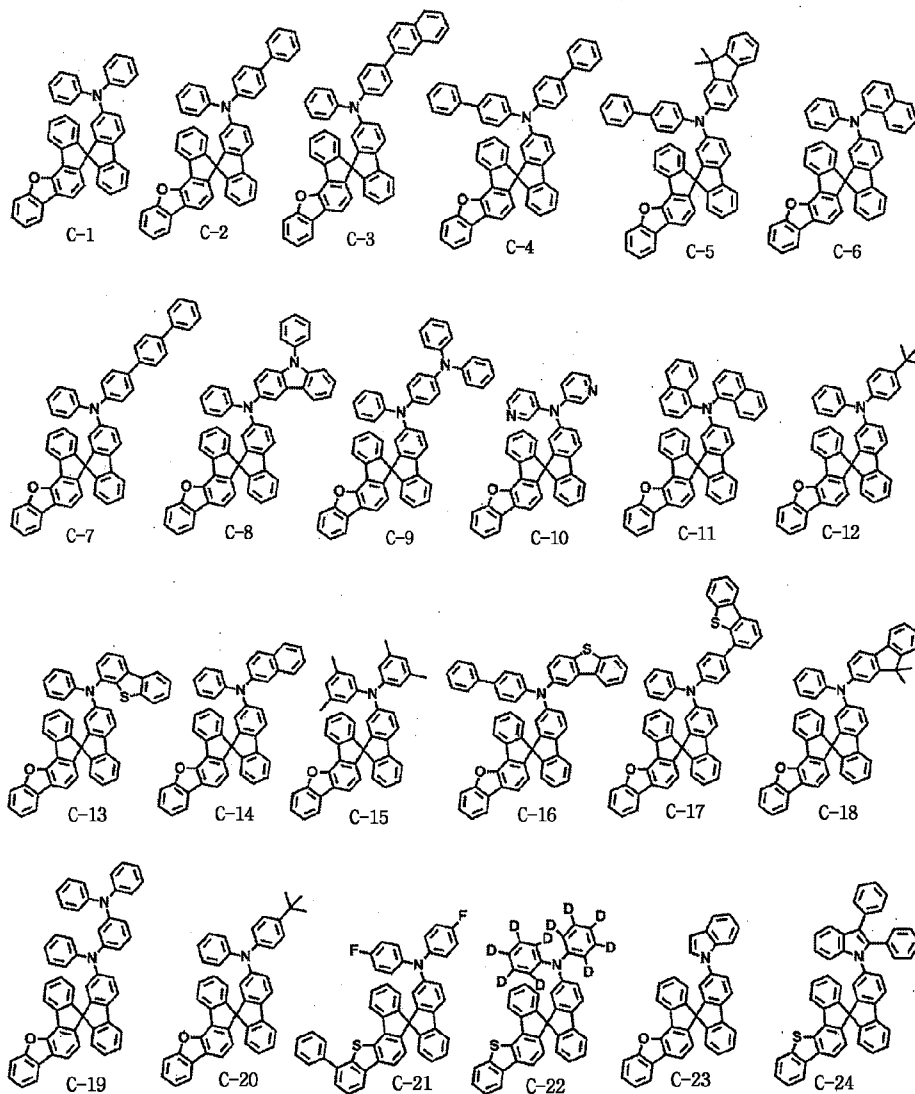
a represents 1 or 4; c and d, each independently, represent an integer of 2 to 4; e represents 3 or 4; where a, c, d, or e is an integer of 2 or more, each of the substituents are the same or different;

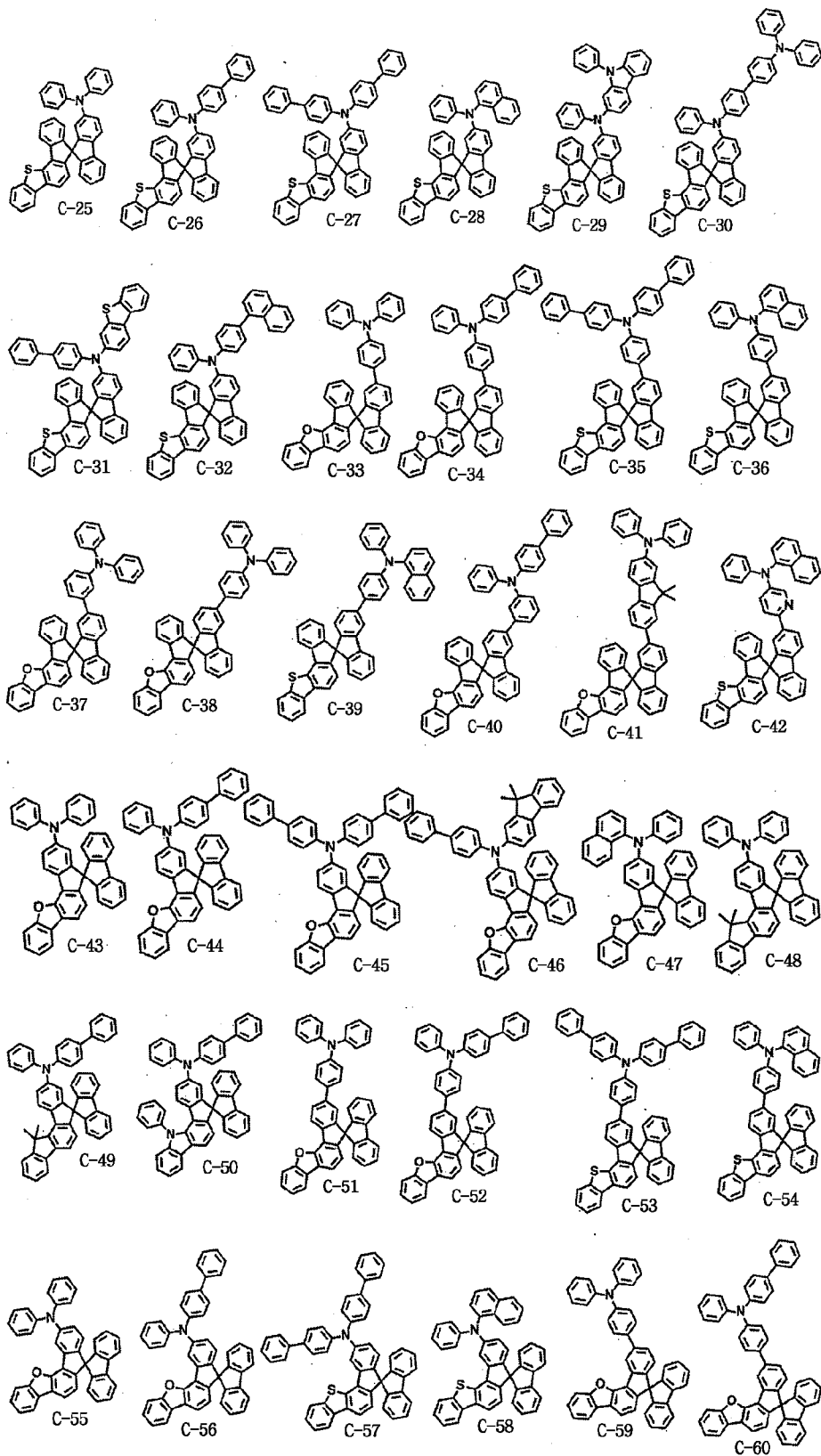
b represents 2;

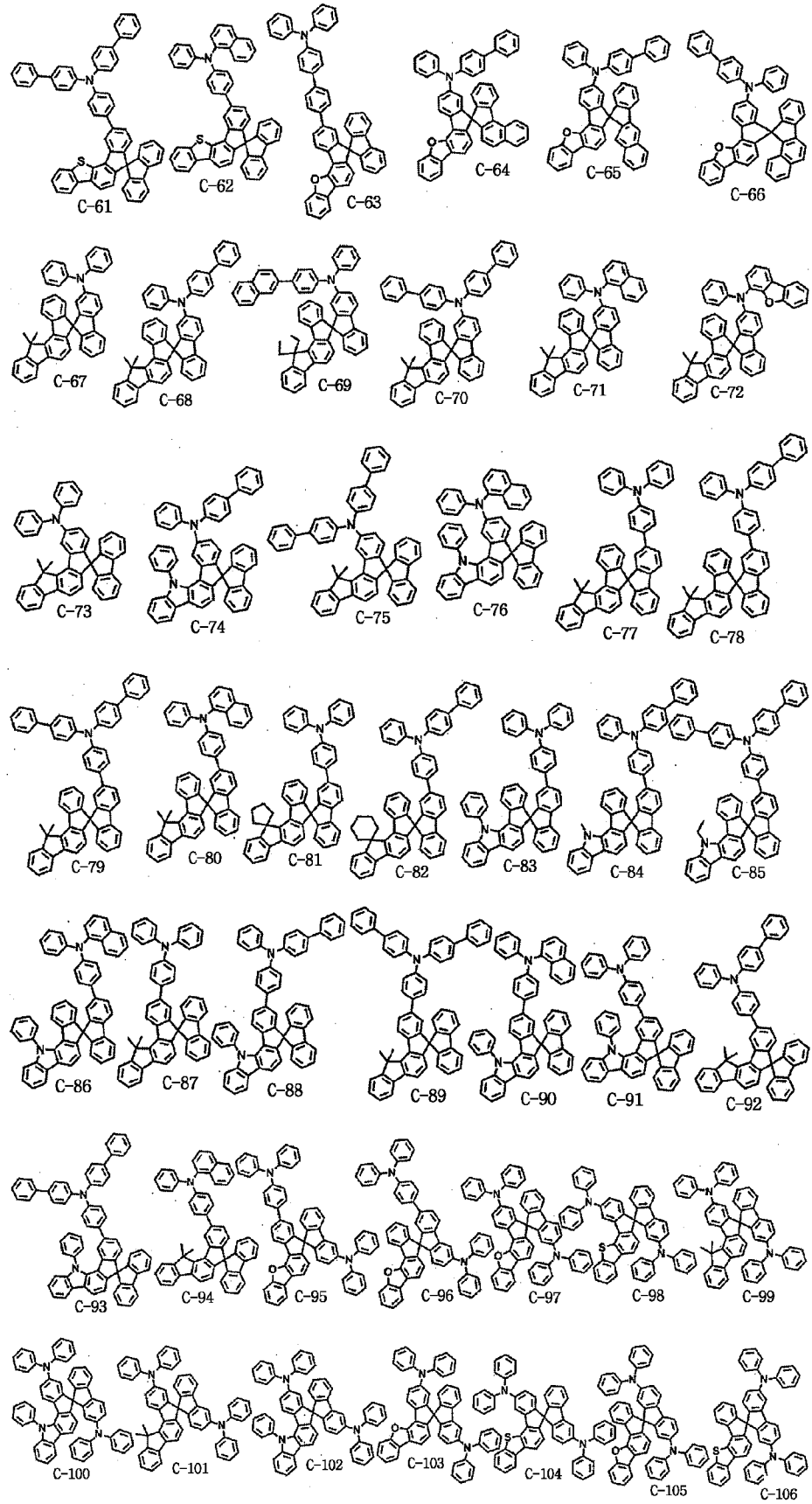
l, m, and n, each independently, represent an integer of 0 to 2; and
 $l+m+n$ is 1 or 2.

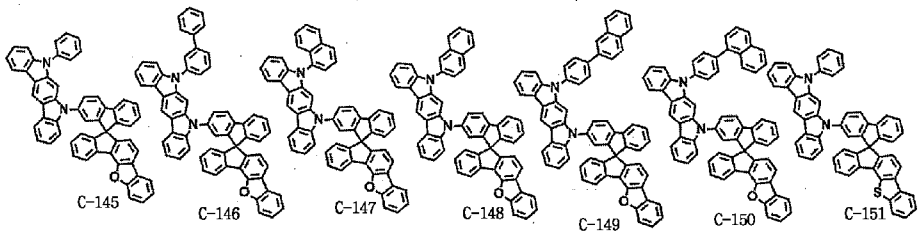
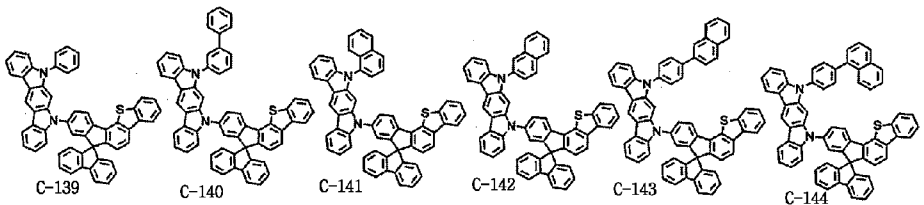
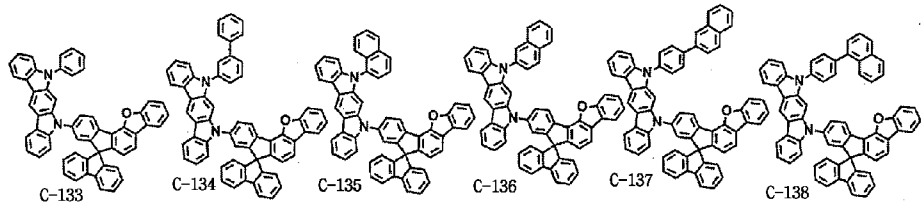
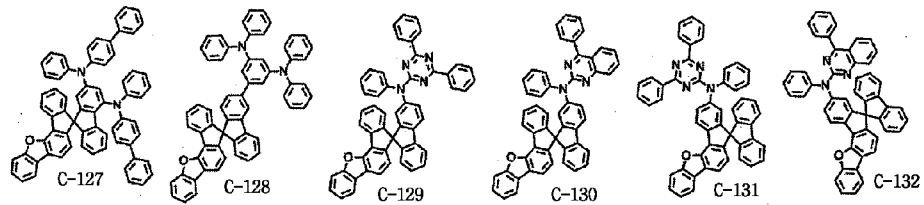
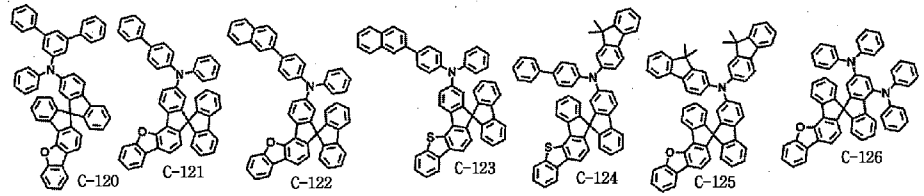
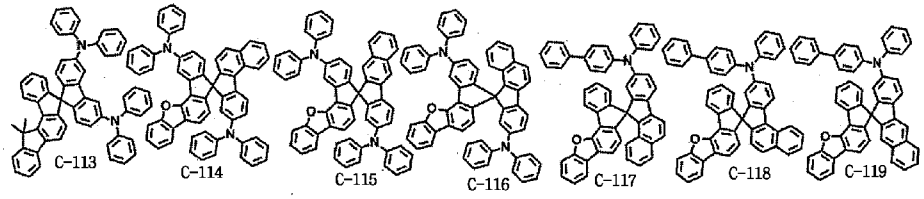
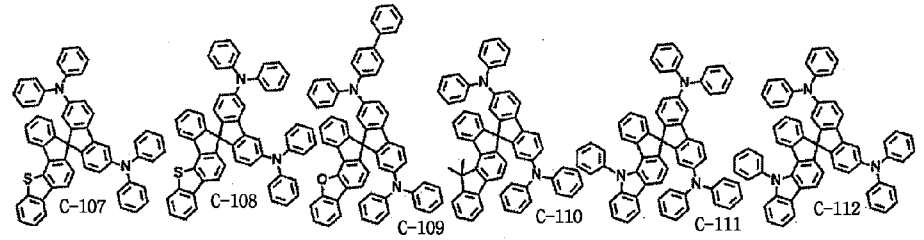
[Claim 9]

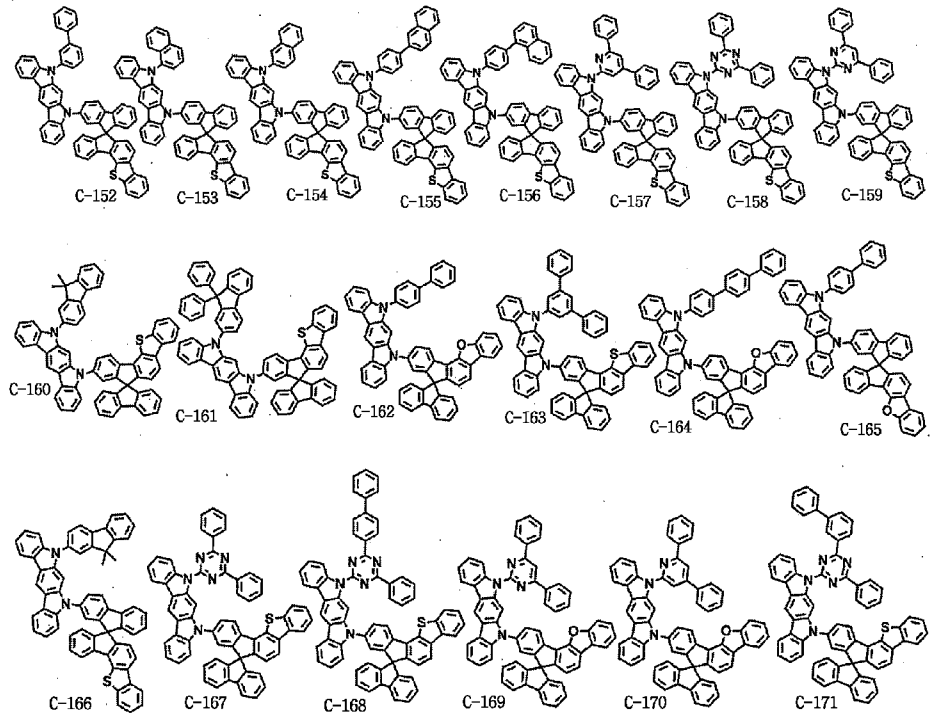
The organic electroluminescent compound according to claim 1, wherein the compound represented by formula 1 is selected from the group consisting of:











[Claim 10]

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

INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2014/001437

A. CLASSIFICATION OF SUBJECT MATTER

*C07D409/12 INVALID C07D 405/12 (2006.01) C07D 333/76 (2006.01) C07D 405/04 (2006.01) C07D 409/04 (2006.01)
C07D 209/96 (2006.01) C07D 307/94 (2006.01) C07D 409/14 (2006.01) C07D 405/14 (2006.01) H01L 51/54 (2006.01)
C07C 211/61 (2006.01) C07F 13/00 (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)

Registry and Caplus: substructure search based on formula 1 of 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

* Special categories of cited documents:		
"A" document defining the general state of the art which is not considered to be of particular relevance	"I" 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 4 April 2014	Date of mailing of the international search report 04 April 2014
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaaustralia.gov.au Facsimile No.: +61 2 6283 7999	Authorised officer Ansari Samad AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. 0262832718

INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation).		PCT/KR2014/001437
DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2009-266927 A (KONICA MINOLTA HOLDINGS INC.) 12 November 2009 & ABSTRACT FROM PATENT ABSTRACTS OF JAPAN FOR JP 2009-266927 A Abstract, page 218, compounds F3-25, F3-26, page 221, compound F3-31, page 222, compound F3-33, F3-34, page 223, compounds F3-35, F3-36, compounds of formulae F1 to F3.	1-10
X	MAKOTO KIMURA <i>et. al.</i> "New 9-fluorene-type trispirocyclic compounds for thermally stable hole transport materials in OLEDs", <i>J. Materials Chem.</i> , 2005, 15, 2393-2398. Figure 1, compound 6	1-8, 10
X	SOON OK JEON <i>et. al.</i> "Fluorenebenzofuran as the core structure of high triplet energy host materials for green phosphorescent organic light-emitting diodes", <i>J. Materials Chem.</i> , 2012, 22, 10537-10541. Scheme 1, compound BFF1	1-8, 10
X	TING LEI <i>et. al.</i> "Highly stable blue light-emitting materials with a three-dimensional architecture: improvement of charge injection and electroluminescence performance", <i>New Journal of Chemistry</i> , 2010, 34, 699-707. Scheme 3, compounds FOMe and FOH	1-8
P,X	WO 2013/100467 A1 (CHEIL INDUSTRIES INC.) 04 July 2013 Abstract, compounds on pages 6-11	1-10
<p>Form PCT/ISA/210 (fifth sheet) (July 2009)</p>		

INTERNATIONAL SEARCH REPORT Information on patent family members		International application No. PCT/KR2014/001437	
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
JP 2009-266927 A	12 Nov 2009	JP 5304010 B2	02 Oct 2013
WO 2013/100467 A1	04 Jul 2013	WO 2013100467 A1	04 Jul 2013
End of Annex			
<p>Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001. Form PCT/ISA/210 (Family Annex)(July 2009)</p>			