



(43) International Publication Date
2 July 2015 (02.07.2015)

(10) International Publication Number
WO 2015/099485 A1

(51) International Patent Classification:

C07C 211/61 (2006.01) *C07F 7/02* (2006.01)
C07D 209/82 (2006.01) *C09K 11/06* (2006.01)
C07D 213/02 (2006.01) *H01L 51/54* (2006.01)
C07D 307/91 (2006.01) *H05B 33/14* (2006.01)
C07D 333/76 (2006.01)

(21) International Application Number:

PCT/KR2014/012893

(22) International Filing Date:

26 December 2014 (26.12.2014)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

10-2013-0163754

26 December 2013 (26.12.2013)

KR

(71) Applicant: **ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.** [KR/KR]; 56, 3gongdan 1-ro, Seobuk-gu, Cheonan-si, Chungcheongnam-do 331-980 (KR).

(72) Inventors: **AHN, Hee-Choon**; 62, Yeongtong-ro 174beon-gil, Yeongtong-gu, Suwon-si, Gyeonggi-do 443-400 (KR). **LEE, Tae-Jin**; 10-1, Seongnam-daero 1148beon-gil, Jungwon-gu, Seongnam-si, Gyeonggi-do 462-827 (KR). **KIM, Chi-Sik**; 441-1201, 71, Dongtanbanseok-ro, Hwaseong-si, Gyeonggi-do 445-752 (KR). **CHO, Young-Jun**; 204-701, 393, Pangyo-ro, Bundang-gu, Seongnam-si, Gyeonggi-do

463-400 (KR). **LEE, Kyung-Joo**; 210-1001, 72, Saechang-ro 8-gil, Mapo-gu, Seoul 121-773 (KR).

(74) Agent: **CHANG, Hoon**; Central Intellectual Property & Law, Korean Re Bldg. 5F, 68, Jong-ro 5-gil, Jongno-gu, Seoul 110-733 (KR).

(81) Designated States (*unless otherwise indicated, for every kind of national protection available*): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (*unless otherwise indicated, for every kind of regional protection available*): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Published:

— with international search report (Art. 21(3))

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

(57) Abstract: The present invention relates to a novel organic electroluminescent compound and an organic electroluminescent device comprising the same. By using the organic electroluminescent compound according to the present invention, it is possible to produce an organic electroluminescent device having excellent current efficiency and lifespan characteristics.



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Description

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

Technical Field

- [1] The present invention relates to an organic electroluminescent compound 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-quinolate)(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, and the lifespan of the device decreases. (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, in order to enhance its efficiency and stability, an organic EL device has a structure of a multilayer comprising a hole injection layer, a hole transport layer, a light-emitting layer, an electron transport layer, and an electron injection layer. The selection of a compound comprised in the hole transport layer is known as a method for improving the characteristics of a device such as hole transport efficiency to the light-emitting layer, luminous efficiency, lifespan, etc.
- [7] In this regard, copper phthalocyanine (CuPc), 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB), N,N'-diphenyl-N,N'-bis(3-methylphenyl)-(1,1'-biphenyl)-4,4'-diamine (TPD), 4,4',4''-tris(3-methylphenylphenylamino)triphenylamine (MTDATA), etc. were used as a hole injection and transport material. However, 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] Therefore, a hole transport layer for improving durability of an organic EL device still needs to be developed.
- [9] Japanese Patent Appln. Laying-Open No. JP 2001-196177 discloses a compound in which two benzene rings of a fluorene are each substituted with a diarylamine, as an organic electroluminescent compound. However, the above reference does not disclose an organic electroluminescent device using a compound in which one benzene ring of a fluorene is substituted with two diarylamines.

[10]

Disclosure of Invention

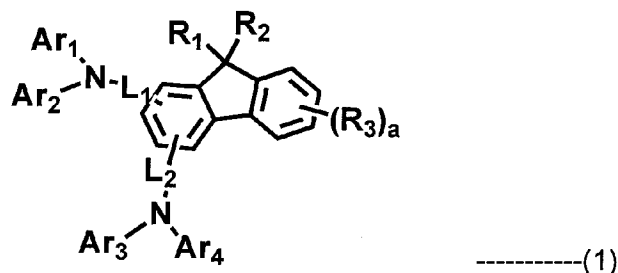
Technical Problem

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

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] L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (3- to 30-membered)heteroarylene;[16] Ar_1 to Ar_4 each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl;[17] R_1 and R_2 each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl; or are linked to each other to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring;[18] R_3 represents 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, 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, $-NR_4R_5$, $-SiR_6R_7R_8$, a cyano, a nitro, or a hydroxyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring;[19] R_4 and R_5 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl;[20] R_6 to R_8 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, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, or a substituted or unsubstituted (C3-C30)cycloalkyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring, whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen, and sulfur;[21] a represents an integer of 1 to 4, where a is an integer of 2 or more, each of R_3 may

be the same or different; and

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

[23]

Advantageous Effects of Invention

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

[25]

Mode for the Invention

- [26] 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.

- [27] 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.

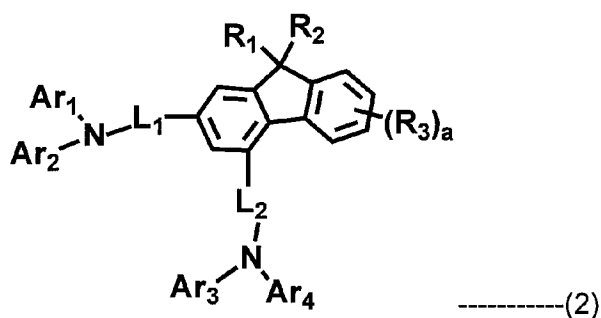
- [28] The organic electroluminescent compound represented by the above formula 1 will be described in detail.

- [29] Herein, “(C1-C30)alkyl” is meant to be a linear or branched alkyl 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.; “(3- to 30-membered)heteroaryl(ene)” is an aryl having 3 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, furazanyl, pyridyl, pyrazinyl, pyrimidinyl, pyridazinyl, etc., and a fused ring-type heteroaryl including benzofuranyl, benzothiophenyl, isobenzofuranyl, dibenzofuranyl, dibenzothiophenyl, benzonaphthothiophenyl, benzimidazolyl, benzothiazolyl, benzoisothiazolyl, benzoisoxazolyl, benzoxazolyl, isoindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolinyl, quinazolinyl, quinoxalinyl, carbazolyl, phenoxazinyl, phenanthridinyl, benzo[dioxolyl, etc. Further, “halogen” includes F, Cl, Br and I.

[30] According to an embodiment of the present invention, the compound of formula 1 can be represented by the following formula 2:

[31]



[32] wherein L₁, L₂, Ar₁ to Ar₄, R₁ to R₃, and a are as defined in formula 1.

[33] Herein, “substituted” in the expression “substituted or unsubstituted” means that a hydrogen atom in a certain functional group is replaced with another atom or group, i.e., a substituent. The substituents of the substituted (C1-C30)alkyl, the substituted (C3-C30)cycloalkyl, the substituted (3- to 7-membered)heterocycloalkyl, the substituted (C6-C30)aryl(ene), the substituted (3- to 30-membered)heteroaryl(ene), and the substituted (C6-C30)aryl(C1-C30)alkyl in L₁, L₂, Ar₁ to Ar₄, and R₁ to R₈ of formula 1 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 (3- to 30-membered)heteroaryl unsubstituted or substituted with a (C6-C30)aryl, a (C6-C30)aryl unsubstituted or substituted with a (3- 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 a (C1-C6)alkyl, a (C6-C25)aryl, and a (5- to 20-membered)heteroaryl.

- [34] In formula 1 above, L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (3- to 30-membered)heteroarylene, preferably each independently represent a single bond, a substituted or unsubstituted (C6-C12)arylene, or a substituted or unsubstituted (5- to 20-membered)heteroarylene, and more preferably each independently represent a single bond, an unsubstituted (C6-C12)arylene, or an unsubstituted (5- to 20-membered)heteroarylene.
- [35] Ar_1 to Ar_4 each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl, preferably each independently represent a substituted or unsubstituted (C6-C15)aryl, and more preferably each independently represent a (C6-C15)aryl unsubstituted or substituted with a (C1-C6)alkyl, a (C6-C15)aryl, or a (5- to 20-membered)heteroaryl.
- [36] R_1 and R_2 each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl; or are linked to each other to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring, preferably each independently represent a substituted or unsubstituted (C1-C6)alkyl, a substituted or unsubstituted (C6-C20)aryl, or a substituted or unsubstituted (5- to 20-membered)heteroaryl; or are linked to each other to form a mono- or polycyclic (C6-C20) alicyclic or aromatic ring, and more preferably each independently represent an unsubstituted (C1-C6)alkyl; a (C6-C20)aryl unsubstituted or substituted with a (C1-C6)alkyl, a (C6-C25)aryl, or a (5- to 20-membered)heteroaryl; or a (5- to 20-membered)heteroaryl

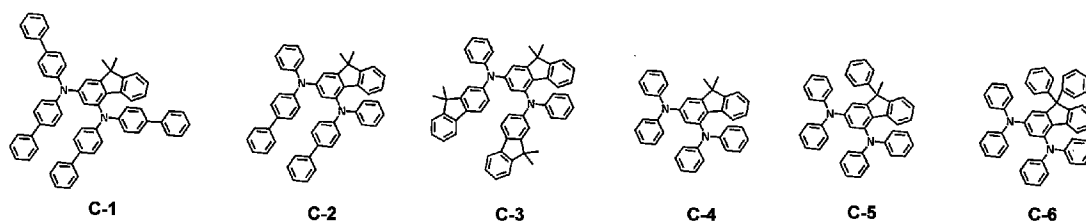
- unsubstituted or substituted with a (C6-C12)aryl; or are linked to each other to form a mono- or polycyclic (C6-C20) aromatic ring.
- [37] R_3 represents hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (3- to 30-membered)heteroaryl, 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, $-NR_4R_5$, $-SiR_6R_7R_8$, a cyano, a nitro, or a hydroxyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring, preferably each independently represent hydrogen, a substituted or unsubstituted (C6-C12)aryl, a substituted or unsubstituted (C5-C12)cycloalkyl, $-NR_4R_5$, or $-SiR_6R_7R_8$; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C6-C12) alicyclic or aromatic ring, and more preferably each independently represent hydrogen, an unsubstituted (C6-C12)aryl, an unsubstituted (C5-C12)cycloalkyl, $-NR_4R_5$, or $-SiR_6R_7R_8$; or are linked to an adjacent substituent(s) to form a monocyclic (C6-C12) aromatic ring.
- [38] R_4 and R_5 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl, preferably each independently represent a substituted or unsubstituted (C6-C12)aryl, and more preferably each independently represent an unsubstituted (C6-C12)aryl.
- [39] R_6 to R_8 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, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, or a substituted or unsubstituted (C3-C30)cycloalkyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) 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 (C1-C6)alkyl, and more preferably each independently represent an unsubstituted (C1-C6)alkyl.
- [40] a represents an integer of 1 to 4, preferably represents an integer of 1 to 2; and where a is an integer of 2 or more, each of R_3 may be the same or different.
- [41] The heteroaryl(ene) and the heterocycloalkyl each independently contain at least one hetero atom selected from B, N, O, S, P(=O), Si, and P.
- [42] According to one embodiment of the present invention, in formula 1 above, L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted (C6-C12)arylene, or a substituted or unsubstituted (5- to 20-membered)heteroarylene; Ar_1 to Ar_4 each independently represent a substituted or unsubstituted (C6-C15)aryl; R_1

and R_2 each independently represent a substituted or unsubstituted (C1-C6)alkyl, a substituted or unsubstituted (C6-C20)aryl, or a substituted or unsubstituted (5- to 20-membered)heteroaryl; or are linked to each other to form a mono- or polycyclic (C6-C20) alicyclic or aromatic ring; R_3 represents hydrogen, a substituted or unsubstituted (C6-C12)aryl, a substituted or unsubstituted (C5-C12)cycloalkyl, $-NR_4R_5$, or $-SiR_6R_7R_8$; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C6-C12) alicyclic or aromatic ring; R_4 and R_5 each independently represent a substituted or unsubstituted (C6-C12)aryl; R_6 to R_8 each independently represent a substituted or unsubstituted (C1-C6)alkyl; and a represents an integer of 1 to 2.

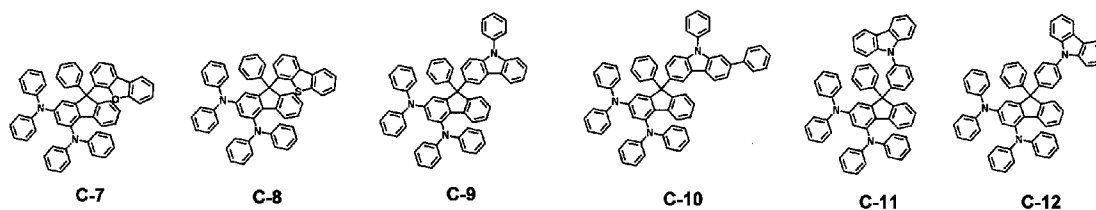
- [43] According to another embodiment of the present invention, in formula 1 above, L_1 and L_2 each independently represent a single bond, an unsubstituted (C6-C12)arylene, or an unsubstituted (5- to 20-membered)heteroarylene; Ar_1 to Ar_4 each independently represent a (C6-C15)aryl unsubstituted or substituted with a (C1-C6)alkyl, a (C6-C15)aryl, or a (5- to 20-membered)heteroaryl; R_1 and R_2 each independently represent an unsubstituted (C1-C6)alkyl; a (C6-C20)aryl unsubstituted or substituted with a (C1-C6)alkyl, a (C6-C25)aryl, or a (5- to 20-membered)heteroaryl; or a (5- to 20-membered)heteroaryl unsubstituted or substituted with a (C6-C12)aryl; or are linked to each other to form a mono- or polycyclic (C6-C20) aromatic ring; R_3 represents hydrogen, an unsubstituted (C6-C12)aryl, an unsubstituted (C5-C12)cycloalkyl, $-NR_4R_5$, or $-SiR_6R_7R_8$; or are linked to an adjacent substituent(s) to form a monocyclic (C6-C12) aromatic ring; R_4 and R_5 each independently represent an unsubstituted (C6-C12)aryl; R_6 to R_8 each independently represent an unsubstituted (C1-C6)alkyl; and a represents an integer of 1 to 2.

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

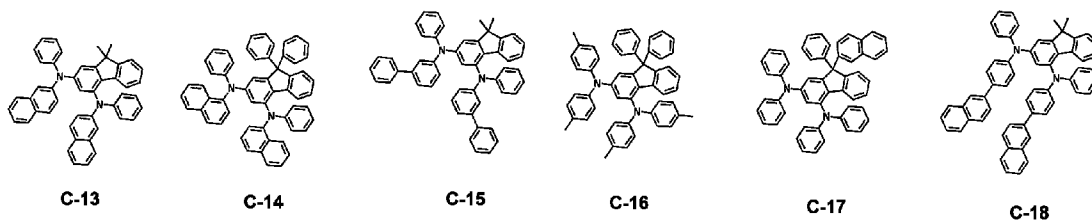
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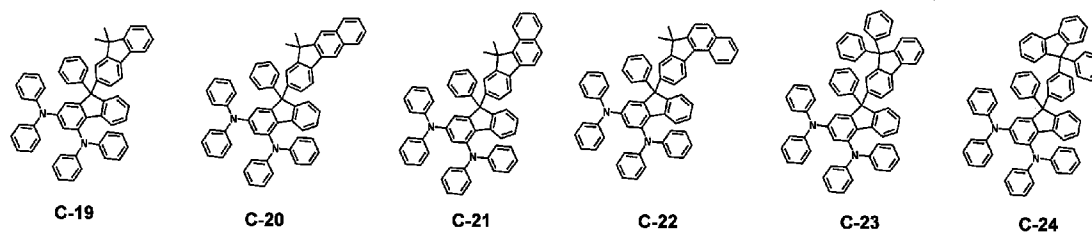
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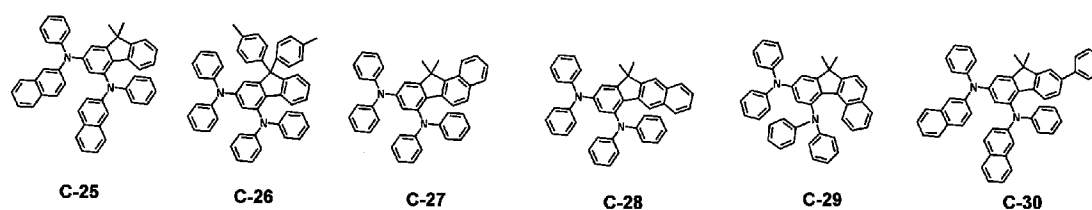
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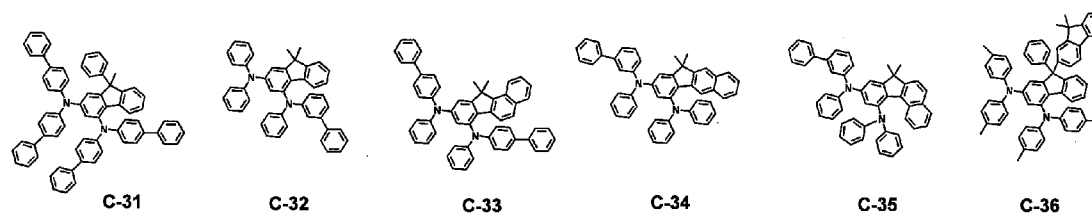
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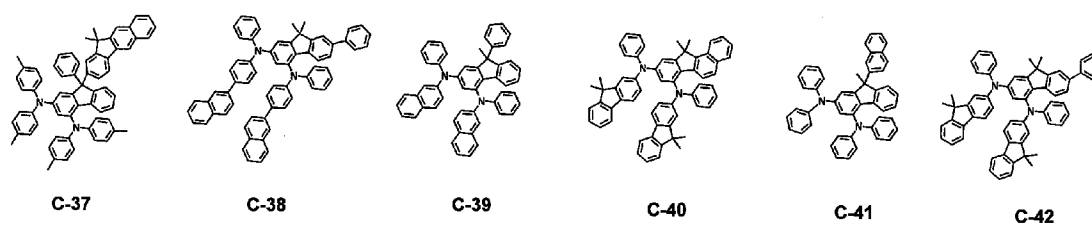
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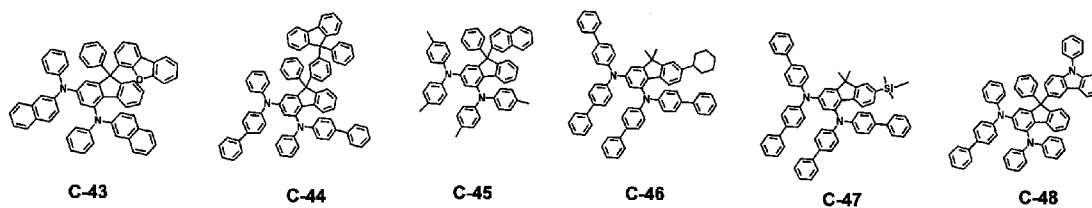
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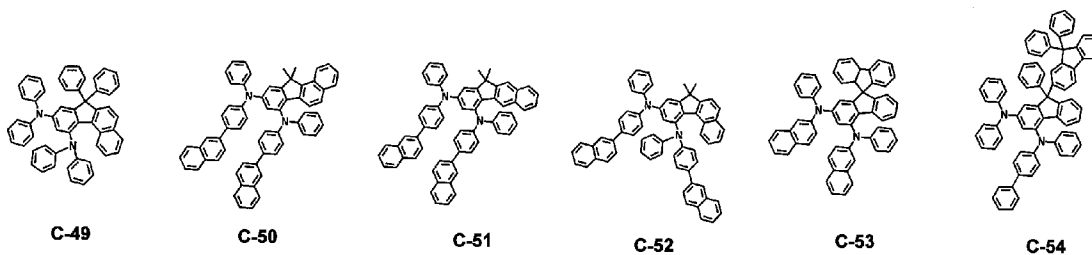
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[52]



[53]



C-55 **C-56** **C-57** **C-58** **C-59** **C-60**

C-61 **C-62** **C-63** **C-64** **C-65** **C-66**

C-67 **C-68** **C-69** **C-70** **C-71** **C-72**

C-79 **C-80** **C-81** **C-82** **C-83** **C-84**

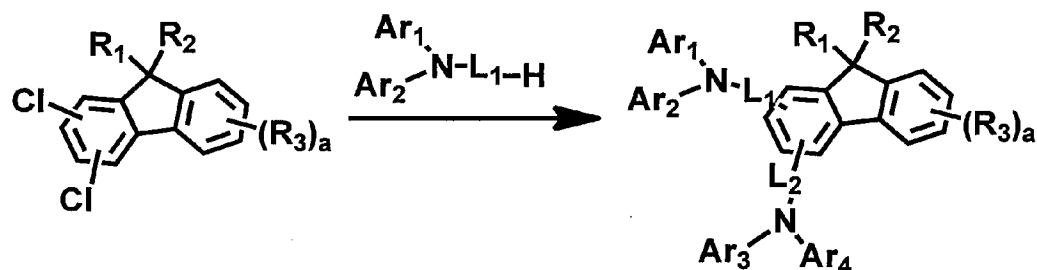
[illegible]

[61] 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.

[62] [Reaction Scheme 1]

[63]



[64] wherein L_1 , L_2 , Ar_1 to Ar_4 , R_1 to R_3 , and a are as defined in formula 1 above.

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

[66] 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.

[67] 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.

[68] One of the first and second electrodes can be an anode, and the other can be 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.

[69] 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 of the present invention can be comprised as a hole transport material. Where used in the light-emitting layer, the organic electroluminescent compound of the present invention can be comprised as a host material.

[70] 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.

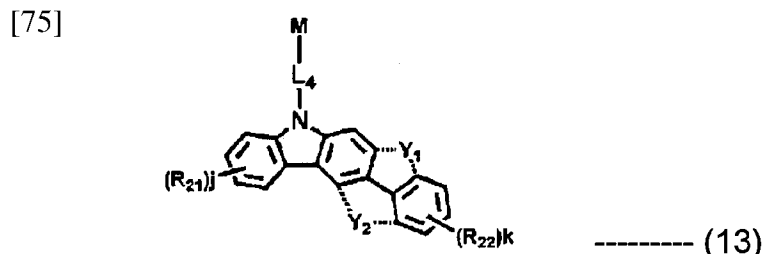
[71] Where the organic electroluminescent compound according to the present invention is comprised as a host material (first host material), the other compound may be comprised as a second host material. Herein, the weight ratio of the first host material

to the second host material is in the range of 1:99 to 99:1.

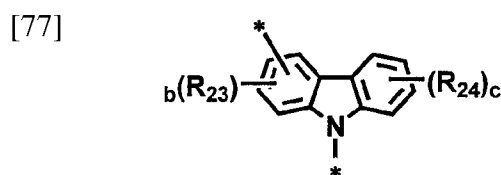
[72] The host material other than the organic electroluminescent compound according to the present invention can be from any of the known phosphorescent hosts. Specifically, the phosphorescent host selected from the group consisting of the compounds of formulae 11 to 13 below is preferable in terms of luminous efficiency.

[73] $H-(Cz-L_4)_h-M$ ----- (11)

[74] $H-(Cz)_i-L_4-M$ ----- (12)



[76] wherein Cz represents the following structure;



[78] R_{21} to R_{24} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (3- to 30-membered)heteroaryl, or $-SiR_{25}R_{26}R_{27}$;

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

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

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

[82] Y_1 and Y_2 each independently represent $-O-$, $-S-$, $-N(R_{31})-$, or $-C(R_{32})(R_{33})-$, provided that Y_1 and Y_2 do not simultaneously exist;

[83] R_{31} to R_{33} each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (5- to 30-membered)heteroaryl, and R_{32} and R_{33} may be the same or different;

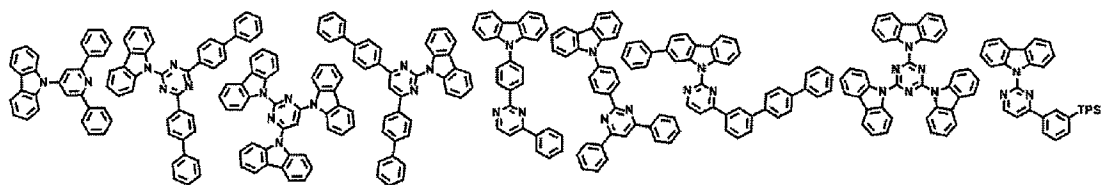
[84] h and i each independently represent an integer of 1 to 3;

[85] j , k , b , and c each independently represent an integer of 0 to 4; and

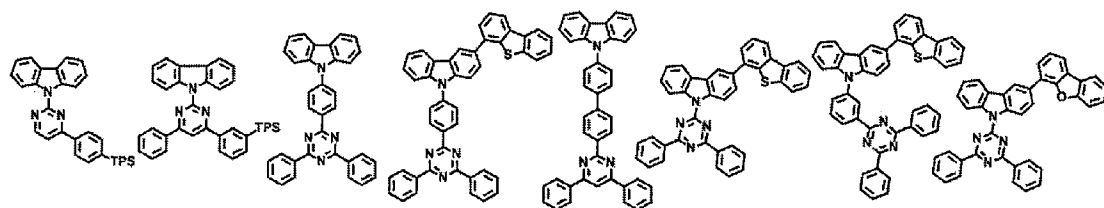
[86] where h , i , j , k , b , or c is an integer of 2 or more, each of $(Cz-L_4)$, each of (Cz) , each of R_{21} , each of R_{22} , each of R_{23} , or each of R_{24} may be the same or different.

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

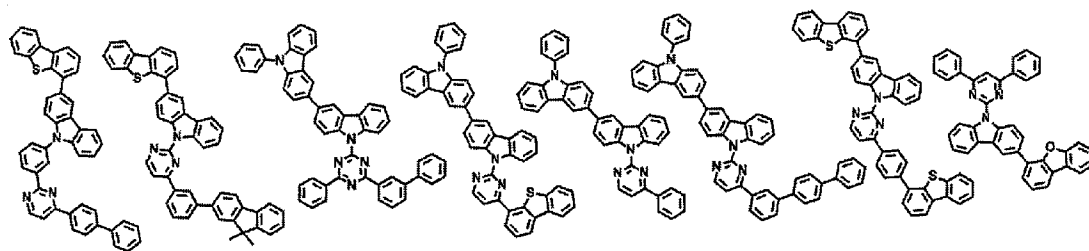
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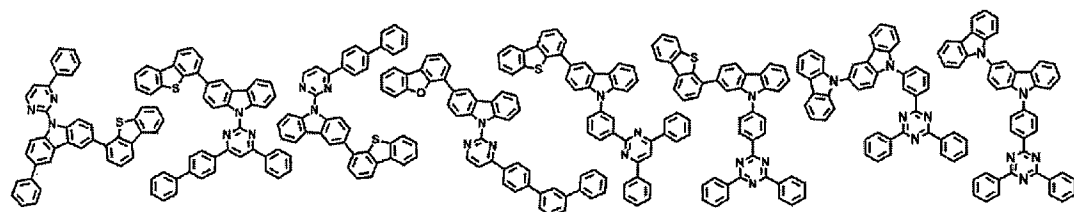
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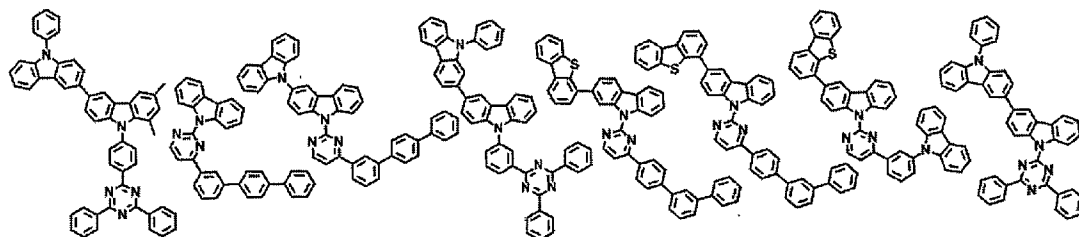
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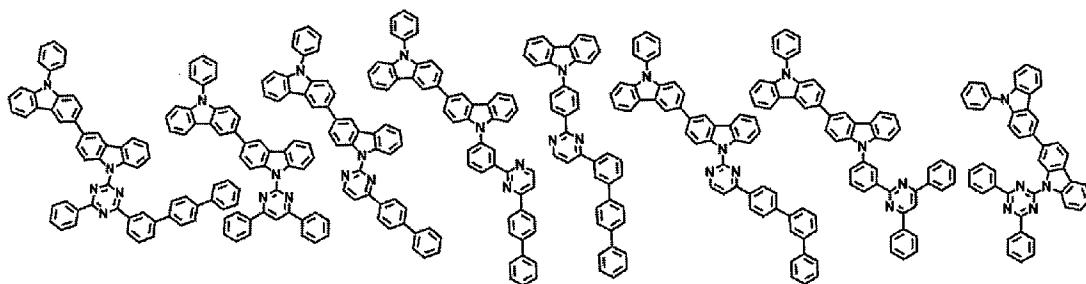
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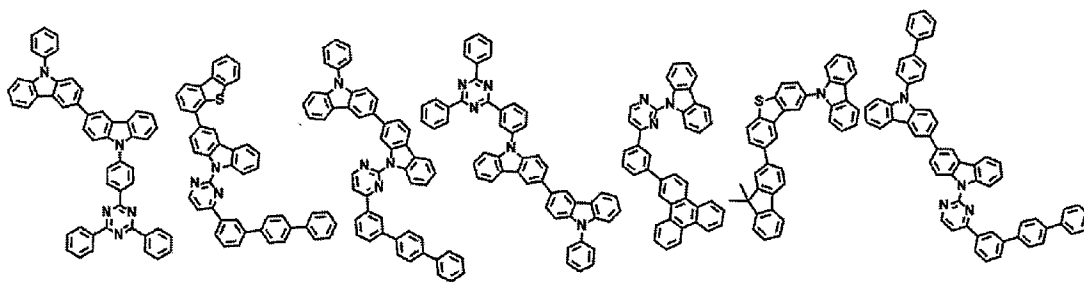
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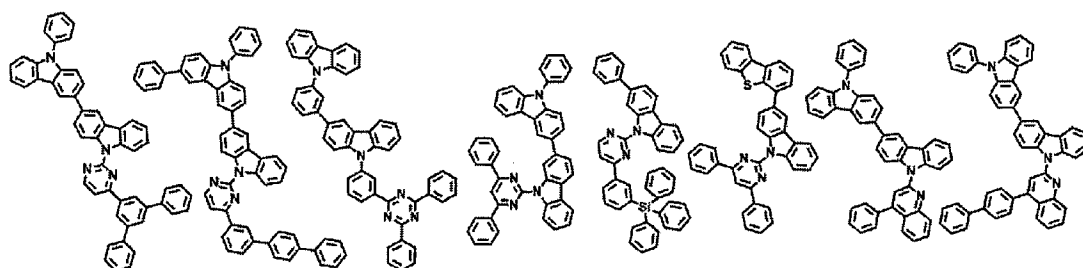
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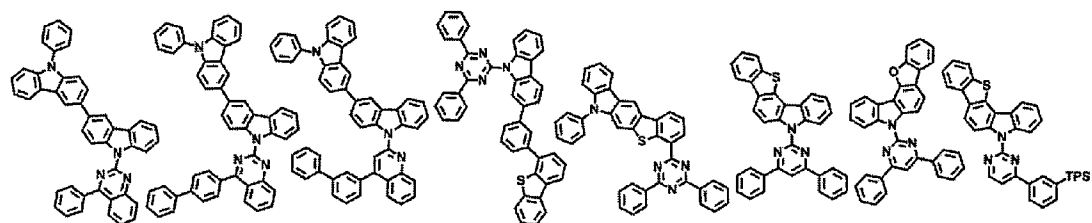
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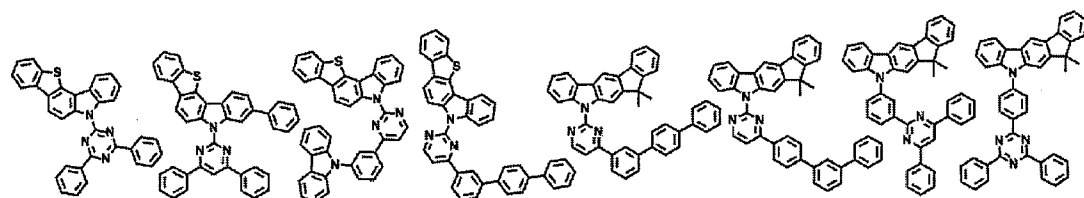
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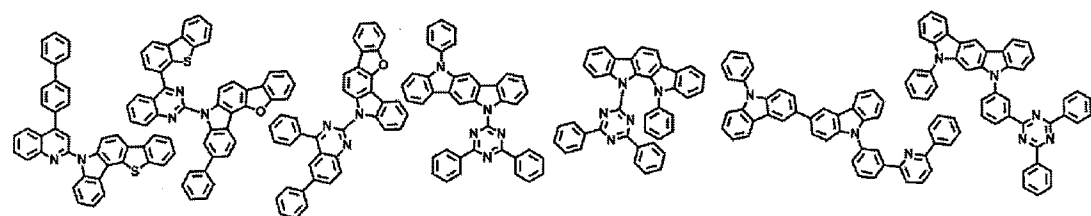
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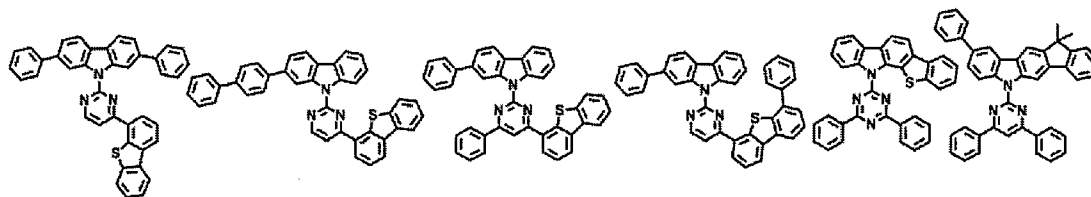
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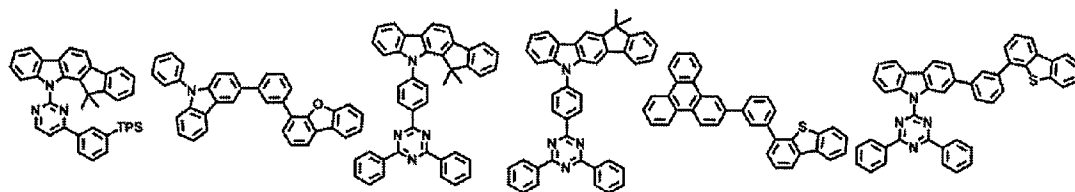
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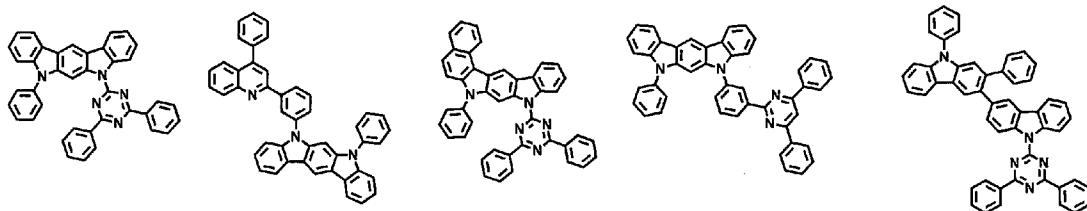
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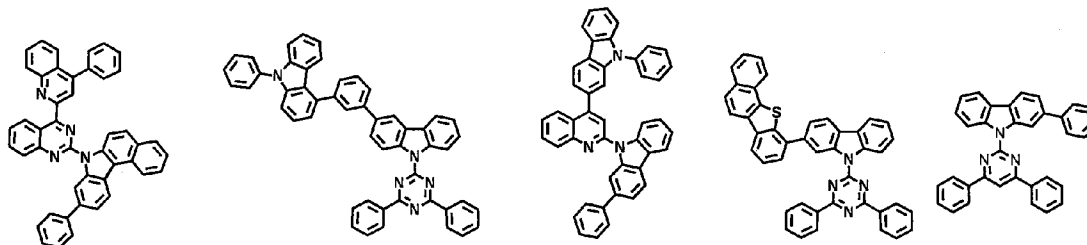
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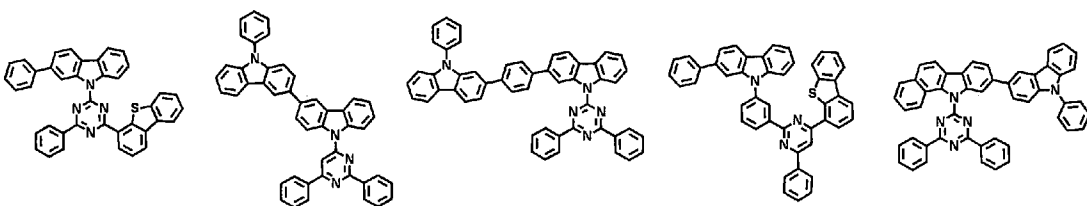
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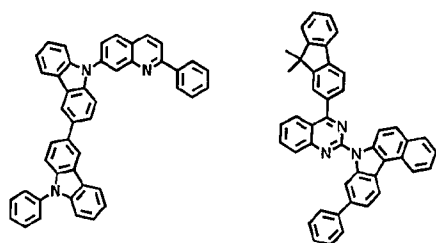
[102]



[103]



[104]

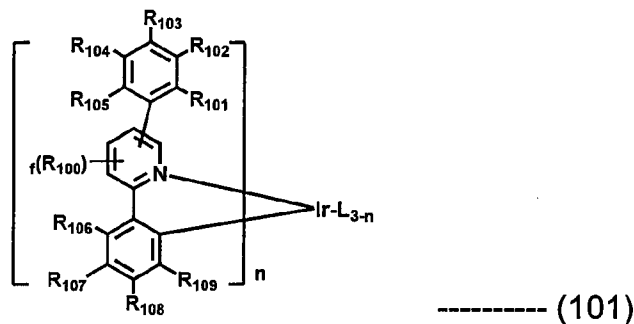


[105] [wherein TPS represents triphenylsilyl]

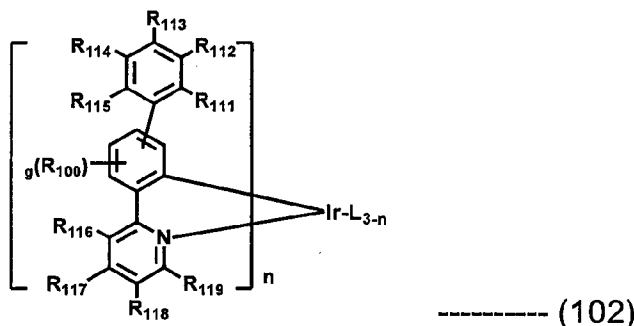
[106] The dopant comprised in the organic electroluminescent device according to the present invention is preferably at least one phosphorescent dopant. The dopant materials applied to the organic electroluminescent device according to the present invention are not limited, but may be preferably selected 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.

[107] The phosphorescent dopants may be preferably selected from compounds represented by the following formulae 101 to 103.

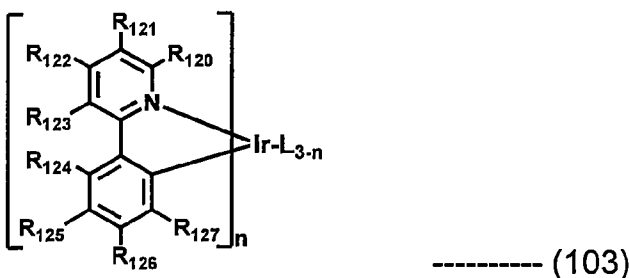
[108]



[109]

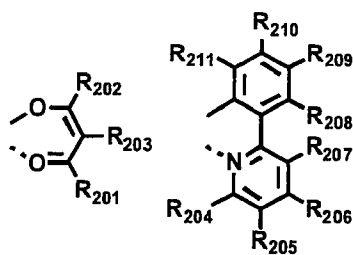


[110]



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

[112]

[113] R₁₀₀ represents hydrogen, a substituted or unsubstituted (C1-C30)alkyl, or a substituted or unsubstituted (C3-C30)cycloalkyl;[114] 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; a substituted or unsubstituted (C6-C30)aryl; or a substituted or unsubstituted (C3-C30)cycloalkyl; R₁₀₆ to R₁₀₉ may be linked to an adjacent substituent(s) to form a substituted or unsubstituted fused ring, e.g. fluorene unsubstituted or substituted with alkyl, dibenzothiophene unsubstituted or substituted with alkyl, or dibenzofuran unsubstituted or substituted with alkyl; and R₁₂₀

to R_{123} may be linked to an adjacent substituent(s) to form a substituted or unsubstituted fused ring, e.g. quinoline unsubstituted or substituted with a halogen, alkyl, or aryl;

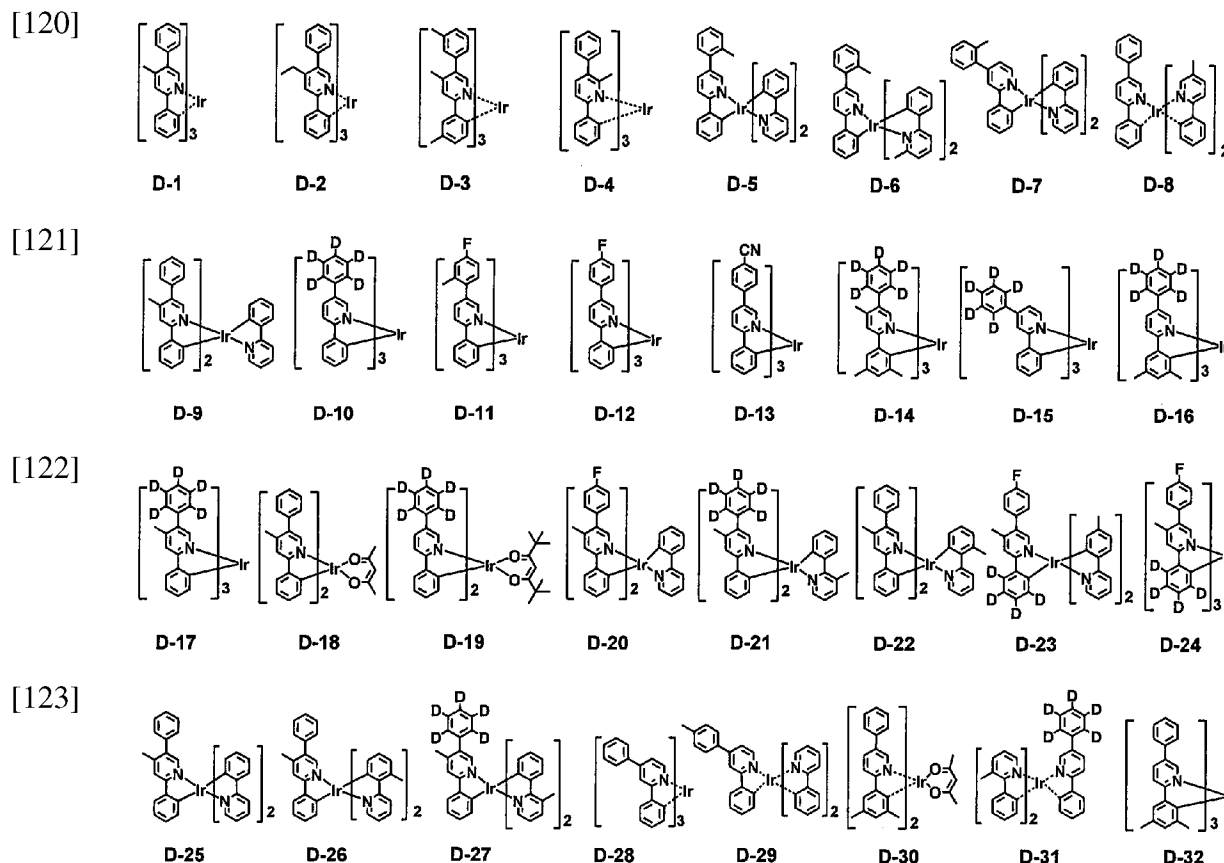
[115] R_{124} to R_{127} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, or a substituted or unsubstituted (C6-C30)aryl; and R_{124} to R_{127} may be linked to an adjacent substituent(s) to form a substituted or unsubstituted fused ring, e.g. fluorene unsubstituted or substituted with alkyl, dibenzothiophene unsubstituted or substituted with alkyl, or dibenzofuran unsubstituted or substituted with alkyl;

[116] R_{201} to R_{211} each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl unsubstituted or substituted with a halogen(s), a substituted or unsubstituted (C3-C30)cycloalkyl, or a substituted or unsubstituted (C6-C30)aryl, and R_{208} to R_{211} may be linked to an adjacent substituent(s) to form a substituted or unsubstituted fused ring, e.g. fluorene unsubstituted or substituted with alkyl, dibenzothiophene unsubstituted or substituted with alkyl, or dibenzofuran unsubstituted or substituted with alkyl;

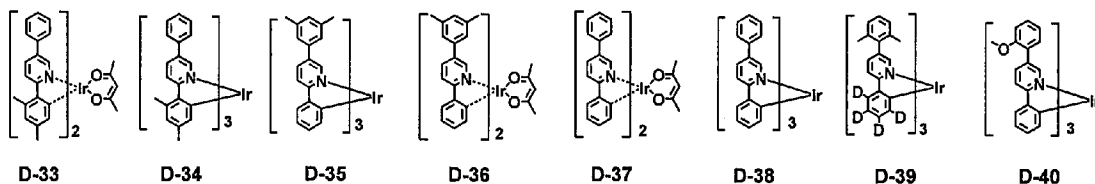
[117] f and g each independently represent an integer of 1 to 3; where f or g is an integer of 2 or more, each of R_{100} may be the same or different; and

[118] n represents an integer of 1 to 3.

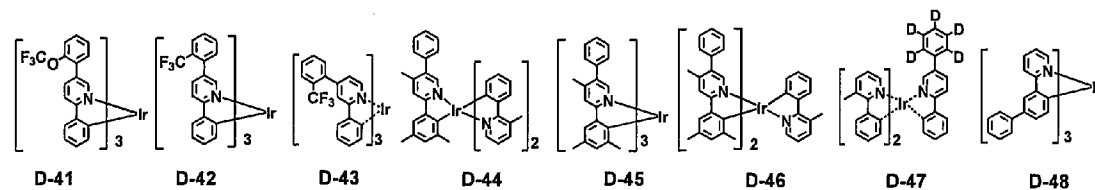
[119] Specifically, the phosphorescent dopant materials include the following:



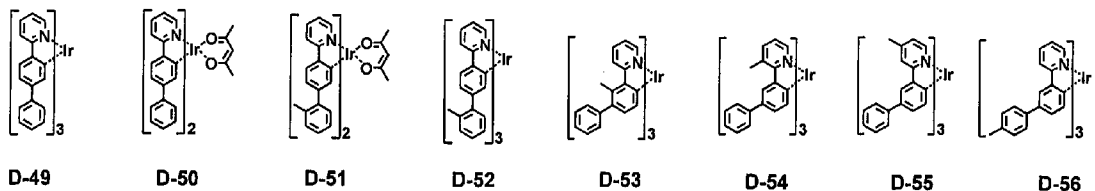
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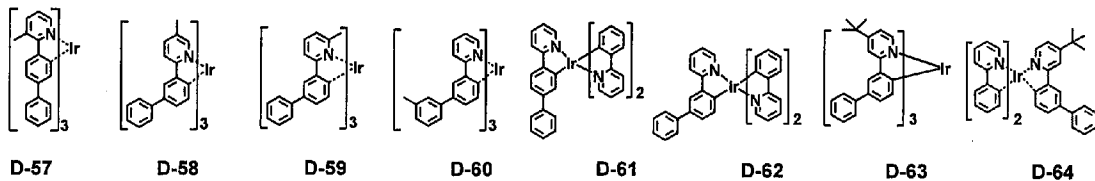
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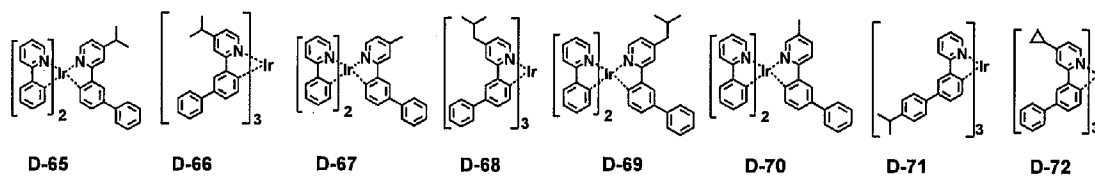
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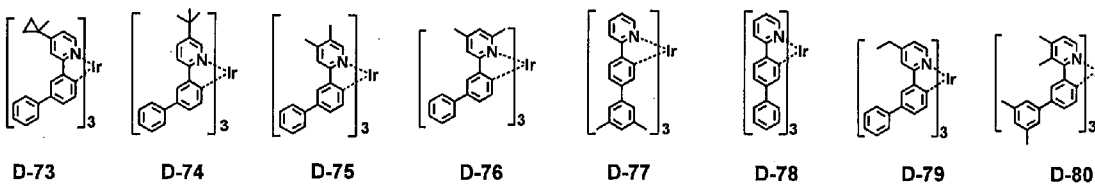
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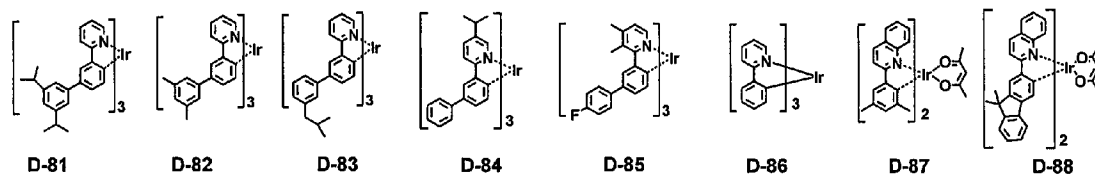
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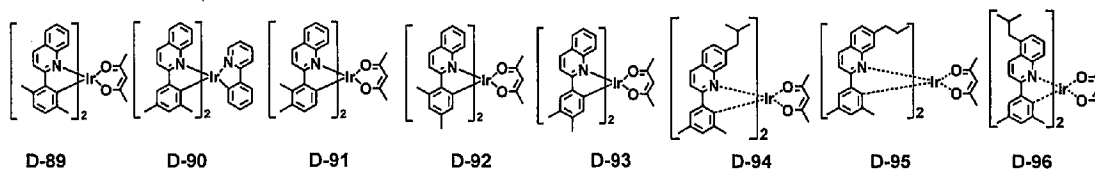
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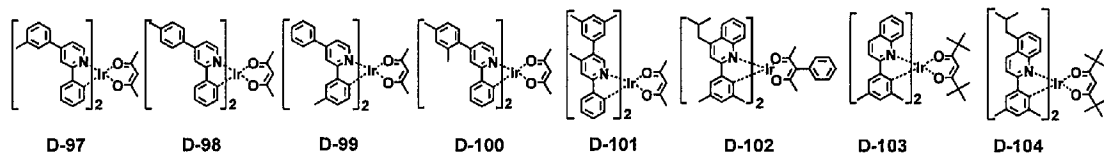
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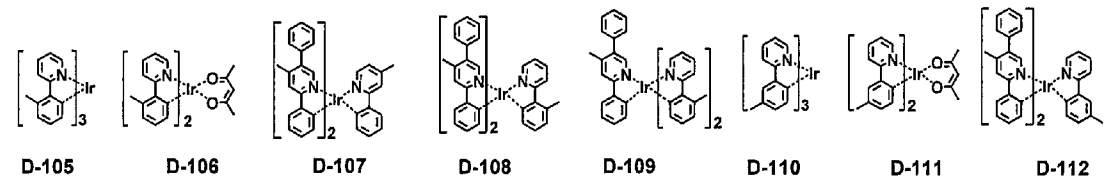
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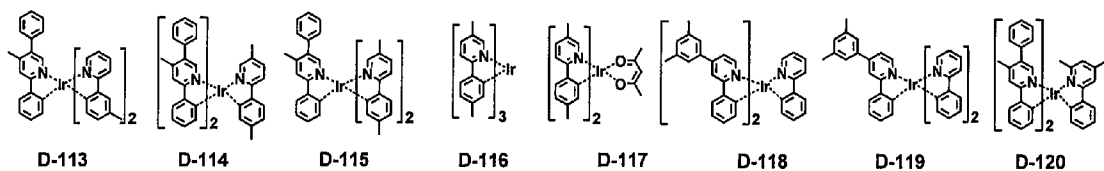
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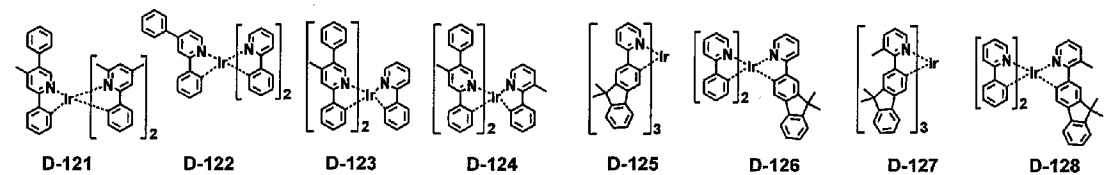
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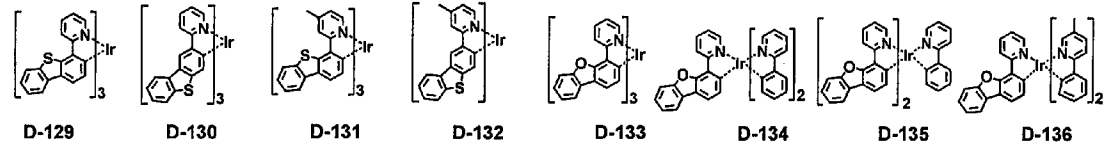
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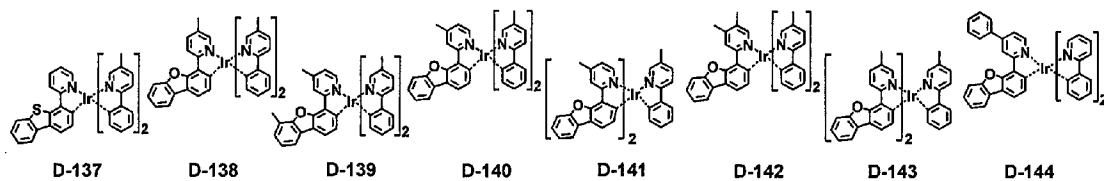
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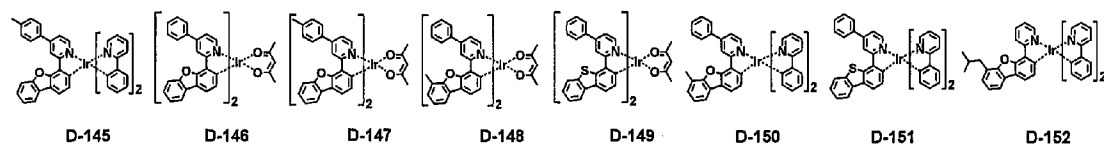
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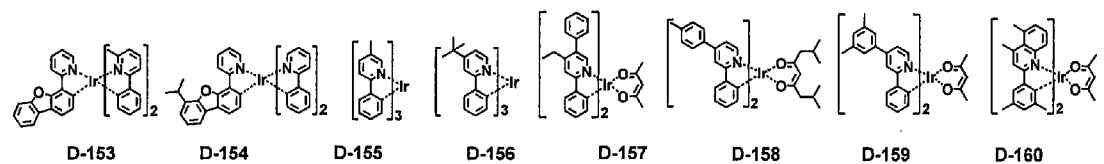
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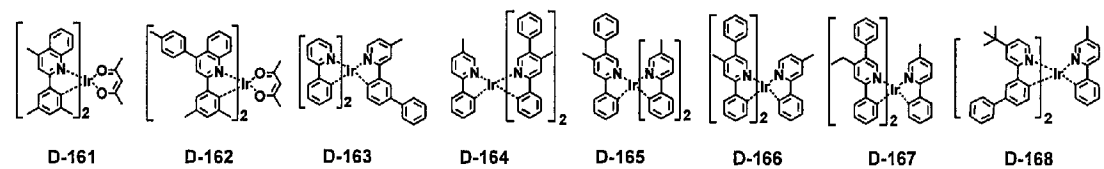
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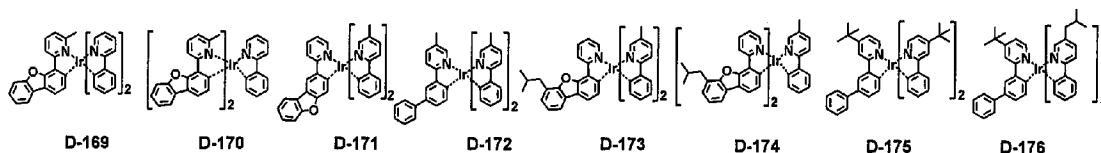
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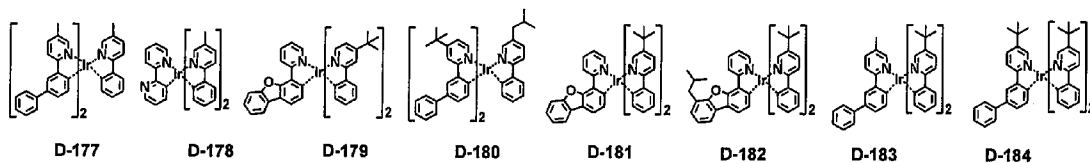
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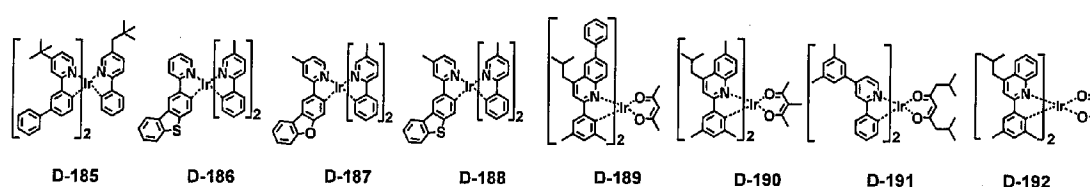
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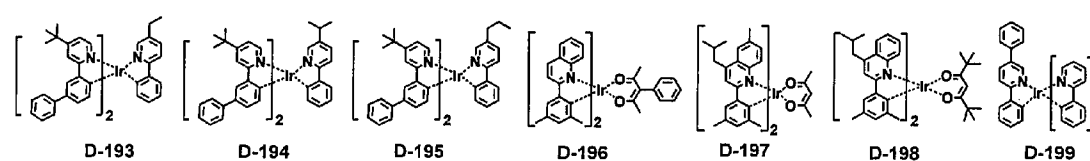
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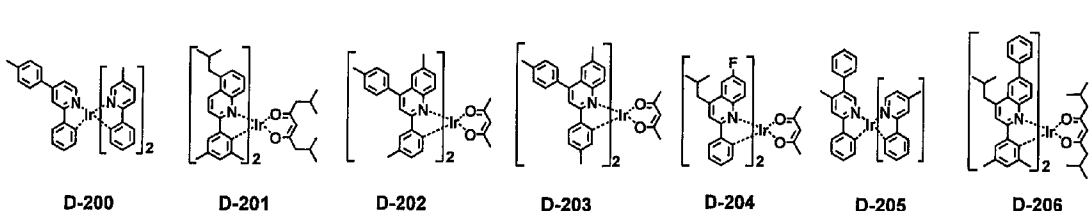
[143]



[144]



[145]



[146] In another embodiment of the present invention, a composition for preparing 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.

[147] 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 preparing the organic electroluminescent device according to the present invention.

[148] 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.

[149] 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.

[150] 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.

[151] 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.

[152] 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.

[153] 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.

[154] When using a wet film-forming method, a thin film can be formed by dissolving or

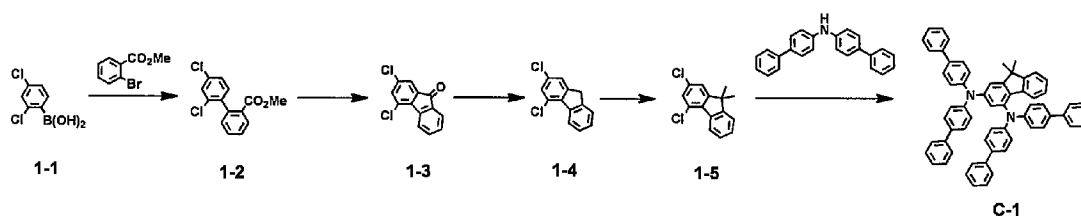
diffusing materials forming each layer into any suitable solvent such as ethanol, chloroform, tetrahydrofuran, dioxane, etc. The solvent can be any solvent where the materials forming each layer can be dissolved or diffused, and where there are no problems in film-formation capability.

[155] 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.

[156]

[157] **Example 1: Preparation of compound C-1**

[158]



[159] **Preparation of compound 1-2**

[160] After introducing 2,4-dichlorophenyl boronic acid (compound 1-1) (64 g, 335 mmol), methyl 2-bromobenzoate (60 g, 279 mmol), tetrakis(triphenylphosphine)palladium (9.5 g, 8.4 mmol), potassium carbonate (96 g, 698 mmol), toluene 600 mL, and ethanol 300 mL in a reaction container, distilled water 300 mL was added to the mixture, and the mixture was stirred at 120°C for 3 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 with magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 1-2 (79 g, 99%).

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

[162] After introducing compound 1-2 (79 g, 279 mmol), Eaton's reagent 110 mL, and chlorobenzene 1 L in a reaction container, the mixture was stirred under reflux overnight. After cooling the reaction solution to room temperature, the reaction was completed with water, and an organic layer was extracted with methylene chloride. The extracted organic layer was dried with magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 1-3 (49 g, 71%).

[163] **Preparation of compound 1-4**

[164] After introducing iodine (18 g, 71 mmol), hypophosphorous acid (35 mL, 315 mmol, 50% aqueous solution), and acetic acid 1 L in a reaction container, the mixture was stirred at 100°C for 30 minutes. Thereafter, compound 1-3 was slowly added dropwise thereto, and the mixture was stirred under reflux overnight. After cooling the reaction

solution to room temperature, the precipitated solid was filtered, and washed with a large amount of hexane. The filtrate was diluted with ethylacetate and washed with water. The extracted organic layer was dried with magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 1-4 (35.7 g, 77%).

[165] Preparation of compound 1-5

[166] After introducing compound 1-4 (35.5 g, 151 mmol), potassium hydroxide (42 g, 760 mmol), potassium iodide (2.5 g, 15 mmol), benzyltriethylammonium chloride (1.7 g, 7.8 mmol), distilled water 700 mL, and dimethyl sulfoxide 700 mL in a reaction container, the mixture was stirred at room temperature for 15 minutes. Thereafter, methyl iodide (25 mL, 378 mmol) was added thereto, and the mixture was stirred at room temperature overnight. The reaction solution was diluted with ethylacetate and washed with distilled water. The extracted organic layer was dried with magnesium sulfate, and the solvent was removed using a rotary evaporator. The remaining substance was then purified with column chromatography to obtain compound 1-5 (32 g, 81%).

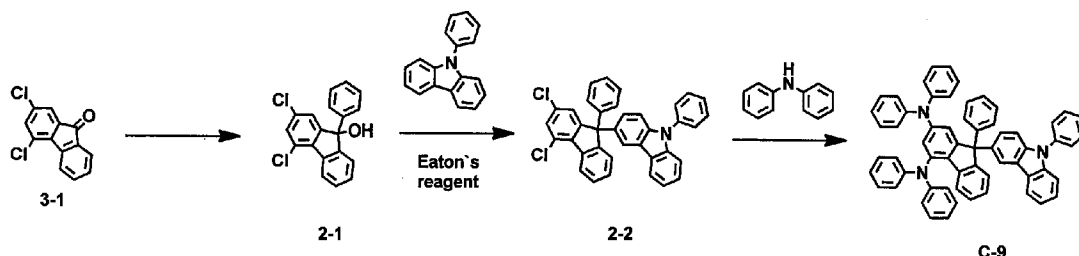
[167] Preparation of compound C-1

[168] After introducing compound 1-5 (7 g, 26.6 mmol), dibiphenyl-4-ylamine (17 g, 52.9 mmol), tris(dibenzylideneacetone)dipalladium(0) (1.9 g, 2.1 mmol), s-phos (1.1 g, 2.7 mmol), sodium t-butoxide (6.4 g, 67 mmol), and o-xylene 150 mL in a reaction container, the mixture was stirred under reflux for 1 hour. Thereafter, the reaction solution cooled to room temperature was diluted with ethylacetate and washed with water several times. The extracted organic layer was then dried with anhydrous magnesium sulfate, distilled under reduced pressure, and purified with column chromatography to obtain compound C-1 (9.6 g, 55%).

[169]

[170] Example 2: Preparation of compound C-9

[171]



[172] Preparation of compound 2-1

[173] After introducing compound 3-1 (20.5 g, 82.3 mol) and tetrahydrofuran 400 mL in a reaction container, the reaction solution was cooled to 0°C, and phenylmagnesium bromide (40 mL, 123 mmol) and 3 M diethylether solution were slowly added dropwise thereto. The reaction solution was stirred at room temperature for 1 hour.

Thereafter, the reaction was completed with ammonium chloride aqueous solution, and the reaction solution was diluted with ethylacetate and washed with water. The extracted organic layer was then dried with anhydrous magnesium sulfate, distilled under reduced pressure, and purified with column chromatography to obtain compound 2-1 (28 g, 99%).

[174] Preparation of compound 2-2

[175] After introducing compound 2-1 (15.8 g, 48.3 mmol), 9-phenylcarbazole (17.6 g, 72.5 mmol), and methylenechloride (MC) 250 mL in a reaction container, the mixture was subjected into nitrogen atmosphere. Eaton's reagent 1.5 mL was then slowly added dropwise thereto, and the mixture was stirred at room temperature for 2 hours. Thereafter, the reaction was completed with distilled water, and the mixture was extracted with methylenechloride. The extracted organic layer was dried with 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 (13.2 g, 49%).

[176] Preparation of compound C-9

[177] After introducing compound 2-2 (13 g, 26.6 mmol), dibiphenylamine (8 g, 47 mmol), tris(dibenzylideneacetone)dipalladium(0) (1.7 g, 1.9 mmol), s-phos (0.96 g, 2.35 mmol), sodium t-butoxide (5.6 g, 58.8 mmol), and o-xylene 120 mL in a reaction container, the mixture was stirred under reflux overnight. Thereafter, the reaction solution cooled to room temperature was diluted with ethylacetate and washed with water several times. The extracted organic layer was then dried with anhydrous magnesium sulfate, distilled under reduced pressure, and purified with column chromatography to obtain compound C-9 (10 g, 52%).

[178]

[179] Device Example 1: Production of an OLED device using the organic
[180] electroluminescent compound according to the present invention

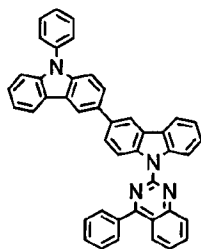
[181] An OLED device was produced using the organic electroluminescent compound according to the present invention. A transparent electrode indium tin oxide (ITO) thin film (10 Ω /sq) on a glass substrate for an organic light-emitting diode (OLED) device (Geomatec, Japan) was subjected to an ultrasonic 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. N^4,N^4 -biphenyl- N^4,N^4 -bis(9-phenyl-9H-carbazol-3-yl)-[1,1'-biphenyl]-4,4'-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 first hole injection layer having a thickness of 80 nm on the ITO

substrate. Then, 1,4,5,8,9,11-hexaazatriphenylenhexacarbonitrile (HAT-CN) 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 second hole injection layer having a thickness of 5 nm on the first hole injection layer. Then, compound **T-1** as below 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 first hole transport layer having a thickness of 10 nm on the second hole injection layer. Then, compound **C-1** 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 second hole transport layer having a thickness of 60 nm on the first hole transport layer. Thereafter, compound **H-1** was introduced into one cell of the vacuum vapor depositing apparatus, as a host, and compound **D-96** was introduced into another cell as a dopant. The two materials were evaporated at different rates and were deposited in a doping amount of 3 wt% based on the total amount of the host and dopant to form a light-emitting layer having a thickness of 40 nm on the second hole transport layer. Then,

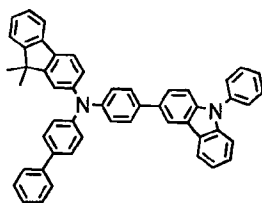
2-(4-(9,10-di(naphthalen-2-yl)anthracen-2-yl)phenyl)-1-phenyl-1H-benzo[d]imidazole was introduced into one cell and lithium quinolate was introduced into another cell. The two materials were evaporated at the same rate and were deposited in a doping amount of 50 wt% each to form an electron transport layer having a thickness of 35 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 80 nm was deposited by another vacuum vapor depositing apparatus on the electron injection layer. Thus, an OLED device was produced. All the materials used for producing the OLED device were purified by vacuum sublimation at 10^{-6} torr prior to use.

[182] The produced OLED device showed a red emission having a luminance of 800 cd/m² and a current density of 2.8 mA/cm². As for the lifespan characteristic, the time period for the luminance to decrease to 90% at 5,000 nits was 800 hours.

[183]



H-1



T-1

[184] **Device Example 2: Production of an OLED device using the organic**

[185] **electroluminescent compound according to the present invention**

[186] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **C-9** to form the second hole transport layer in a thickness of 60 nm.

[187] The produced OLED device showed a red emission having a luminance of 1500 cd/m² and a current density of 5.2 mA/cm². As for the lifespan characteristic, the time period for the luminance to decrease to 90% at 5,000 nits was 750 hours.

[188]

[189] **Device Example 3: Production of an OLED device using the organic**

[190] **electroluminescent compound according to the present invention**

[191] An OLED device was produced in the same manner as in Device Example 1, except for evaporating compound **C-67** to form the second hole transport layer in a thickness of 60 nm.

[192] The produced OLED device showed a red emission having a luminance of 1200 cd/m² and a current density of 4.2 mA/cm². As for the lifespan characteristic, the time period for the luminance to decrease to 90% at 5,000 nits was 780 hours.

[193]

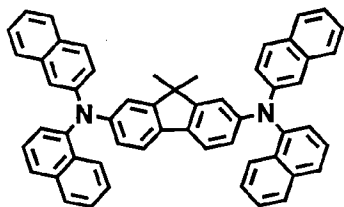
[194] **Comparative Example 1: Production of an OLED device comprising a**

[195] **conventional organic electroluminescent compound**

[196] An OLED device was produced in the same manner as in Device Example 1, except for evaporating the compound below to form the second hole transport layer in a thickness of 60 nm.

[197] The produced OLED device showed a red emission having a luminance of 2000 cd/m² and a current density of 11.2 mA/cm². As for the lifespan characteristic, the time period for the luminance to decrease to 90% at 5,000 nits was 67 hours.

[198]

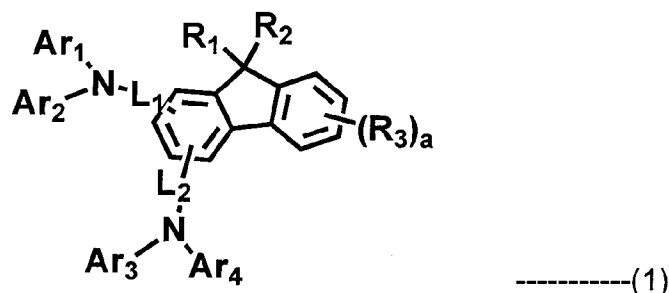


[199] It is verified that the luminous characteristics of the organic electroluminescent compound according to the present invention is superior to the conventional materials. In addition, an organic electroluminescent device using the organic electroluminescent compound according to the present invention has excellent luminous and lifespan characteristics.

Claims

[Claim 1]

An organic electroluminescent compound represented by the following formula 1:



wherein

L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted (C6-C30)arylene, or a substituted or unsubstituted (3- to 30-membered)heteroarylene;

Ar_1 to Ar_4 each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl;

R_1 and R_2 each independently represent a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C3-C30)cycloalkyl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl; or are linked to each other to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring;

R_3 represents 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, 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, $-NR_4R_5$, $-SiR_6R_7R_8$, a cyano, a nitro, or a hydroxyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring;

R_4 and R_5 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, or a substituted or unsubstituted (3- to 30-membered)heteroaryl;

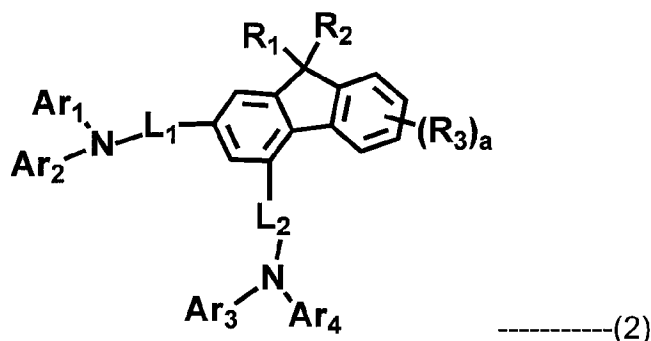
R_6 to R_8 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl, a substituted or unsubstituted (C6-C30)aryl, a substituted or unsubstituted (3- to 30-membered)heteroaryl, a substituted or unsubstituted (3- to 7-membered)heterocycloalkyl, or a substituted or unsubstituted (C3-C30)cycloalkyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C3-C30) alicyclic or aromatic ring, whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen, and sulfur;

a represents an integer of 1 to 4, where a is an integer of 2 or more, each of R_3 may be the same or different; and

the heteroaryl(ene) and the heterocycloalkyl 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 L_1 , L_2 , Ar_1 to Ar_4 , R_1 to R_3 , and a are as defined in claim 1.

[Claim 3]

The organic electroluminescent compound according to claim 1, wherein the substituents of the substituted (C1-C30)alkyl, the substituted (C3-C30)cycloalkyl, the substituted (3- to 7-membered)heterocycloalkyl, the substituted (C6-C30)aryl(ene), the substituted (3- to 30-membered)heteroaryl(ene), and the substituted (C6-C30)aryl(C1-C30)alkyl in L_1 , L_2 , Ar_1 to Ar_4 , and R_1 to R_8 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 (3- to 30-membered)heteroaryl unsubstituted or substituted with a

(C6-C30)aryl, a (C6-C30)aryl unsubstituted or substituted with a (3- 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.

[Claim 4]

The organic electroluminescent compound according to claim 1, wherein

L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted (C6-C12)arylene, or a substituted or unsubstituted (5- to 20-membered)heteroarylene;

Ar_1 to Ar_4 each independently represent a substituted or unsubstituted (C6-C15)aryl;

R_1 and R_2 each independently represent a substituted or unsubstituted (C1-C6)alkyl, a substituted or unsubstituted (C6-C20)aryl, or a substituted or unsubstituted (5- to 20-membered)heteroaryl; or are linked to each other to form a mono- or polycyclic (C6-C20) alicyclic or aromatic ring;

R_3 represents hydrogen, a substituted or unsubstituted (C6-C12)aryl, a substituted or unsubstituted (C5-C12)cycloalkyl, $-NR_4R_5$, or $-SiR_6R_7R_8$; or are linked to an adjacent substituent(s) to form a mono- or polycyclic (C6-C12) alicyclic or aromatic ring;

R_4 and R_5 each independently represent a substituted or unsubstituted (C6-C12)aryl;

R_6 to R_8 each independently represent a substituted or unsubstituted (C1-C6)alkyl; and

a represents an integer of 1 to 2.

[Claim 5]

The organic electroluminescent compound according to claim 1, wherein

L_1 and L_2 each independently represent a single bond, an unsubstituted (C6-C12)arylene, or an unsubstituted (5- to 20-membered)heteroarylene;

Ar_1 to Ar_4 each independently represent a (C6-C15)aryl unsubstituted or substituted with a (C1-C6)alkyl, a (C6-C15)aryl, or a (5- to

20-membered)heteroaryl;

R_1 and R_2 each independently represent an unsubstituted (C1-C6)alkyl; a (C6-C20)aryl unsubstituted or substituted with a (C1-C6)alkyl, a (C6-C25)aryl, or a (5- to 20-membered)heteroaryl; or a (5- to 20-membered)heteroaryl unsubstituted or substituted with a (C6-C12)aryl; or are linked to each other to form a mono- or polycyclic (C6-C20) aromatic ring;

R_3 represents hydrogen, an unsubstituted (C6-C12)aryl, an unsubstituted (C5-C12)cycloalkyl, $-NR_4R_5$, or $-SiR_6R_7R_8$; or are linked to an adjacent substituent(s) to form a monocyclic (C6-C12) aromatic ring;

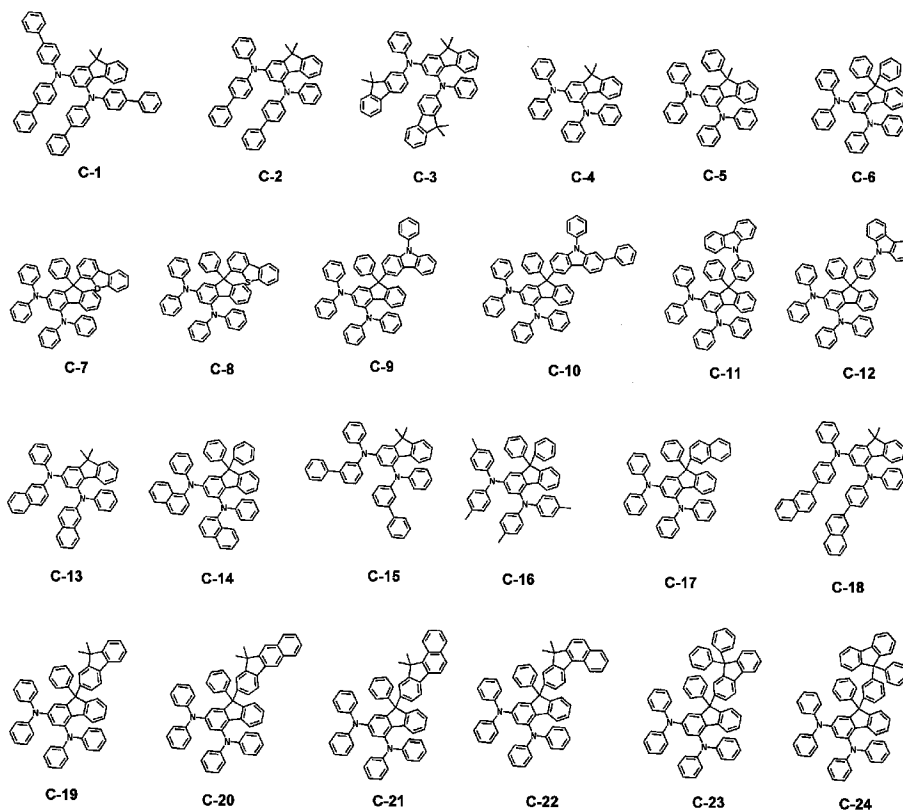
R_4 and R_5 each independently represent an unsubstituted (C6-C12)aryl;

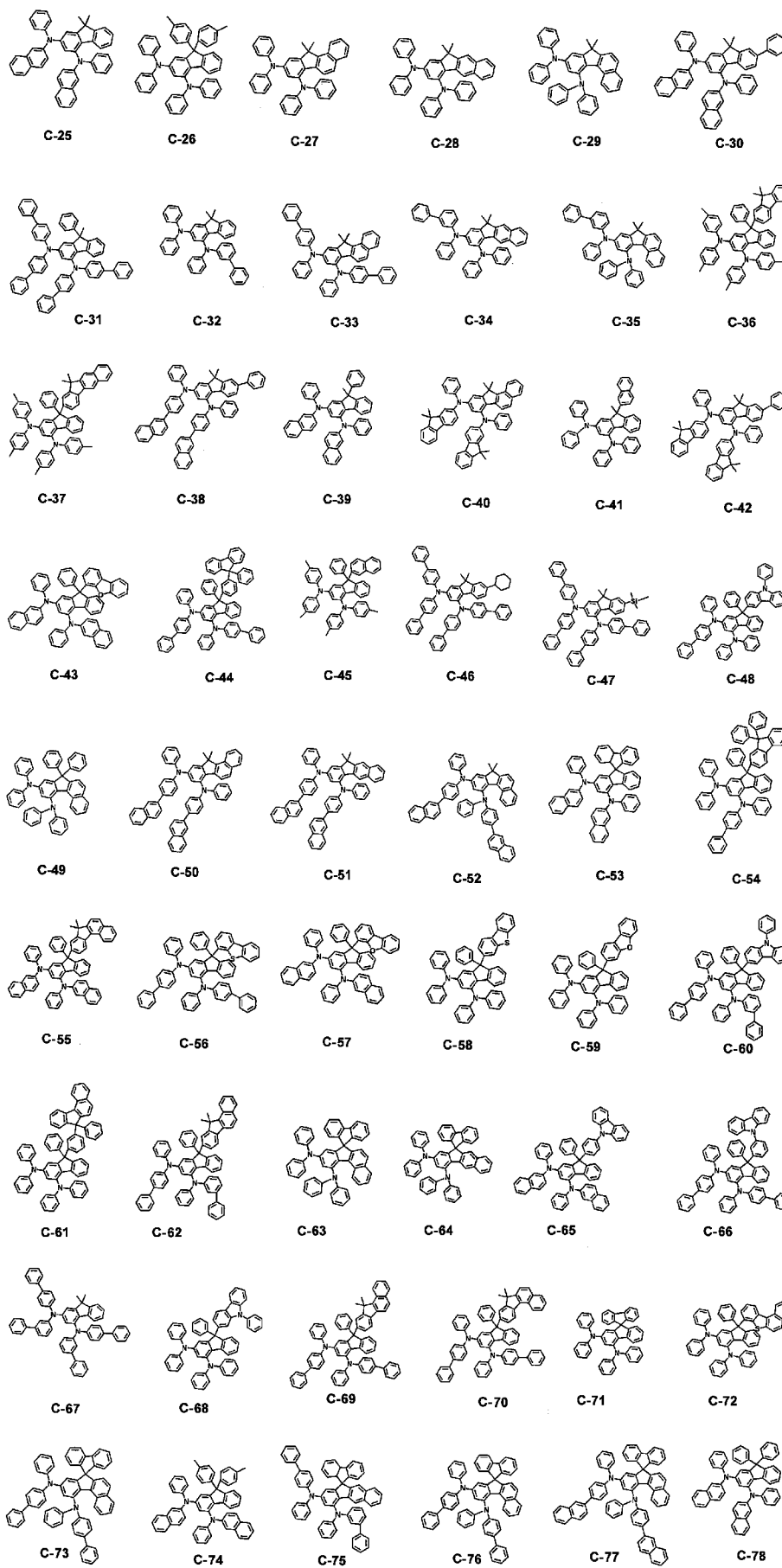
R_6 to R_8 each independently represent an unsubstituted (C1-C6)alkyl; and

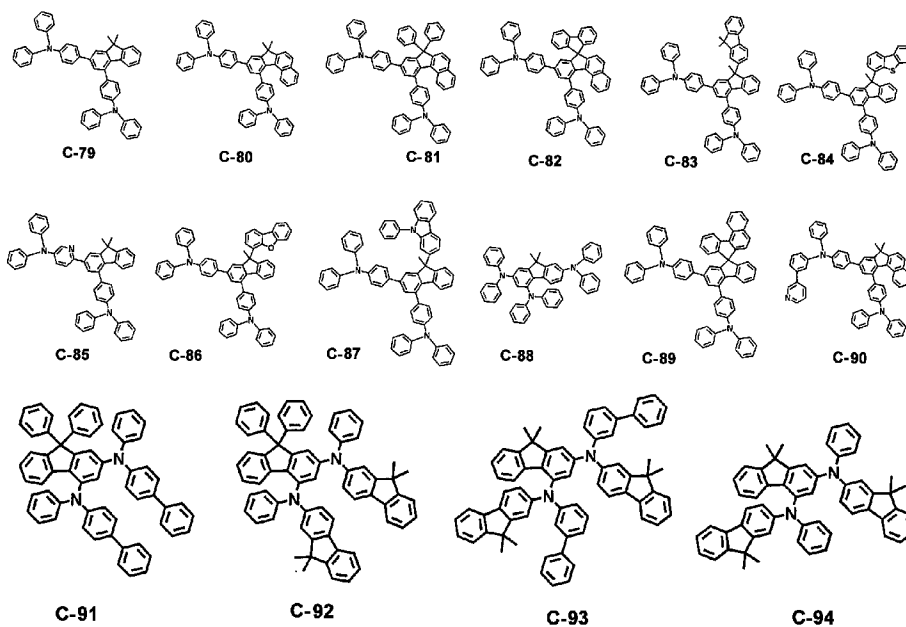
a represents an integer of 1 to 2.

[Claim 6]

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







[Claim 7]

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

INTERNATIONAL SEARCH REPORT

 International application No.
PCT/KR2014/012893

A. CLASSIFICATION OF SUBJECT MATTER

C07C 211/61 (2006.01) C07D 209/82 (2006.01) C07D 213/02 (2006.01) C07D 307/91 (2006.01) C07D 333/76 (2006.01)
C07F 7/02 (2006.01) C09K 11/06 (2006.01) H01L 51/54 (2006.01) H05B 33/14 (2006.01)

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

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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

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

STN: Chemical Abstracts Registry and CAPLUS. Substructure search of general formula (1) in claim 1.
 Applicant and inventor name search: Patentscope and Auspat

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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



Further documents are listed in the continuation of Box C



See patent family annex

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

Date of the actual completion of the international search 20 February 2015	Date of mailing of the international search report 20 February 2015
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaustalia.gov.au	Authorised officer Scott Makin AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. +61 2 6283 7971

INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2014/012893
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	CHEN, X.-H. et al, "Synthesis and Properties of 1-(4-Aminophenyl)-2,4-dicyano-3-diethylamino-9,9-diethylfluorenes: Potential Fluorescent Material", Chemistry Letters, 2008, vol. 37(6), pp. 570-571 Abstract; Table 1 and cpds. 1-4, fig. 1, p. 570; Figs. 2 and 3, p. 571	1-7
P,X	WO 2014/200260 A1 (SK CHEMICALS CO., LTD.) 18 December 2014 Abstract; Claims; Compound 5, p. 9 and claim 6	1, 3-5, 7
P,X	WO 2014/171779 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 23 October 2014 Abstract; Claims; Compound C-92, p. 12 and claim 6	1-3, 7
P,X	WO 2014/129846 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 28 August 2014 Abstract; Claims; Compounds C-127, C-128, p. 13 and claim 9	1-4, 7
P,X	WO 2014/021572 A1 (SK CHEMICALS CO., LTD.) 06 February 2014 Abstract; Claims; Compounds 12-16, p. 10 and claim 7	1, 3-5, 7
P,X	WO 2014/015935 A2 (MERCK PATENT GMBH) 30 January 2014 Abstract; Claims; Formula 245, p. 53	1, 3-5, 7

INTERNATIONAL SEARCH REPORT Information on patent family members		International application No. PCT/KR2014/012893	
This Annex lists known patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.			
Patent Document/s Cited in Search Report		Patent Family Member/s	
Publication Number	Publication Date	Publication Number	Publication Date
WO 2014/200260 A1	18 December 2014		
WO 2014/171779 A1	23 October 2014	KR 20140125576 A	29 Oct 2014
WO 2014/129846 A1	28 August 2014	KR 20140104895 A	29 Aug 2014
		TW 201439277 A	16 Oct 2014
WO 2014/021572 A1	06 February 2014	KR 20140018789 A	13 Feb 2014
		TW 201408626 A	01 Mar 2014
WO 2014/015935 A2	30 January 2014	TW 201425275 A	01 Jul 2014
End of Annex			
Due to data integration issues this family listing may not include 10 digit Australian applications filed since May 2001. Form PCT/ISA/210 (Family Annex)(July 2009)			