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

(57) **Abstract:** The present invention relates to a novel organic electroluminescent compound and an organic electroluminescent device containing the same. Since the organic electroluminescent compounds according to the present invention have high efficiency in transporting electrons, crystallization may be prevented when manufacturing a device. Further, the compounds have good layer formability and improve the current characteristics of the device. Therefore, they can produce an organic electroluminescent device having lowered driving voltages and enhanced power efficiency.



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Description

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

Technical Field

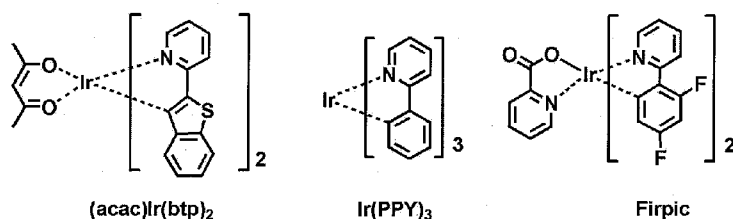
- [1] The present invention relates to novel organic electroluminescent compounds and an organic electroluminescent device using the same.

[2]

Background Art

- [3] An electroluminescent (EL) device is a self-light-emitting device which has advantages over other types of display devices in that it provides a wider viewing angle, a greater contrast ratio, and has a faster response time. An organic EL device was first developed by Eastman Kodak, by using small molecules which are aromatic diamines, and aluminum complexes as a material for forming a light-emitting layer [Appl. Phys. Lett. 51, 913, 1987].
- [4] The most important factor to determine luminous efficiency in an organic EL device is a light-emitting material. Until now, fluorescent materials have been widely used as a light-emitting material. However, in view of electroluminescent mechanisms, phosphorescent materials theoretically show four (4) times higher luminous efficiency than fluorescent materials. Thus, recently, phosphorescent materials have been investigated. Iridium(III) complexes have been widely known as phosphorescent materials, including bis(2-(2'-benzothienyl)-pyridinato-N,C3')iridium(acetylacetonate) ((acac)Ir(btp)₂), tris(2-phenylpyridine)iridium (Ir(ppy)₃) and bis(4,6-difluorophenylpyridinato-N,C2)picolinate iridium (Firpic) as red, green and blue materials, respectively. Especially, a lot of phosphorescent materials are being researched in Japan, Europe and U.S.A recently.

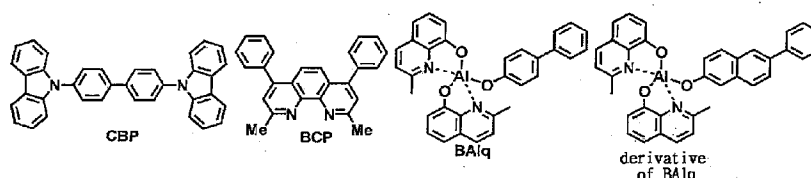
[5]



- [6] At present, 4,4'-N,N'-dicarbazol-biphenyl (CBP) is the most widely known host material for phosphorescent substances. Further, an organic EL device having high efficiency using bathocuproine (BCP) and aluminum(III)bis(2-methyl-8-quinolate)(4-phenylphenolate) (BALq) for a hole blocking layer is known, and Pioneer (Japan) et al. developed a high performance

organic EL device employing a derivative of BA1q as a host material.

[7]



[8]

Though these phosphorous host materials provide good light-emitting characteristics, they have the following disadvantages: (1) Due to their low glass transition temperature and poor thermal stability, their degradation may occur during a high-temperature deposition process in a vacuum. (2) The power efficiency of an organic EL device is given by $[(\pi/\text{voltage}) \times \text{current efficiency}]$, and the power efficiency is inversely proportional to the voltage, and thus the power efficiency should be high in order to reduce power consumption. Although an organic EL device comprising phosphorescent materials provides higher current efficiency (cd/A) than one comprising fluorescent materials, when the conventional materials such as BA1q or CBP are used as phosphorescent host materials, a significantly high driving voltage is necessary compared to an organic EL device using a fluorescent material. Thus, there is no merit in terms of power efficiency (lm/W). (3) Further, the operation lifetime of an organic EL device is short and luminous efficiency is still required to be improved.

[9]

International Patent Publication No. WO 2006/049013 discloses compounds for organic electroluminescent materials having a condensed bicyclic group as a backbone structure. However, it does not disclose a compound having a nitrogen-containing condensed bicyclic group together with a carbazole group which is fused with an aromatic ring-fused heterocycloalkyl or cycloalkyl group.

[10]

Disclosure of Invention

Technical Problem

[11]

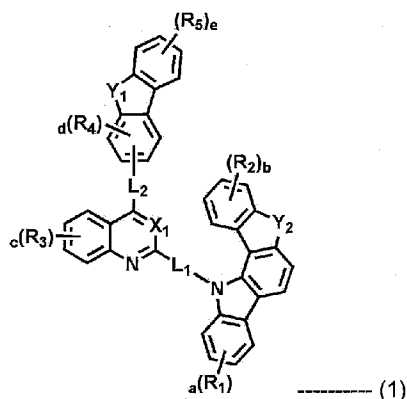
The objective of the present invention is to provide an organic electroluminescent compound which has an excellent structure imparting high luminous efficiency and a long operation lifetime to a device, and having proper color coordination; and an organic electroluminescent device having high efficiency and a long lifetime, using said compounds.

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 3- to 30-membered heteroarylene group, a substituted or unsubstituted (C6-C30)arylene group, or a substituted or unsubstituted (C3-C30)cycloalkylene group;

[16] X_1 represents CH or N;

[17] Y_1 and Y_2 each independently represent -O-, -S-, -CR₆R₇- or -NR₈-;

[18] R_1 to R_5 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted 5- to 7-membered heterocycloalkyl group, a substituted or unsubstituted (C6-C30)aryl(C1-C30)alkyl group, -NR₁₁R₁₂, -SiR₁₃R₁₄R₁₅, -SR₁₆, -OR₁₇, a cyano group, a nitro group, or a hydroxyl group; or R_4 and R_5 each independently are linked to an adjacent substituent(s) via a substituted or unsubstituted (C3-C30)alkylene or a substituted or unsubstituted (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one heteroatom selected from nitrogen, oxygen and sulfur;

[19] R_6 to R_8 and R_{11} to R_{17} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted 5- to 7-membered heterocycloalkyl group, or a substituted or unsubstituted (C3-C30)cycloalkyl group; or are linked to an adjacent substituent(s) via a substituted or unsubstituted (C3-C30)alkylene or a substituted or unsubstituted (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one heteroatom selected from nitrogen, oxygen and sulfur;

[20] a, b, c and e each independently represent an integer of 1 to 4; where a, b, c or e is an integer of 2 or more, each of R_1 , each of R_2 , each of R_3 or each of R_5 is the same or

different;

[21] d represents an integer of 1 to 3; where d is an integer of 2 or more, each of R₄ is the same or different; and

[22] the heterocycloalkyl group and the heteroaryl(ene) group contain at least one heteroatom selected from B, N, O, S, P(=O), Si and P.

[23]

Advantageous Effects of Invention

[24] The organic electroluminescent compounds according to the present invention can manufacture an organic electroluminescent device which has high luminous efficiency and a long operation lifetime.

[25] In addition, since the organic electroluminescent compounds according to the present invention have high efficiency in transporting electrons, crystallization could be prevented when manufacturing a device. Further, the compounds have good layer formability and improve the current characteristics of the device. Therefore, they can produce an organic electroluminescent device having lowered driving voltages and enhanced power efficiency.

[26]

Mode for the Invention

[27] 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.

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

[29] Herein, “(C1-C30)alkyl(ene)” is meant to be a linear or branched alkyl(ene) having 1 to 30 carbon atoms, in which the number of carbon atoms is preferably 1 to 20, more preferably 1 to 10, and includes methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, etc., but are not limited thereto.

[30] Herein, “(C2-C30) alkenyl(ene)” is meant to be a linear or branched alkenyl(ene) 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., but are not limited thereto.

[31] Herein, “(C2-C30)alkynyl” is a linear or branched alkynyl having 2 to 30 carbon atoms, in which the number of carbon atoms is preferably 2 to 20, more preferably 2 to 10, and includes ethynyl, 1-propynyl, 2-propynyl, 1-butyne, 2-butyne, 3-butyne, 1-methylpent-2-ynyl, etc., but are not limited thereto.

[32] Herein, “(C1-C30)alkoxy” is a linear or branched alkoxy having 1 to 30 carbon

atoms, in which the number of carbon atoms is preferably 1 to 20, more preferably 1 to 10, and includes methoxy, ethoxy, propoxy, isopropoxy, 1-ethylpropoxy, etc., but are not limited thereto.

- [33] Herein, “(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., but are not limited thereto.
- [34] Herein, “(C3-C30)cycloalkylene” is one formed by removing a hydrogen from cycloalkyl having 3 to 30, preferably 3 to 20, more preferably 3 to 7 carbon atoms.
- [35] Herein, “5- to 7-membered heterocycloalkyl” is a cycloalkyl having at least one heteroatom selected from B, N, O, S, P(=O), Si and P, preferably N, O and S, and 5 to 7 ring backbone atoms, and includes tetrahydrofurane, pyrrolidine, thiolan, tetrahydropyran, etc., but are not limited thereto.
- [36] Herein, “halogen” includes F, Cl, Br and I.
- [37] Herein, “(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 12, and includes phenyl, biphenyl, terphenyl, naphthyl, fluorenyl, phenanthrenyl, anthracenyl, indenyl, triphenylenyl, pyrenyl, tetracenyl, perylenyl, chrysenyl, naphthacenyl, fluoranthenyl, etc., but are not limited thereto. The naphthyl includes 1-naphthyl, 2-naphthyl, etc.; the anthracenyl includes 1-anthracenyl, 2-anthracenyl, 9-anthracenyl, etc.; the phenanthrenyl includes 1-phenanthrenyl, 2-phenanthrenyl, 3-phenanthrenyl, 4-phenanthrenyl, 9-phenanthrenyl, etc.; the naphthacenyl includes 1-naphthacenyl, 2-naphthacenyl, 9-naphthacenyl, etc.; the pyrenyl includes 1-pyrenyl, 2-pyrenyl, 4-pyrenyl, etc.; the biphenyl includes 2-biphenyl, 3-biphenyl, 4-biphenyl, etc.; the terphenyl includes p-terphenyl-4-yl, p-terphenyl-3-yl, p-terphenyl-2-yl, m-terphenyl-4-yl, m-terphenyl-3-yl, m-terphenyl-2-yl, etc.; the fluorenyl includes 1-fluorenyl, 2-fluorenyl, 3-fluorenyl, 4-fluorenyl, 9-fluorenyl, etc.
- [38] Herein, “3- to 30-membered heteroaryl(ene)” is an aryl having at least one, preferably 1 to 4 heteroatoms selected from the group consisting of B, N, O, S, P(=O), Si and P, and 3 to 30 ring backbone atoms; is a monocyclic ring, or a fused ring condensed with at least one benzene ring; has preferably 5 to 21, more preferably 5 to 12 ring backbone atoms; may be partially saturated; may be one formed by linking at least one heteroaryl or aryl group to a heteroaryl group via a single bond(s). The heteroaryl(ene) includes a divalent aryl group, which forms an N-oxide, a quaternary salt, etc., by oxidation or quaternarization of a heteroatom existing in a ring. The heteroaryl(ene) 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., a fused ring-type heteroaryl including benzofuranyl, benzothiophenyl, dibenzofuranyl, dibenzothiophenyl, isobenzofuranyl, benzoimidazolyl, benzothiazolyl, benzoisothiazolyl, benzoisoxazolyl, benzoxazolyl, isoindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolyl, quinazolinyl, quinoxalinyl, carbazolyl, phenanthridinyl, benzodioxolyl, acridinyl, phenanthrolinyl, phenazinyl, phenothiazinyl, phenoxazinyl, etc., and N-oxides (for example, pyridyl N-oxide and quinolyl N-oxide) and quaternary salts thereof, but are not limited thereto. The pyrrolyl includes 1-pyrrolyl, 2-pyrrolyl, 3-pyrrolyl, etc.; the pyridyl includes 2-pyridyl, 3-pyridyl, 4-pyridyl, etc.; the indolyl includes 1-indolyl, 2-indolyl, 3-indolyl, 4-indolyl, 5-indolyl, 6-indolyl, 7-indolyl, etc.; the isoindolyl includes 1-isoindolyl, 2-isoindolyl, 3-isoindolyl, 4-isoindolyl, 5-isoindolyl, 6-isoindolyl, 7-isoindolyl, etc.; the furyl includes 2-furyl, 3-furyl, etc.; the benzofuranyl includes 2-benzofuranyl, 3-benzofuranyl, 4-benzofuranyl, 5-benzofuranyl, 6-benzofuranyl, 7-benzofuranyl, etc.; isobenzofuranyl includes 1-isobenzofuranyl, 3-isobenzofuranyl, 4-isobenzofuranyl, 5-isobenzofuranyl, 6-isobenzofuranyl, 7-isobenzofuranyl, etc.; quinolyl includes 3-quinolyl, 4-quinolyl, 5-quinolyl, 6-quinolyl, 7-quinolyl, 8-quinolyl, etc.; the isoquinolyl includes 1-isoquinolyl, 3-isoquinolyl, 4-isoquinolyl, 5-isoquinolyl, 6-isoquinolyl, 7-isoquinolyl, 8-isoquinolyl, etc.; the quinoxalinyl includes 2-quinoxalinyl, 5-quinoxalinyl, 6-quinoxalinyl, etc.; the carbazolyl includes 1-carbazolyl, 2-carbazolyl, 3-carbazolyl, 4-carbazolyl, 9-carbazolyl, etc.; the phenanthridinyl includes 1-phenanthridinyl, 2-phenanthridinyl, 3-phenanthridinyl, 4-phenanthridinyl, 6-phenanthridinyl, 7-phenanthridinyl, 8-phenanthridinyl, 9-phenanthridinyl, 10-phenanthridinyl, etc.; the acridinyl includes 1-acridinyl, 2-acridinyl, 3-acridinyl, 4-acridinyl, 9-acridinyl, etc.; the phenanthrolinyl includes 1,7-phenanthroline-2-yl, 1,7-phenanthroline-3-yl, 1,7-phenanthroline-4-yl, 1,7-phenanthroline-5-yl, 1,7-phenanthroline-6-yl, 1,7-phenanthroline-8-yl, 1,7-phenanthroline-9-yl, 1,7-phenanthroline-10-yl, 1,8-phenanthroline-2-yl, 1,8-phenanthroline-3-yl, 1,8-phenanthroline-4-yl, 1,8-phenanthroline-5-yl, 1,8-phenanthroline-6-yl, 1,8-phenanthroline-7-yl, 1,8-phenanthroline-9-yl, 1,8-phenanthroline-10-yl, 1,9-phenanthroline-2-yl, 1,9-phenanthroline-3-yl, 1,9-phenanthroline-4-yl, 1,9-phenanthroline-5-yl, 1,9-phenanthroline-6-yl, 1,9-phenanthroline-7-yl, 1,9-phenanthroline-8-yl, 1,9-phenanthroline-10-yl, 1,10-phenanthroline-2-yl, 1,10-phenanthroline-3-yl, 1,10-phenanthroline-4-yl, 1,10-phenanthroline-5-yl, 2,9-phenanthroline-1-yl, 2,9-phenanthroline-3-yl, 2,9-phenanthroline-4-yl, 2,9-phenanthroline-5-yl, 2,9-phenanthroline-6-yl, 2,9-phenanthroline-7-yl, 2,9-phenanthroline-8-yl, 2,9-phenanthroline-10-yl, 2,8-phenanthroline-1-yl, 2,8-phenanthroline-3-yl, 2,8-phenanthroline-4-yl,

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[39] 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.

[40] Substituents of the substituted alkyl(ene) group, the substituted alkenyl group, the substituted cycloalkyl(ene) group, the substituted heterocycloalkyl group, the substituted aryl(ene) group, the substituted heteroaryl(ene) group, the substituted arylalkyl group and the substituted aromatic ring in L₁, L₂, R₁ to R₈, and R₁₁ to R₁₇ groups each independently are preferably at least one selected from the group consisting of deuterium, a halogen, a (C1-C30)alkyl group substituted or unsubstituted with a halogen, a (C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a 3- to 30-membered heteroaryl group substituted with a (C1-C30)alkyl, a 3- to 30-membered heteroaryl group substituted with a (C6-C30)aryl, a (C3-C30)cycloalkyl group, a 5- to 7-membered heterocycloalkyl group, a tri(C1-C30)alkylsilyl group, a tri(C6-C30)arylsilyl group, a di(C1-C30)alkyl(C6-C30)arylsilyl group, a (C1-C30)alkyldi(C6-C30)arylsilyl group, a (C2-C30)alkenyl group, a (C2-C30)alkynyl group, a cyano group, a N-carbazolyl group, a di(C1-C30)alkylamino group, a di(C6-C30)arylamino group, a (C1-C30)alkyl(C6-C30)arylamino group, a di(C6-C30)arylboronyl group, a di(C1-C30)alkylboronyl group, a (C1-C30)alkyl(C6-C30)arylboronyl group, a (C6-C30)aryl(C1-C30)alkyl group, a (C1-C30)alkyl(C6-C30)aryl group, a carboxyl group, a nitro group and a hydroxyl group; more preferably at least one selected from the group consisting of deuterium, a halogen, a (C1-C20)alkyl group substituted or unsubstituted with a halogen and a (C6-C20)aryl group; even more preferably at least one selected from the group consisting of deuterium, fluorine, methyl, phenyl and naphthyl.

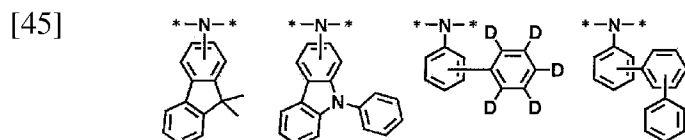
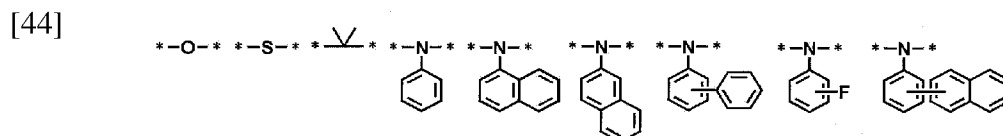
[41] Specifically, in the above formula 1, L₁ and L₂ each independently represent a single

bond, a 3- to 30-membered heteroarylene group, a (C6-C30)arylene group, or a (C6-C30)cycloalkylene group; X_1 is CH or N; Y_1 and Y_2 each independently represent -O-, -S-, -CR₆R₇- or -NR₈-; R_1 to R_5 each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group, a (C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a N-carbazolyl group, -NR₁₁R₁₂, or -SiR₁₃R₁₄R₁₅; or R_4 and R_5 each independently are linked to an adjacent substituent(s) via a (C3-C30)alkylene or a (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one heteroatom selected from nitrogen, oxygen and sulfur; R_6 to R_8 each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group; R_{11} to R_{15} each independently represent a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group; and the arylene group, the heteroarylene group and the cycloalkylene group in L_1 and L_2 , and the alkyl group, the aryl group and the heteroaryl group in R_1 to R_8 and R_{11} to R_{15} can be substituted with at least one selected from the group consisting of deuterium, a halogen, a (C1-C30)alkyl group substituted or unsubstituted with a halogen, a (C6-C30)aryl group, a (C1-C30)alkyl(C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a 3- to 30-membered heteroaryl group substituted with a (C6-C30)aryl, a (C3-C30)cycloalkyl group and a (C6-C30)aryl(C1-C30)alkyl group.

- [42] In the above formula 1, L_1 and L_2 each independently are preferably selected from the group consisting of a single bond, cyclopropylene, cyclobutylene, cyclopentylene, cyclohexylene, cycloheptylene, cyclooctylene, phenylene, methylphenylene, naphthylene, biphenylene, terphenylene, anthracenylene, indenylene, fluorenylene, phenanthrenylene, triphenylenylene, pyrenylene, phenylenylene, crycenylene, naphthacenylene, fluoranthenyl, furylene, thiophenylene, pyrrolylene, imidazolylene, pyrazolylene, thiazolylene, thiadiazolylene, isothiazolylene, isoxazolylene, oxazolylene, oxadiazolylene, triazinylene, tetrazinylene, triazolylene, furazanylene, pyridylene, pyrazinylene, pyrimidinylene, pyridazinylene, benzofuranylene, benzothiophenylene, isobenzofuranylene, benzoimidazolylene, benzothiazolylene, benzoisothiazolylene, benzoisoxazolylene, benzoxazolylene, isoindolylene, indolylene, indazolylene, benzothiadiazolylene, quinolylene, isoquinolylene, cinnolinylene, quinazolinylene, quinoxalinylene, carbazolylene, phenanthridinylene, benzodioxolylene, dibenzofuranylene and dibenzothiophenylene, and the L_1 and L_2 can be substituted with at least one selected from the group consisting of deuterium, a halogen, (C1-C30)alkyl group substituted or unsubstituted with a halogen, a (C6-C30)aryl group, a (C1-C30)alkyl(C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a 3- to 30-membered heteroaryl group substituted with a (C6-C30)aryl, a (C3-C30)cycloalkyl group and a (C6-C30)aryl(C1-C30)alkyl. In the above formula 1, L_1 and L_2 each inde-

pendently are more preferably selected from the group consisting of a single bond, phenylene, methylphenylene and cyclohexylene.

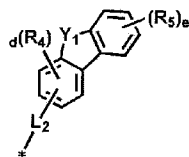
[43] In the above formula 1, Y_1 and Y_2 each independently selected from the following structures:



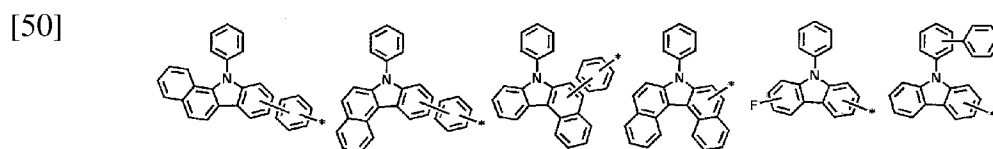
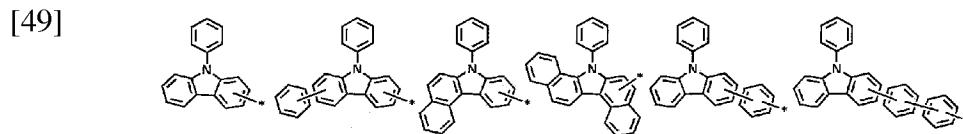
[46] In the above formula 1, R_1 to R_5 each independently are preferably a (C6-C20)aryl group, an 5- to 21-membered heteroaryl group, $-NR_{11}R_{12}$ or $-SiR_{13}R_{14}R_{15}$; or are linked to an adjacent substituent(s) via a substituted or unsubstituted (C3-C30)alkylene or a substituted or unsubstituted (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring, more preferably phenyl, carbazolyl, diphenylamino or methyldiphenylsilyl; or are linked to an adjacent substituent(s) via a substituted or unsubstituted (C3-C30)alkylene or a substituted or unsubstituted (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring. The R_{11} to R_{15} each independently are preferably a (C1-C6)alkyl group or a (C6-C20)aryl group, more preferably methyl or phenyl.

[47] In the above formula 1, R_6 to R_8 each independently are preferably hydrogen, a (C1-C6)alkyl group, a substituted or unsubstituted (C6-C20)aryl group, or a substituted or unsubstituted 5- to 21-membered heteroaryl group, more preferably hydrogen; methyl; phenyl; biphenyl; naphthyl; phenyl substituted with deuterium; phenyl substituted with fluorine; fluorenyl substituted with methyl; or naphthylphenyl.

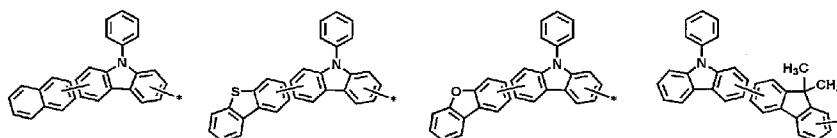
[48] In the above formula 1,



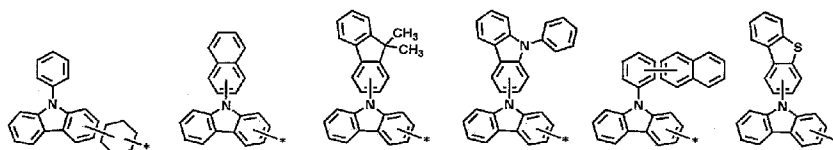
are not limited thereto:



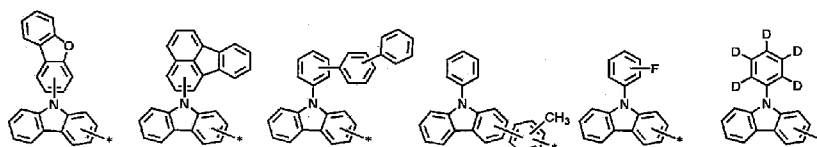
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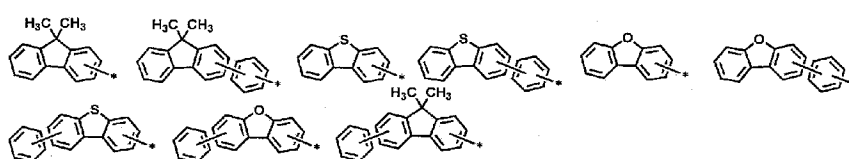
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[53]

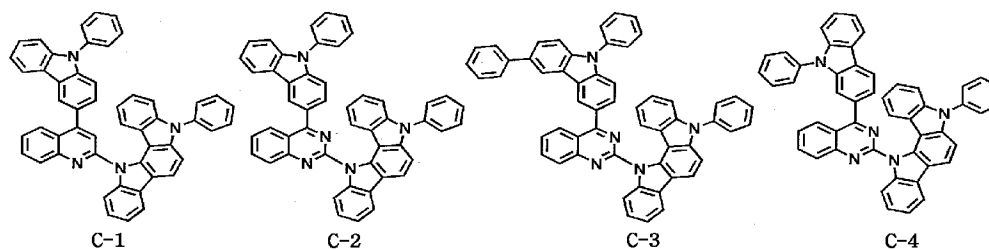


[54]

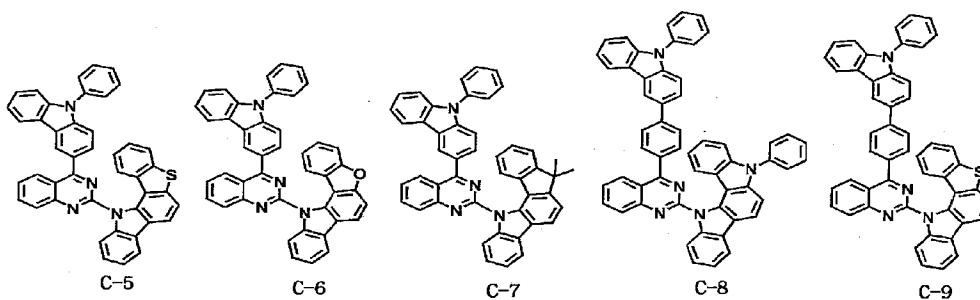


[55] The representative organic electroluminescent compounds of the present invention include the following compounds, but are not limited thereto:

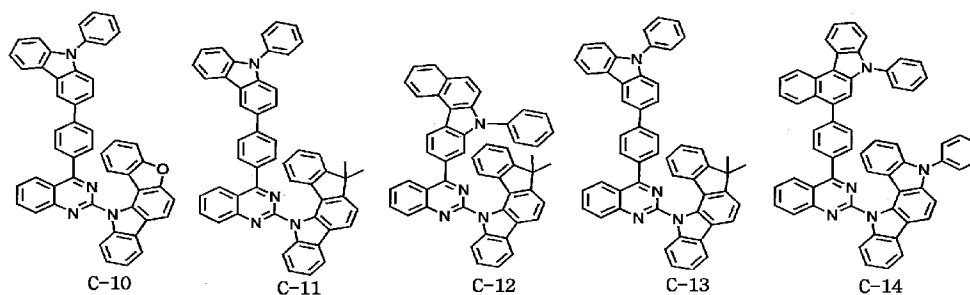
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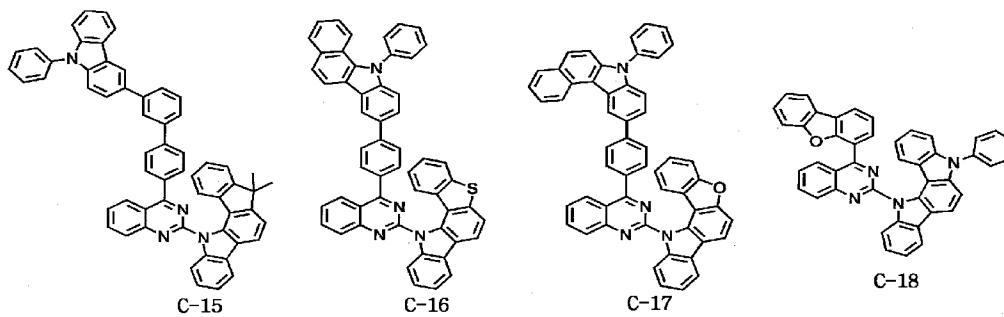
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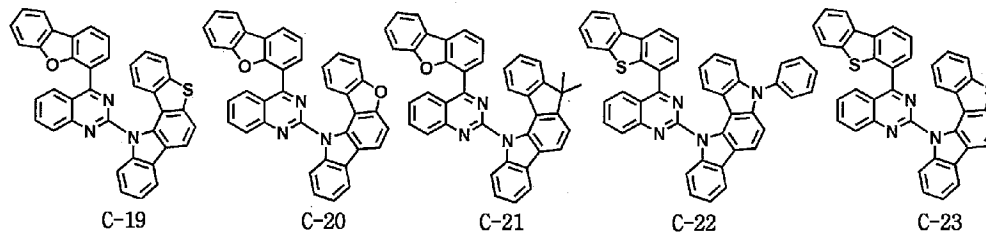
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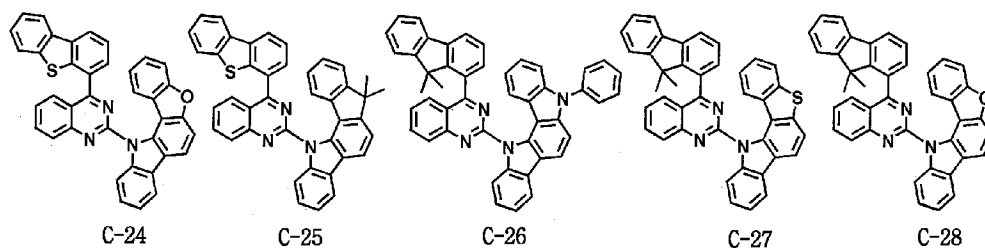
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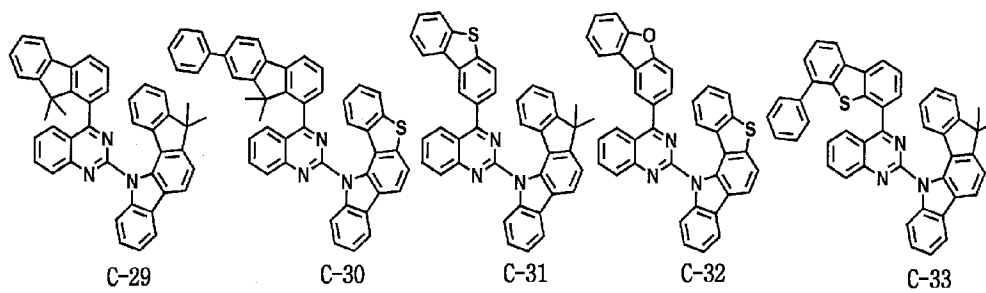
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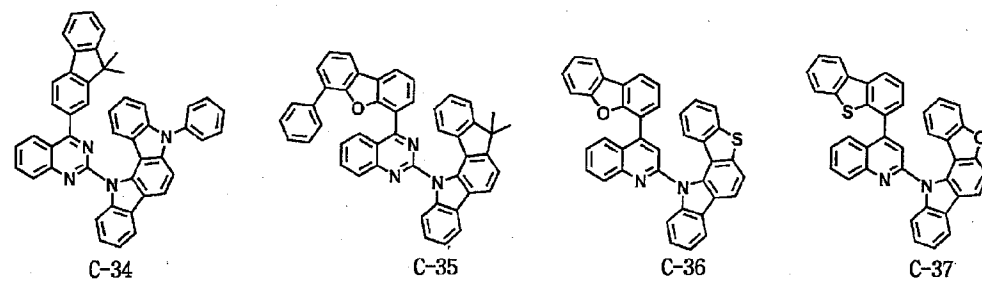
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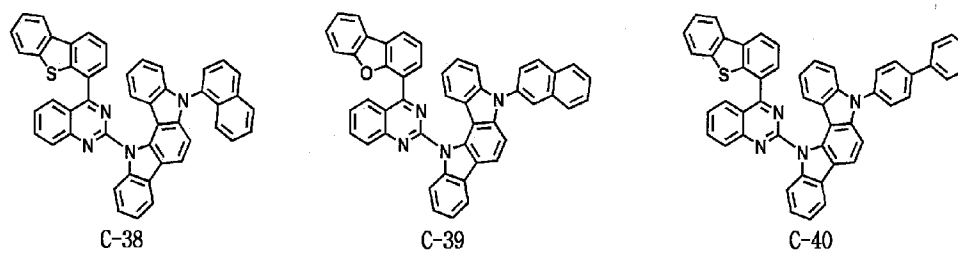
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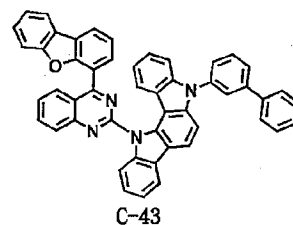
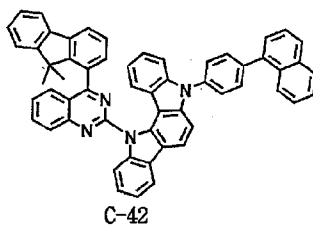
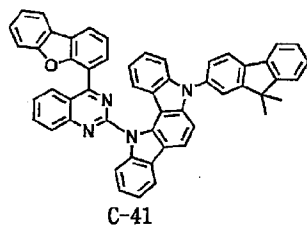
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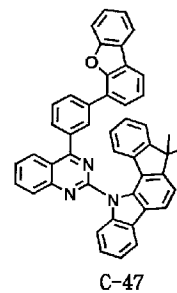
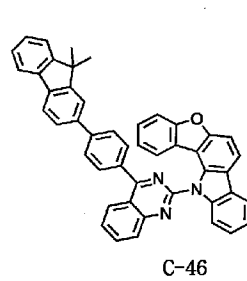
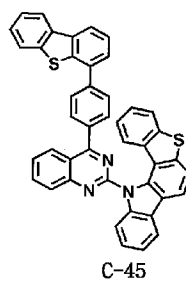
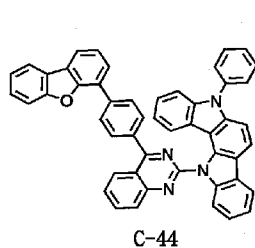
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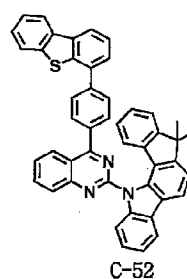
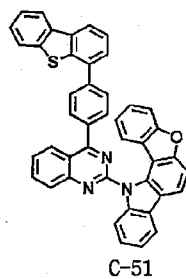
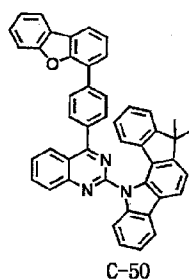
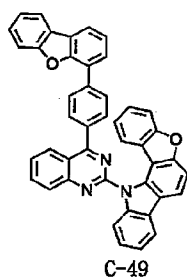
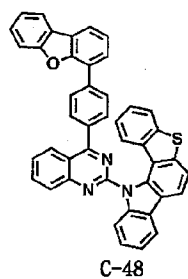
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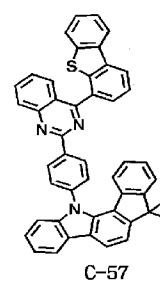
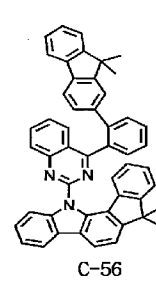
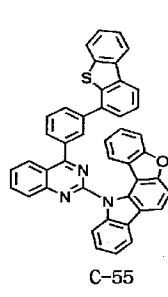
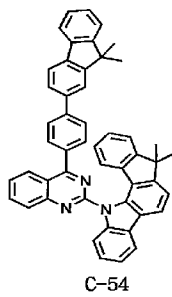
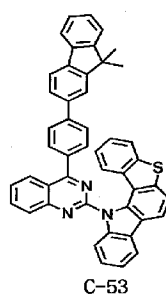
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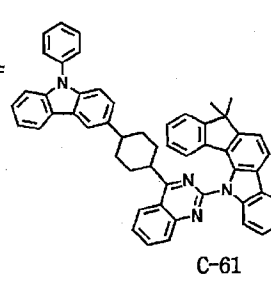
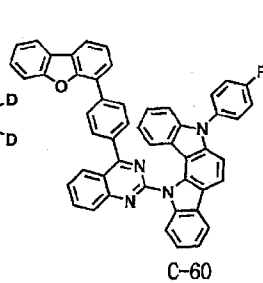
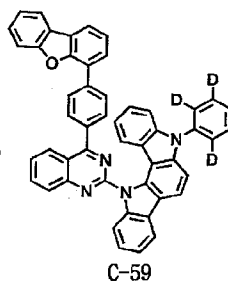
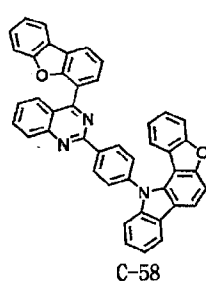
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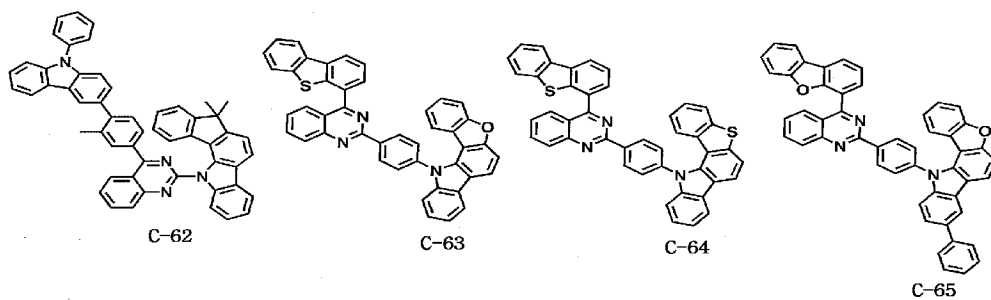
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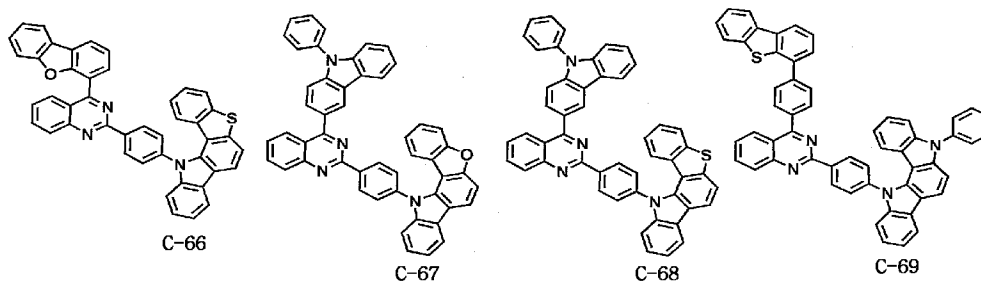
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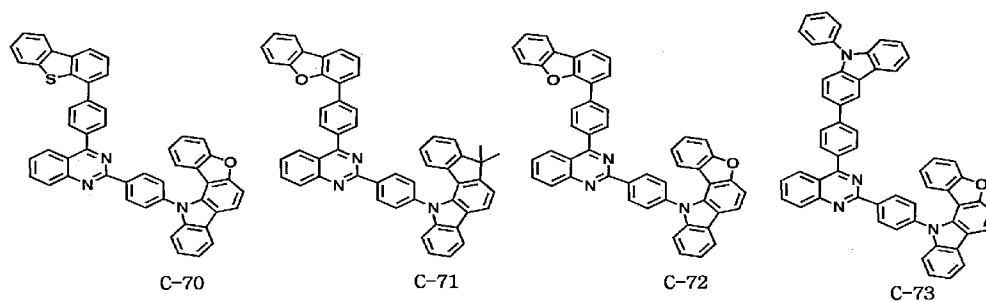
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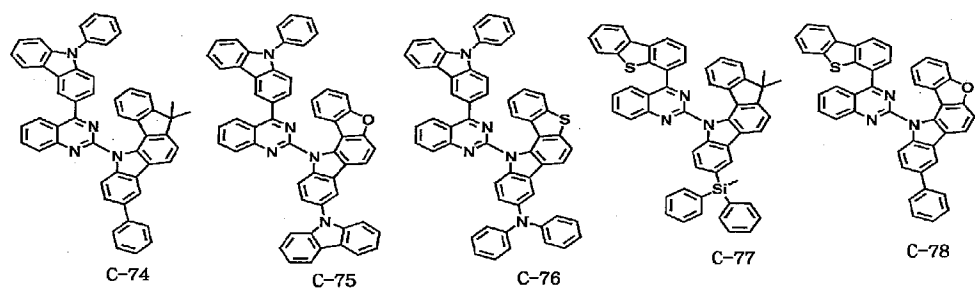
[71]



[72]



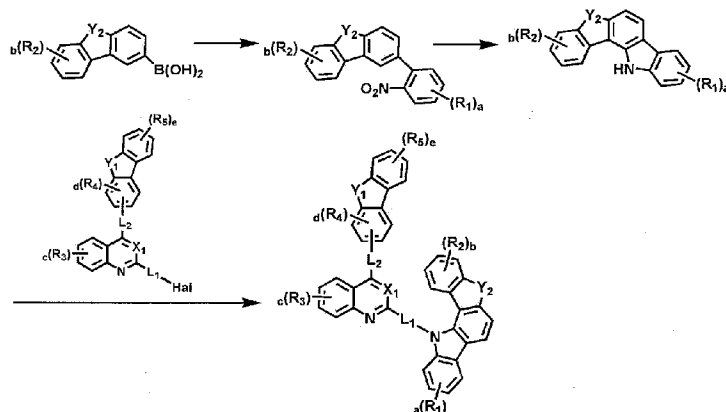
[73]



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

[75] [Reaction Scheme 1]

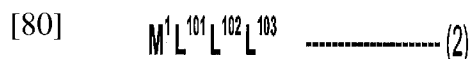
[76]



[77] wherein L_1 , L_2 , X_1 , Y_1 , Y_2 , R_1 to R_5 , a , b , c , d and e are as defined in formula 1 above, and Hal represents a halogen.

[78] In addition, the present invention provides an organic electroluminescent material comprising the compound of formula 1 and an organic electroluminescent device comprising the material. 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. Said organic electroluminescent device comprises a first electrode, a second electrode, and at least one organic layer between said first and second electrodes. Said organic layer comprises at least one organic electroluminescent compound of formula 1 according to the present invention. Further, said organic layer comprises a light-emitting layer in which the organic electroluminescent compound of formula 1 is comprised as a host material.

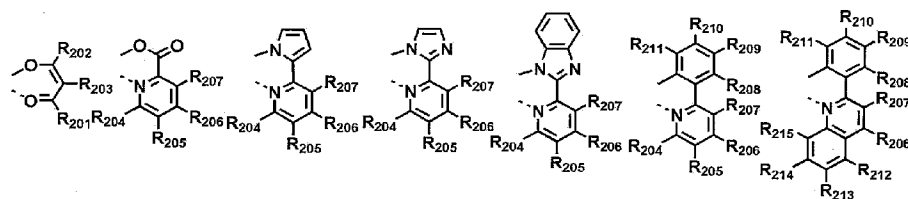
[79] In addition, a phosphorescent dopant, which is used for an organic electroluminescent device together with the host material according to the present invention, may be selected from compounds represented by the following formula 2:



[81] wherein M^1 is selected from the group consisting of Ir, Pt, Pd and Os; and

[82] L^{101} , L^{102} and L^{103} are each independently selected from the following structures:

[83]



[84]

[88] R₂₀₄ to R₂₁₉ each independently represent hydrogen, deuterium, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C1-C30)alkoxy group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted (C2-C30)alkenyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted mono- or di-(C1-C30)alkylamino group, a substituted or unsubstituted mono- or di-(C6-C30)arylamino group, SF₅, a substituted or unsubstituted tri(C1-C30)alkylsilyl group, a substituted or unsubstituted di(C1-C30)alkyl(C6-C30)arylsilyl group, a substituted or unsubstituted tri(C6-C30)arylsilyl group, a cyano group or a halogen;

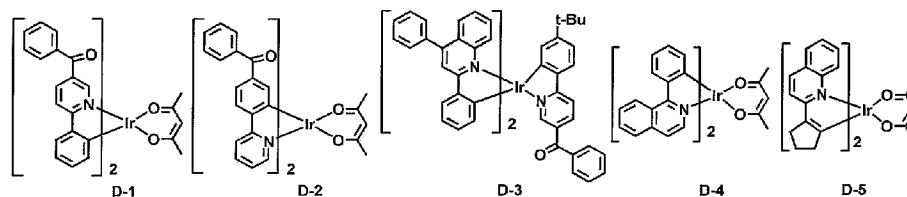
[92] R_{227} to R_{229} each independently represent hydrogen, deuterium, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group or a halogen;

[93] Q represents $R_{231}R_{232}$, $R_{233}R_{234}$ or $R_{237}R_{238}R_{241}R_{242}$; R_{231} to R_{242} each independently

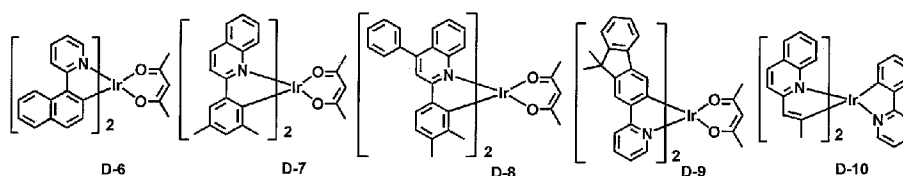
represent hydrogen, deuterium, a (C1-C30)alkyl group unsubstituted or substituted with halogen(s), a (C1-C30)alkoxy group, a halogen, a substituted or unsubstituted (C6-C30)aryl group, a cyano group, or a substituted or unsubstituted (C5-C30)cycloalkyl group, or each of R_{231} to R_{242} may be linked to an adjacent substituent via (C2-C30)alkylene group or (C2-C30)alkenylene group to form a spiro ring or a fused ring or may be linked to R_{207} or R_{208} via a (C2-C30)alkylene group or (C2-C30)alkenylene group to form a saturated or unsaturated fused ring.

[94] The dopants of formula 2 include the following, but are not limited thereto:

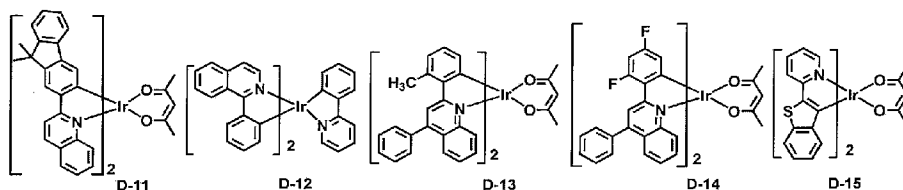
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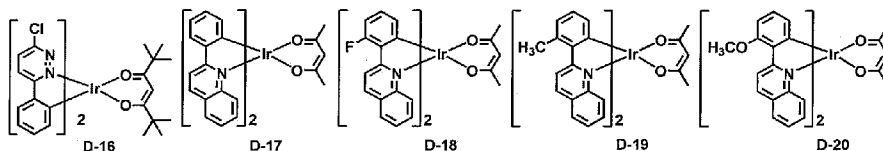
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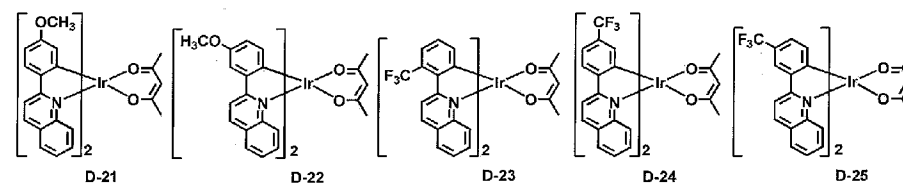
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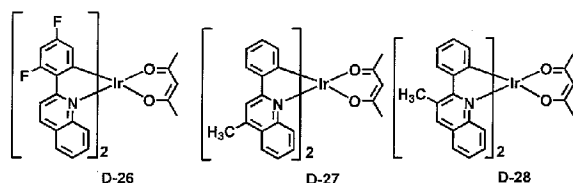
[98]



[99]



[100]



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

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

[103] In addition, the organic electroluminescent device may emit white light by further comprising at least one light-emitting layer which comprises a blue electroluminescent compound, a red electroluminescent compound or a green electroluminescent compound, besides the organic electroluminescent compound according to the present invention.

[104] Preferably, in the organic electroluminescent device according to the present invention, at least one layer (hereinafter, "a surface layer") selected from a chalcogenide layer, a metal halide layer and a metal oxide layer may be placed on an inner surface(s) of one or both electrode(s). Specifically, it is preferred that a chalcogenide(includes oxides) layer of silicon or aluminum is placed on an anode surface of an electroluminescent medium layer, and a metal halide layer or metal oxide layer is placed on a cathode surface of an electroluminescent medium layer. Such a surface layer provides operation stability for the organic electroluminescent device. Preferably, said chalcogenide includes SiOX($1 \leq X \leq 2$), AlOX($1 \leq X \leq 1.5$), SiON, SiAlON, etc.; said metal halide includes LiF, MgF₂, CaF₂, a rare earth metal fluoride, etc.; and said metal oxide includes Cs₂O, Li₂O, MgO, SrO, BaO, CaO, etc.

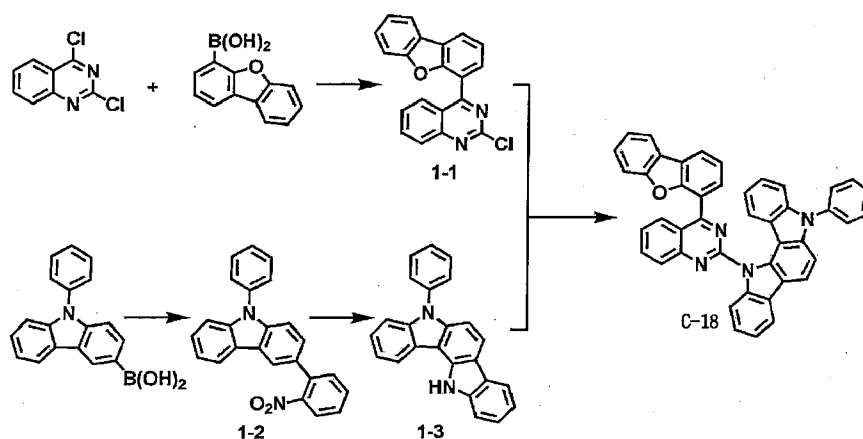
[105] Preferably, in the organic electroluminescent device according to the present invention, a mixed region of an electron transport compound and an reductive dopant, or a mixed region of a hole transport compound and an oxidative dopant may be placed on at least one surface of a pair of electrodes. In this case, the electron transport compound is reduced to an anion, and thus it becomes easier to inject and transport electrons from the mixed region to an electroluminescent medium. Further, the hole

transport compound is oxidized to a cation, and thus it becomes easier to inject and transport holes from the mixed region to the electroluminescent medium. Preferably, the oxidative dopant includes various Lewis acids and acceptor compounds; and the reductive dopant includes alkali metals, alkali metal compounds, alkaline earth metals, rare-earth metals, and mixtures thereof. A reductive dopant layer may be employed as a charge generating layer to prepare an electroluminescent device having two or more electroluminescent layers and emitting white light.

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

[107] Example 1: Preparation of compound C-18

[108]



[109] Preparation of compound 1-1

[110] After dissolving 2,4-dichloroquinazoline (50 g, 251 mmol) and dibenzo[*b,d*]furan-4-yl boronic acid (53.2 g, 251 mmol) in a mixture of toluene (1 L) and purified water (200 mL), $\text{Pd(PPh}_3)_4$ (14.5 g, 12.5 mmol) and Na_2CO_3 (80 g, 755 mmol) was added to the mixture. The mixture was stirred for 20 hours at 80°C. The reaction mixture was cooled to room temperature, and the reaction was terminated with ammonium chloride aqueous solution (200 mL). The reaction mixture was extracted with ethyl acetate (EA) 1 L and an aqueous layer was further extracted with dichloromethane (1 L). The obtained organic layer was dried with anhydrous magnesium sulfate, and the organic solvent was removed under reduced pressure. The obtained solid was filtered through silica gel, and the solvent was removed under reduced pressure. The obtained solid was washed with EA (100 mL) to obtain compound 1-1 (50 g, 74%).

[111]

[112] Preparation of compound 1-2

[113] After dissolving 9-phenyl-9H-carbazol-3-yl boronic acid (30 g, 149 mmol),

1-bromo-2-nitrobenzene (51 g, 178.8 mmol), K_2CO_3 (52 g, 372.5 mmol) and $Pd(PPh_3)_4$ (6.8 g, 5.8 mmol) in a mixture of toluene (600 mL), EtOH (150 mL) and purified water (150 mL), the reaction mixture was stirred under reflux for 24 hours. After terminating the reaction, the reaction mixture was cooled to room temperature, and an aqueous layer was removed from the mixture by a gravity separation. The obtained organic layer was concentrated and triturated with methylene chloride (MC), and then was filtered to obtain compound 1-2 (50 g, 92%).

[114]

[115] Preparation of compound 1-3

[116] After dissolving compound 1-2 (50 g, 137 mmol) in a mixture of $P(OEt)_3$ (300 mL) and 1,2-dichlorobenzene (300 mL), the reaction mixture was stirred for 24 hours at $150^\circ C$. After terminating the reaction, the reaction mixture was concentrated under reduced pressure and extracted with EA. Then, the organic layer was concentrated and purified through silica column to obtain compound 1-3 (32 g, 70%).

[117]

[118] Preparation of compound C-18

[119] After suspending compound 1-1 (5.5 g, 16.5 mmol) and compound 1-3 (5.0 g, 15 mmol) in dimethyl formamide (DMF) 80mL, 60% NaH (930 mg, 23.2 mmol) was added to the mixture at room temperature. The reaction mixture was stirred for 12 hours. After adding purified water (1 L), the mixture was filtered under reduced pressure. The obtained solid was triturated with MeOH/EA, was triturated with DMF, and was triturated with EA/tetrahydrofuran (THF). It was dissolved in MC and was filtered through silica. Then, it was triturated with MeOH/EA to obtain compound C-18 (4.6 g, 48.9%).

[120] MS/FAB found 627; calculated 626.70

[121]

[122] Device Example 1: Production of an organic light-emitting diode (OLED) device using the organic electroluminescent compound according to the present invention

[123] An OLED device was produced using the compound according to the present invention. A transparent electrode indium tin oxide (ITO) thin film ($15 \Omega/sq$) on a glass substrate for an OLED device (Samsung Corning, Republic of Korea) was subjected to an ultrasonic washing with trichloroethylene, acetone, ethanol and distilled water, sequentially, and then was stored in isopropanol. Then, the ITO substrate was mounted on a substrate holder of a vacuum vapor depositing apparatus. $N^1N^{1'}-([1,1'-biphenyl]-4,4'-diyl)bis(N^1-(naphthalen-1-yl)-N^4,N^4-diphenylbenzene-1,4-diamine)$ 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, N,N'-di(4-biphenyl)-N,N'-di(4-biphenyl)-4,4'-diaminobiphenyl was introduced into another cell of said vacuum vapor depositing apparatus, and was evaporated by applying electric current to the cell, thereby forming a hole transport layer having a thickness of 20 nm on the hole injection layer. Thereafter, compound C-18 was introduced into one cell of the vacuum vapor depositing apparatus, as a host material, and compound D-7 was introduced into another cell as a dopant. The two materials were evaporated at different rates and were deposited in a doping amount of 4 wt% of dopant with respect to the total amount of the host material and the 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-benzo[d]imidazole was introduced into one cell and lithium quinolate was introduced into another cell. The two materials were evaporated at the same rates and were deposited in a doping amount of 50 wt% for each material 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 1 to 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.

[124] The produced OLED device showed red emission having a luminance of 3,400 cd/m² and a current density of 44.7 mA/cm² at a driving voltage of 5.6 V.

[125]

[126] Comparative Example 1: Production of an OLED device using conventional
[127] electroluminescent material

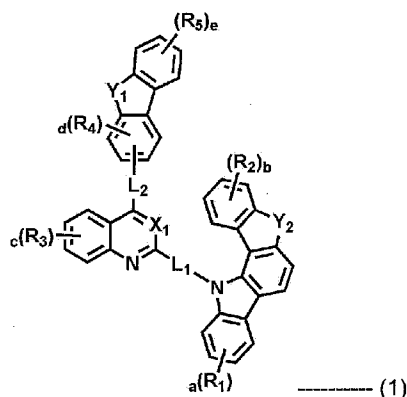
[128] An OLED device was produced in the same manner as that of Device Example 1, except that a light-emitting layer was deposited on the hole transport layer by using 4,4'-N,N'-dicarbazol-biphenyl (CBP) as a host material and compound D-11 as a dopant and that a hole blocking layer having a thickness of 10 nm was deposited between the light-emitting layer and a electron transport layer by using aluminum(III) bis(2-methyl-8-quinolinato)-4-phenylphenolate.

[129] The produced OLED device showed red emission having a luminance of 1,000 cd/m² and a current density of 20.0 mA/cm² at a driving voltage of 8.2 V.

Claims

[Claim 1]

An organic electroluminescent compound represented by the following formula 1:



wherein

L_1 and L_2 each independently represent a single bond, a substituted or unsubstituted 3- to 30-membered heteroarylene group, a substituted or unsubstituted (C6-C30)arylene group, or a substituted or unsubstituted (C3-C30)cycloalkylene group;

X_1 represents CH or N;

Y_1 and Y_2 each independently represent -O-, -S-, $-CR_6R_7$ - or $-NR_8$ -;

R_1 to R_5 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted (C3-C30)cycloalkyl group, a substituted or unsubstituted 5- to 7-membered heterocycloalkyl group, a substituted or unsubstituted (C6-C30)aryl(C1-C30)alkyl group, $-NR_{11}R_{12}$, $-SiR_{13}R_{14}R_{15}$, $-SR_{16}$, $-OR_{17}$, a cyano group, a nitro group, or a hydroxyl group; or R_4 and R_5 each independently are linked to an adjacent substituent(s) via a substituted or unsubstituted (C3-C30)alkylene or a substituted or unsubstituted (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one heteroatom selected from nitrogen, oxygen and sulfur;

R_6 to R_8 and R_{11} to R_{17} each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted 5- to 7-membered heterocycloalkyl group, or a sub-

stituted or unsubstituted (C3-C30)cycloalkyl group; or are linked to an adjacent substituent(s) via a substituted or unsubstituted (C3-C30)alkylene or a substituted or unsubstituted (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one heteroatom selected from nitrogen, oxygen and sulfur;

a, b, c and e each independently represent an integer of 1 to 4; where a, b, c or e is an integer of 2 or more, each of R₁, each of R₂, each of R₃ or each of R₅ is the same or different;

d represents an integer of 1 to 3; where d is an integer of 2 or more, each of R₄ is the same or different; and

the heterocycloalkyl group and the heteroaryl(ene) group contain at least one heteroatom selected from B, N, O, S, P(=O), Si and P.

[Claim 2]

The organic electroluminescent compound according to claim 1, wherein substituents of the substituted alkyl(ene) group, the substituted alkenylene group, the substituted cycloalkyl(ene) group, the substituted heterocycloalkyl group, the substituted aryl(ene) group, the substituted heteroaryl(ene) group, the substituted arylalkyl group and the substituted aromatic ring in said L₁ and L₂, R₁ to R₈ and R₁₁ to R₁₇ groups each independently are at least one selected from the group consisting of deuterium, a halogen, a (C1-C30)alkyl group substituted or unsubstituted with a halogen, a (C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a 3- to 30-membered heteroaryl group substituted with a (C1-C30)alkyl, a 3- to 30-membered heteroaryl group substituted with a (C6-C30)aryl, a (C3-C30)cycloalkyl group, a 5- to 7-membered heterocycloalkyl group, a tri(C1-C30)alkylsilyl group, a tri(C6-C30)arylsilyl group, a di(C1-C30)alkyl(C6-C30)arylsilyl group, a (C1-C30)alkyldi(C6-C30)arylsilyl group, a (C2-C30)alkenyl group, a (C2-C30)alkynyl group, a cyano group, a N-carbazolyl group, a di(C1-C30)alkylamino group, a di(C6-C30)arylamino group, a (C1-C30)alkyl(C6-C30)arylamino group, a di(C6-C30)arylboronyl group, a di(C1-C30)alkylboronyl group, a (C1-C30)alkyl(C6-C30)arylboronyl group, a (C6-C30)aryl(C1-C30)alkyl group, a (C1-C30)alkyl(C6-C30)aryl group, a carboxyl group, a nitro group and a hydroxyl group.

[Claim 3]

The organic electroluminescent compound according to claim 1, wherein L₁ and L₂ each independently represent a single bond, a 3- to

30-membered heteroarylene group, a (C6-C30)arylene group, or a (C6-C30)cycloalkylene group;

X₁ is CH or N;

Y₁ and Y₂ each independently represent -O-, -S-, -CR₆R₇- or -NR₈-;

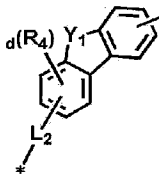
R₁ to R₅ each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group, a (C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a N-carbazolyl group, -NR₁₁R₁₂, or -SiR₁₃R₁₄R₁₅; or R₄ and R₅ each independently are linked to an adjacent substituent(s) via a (C3-C30)alkylene or a (C3-C30)alkenylene group to form a mono- or polycyclic, alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one heteroatom selected from nitrogen, oxygen and sulfur;

R₆ to R₈ each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group;

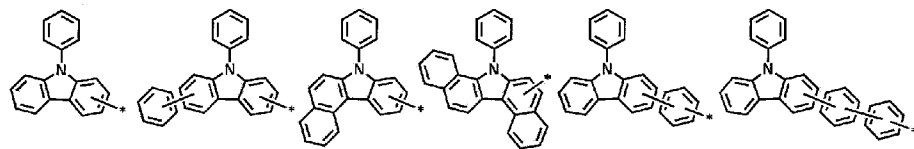
R₁₁ to R₁₅ each independently represent a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group; and the arylene group, the heteroarylene group and the cycloalkylene group in L₁ and L₂, and the alkyl group, the aryl group and the heteroaryl group in R₁ to R₈ and R₁₁ to R₁₅ can be substituted with at least one selected from the group consisting of deuterium, a halogen, a (C1-C30)alkyl group substituted or unsubstituted with a halogen, a (C6-C30)aryl group, a (C1-C30)alkyl(C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a 3- to 30-membered heteroaryl group substituted with a (C6-C30)aryl, a (C3-C30)cycloalkyl group and a (C6-C30)aryl(C1-C30)alkyl group.

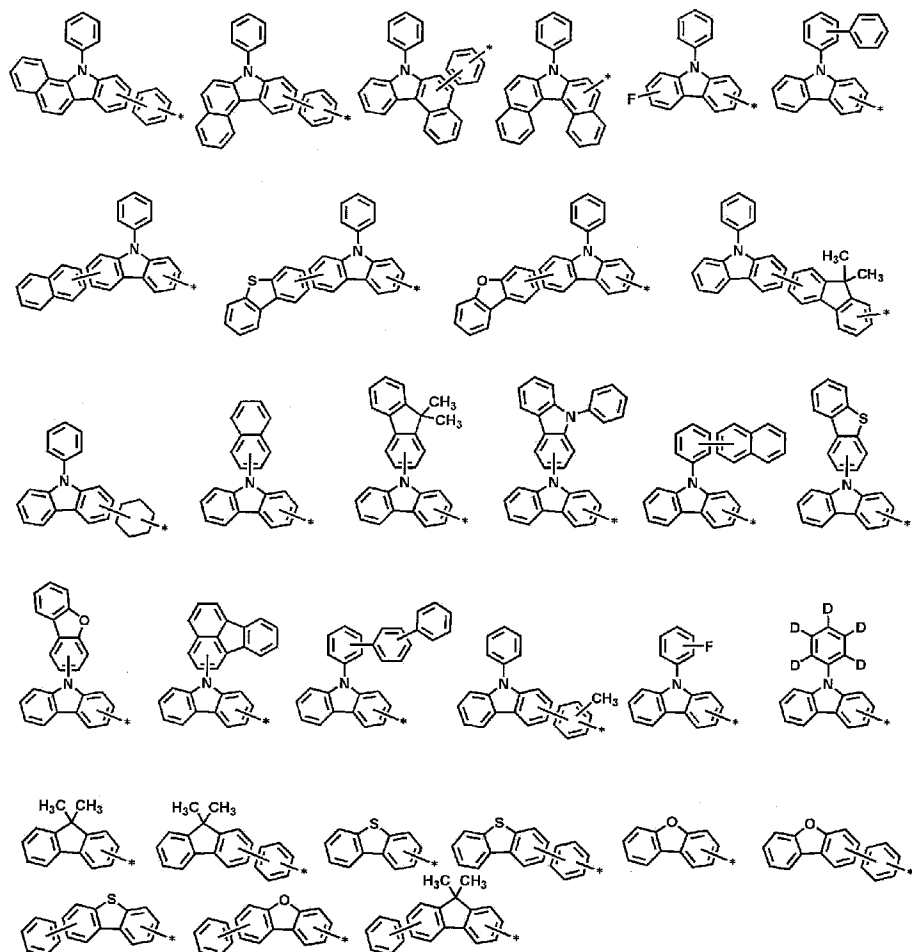
[Claim 4]

The organic electroluminescent compound according to claim 1, wherein

the moiety:  in formula 1 is selected from the

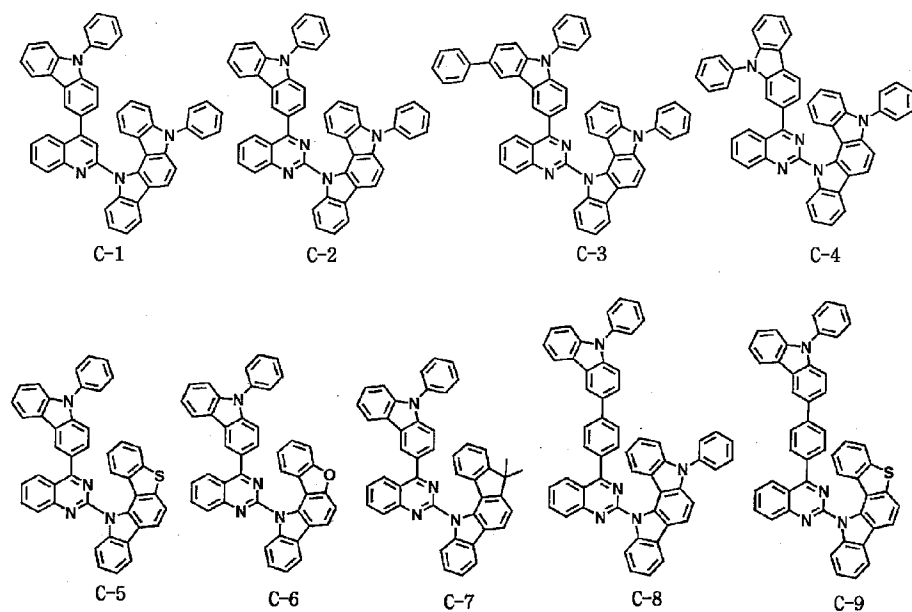
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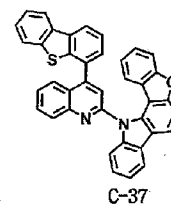
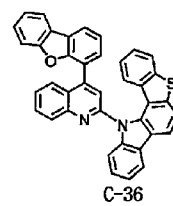
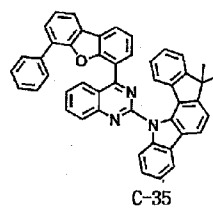
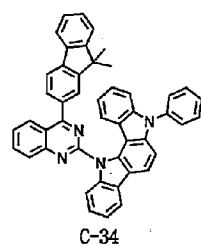
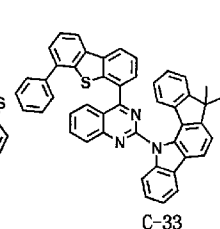
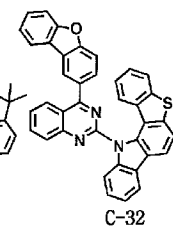
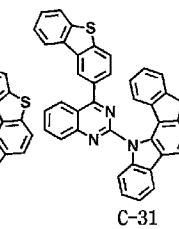
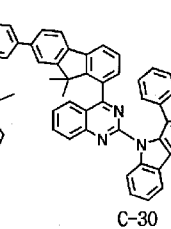
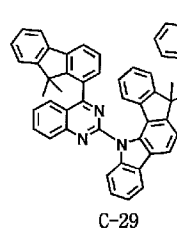
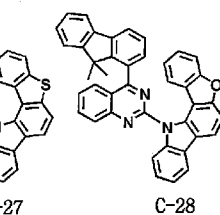
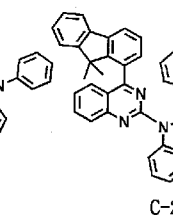
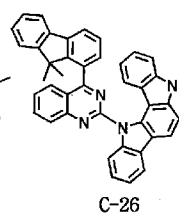
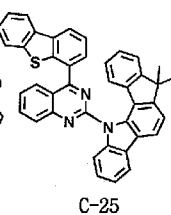
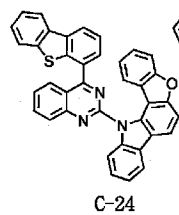
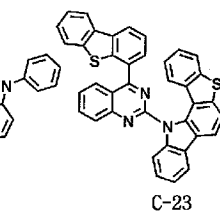
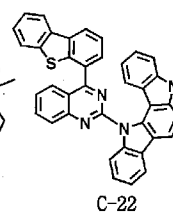
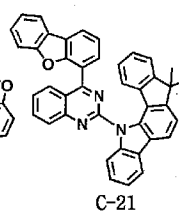
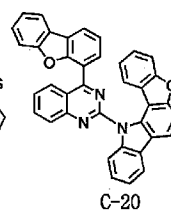
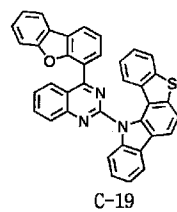
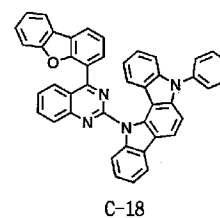
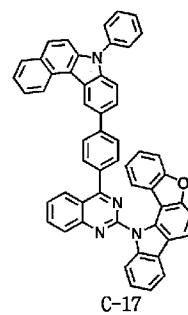
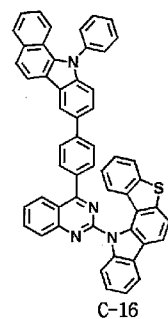
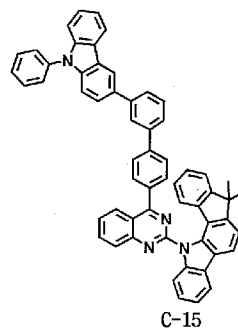
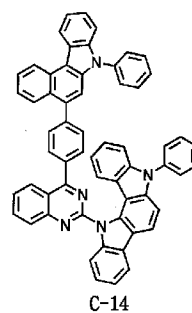
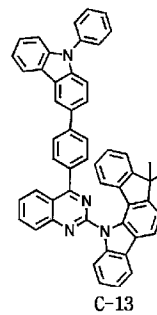
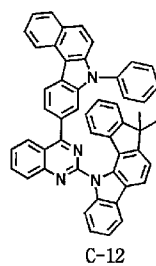
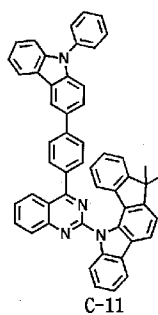
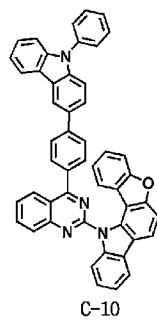


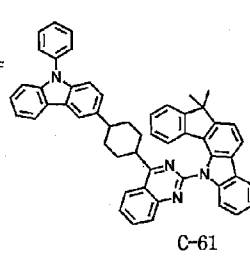
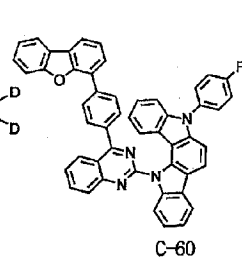
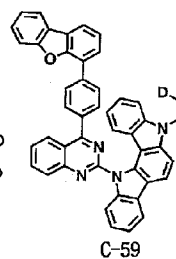
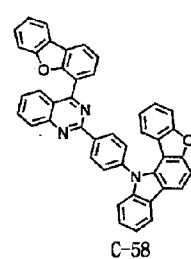
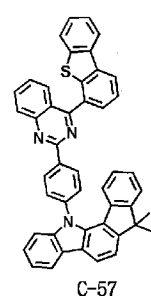
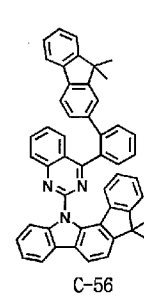
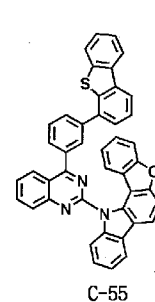
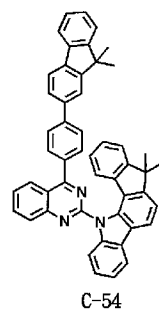
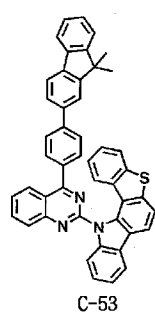
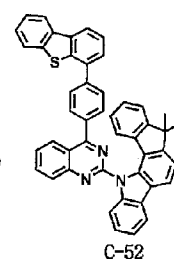
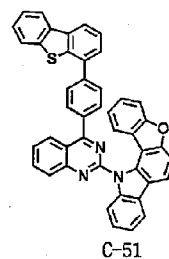
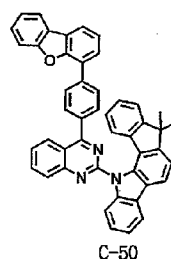
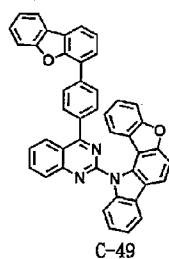
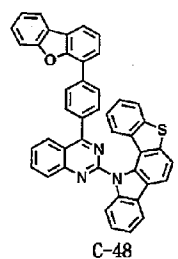
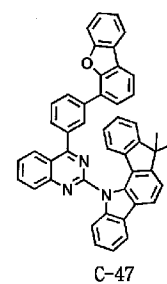
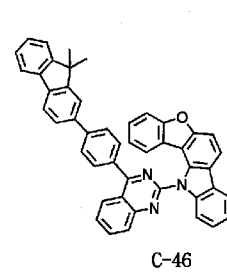
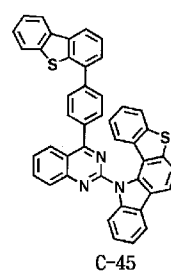
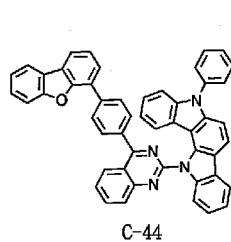
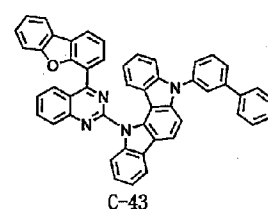
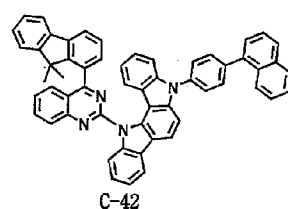
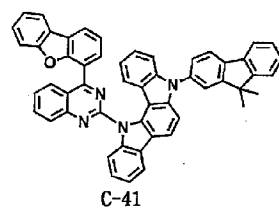
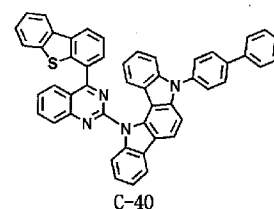
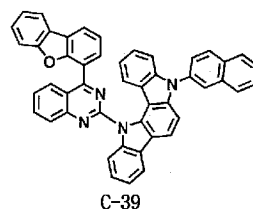
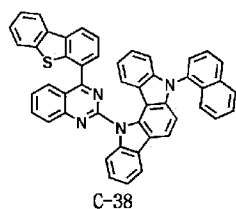


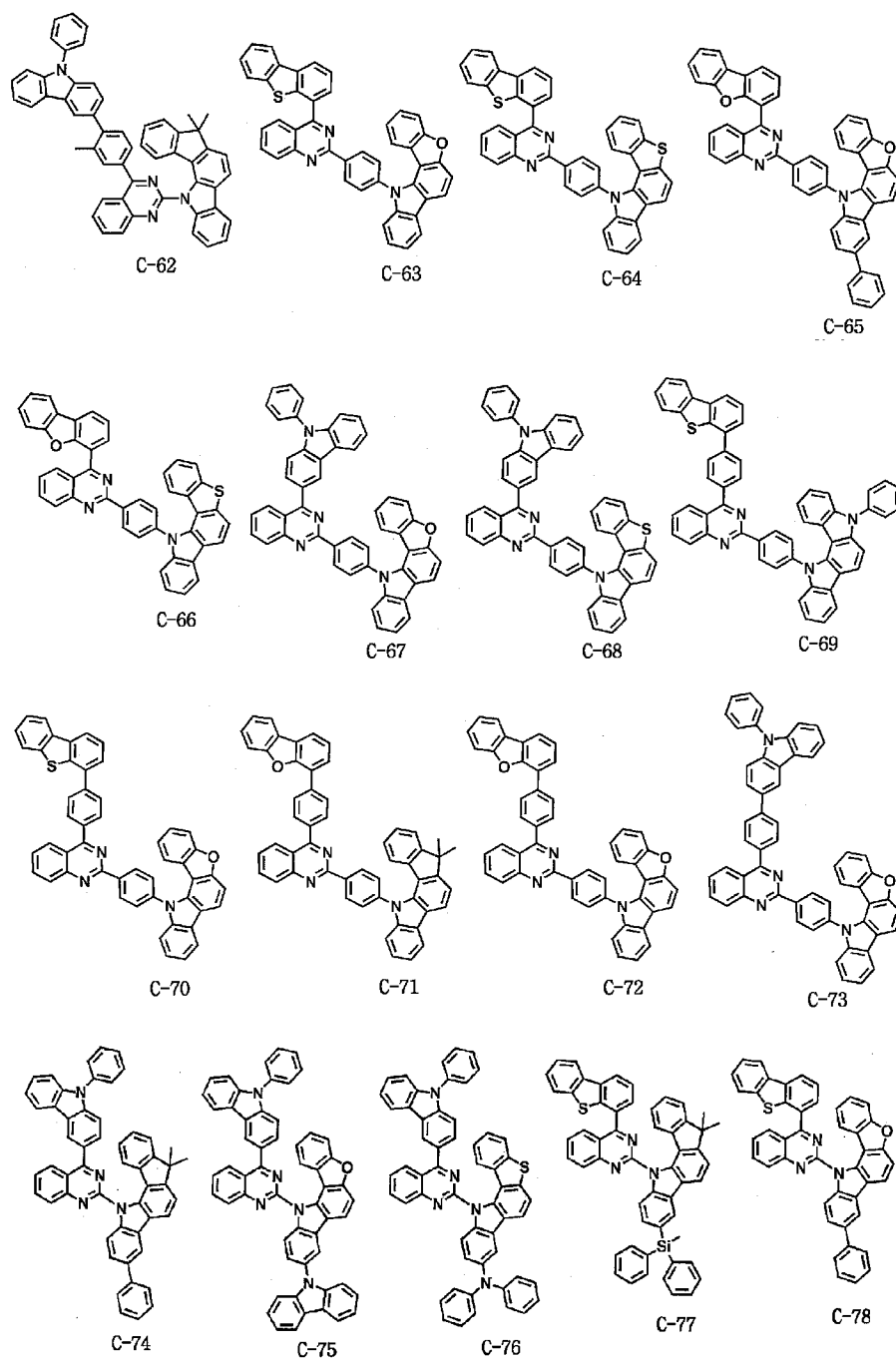
[Claim 5]

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









[Claim 6]

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

INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2012/007008

A. CLASSIFICATION OF SUBJECT MATTER

C07D 487/04 (2006.01) C07D 495/04 (2006.01) C07D 491/04 (2006.01) C07D 403/04 (2006.01) C07D 409/14 (2006.01)
 C07D 403/10 (2006.01) C07D 405/14 (2006.01) C09K 11/06 (2006.01) H01L 51/54 (2006.01) H01L 27/32 (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)

Registry, CAPLUS: substructure search based on claim 1.

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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

☒ Further documents are listed in the continuation of Box C☒ See patent family annex

* Special categories of cited documents:	
"A" document defining the general state of the art which is not considered to be of particular relevance	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle of theory underlying the invention
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INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2012/007008
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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P,X	WO 2012/050371 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 19 April 2012 See abstract; chemical formula 1; pp25-26.	1-6
P,X	WO 2012/050347 A1 (ROHM AND HAAS ELECTRONIC MATERIALS KOREA LTD.) 19 April 2012 See abstract; chemical formulae 3 & 4; pp20-21.	1-6

INTERNATIONAL SEARCH REPORT Information on patent family members		International application No. PCT/KR2012/007008	
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.			
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End of Annex			