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

(57) **Abstract:** The present invention relates to novel organic electroluminescence compounds and an organic electroluminescence device containing the same. The compounds according to the present invention have high luminous efficiency and long operation lifetime. Therefore, they can produce an organic electroluminescence device having enhanced power consumption efficiency.



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Description

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

Technical Field

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

Background Art

- [2] An electroluminescence (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].
- [3] 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, developing phosphorescent materials is one of the best methods to theoretically enhance the luminous efficiency by four (4) times. 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. Especially, many phosphorescent materials are being researched in Japan, Europe and U.S.A. recently.
- [4] Until now, 4,4'-N,N'-dicarbazol-biphenyl (CBP) is the most widely known host material for phosphorescent substances. Further, an organic EL device using bathocuproine (BCP) and aluminum(III)bis(2-methyl-8-quinolate)(4-phenylphenolate) (BALq) for a hole blocking layer is known, and Pioneer (Japan) et al. developed a high performance organic EL device employing a derivative of BALq as a host material.
- [5] Though these materials provide good light-emitting characteristics, they have the following disadvantages. Due to their low glass transition temperature and poor thermal stability, their degradation may occur during a high-temperature deposition process in a vacuum. The power efficiency of an organic EL device is given by $[(\pi/\text{voltage}) \times \text{current efficiency}]$, and the power efficiency is inversely proportional to

voltage, and thus in order to lower the power consumption, the power efficiency should be raised. Although an organic EL device comprising phosphorescent materials provides much higher current efficiency (cd/A) than one comprising fluorescent materials, an organic EL device using conventional phosphorescent materials such as BAq or CBP has a higher driving voltage than that using fluorescent materials. Thus, the EL device using the conventional phosphorescent materials has no advantage in terms of power efficiency (lm/W). Further, the operation lifetime of the organic EL device is short.

- [6] Korean Patent No. KR 0948700 discloses as compounds for an organic EL device an arylcarbazole compound substituted with a heteroaryl group comprising a nitrogen atom. However, it does not disclose a fused carbazole compound which is, at the nitrogen position, directly or indirectly linked to a heteroaryl group substituted with a biphenyl or terphenyl group.

Disclosure of Invention

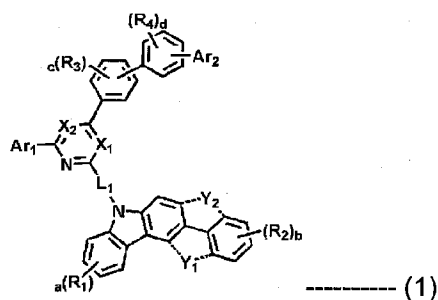
Technical Problem

- [7] The objective of the present invention is to provide an organic electroluminescence compound imparting high luminous efficiency and a long operation lifetime to a device, and having proper color coordination; and an organic electroluminescence device having high efficiency and a long lifetime, using said compound as a light-emitting material.

Solution to Problem

- [8] The present inventors found that the above objective can be achieved by a compound represented by the following formula 1:

[9]



- [10] wherein

- [11] L_1 represents 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;

- [12] X_1 and X_2 each independently represent CH or N;

- [13] Y_1 and Y_2 each independently represent -O-, -S-, -CR₅R₆- or -NR₇-, provided that Y_1 and Y_2 do not simultaneously exist;

- [14] Ar_1 and Ar_2 each independently represent hydrogen, deuterium, a halogen, a sub-

stituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30-membered heteroaryl group;

[15] R₁ to R₄ 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 are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

[16] R₅ to R₇ and R₁₁ to R₁₇ each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 3- to 30-membered heteroaryl group, a substituted or unsubstituted 5- to 7-membered heterocycloalkyl group, or a substituted or unsubstituted (C3-C30)cycloalkyl group; or are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

[17] a, b, c and d each independently represent an integer of 1 to 4; where a, b, c or d is an integer of 2 or more, each of R₁, each of R₂, each of R₃ or each of R₄ is the same or different;

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

Advantageous Effects of Invention

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

[20] In addition, since the 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 characteristic of the device. Therefore, they can produce an organic electroluminescence device having lowered driving voltages and enhanced power efficiency.

Mode for the Invention

[21] 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.

- [22] The present invention relates to an organic electroluminescence compound represented by the above formula 1 and an organic electroluminescence device comprising the compound.
- [23] Herein, “(C1-C30)alkyl(ene)” is meant to be a linear or branched alkyl(ene) having 1 to 30 carbon atoms, in which the number of carbon atoms is preferably 1 to 20, more preferably 1 to 10, and includes methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, etc.; “(C2-C30) alkenyl” is meant to be a linear or branched alkenyl having 2 to 30 carbon atoms, in which the number of carbon atoms is preferably 2 to 20, more preferably 2 to 10, and includes vinyl, 1-propenyl, 2-propenyl, 1-butenyl, 2-butenyl, 3-butenyl, 2-methylbut-2-enyl, etc.; “(C2-C30)alkynyl” is a linear or branched alkynyl having 2 to 30 carbon atoms, in which the number of carbon atoms is preferably 2 to 20, more preferably 2 to 10, and includes ethynyl, 1-propynyl, 2-propynyl, 1-butylnyl, 2-butylnyl, 3-butylnyl, 1-methylpent-2-ynyl, etc.; “(C3-C30)cycloalkyl” is a mono- or polycyclic hydrocarbon having 3 to 30 carbon atoms, in which the number of carbon atoms is preferably 3 to 20, more preferably 3 to 7, and includes cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, etc.; “3- to 7-membered heterocycloalkyl” is a cycloalkyl having at least one heteroatom selected from B, N, O, S, P(=O), Si and P, preferably O, S and N, and 3 to 7 ring backbone atoms, and includes tetrahydrofurane, pyrrolidine, thiolan, tetrahydropyran, etc.; “(C6-C30)aryl(ene)” is a monocyclic or fused ring derived from an aromatic hydrocarbon having 6 to 30 carbon atoms, in which the number of carbon atoms is preferably 6 to 20, more preferably 6 to 15, and includes phenyl, biphenyl, terphenyl, naphthyl, fluorenyl, phenanthrenyl, anthracenyl, indenyl, triphenylenyl, pyrenyl, tetracenyl, perylenyl, chrysenyl, naphthacenyl, fluoranthenyl, etc.; “3- to 30-membered heteroaryl(ene)” is an aryl group having at least one, preferably 1 to 4 heteroatom selected from the group consisting of B, N, O, S, P(=O), Si and P, and 3 to 30 ring backbone atoms; is a monocyclic ring, or a fused ring condensed with at least one benzene ring; has preferably 5 to 20, more preferably 5 to 15 ring backbone atoms; may be partially saturated; may be one formed by linking at least one heteroaryl or aryl group to a heteroaryl group via a single bond(s); and includes a monocyclic ring-type heteroaryl including furyl, thiophenyl, pyrrolyl, imidazolyl, pyrazolyl, thiazolyl, thiadiazolyl, isothiazolyl, isoxazolyl, oxazolyl, oxadiazolyl, triazinyl, tetrazinyl, triazolyl, tetrazolyl, furazanyl, pyridyl, pyrazinyl, pyrimidinyl, pyridazinyl, etc., and a fused ring-type heteroaryl including benzofuranyl, benzothiophenyl, isobenzofuranyl, dibenzofuranyl, dibenzothiophenyl, benzoimidazolyl, benzothiazolyl, benzoisothiazolyl, benzoisoxazolyl, benzoxazolyl, isoindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolinyl, quinazolinyl,

quinoxaliny, carbazolyl, phenoxaziny, phenanthridiny, benzodioxoly, etc. Further, "Halogen" includes F, Cl, Br and I.

- [24] Herein, "substituted" in the expression "substituted or unsubstituted" means that a hydrogen atom in a certain functional group is replaced with another atom or group, i.e., a substituent.
- [25] Substituents of the substituted alkyl group, the substituted aryl(ene) group, the substituted heteroaryl(ene) group, the substituted cycloalkyl(ene) group, the substituted heterocycloalkyl group and the substituted aralkyl group in L_1 , Ar_1 , Ar_2 , R_1 to R_7 and R_{11} to R_{17} groups of formula 1, 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 substituted or unsubstituted with a (C1-C30)alkyl or 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; an 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, preferably are at least one selected from the group consisting of deuterium, a halogen, a (C1-C6)alkyl group, a (C6-C20)aryl group and a di(C6-C20)arylamino group, more preferably are at least one selected from the group consisting of deuterium, fluorine, methyl, phenyl and diphenylamino.
- [26] In the above formula 1, L_1 represents a single bond, a 3- to 30-membered heteroarylene group or a (C6-C30)arylene group; X_1 and X_2 each independently represent CH or N; Y_1 and Y_2 each independently represent -O-, -S-, -CR₅R₆- or -NR₇-, provided that Y_1 and Y_2 do not simultaneously exist; Ar_1 and Ar_2 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_1 to R_4 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 carbazolyl group, or -SiR₁₃R₁₄R₁₅; or R_4 is linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur; R_5 to R_7 each independently represent a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group; or R_5 and R_6 are linked to each other to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from

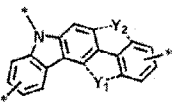
nitrogen, oxygen and sulfur; R_{13} to R_{15} each independently represent a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group; and the arylene and heteroarylene groups in L_1 , the alkyl, aryl, heteroaryl and carbazolyl groups in Ar_1 and Ar_2 , and the alkyl, aryl and heteroaryl groups in R_1 to R_7 and R_{13} 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 3- to 30-membered heteroaryl group substituted or unsubstituted with a (C1-C30)alkyl or a (C6-C30)aryl; a (C3-C30)cycloalkyl group; a 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 carbazolyl group; a di(C1-C30)alkylamino group; a di(C6-C30)arylamino group; a (C1-C30)alkyl(C6-C30)arylamino group; a (C6-C30)aryl(C1-C30)alkyl group; and a (C1-C30)alkyl(C6-C30)aryl group.

- [27] In the above formula 1, L_1 is preferably a single bond or a (C6-C20)arylene group substituted or unsubstituted with a (C1-C6)alkyl, more preferably a single bond, phenylene, or phenylene substituted with methyl.
- [28] Y_1 and Y_2 each independently represent -O-, -S-, -CR₅R₆- or -NR₇-, wherein R_5 to R_7 each independently are preferably an unsubstituted (C1-C6)alkyl group or an unsubstituted (C6-C20)aryl group, or are linked together to form a mono- or polycyclic, (C5-C20)alicyclic or aromatic ring, more preferably methyl, phenyl or are spiro-linked together.
- [29] Ar_1 and Ar_2 each independently are preferably hydrogen; a halogen; an unsubstituted (C1-C6)alkyl group; a (C6-C20)aryl group substituted or unsubstituted with deuterium, halogen or a (C1-C6)alkyl; or an unsubstituted 5- to 15-membered heteroaryl group, more preferably hydrogen; fluorine; methyl; phenyl; phenyl substituted with deuterium; phenyl substituted with fluorine; naphthyl; biphenyl; fluorenyl substituted with methyl; or carbazolyl.
- [30] R_1 to R_4 each independently are preferably hydrogen, an unsubstituted (C1-C6)alkyl group, a (C6-C20)aryl group substituted or unsubstituted with a di(C6-C20)arylamino group, an unsubstituted 5- to 15-membered heteroaryl group, or -SiR₁₃R₁₄R₁₅; or are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring, more preferably hydrogen; methyl; phenyl; phenyl substituted with diphenylamino; carbazolyl; or triphenylsilyl; or phenyl substituted with fluorine; naphthyl; biphenyl; fluorenyl substituted with methyl; or carbazolyl; or are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring.
- [31] R_{11} to R_{17} each independently are preferably an unsubstituted (C6-C20)aryl group, more preferably phenyl.
- [32] In the above formula 1, preferably, L_1 is a single bond or a (C6-C20)arylene group

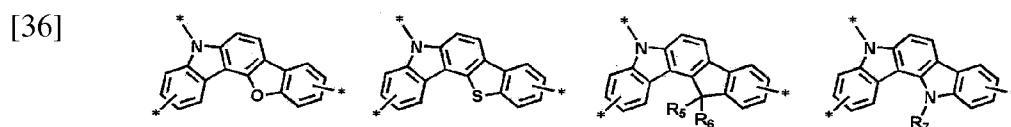
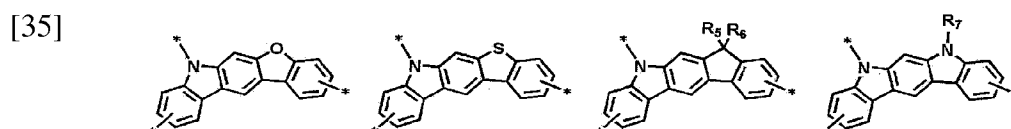
substituted or unsubstituted with a (C1-C6)alkyl; X_1 and X_2 each independently represent CH or N; Y_1 and Y_2 each independently represent -O-, -S-, $-CR_5R_6-$ or $-NR_7-$, wherein R_5 to R_7 each independently represent an unsubstituted (C1-C6)alkyl group or an unsubstituted (C6-C20)aryl group; Ar_1 and Ar_2 each independently represent hydrogen, a halogen, an unsubstituted (C1-C6)alkyl group, a (C6-C20)aryl group substituted or unsubstituted with deuterium, halogen or a (C1-C6)alkyl, or an unsubstituted 5- to 15-membered heteroaryl group; R_1 to R_4 each independently represent hydrogen, an unsubstituted (C1-C6)alkyl group, a (C6-C20)aryl group substituted or unsubstituted with a di(C6-C20)arylamino group, an unsubstituted 5- to 15-membered heteroaryl group, or $-SiR_{13}R_{14}R_{15}$, or are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring, wherein R_{13} to R_{15} each independently represent an unsubstituted (C6-C20)aryl group.

- [33] More specifically, L_1 represents a single bond, phenylene, biphenylene, terphenylene, indenylene, fluorenylene, triphenylenylene, pyrenylene, perylenylene, crysenylene, naphthacenylenylene, fluoranthenylenylene, thiophenylenylene, pyrrolylene, pyrazolylenylene, thiazolylenylene, oxazolylenylene, oxadiazolylenylene, triazinylene, tetrazinylenylene, triazolylenylene, furazanylenylene, pyridylene, benzofuranylenylene, benzothiophenylenylene, indolylenylene, benzoimidazolylenylene, benzothiazolylenylene, benzoisothiazolylenylene, benzoisoxazolylenylene, benzoxazolylenylene, benzothiadiazolylenylene, dibenzofuranylenylene or dibenzothiophenylenylene; Ar_1 and Ar_2 each independently represent hydrogen, methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl, n-pentyl, i-pentyl, n-hexyl, n-heptyl, n-octyl, 2-ethylhexyl, n-nonyl, decyl, dodecyl, hexadecyl, trifluoromethyl, perfluoroethyl, trifluoroethyl, perfluoropropyl, perfluorobutyl, phenyl, biphenyl, fluorenyl, fluoranthenyl, terphenyl, pyrenyl, crysenyl, naphthacenyl, perylenyl, pyridyl, pyrrolyl, furanyl, thiophenyl, imidazolyl, benzoimidazolyl, quinolyl, triazinyl, benzofuranyl, dibenzofuranyl, benzothiophenyl, dibenzothiophenyl, pyrazolyl, indolyl, carbazolyl, thiazolyl, oxazolyl, benzothiazolyl, benzoxazolyl, phenanthrolinyl, quinoxalinyl, or N-carbazolyl; R_1 to R_4 each independently represent hydrogen, deuterium, chloro, fluoro, methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl, n-pentyl, i-pentyl, n-hexyl, n-heptyl, n-octyl, 2-ethylhexyl, n-nonyl, decyl, dodecyl, hexadecyl, trifluoromethyl, perfluoroethyl, trifluoroethyl, perfluoropropyl, perfluorobutyl, phenyl, naphthyl, anthryl, biphenyl, fluorenyl, fluoranthenyl, triphenylenyl, pyrenyl, crysenyl, naphthacenyl, perylenyl, pyridyl, pyrrolyl, furanyl, thiophenyl, imidazolyl, benzoimidazolyl, indenyl, pyrazinyl, pyrimidinyl, pyridazinyl, quinolyl, triazinyl, benzofuranyl, dibenzofuranyl, benzothiophenyl, dibenzothiophenyl, pyrazolyl, indolyl, carbazolyl, thiazolyl, oxazolyl, benzothiazolyl, benzoxazolyl, phenanthrolinyl, trimethylsilyl, triethylsilyl, tripropylsilyl, tri(t-butyl)silyl, t-butyl dimethylsilyl, dimethylphenylsilyl, methyl diphenylsilyl or triphenylsilyl; the substituent in L_1 , Ar_1 , Ar_2 and R_1 to R_4 each independently can be sub-

stituted with at least one selected from the group consisting of deuterium, chloro, fluoro, methyl, ethyl, n-propyl, i-propyl, n-butyl, i-butyl, t-butyl, n-pentyl, i-pentyl, n-hexyl, n-heptyl, n-octyl, 2-ethylhexyl, n-nonyl, decyl, dodecyl, hexadecyl, trifluoromethyl, perfluoroethyl, trifluoroethyl, perfluoropropyl, perfluorobutyl, phenyl, naphthyl, biphenyl, 9,9-dimethylfluorenyl, 9,9-diphenylfluorenyl, fluoranthenyl, triphenylenyl, pyrenyl, crysensyl, naphthacensyl, perylenyl, dimethylamino, diethylamino, methylphenylamino, diphenylamino, trimethylsilyl, triethylsilyl, tripropylsilyl, tri(t-butyl)silyl, t-butyl dimethylsilyl, dimethylphenylsilyl, methyl diphenylsilyl, triphenylsilyl, N-carbazolyl and N-phenylcarbazolyl.

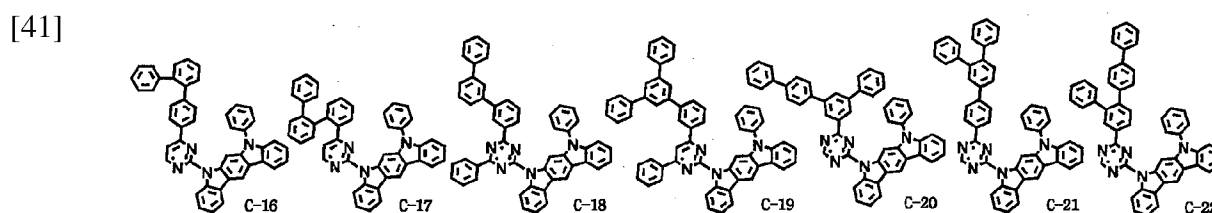
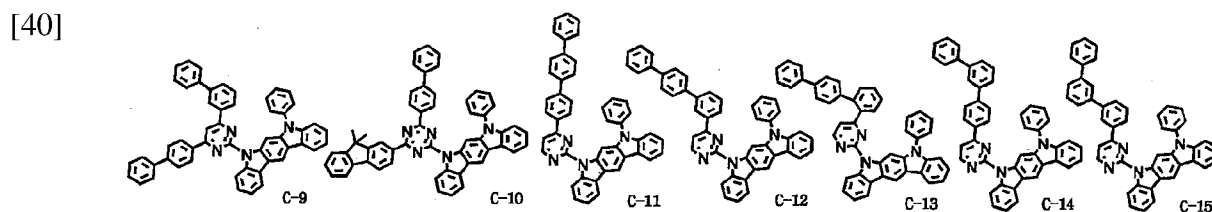
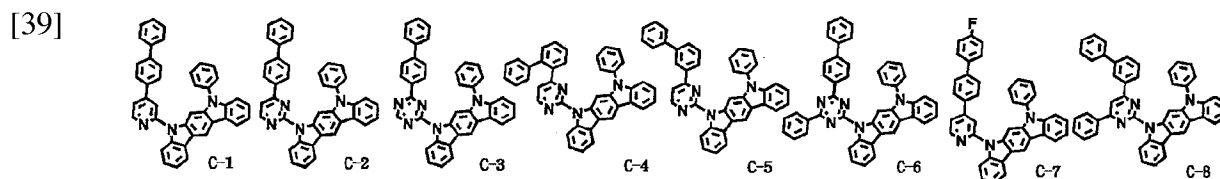
[34] In the above formula 1, the moiety,  is selected from the following

structures, but are not limited thereto:

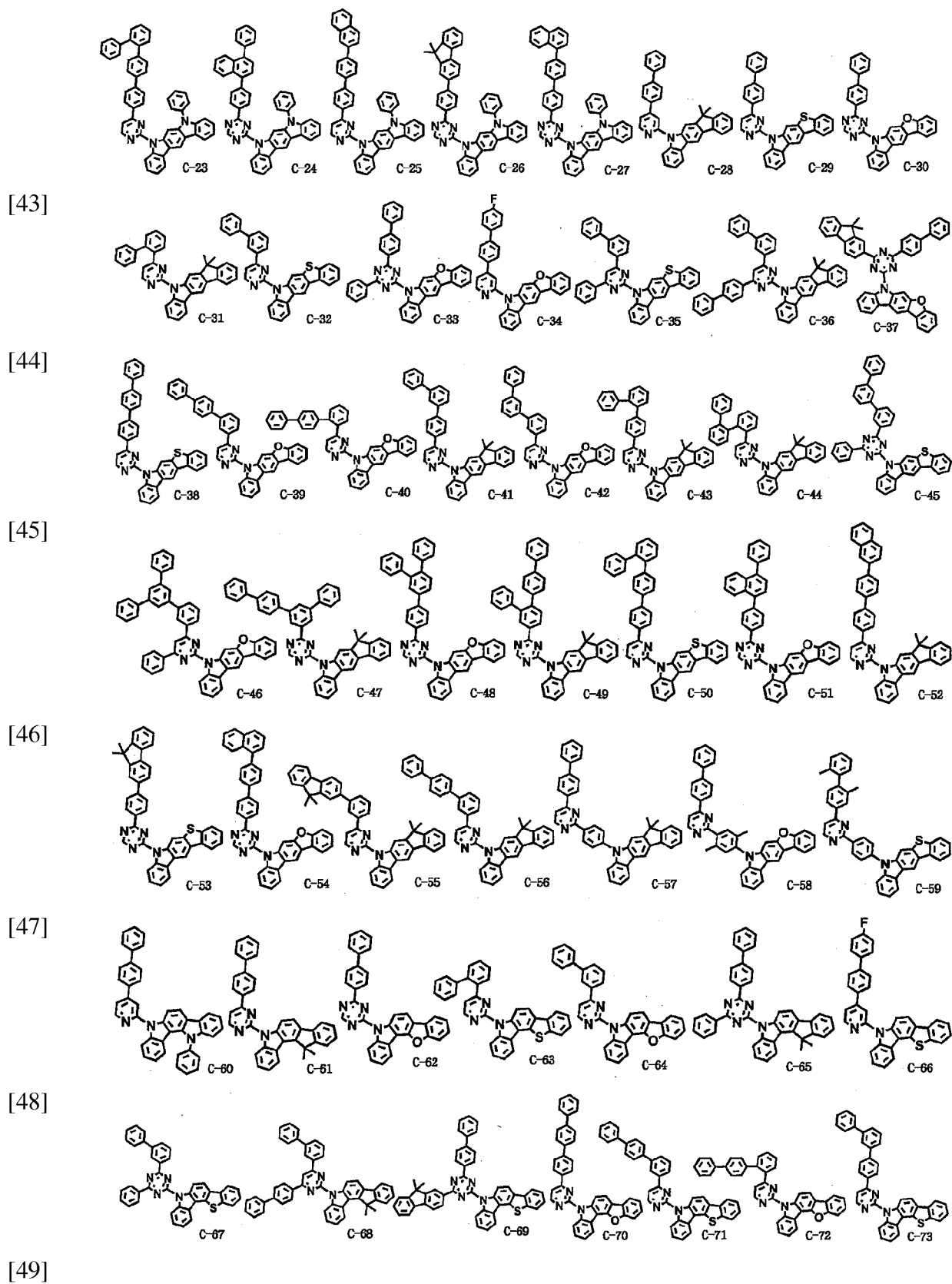


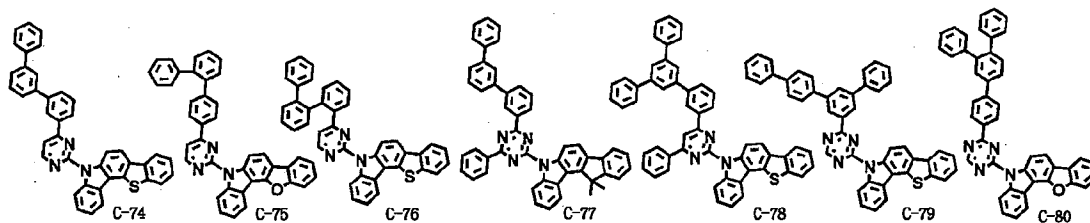
[37] wherein R₅, R₆ and R₇ are as defined in formula 1 above.

[38] The representative compounds of the present invention include the following compounds:

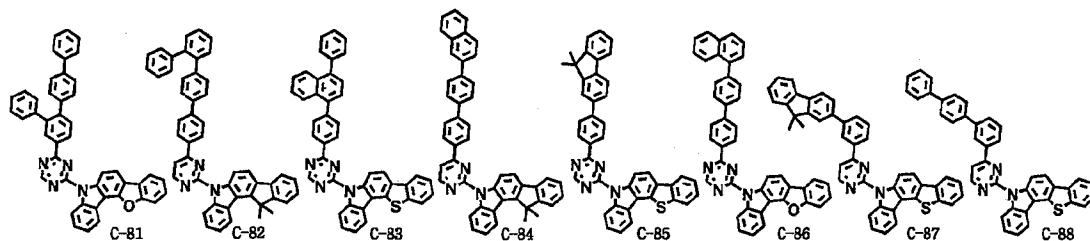


[42]

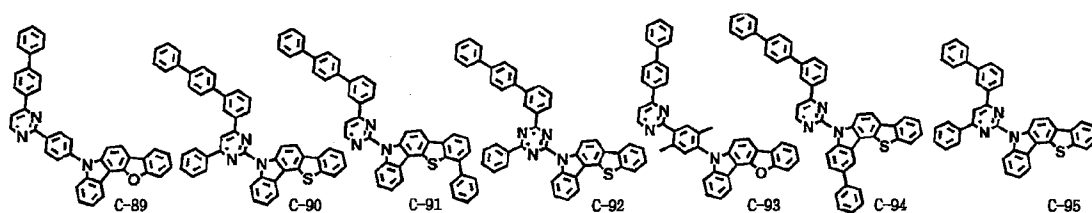




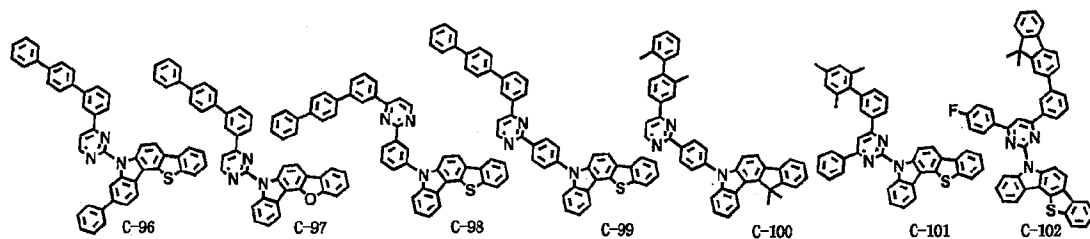
[50]



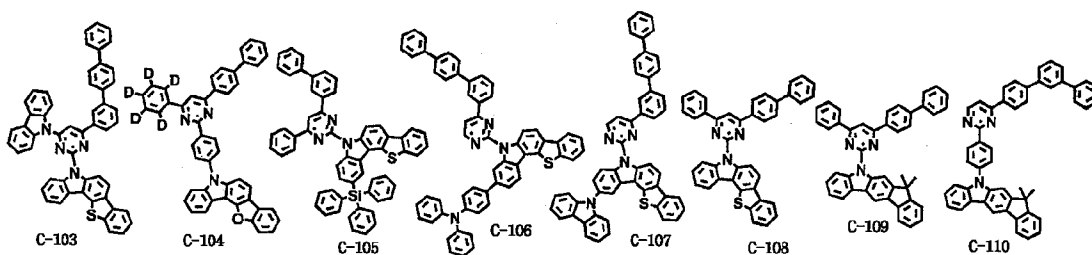
[51]



[52]



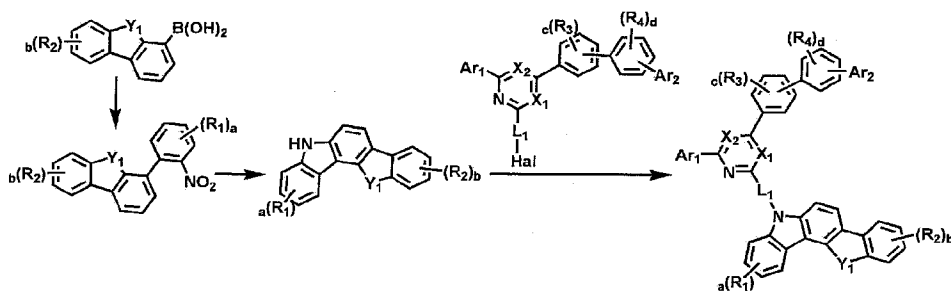
[53]



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

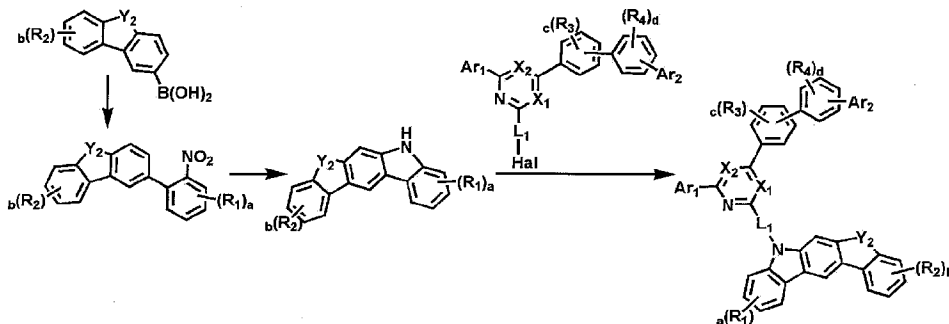
[55] [Reaction Scheme 1]

[56]

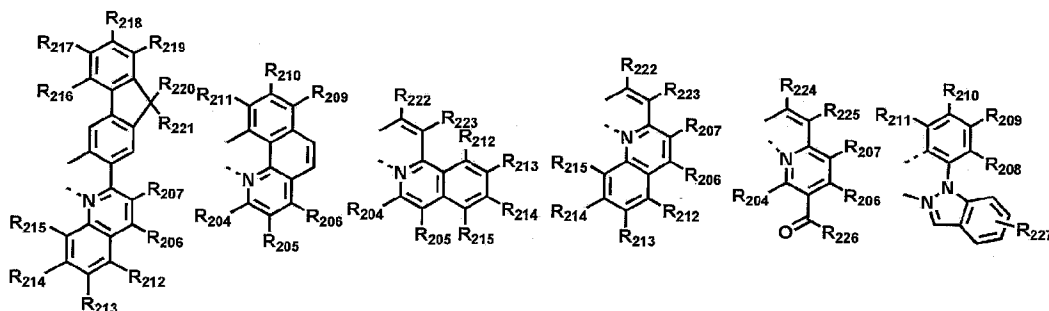


[57] [Reaction Scheme 2]

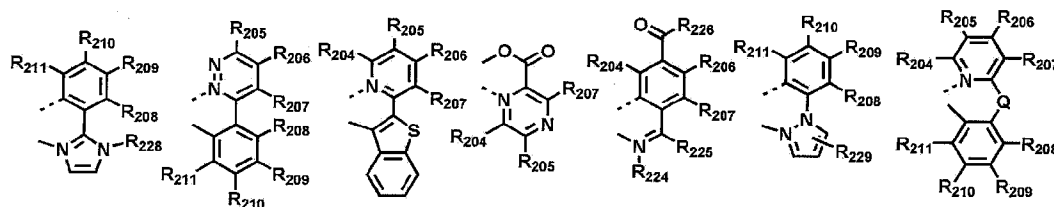
[58]



[66]



[67]



[68] R₂₀₁ to R₂₀₃ each independently represent hydrogen, deuterium, a (C1-C30)alkyl group unsubstituted or substituted with halogen(s), a (C6-C30)aryl group unsubstituted or substituted with (C1-C30)alkyl group(s), or a halogen;

[69] 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;

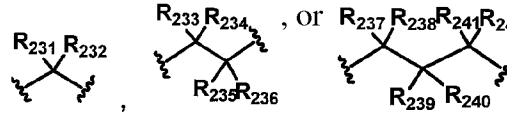
[70] R₂₂₀ to R₂₂₃ each independently represent hydrogen, deuterium, a (C1-C30)alkyl group unsubstituted or substituted with halogen(s), or a (C6-C30)aryl group unsubstituted or substituted with (C1-C30)alkyl group(s);

[71] R₂₂₄ and R₂₂₅ each independently represent hydrogen, deuterium, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a halogen, or R₂₂₄ and R₂₂₅ are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring;

[72] R₂₂₆ represents a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, a substituted or unsubstituted 5- or 30-membered heteroaryl group or a halogen;

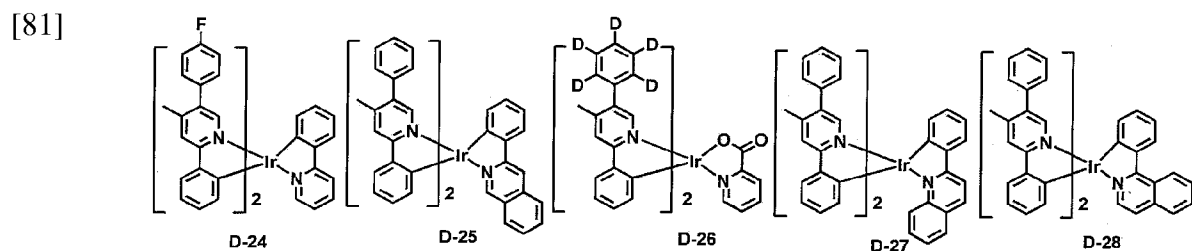
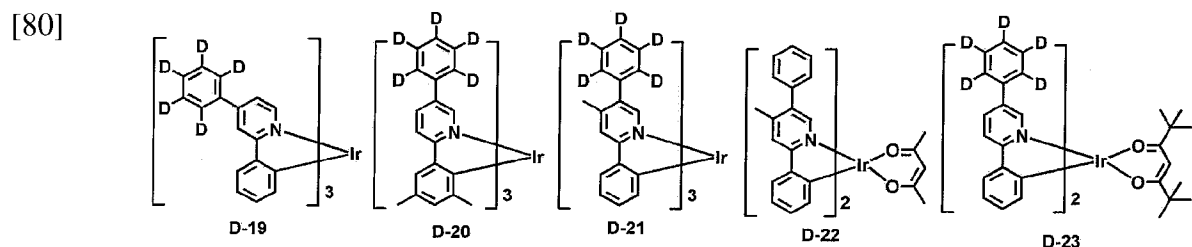
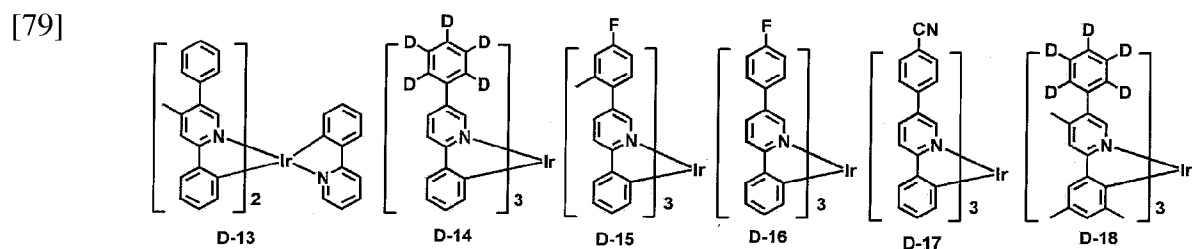
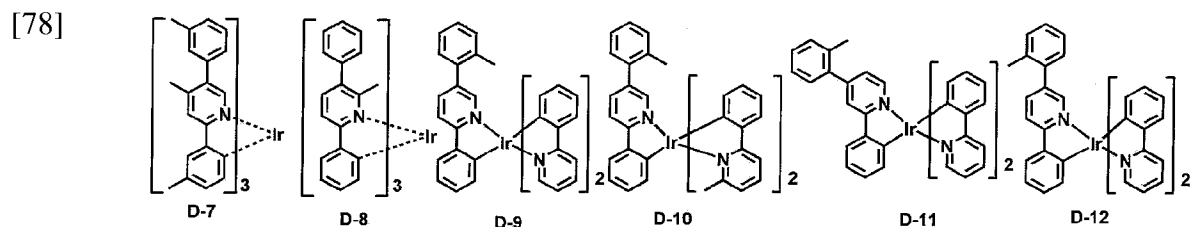
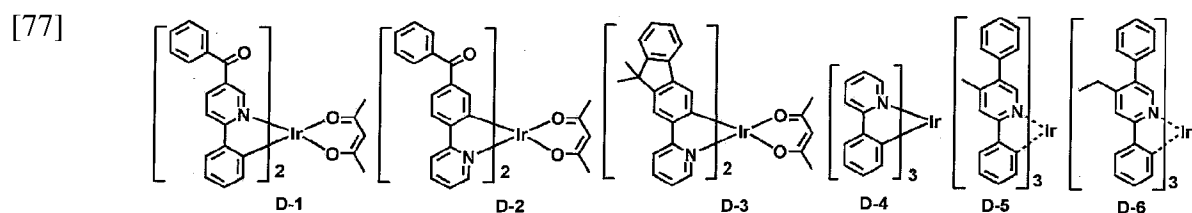
[73] R₂₂₇ to R₂₂₉ each independently represent hydrogen, deuterium, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group or a halogen;

[74] Q represents

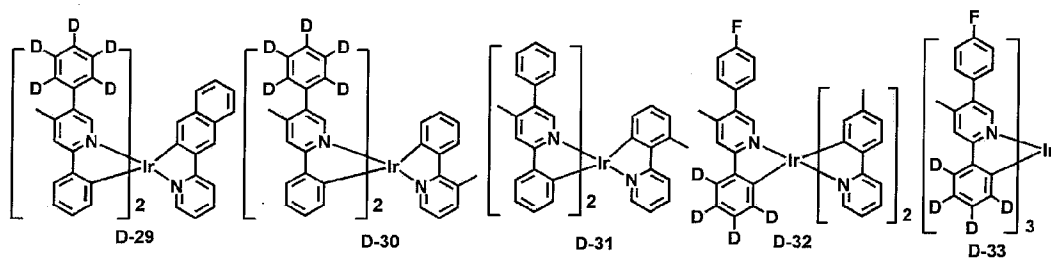
[75]  , or $R_{237}R_{238}R_{241}R_{242}$; R_{231} to R_{242} each independently represent

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 alkylene group or alkenylene group to form a spiro ring or a fused ring or may be linked to R_{207} or R_{208} via alkylene group or alkenylene group to form a saturated or unsaturated fused ring.

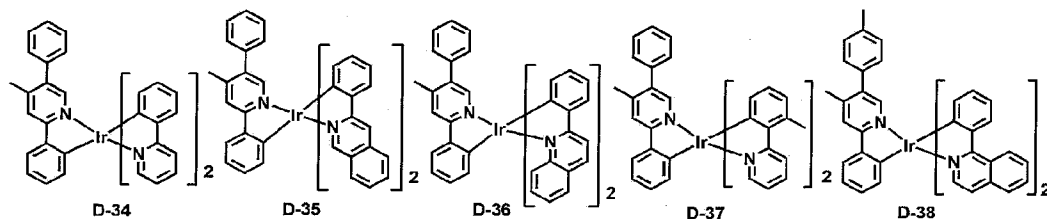
[76] The dopants of formula 2 include the following, but are not limited thereto:



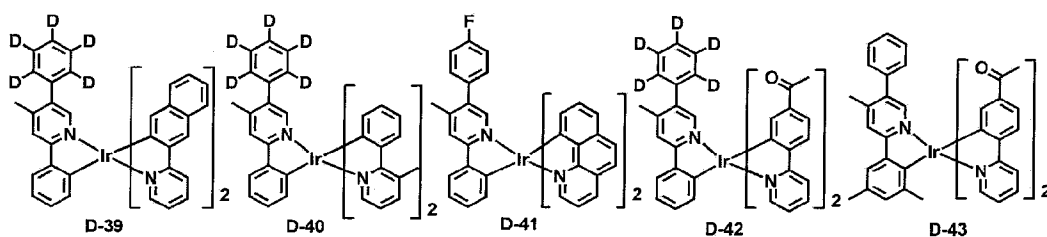
[82]



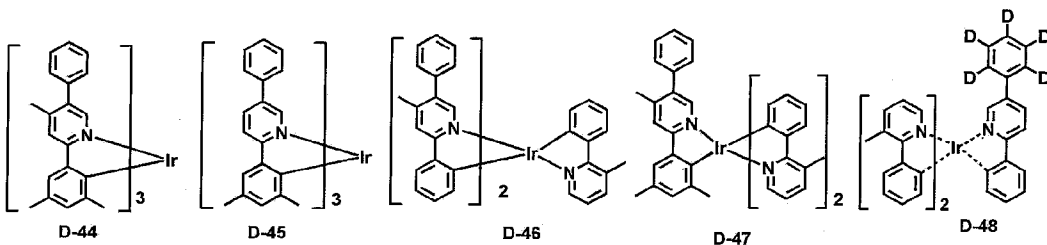
[83]



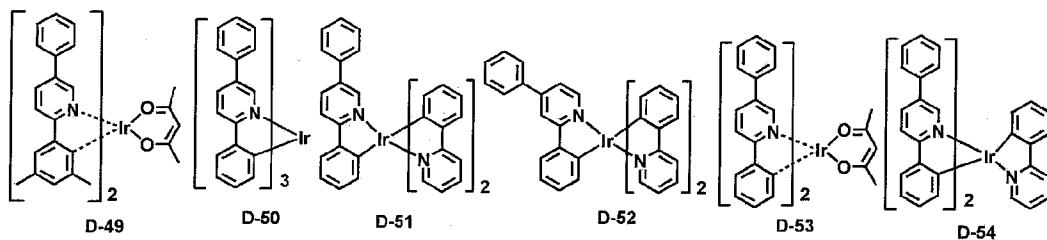
[84]



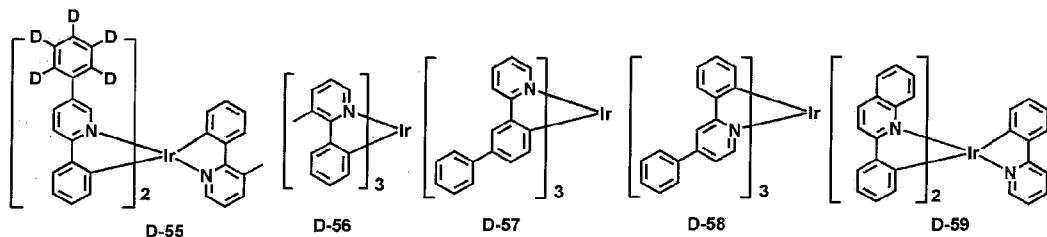
[85]



[86]



[87]



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

[89] In the organic electroluminescence device according to the present invention, the

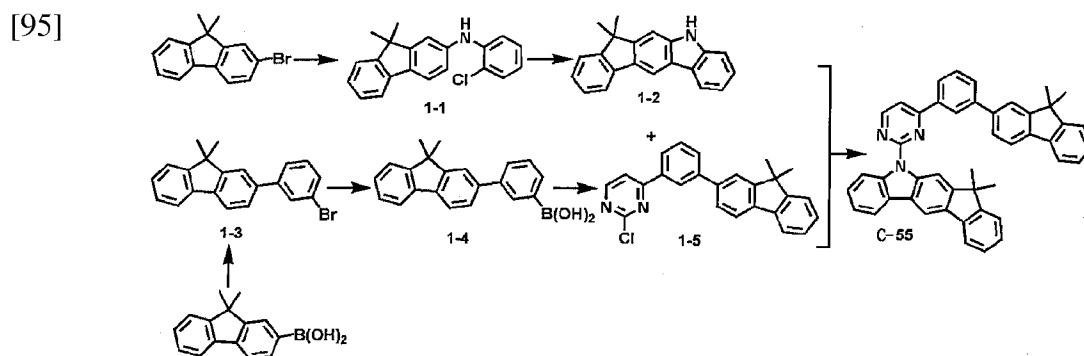
organic layer may further comprise at least one metal selected from the group consisting of metals of Group 1, metals of Group 2, transition metals of the 4th period, transition metals of the 5th period, lanthanides and organic metals of d-transition elements of the Periodic Table, or at least one complex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

[90] In addition, the organic electroluminescence 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 compound according to the present invention.

[91] Preferably, in the organic electroluminescence 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 electroluminescence 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.

[92] Preferably, in the organic electroluminescence 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 electroluminescence device having two or more electroluminescent layers and emitting white light.

[93] Hereinafter, the organic electroluminescence 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:

[94] Example 1: Preparation of compound C-55[96] Preparation of compound 1-1

[97] After mixing 2-bromo-9,9-dimethyl-9H-fluorene (50 g, 0.183 mol), 2-chloroaniline (57 mL, 0.549 mol), Pd(OAc)₂ (1.6 g, 0.007 mol), P(t-Bu)₃ (7.2 mL, 0.0146 mol), NaOt-Bu (44 g, 0.458 mol) and toluene (500 mL), the reaction mixture was stirred for 12 hours at 120°C. After terminating the reaction, the reaction mixture was extracted with ethyl acetate (EA). The obtained organic layer was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 1-1 (32 g, 55 %).

[98] Preparation of compound 1-2

[99] After mixing compound 1-1 (32 g, 0.1 mol), Pd(OAc)₂ (1.1 g, 0.005 mol), di-t-butyl(methyl)phosphoniumtetrafluoroborate (2.48 g, 0.01 mol), K₂CO₃ (42 g, 0.30 mol) and DMA (550 mL), the reaction mixture was stirred for 12 hours at 200°C. After terminating the reaction, the reaction mixture was extracted with EA. The obtained organic layer was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 1-2 (14 g, 47 %).

[100] Preparation of compound 1-3

[101] After mixing 9,9-dimethylfluorene-2-boronic acid (30g, 126.0 mmol), 1,3-dibromobenzene (30.45 mL, 252.00 mmol), PdCl₂(PPh₃)₂ (2.6 g, 3.78 mmol), 2M Na₂CO₃ (160 mL) and toluene (500 mL), the reaction mixture was heated at 100°C. After 5 hours, the reaction mixture was cooled to room temperature, was extracted with EA, was washed with distilled water, was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 1-3 (80 g, 67.46 %).

[102] Preparation of compound 1-4

[103] After dissolving compound 1-3 (30 g, 85.89 mmol) in tetrahydrofuran (THF) (500 mL), and adding 2.5 M n-BuLi in hexane (41.23 mL, 103.07 mmol) to the reaction mixture in -78°C, the reaction mixture was stirred for 1 hour. The reaction mixture was stirred for 12 hours at room temperature with adding B(OMe)₃ (14.36 mL, 128.84

mmol) slowly to the reaction mixture, and with slowly increasing the temperature. After extracting with EA, the obtained organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was recrystallized with EA and hexane to obtain compound 1-4 (24 g, 68.93 %).

[104] Preparation of compound 1-5

[105] After mixing 2,4-dichloropyrimidine (7 g, 46.98 mmol), compound 1-4 (16.2 g, 51.68 mmol), $\text{Pd}(\text{PPh}_3)_4$ (1.62 g, 1.40 mmol), 2M Na_2CO_3 (60 mL) and dimethylether (DME) (400 mL), the reaction mixture was stirred at 90°C. After 4 hours, the reaction mixture was cooled to room temperature, distilled water was added, and the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 1-5 (14 g, 77.83 %).

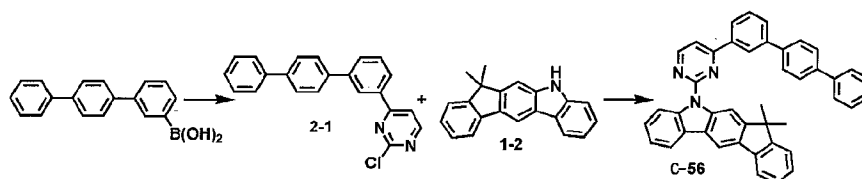
[106] Preparation of compound C-55

[107] After dissolving compound 1-2 (4.5 g, 15.89 mmol) and compound 1-5 (7.3 g, 19.07 mmol) in dimethylformamide (DMF) (130 mL), 60% NaH in mineral oil (0.76 g, 19.07 mmol) was slowly added to the mixture, and was stirred for 12 hours at room temperature. After terminating the reaction, MeOH was added to the reaction mixture. The obtained solid was filtered through silica gel, was recrystallized with EA and DMF to obtain compound C-55 (1.8 g, 18 %).

[108] MS/FAB found 630; calculated 629.79

[109] Example 2: Preparation of compound C-56

[110]



[111] Preparation of compound 2-1

[112] After mixing 3-terphenylboronic acid (8.2 g, 29.9 mmol), 2,4-dichloropyrimidine (6.6 g, 44.9 mmol), $\text{Pd}(\text{PPh}_3)_4$ (1.7 g, 1.5 mmol), Na_2CO_3 (7.9 g, 74.8 mmol), toluene (100 mL), EtOH (25 mL) and purified water (25 mL), the reaction mixture was stirred for 5 hours under reflux. After terminating the reaction, the reaction mixture was cooled to room temperature, and was filtered. The obtained organic layer was dried with MgSO_4 , was concentrated under reduced pressure, was filtered through silica gel. The remaining solution was concentrated under reduced pressure to obtain compound 2-1 (7 g, 70 %).

[113] Preparation of compound C-56

[114] After dissolving compound 1-2 (3.27 g, 11.5 mmol) in DMF (80 mL), NaH (507 mg,

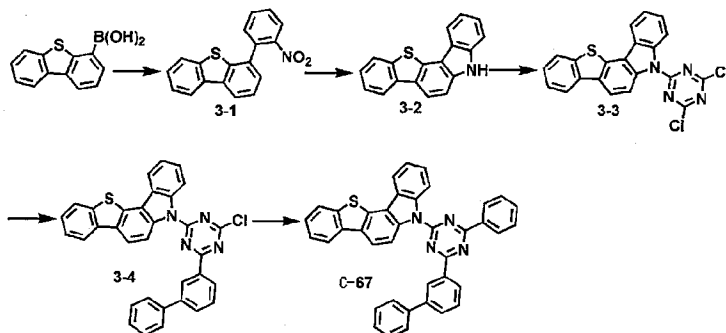
12.7 mmol) was added at 0°C, and was stirred for 10 minutes. Compound 2-1 (4.2 g, 12.1 mmol) was added to the reaction mixture, and was stirred for 7 hours. After terminating the reaction, MeOH was added to the reaction mixture. The obtained solid was filtered through silica gel, was recrystallized with DMF to obtain compound **C-56** (3.4 g, 50 %).

[115] MS/FAB found 590; calculated 589.73

[116]

[117] Example 3: Preparation of compound **C-67**

[118]



[119] Preparation of compound 3-1

[120] After mixing 4-dibenzothiopheneboronic acid (10 g, 43.84 mmol), 2-bromonitrobenzene (10.6 g, 52.61 mmol), Pd(PPh₃)₄ (2.53 g, 2.19 mmol), 2M K₂CO₃ (50 mL), toluene (200 mL) and ethanol (50 mL), the reaction mixture was stirred under reflux. After 4 hours, the reaction mixture was cooled to room temperature and distilled water was added, and was extracted with EA. The obtained organic layer was dried with anhydrous MgSO₄ to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 3-1 (11 g, 82.17 %).

[121] Preparation of compound 3-2

[122] After mixing compound 3-1 (11 g, 36.02 mmol), P(OEt)₃ (100 mL) and 1,2-dichlorobenzene (100 mL), the reaction mixture was stirred at 150°C under reflux. After 5 hours, the reaction mixture was cooled to room temperature, was distilled under reduced pressure, and was filtered through column to obtain compound 3-2 (7 g, 25.60 mmol, 71.13 %).

[123] Preparation of compound 3-3

[124] After dissolving 2,4,6-trichloro-1,3,5-triazine (43.5 g, 236 mmol) in THF (230 mL), the reaction mixture was stirred at -78°C (reaction mixture A). After dissolving compound 3-2 (22 g, 79 mmol) in THF (150 mL), the reaction mixture was putted into NaH (4.7 g, 118 mmol) (reaction mixture B). After putting reaction mixture B into reaction mixture A slowly, the reaction mixture was stirred for 4 hours. After stirring, distilled water (500 mL) was added slowly, and was stirred for 30 minutes. The

obtained solid was filtered through column chromatography and recrystallized to obtain compound 3-3 (31 g, 92 %).

[125] Preparation of compound 3-4

[126] After mixing compound 3-3 (12 g, 28 mmol), biphenyl-3-ylboronic acid (6 g, 31 mmol), Na_2CO_3 (8 g, 71 mmol), toluene (370 mL), distilled water (36 mL) and $\text{Pd}(\text{PPh}_3)_4$ (1 g, 0.9 mmol), the reaction mixture was stirred at 120°C . After 5 hours, the reaction mixture was cooled to room temperature, distilled water was added, and the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, was distilled under reduced pressure to remove the solvent. The obtained solid was washed with methanol, was filtered, was dried, and was filtered through column chromatography and recrystallized to obtain compound 3-4 (15 g, 98 %).

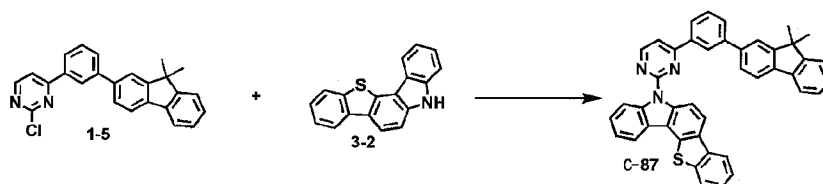
[127] Preparation of compound C-67

[128] After mixing compound 3-4 (8.5 g, 16 mmol), phenylboronic acid (3 g, 23 mmol), Na_2CO_3 (5 g, 47 mmol), toluene (168 mL), distilled water (24 mL), ethanol (24 mL) and $\text{Pd}(\text{PPh}_3)_4$ (1 g, 0.9 mmol), the reaction mixture was stirred at 120°C . After 5 hours, the reaction mixture was cooled to room temperature, distilled water was added, and the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous MgSO_4 to remove the remaining moisture, was distilled under reduced pressure to remove the solvent. The obtained solid was washed with methanol, was filtered, was dried, and was filtered through column chromatography and recrystallized to obtain compound C-67 (6.1 g, 66 %).

[129] MS/FAB found 581; calculated 580.70

[130] Example 4: Preparation of compound C-87

[131]



[132] Preparation of compound C-87

[133] After dissolving compound 1-5 (10.5 g, 27.44 mmol) and compound 3-2 (5 g, 18.29 mmol) in DMF (100 mL), 60% NaH in mineral oil (1.08 g, 27.44 mmol) was slowly added to the mixture, and was stirred for 12 hours at room temperature. After terminating the reaction, distilled water was added to the reaction mixture. The obtained solid was filtered under reduced pressure, and the obtained solid was dissolved in CHCl_3 , and was filtered through column to obtain compound C-87 (7.5 g, 67.25 %).

[134] MS/FAB found 620; calculated 619.78

[135] Example 5: Preparation of compound C-88

Chemical reaction scheme showing the synthesis of compound C-8B. Reactant 2-1 (4-(4-phenylphenyl)-2-chloropyrimidin-5-yl) reacts with reactant 3-2 (indole-3-carboxamide derivative) to form product C-8B (a complex fused heterocycle system).

[138] After dissolving compound 3-2 (3.5 g, 12.83 mmol) and compound 2-1 (4 g, 11.66 mmol) in DMF (150 mL), 60% NaH in mineral oil (0.7 g, 17.50 mmol) was slowly added to the mixture, and was stirred for 12 hours at room temperature. After terminating the reaction, distilled water was added to the reaction mixture. The obtained solid was filtered under reduced pressure, and the obtained solid was washed with EA, DMF and THF sequentially, to obtain compound **C-88** (3.7 g, 49.74 %).

[140] Example 6: Preparation of compound C-91

Chemical reaction scheme for the synthesis of compound C-91:

Fluorene reacts with $B(OH)_3$ to form intermediate 6-1.

Intermediate 6-1 reacts with $B(OH)_3$ to form intermediate 6-2.

Intermediate 6-2 reacts with 4-nitrophenylboronic acid to form intermediate 6-3.

Intermediate 6-3 reacts with 2-phenyl-1H-indole to form intermediate 6-4.

Intermediate 6-4 reacts with 2-chloro-6-(4-phenylphenyl)pyridine (2-1) to form the final product C-91.

[143] After mixing 4-dibenzothiopheneboronic acid (40 g, 175.36 mmol), bromobenzene (36.8 mL, 350.73 mmol), Pd(PPh₃)₄ (4.05 g, 3.50 mmol), 2M Na₂CO₃ (220 mL), toluene (600 mL) and ethanol (60 mL), the reaction mixture was stirred at 100°C. After 3 hours, the reaction mixture was cooled to room temperature and distilled water was added, and was extracted with EA. The obtained organic layer was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 6-1 (35 g, 76.57 %).

[145] After dissolving compound 6-1 (35 g, 134.43 mmol) in THF (500 mL), and adding 2.5 M n-BuLi in hexane (107.5 mL, 268.8 mmol) to the reaction mixture in -78°C, the reaction mixture was stirred for 1 hour at -78°C, was heated to room temperature, and was stirred for 2 hours. B(OMe)₃ (41.9 mL, 403.30 mmol) was added slowly in -78°C, the reaction mixture was stirred for 12 hours while slowly heated to room temperature. After extracting with EA, the obtained organic layer was dried with anhydrous MgSO₄ to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was recrystallized with methylchloride (MC) and hexane to obtain

compound 6-2 (26 g, 63.78 %).

[146] Preparation of compound 6-3

[147] After mixing compound 6-2 (26 g, 85.47 mmol), 2-bromonitrobenzene (18.9 g, 52.61 mmol), Pd(PPh₃)₄ (1.9 g, 1.70 mmol), 2M Na₂CO₃ (100 mL), toluene (250 mL) and ethanol (50 mL), the reaction mixture was stirred under reflux. After 4 hours, the reaction mixture was cooled to room temperature and distilled water was added, and was extracted with EA. The obtained organic layer was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 6-3 (32 g, 98.15 %).

[148] Preparation of compound 6-4

[149] After mixing compound 6-3 (32 g, 83.87 mmol), P(OEt)₃ (200 mL) and 1,2-dichlorobenzene (100 mL), the reaction mixture was stirred at 150°C under reflux. After 12 hours, the reaction mixture was cooled to room temperature, was distilled under reduced pressure, and was filtered through column to obtain compound 6-4 (9 g, 30.70 %).

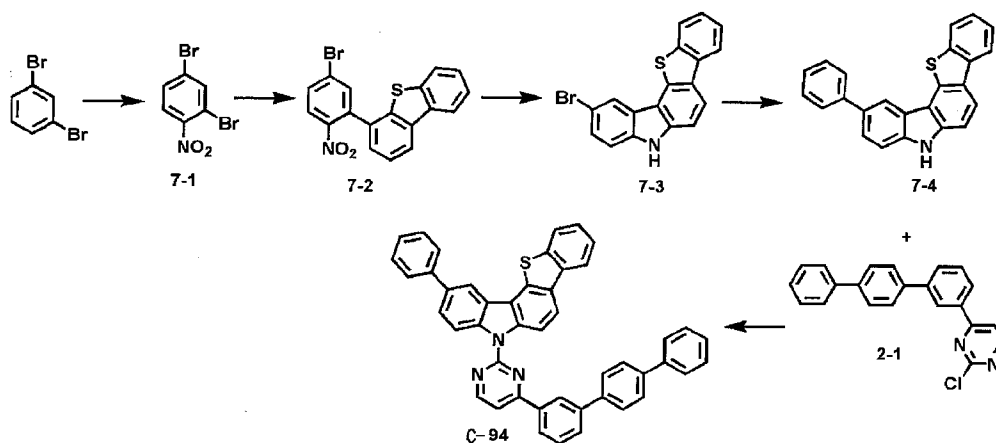
[150] Preparation of compound C-91

[151] After dissolving compound 6-4 (4 g, 11.44 mmol) and compound 2-1 (4.7 g, 13.73 mmol) in DMF (190 mL), 60% NaH in mineral oil (0.68 g, 17.16 mmol) was slowly added to the mixture, and was stirred for 12 hours at room temperature. After terminating the reaction, distilled water was added to the reaction mixture. The obtained solid was filtered under reduced pressure, and the obtained solid was washed with EA, DMF and THF sequentially, to obtain compound C-91 (2.0 g, 26.65 %).

[152] MS/FAB found 656; calculated 655.81

[153] Example 7: Preparation of compound C-94

[154]



[155] Preparation of compound 7-1

[156] After making a bath of -4°C putting ice in aqueous solution of saturated NaCl, nitric acid (1.5 equivalents) was putted into sulfuric acid solvent, and was waited until the temperature of the reaction solvent became 0°C. 1,3-dibromobenzene (10 g, 42.4

mmol) was putted into a dropping funnel, and was dropwised to not exceed 10°C. After 30 minutes, the reaction mixture was quenched with water. The obtained solid was filtered, was washed with purified water several times, and was filtered through column to obtain compound 7-1 (7 g, 59 %).

[157] Preparation of compound 7-2

[158] After mixing 4-biphenylthiopheneboronic acid (41 g, 181 mmol), compound 7-1 (71 g, 254.5 mmol), 2M Na₂CO₃ (270 mL), toluene (900 mL) and ethanol (300 mL), the reaction mixture was stirred under reflux. After 5 hours, the reaction mixture was cooled to room temperature, and was extracted with EA. The obtained organic layer was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 7-2 (34 g, 35 %).

[159] Preparation of compound 7-3

[160] After mixing compound 7-2 (34 g, 89.52 mmol), P(OEt)₃ (350 mL) and 1,2-dichlorobenzene (350 mL), the reaction mixture was stirred at 150°C. After 7 hours, the reaction mixture was cooled to room temperature, was distilled under reduced pressure, and was recrystallized with EA to obtain compound 7-3 (11 g, 35 %).

[161] Preparation of compound 7-4

[162] After putting compound 7-3 (7 g, 25.60 mmol), iodobenzene (10.44 g, 51.21 mmol), CuI (2.5 g, 12.80 mmol), K₃PO₄ (16.30 g, 76.82 mmol) and toluene (200 mL), the reaction mixture was heated to 50°C, ethylenediamine (1.72 mL, 25.60 mmol) was added, and was stirred under reflux. After 12 hours, the reaction mixture was cooled to room temperature, was extracted with EA, was washed with aqueous solution of NaHCO₃, was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 7-4 (8 g, 89.41 %).

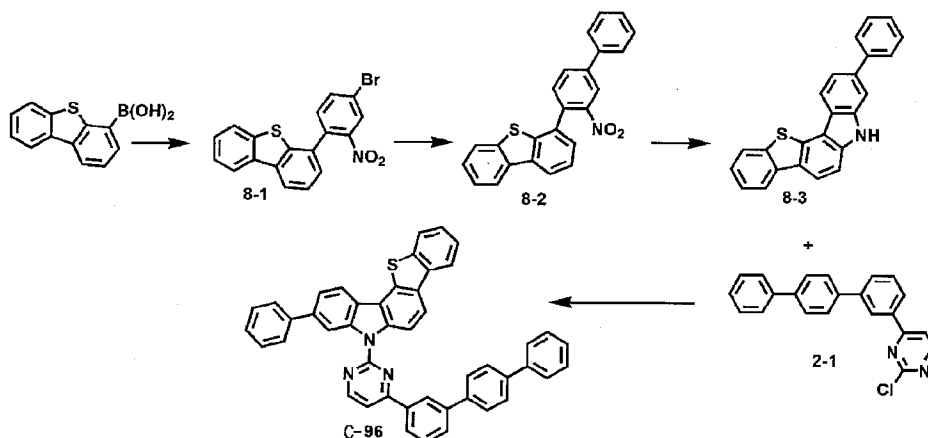
[163] Preparation of compound C-94

[164] After dissolving NaH (600 mg) in DMF (40 mL), the solution was stirred. After dissolving compound 7-4 (3.5 g, 10.015 mmol) in DMF (30 mL), the solution was added to the above NaH solution, and was stirred for 1 hour. After dissolving compound 2-1 (4.1 g, 12.0189 mmol) in DMF (30 mL), and stirring, the mixed solution of compound 7-4 was added to the mixed solution of compound 2-1, and was stirred for 12 hours. After terminating the reaction, purified water was added to the reaction mixture. The obtained solid was filtered, and was filtered through column to obtain compound C-94 (3 g, 34 %).

[165] MS/FAB found 656; calculated 655.81

[166] Example 8: Preparation of compound C-96

[167]



[168] Preparation of compound 8-1

[169] After mixing 2,5-dibromonitrobenzene (30 g, 106.8 mmol), dibenzothiophenylboronic acid (20.3 g, 88.9 mmol), $Pd(PPh_3)_4$ (5.1 g, 4.45 mmol), Na_2CO_3 (27.9 g, 267 mmol), toluene (600 mL) and EtOH (100 mL), the reaction mixture was stirred for 3 hours at 90°C. After stirring, purified water was slowly added to terminate the reaction. The reaction mixture was extracted with EA. The obtained organic layer was dried with $MgSO_4$, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 8-1 (24 g, 59 %).

[170] Preparation of compound 8-2

[171] After mixing compound 8-1 (23 g, 59.85 mmol), phenylboronic acid (8.8 g, 71.83 mmol), $Pd(PPh_3)_4$ (3.46 g, 2.99 mmol), Na_2CO_3 (19 g, 179.5 mmol), toluene (180 mL) and EtOH (90 mL), the reaction mixture was stirred for 3 hours at 120°C under reflux. After stirring, purified water was slowly added to terminate the reaction. The reaction mixture was extracted with EA. The obtained organic layer was dried with $MgSO_4$, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 8-2 (22 g, 96 %).

[172] Preparation of compound 8-3

[173] After mixing compound 8-2 (15 g, 39.3 mmol), and $P(OEt)_3$ (150 mL), the reaction mixture was stirred for 24 hours at 150°C. After stirring, the remaining solvent was removed using distillation column, and the reaction mixture was filtered through column to obtain compound 8-3 (6 g, 46 %).

[174] Preparation of compound C-96

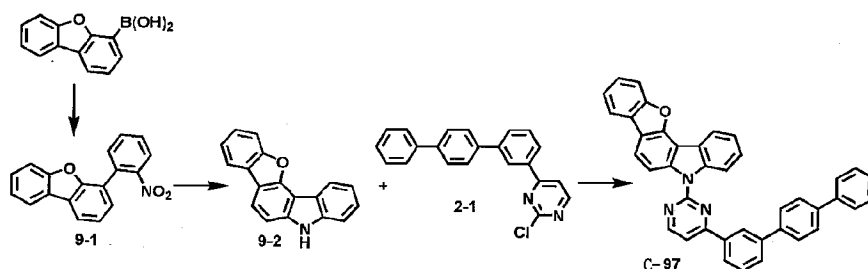
[175] After dissolving NaH (1 g, 30.6 mmol) in DMF (20 mL), the solution was stirred. After dissolving compound 8-3 (8.56 g, 24.5 mmol) in DMF (300 mL), the solution was added to the above NaH solution, and was stirred for 1 hour. After dissolving compound 2-1 (7 g, 20.4 mmol) in DMF (250 mL), and stirring, the mixed solution of compound 8-3 was added to the mixed solution of compound 2-1, and was stirred for 12 hours. After terminating the reaction, purified water was added to the reaction

mixture. The obtained solid was filtered, and was filtered through column to obtain compound **C-96** (8.9 g, 67 %)

[176] MS/FAB found 656; calculated 655.81

[177] Example 9: Preparation of compound C-97

[178]



[179] Preparation of compound 9-1

[180] After putting dibenzo[*b,d*]furan-4-ylboronic acid (20 g, 94.3 mmol), 2-bromonitrobenzene (17 g, 84.9 mmol), Pd(PPh₃)₄ (4.9 g, 4.24 mmol), K₂CO₃ (23.5 g, 169.8 mmol), toluene (100 mL), EtOH (50 mL), and purified water (50 mL), the reaction mixture was stirred for 3 hours under reflux. After terminating the reaction, the reaction mixture was cooled to room temperature, the aqueous layer was removed, and the organic layer was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through silica gel. The remaining solution was concentrated under reduced pressure to obtain compound 9-1 (24 g, 98 %).

[181] Preparation of compound 9-2

[182] After mixing compound 9-1 (24 g, 83.0 mmol), PPh₃ (65 g, 248.9 mmol) and 1,2-dichlorobenzene (250 mL), the reaction mixture was stirred for 18 hours under reflux. After terminating the reaction, the reaction mixture was cooled to room temperature, was washed with distilled water, and was extracted with dichloromethane (DCM). The extracted DCM layer was washed with 5% NH₄Cl aqueous solution, was dried with MgSO₄, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through silica gel. The remaining solution was concentrated under reduced pressure, was triturated with MeOH to obtain compound 9-2 (11.2 g, 52 %).

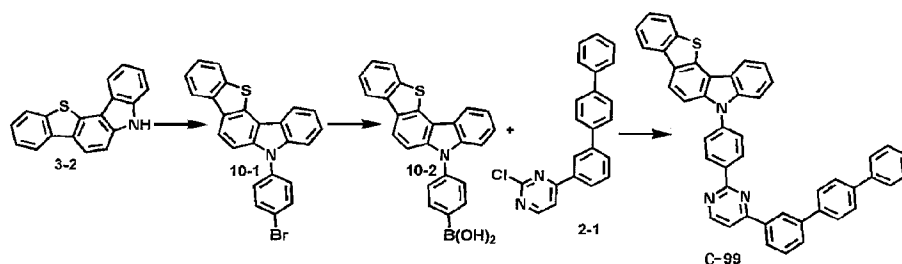
[183] Preparation of compound C-97

[184] After dissolving compound 9-2 (6 g, 23.3 mmol) in DMF (60 mL), NaH (1.1 g, 28.0 mmol) was added at 0°C, and was stirred for 10 minutes. Compound 2-1 (8.8 g, 25.7 mmol) was added to the reaction mixture, and was stirred for 19 hours. After terminating the reaction, MeOH was added to the reaction mixture. The obtained solid was filtered through silica gel, was recrystallized with DMF to obtain compound **C-97** (7 g, 53 %).

[185] MS/FAB found 656; calculated 655.81

[186] Example 10: Preparation of compound C-99

[187]



[188] Preparation of compound 10-1

[189] After mixing compound 3-2 (30 g, 109.7 mmol), 4-bromiodobenzene (62 g, 219.4 mmol), CuI (20.9 g, 109.7 mmol), ethylenediamine (14.7 mL, 219.4 mmol), K_3PO_4 (58 g, 274.3 mmol) and toluene (600 mL), the reaction mixture was stirred under reflux. After 15 hours, the reaction mixture was cooled to room temperature, CuI and K_3PO_4 was removed by filtering under reduced pressure. The remaining solution was washed with distilled water, was extracted with MC. The obtained organic layer was dried with $MgSO_4$, was filtered, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 10-1 (37 g, 78.74 %).

[190] Preparation of compound 10-2

[191] After dissolving compound 10-1 (25 g, 58.36 mmol) in THF (500 mL), and adding 2.5 M n-BuLi in hexane (23.3 mL, 58.36 mmol) to the reaction mixture in $-78^\circ C$, the reaction mixture was stirred. After 1 hour, $B(Oi-Pr)_3$ (20.1 mL, 87.54 mmol) was added slowly, and the reaction mixture was stirred for 1 hour. The reaction mixture was stirred for 4 hours at room temperature, with slowly increasing the temperature. After stirring, distilled water was added, and the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous $MgSO_4$ to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was recrystallized with EA and hexane to obtain compound 10-2 (19 g, 82.78 %).

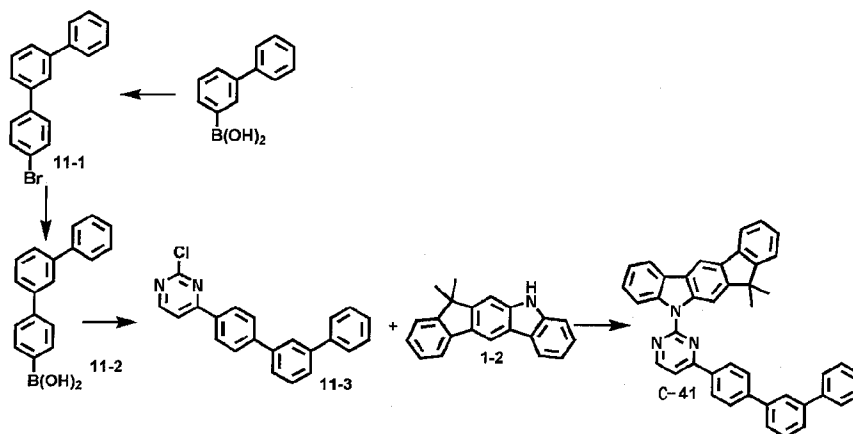
[192] Preparation of compound C-99

[193] After mixing compound 2-1 (6.1 g, 17.79 mmol), compound 10-2 (7 g, 17.79 mmol), $Pd(PPh_3)_4$ (1.0 g, 0.88 mmol), 2M K_3PO_4 aqueous solution (22 mL), toluene (130 mL) and ethanol (66 mL), the reaction mixture was stirred under reflux. After 4 hours, methanol was added to the reaction mixture. The obtained solid was filtered under reduced pressure, and the filtered solid was filtered through column to obtain compound C-99 (4.0 g, 34.28 %).

[194] MS/FAB found 656; calculated 655.81

[195] Example 11: Preparation of compound C-41

[196]



[197] Preparation of compound 11-1

[198] After mixing (1,1'-biphenyl)-3-ylboronic acid (25 g, 0.1 mol), 1-bromo-4-iodobenzene (89.3 g, 0.32 mol), Pd(PPh₃)₂Cl₂ (2.7 g, 0.038 mol), Na₂CO₃ (26 g, 0.25 mol), toluene (150 mL), EtOH (38 mL) and distilled water (150 mL), the reaction mixture was stirred for 3 hours at 110°C. After terminating the reaction, the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous MgSO₄ to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 11-1 (31 g, 80 %).

[199] Preparation of compound 11-2

[200] After dissolving compound 11-1 (31 g, 0.1 mol) in THF (750 mL), and adding 2.25 M n-BuLi in hexane (60 mL) to the reaction mixture in -78°C, the reaction mixture was stirred. After 1 hour, B(Oi-Pr)₃ (46 mL, 0.2 mol) was added slowly, and the reaction mixture was stirred for 12 hours at room temperature, with slowly increasing the temperature. After stirring, distilled water was added, and the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous MgSO₄ to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was filtered through column to obtain compound 11-2 (21 g, 76 %).

[201] Preparation of compound 11-3

[202] After mixing 2,4-dichloropyrimidine (7.6 g, 0.05 mol), compound 11-2 (10.7 g, 0.39 mol), Pd(PPh₃)₄ (2.3 g, 0.02 mol), Na₂CO₃ (10 g, 0.089 mol), toluene (200 mL), EtOH (50 mL) and purified water (50 mL), the reaction mixture was stirred for 12 hours at 120°C. After terminating the reaction, the reaction mixture was extracted with EA. The obtained organic layer was dried with anhydrous MgSO₄ to remove the remaining moisture, was distilled under reduced pressure to remove the solvent, and was recrystallized to obtain compound 11-3 (9.4 g, 70 %).

[203] Preparation of compound C-41

[204] After dissolving 60% NaH (1.0 g, 0.025 mol) in DMF (40 mL), the solution was

stirred. After dissolving compound 1-2 (5.0 g, 0.017 mol) in DMF (40 mL), the solution was added to the above NaH solution, and was stirred for 1 hour. After dissolving compound 11-3 (7.2 g, 0.017 mol) in DMF (20 mL), and stirring, the mixed solution of compound 1-2 was added to the mixed solution of compound 11-3, and was stirred for 12 hours. The obtained yellow solid was filtered, was washed with MeOH, and was recrystallized to obtain compound **C-41** (5 g, 48 %)

[205] MS/FAB found 590; calculated 589.73

[206] Experimental Example 1: Production of an OLED device using the compound according to the present invention

[207] An OLED device was produced using the compound according to the present invention. A transparent electrode indium tin oxide (ITO) thin film (15 Ω /sq) on a glass substrate for an organic light-emitting diode (OLED) device (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. $N^1, N^{1'}-([1, 1'-biphenyl]-4, 4'-diyl)bis(N^1-(naphthalen-1-yl)-N^4, N^4-diphenylbenzene-1, 4-diamine)$ 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-67** was introduced into one cell of the vacuum vapor depositing apparatus, as a host material, and compound **D-56** was introduced into another cell as a dopant. The two materials were evaporated at different rates and were deposited in a doping amount of 15 wt% to form a light-emitting layer having a thickness of 30 nm on the hole transport layer. Then, 2-(4-(9,10-di(naphthalen-2-yl)anthracen-2-yl)phenyl)-1-phenyl-1H-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% to form an electron transport layer having a thickness of 30nm on the light-emitting layer. Then, after depositing lithium quinolate as an electron injection layer having a thickness of 2 nm on the electron transport layer, an Al cathode having a thickness of 150 nm was deposited by another vacuum vapor deposition apparatus on the electron injection layer. Thus, an OLED device was produced. All the material used for producing the OLED device were purified by

vacuum sublimation at 10^{-6} torr prior to use.

[208] The produced OLED device showed green emission having a luminance of 1,040 cd/m² and a current density of 2.29 mA/cm² at a driving voltage of 3.0 V.

[209] Comparative Example 1: Production of an OLED device using conventional electro-luminescent compounds

[210] An OLED device was produced in the same manner as that of Example 1, except that 4,4'-bis(carbazol-9-yl)biphenyl (CBP) was used as a host material and compound **D-5** was used as a dopant, and introduced into a cell of the vacuum vapor depositing apparatus, and that a hole blocking layer having a thickness of 10 nm was deposited by using bis(2-methyl-8-quinolinato)(p-phenylphenolato)aluminum(III) (Balq).

[211] Comparative Example 2: Production of an OLED device using conventional electro-luminescent compounds

[212] An OLED device was produced in the same manner as that of Example 1, except that 4,4'-bis(carbazol-9-yl)biphenyl (CBP) was used as a host material and compound **D-58** was used as a dopant, and introduced into a cell of the vacuum vapor depositing apparatus, and that a hole blocking layer having a thickness of 10 nm was deposited by using bis(2-methyl-8-quinolinato)(p-phenylphenolato)aluminum(III) (Balq).

[213] The results of experimental examples 1-15 and comparative examples 1-2 are shown in the following Table.

[214]

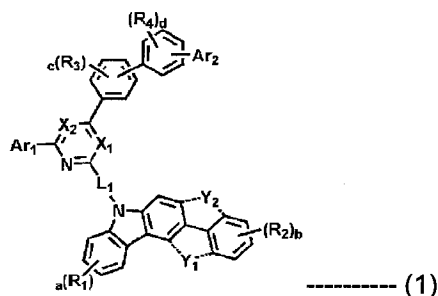
No. of Experimental Examples	Host Material	Dopant	Driving Voltage (V)	Current Density (mA/cm ²)	Luminance (cd/m ²) /Color
1	C-67	D-56	3.0	2.29	1040/Green
2	C-41	D-5	3.2	2.60	1030/Green
3	C-55	D-57	3.9	2.35	1050/Green
4	C-56	D-57	4.0	2.09	1040/Green
5	C-56	D-58	3.0	2.67	1060/Orange
6	C-71	D-34	3.8	2.51	1020/Green
7	C-73	D-34	3.3	2.17	1000/Green
8	C-87	D-5	3.5	2.52	1030/Green
9	C-87	D-59	3.2	6.34	1040/Dark Orange
10	C-90	D-5	3.1	2.45	1050/Green
11	C-91	D-5	3.7	2.18	1050/Green
12	C-94	D-37	4.3	3.15	1020/Green
13	C-97	D-34	4.1	2.30	1070/Green
14	C-98	D-5	3.1	2.39	1050/Green
15	C-99	D-5	3.2	2.66	1040/Green
Comparative Example 1	CBP	D-5	4.9	2.86	1000/Green
Comparative Example 2	CBP	D-58	4.6	3.05	1000/Orange

- [215] The organic electroluminescence compounds of the present invention have superior luminous characteristics than the conventional materials. In addition, a device using the compounds according to the present invention as a green or orange light emitting host material not only has excellent luminous characteristics, but also induces an increase in power efficiency by reducing the driving voltage.

Claims

[Claim 1]

A compound represented by the following formula 1:



wherein

L_1 represents 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 and X_2 each independently represent CH or N;

Y_1 and Y_2 each independently represent -O-, -S-, -CR₅R₆- or NR₇-, provided that Y_1 and Y_2 do not simultaneously exist;

Ar_1 and Ar_2 each independently represent hydrogen, deuterium, a halogen, a substituted or unsubstituted (C1-C30)alkyl group, a substituted or unsubstituted (C6-C30)aryl group, or a substituted or unsubstituted 3- to 30-membered heteroaryl group;

R_1 to R_4 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 are linked to an adjacent substituent(s) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

R_5 to R_7 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) to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

a, b, c and d each independently represent an integer of 1 to 4; where a, b, c or d is an integer of 2 or more, each of R_1 , each of R_2 , each of R_3 or each of R_4 is the same or different;

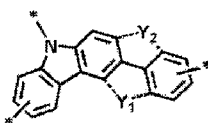
the heterocycloalkyl group and the heteroaryl(ene) group contain at least one hetero atom selected from B, N, O, S, P(=O), Si and P.

[Claim 2]

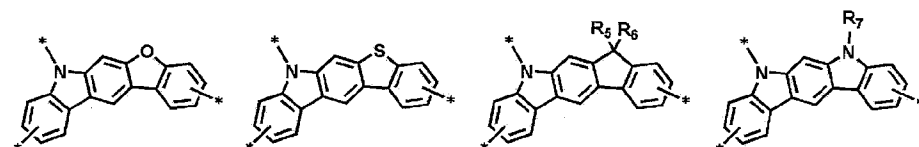
The compound according to claim 1, wherein the substituents of the substituted alkyl group, the substituted aryl(ene) group, the substituted heteroaryl(ene) group, the substituted cycloalkyl(ene) group, the substituted heterocycloalkyl group and the substituted aralkyl group L_1 , Ar_1 , Ar_2 , R_1 to R_7 and R_{11} to R_{17} 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 substituted or unsubstituted with a (C1-C30)alkyl or 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; an 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.

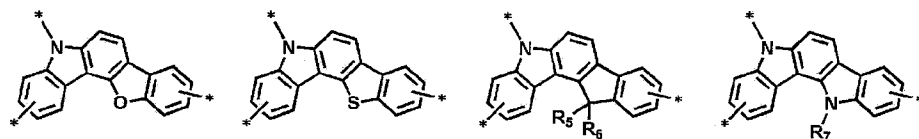
[Claim 3]

The compound according to claim 1, wherein

the moiety,  in formula 1 is selected from the following

structures:





wherein R_5 , R_6 and R_7 are as defined in claim 1.

[Claim 4]

The compound according to claim 1, wherein

L_1 represents a single bond, a 3- to 30-membered heteroarylene group or a (C6-C30)arylene group;

X_1 and X_2 each independently represent CH or N;

Y_1 and Y_2 each independently represent -O-, -S-, -CR₅R₆- or -NR₇-, provided that Y_1 and Y_2 do not simultaneously exist;

Ar_1 and Ar_2 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;

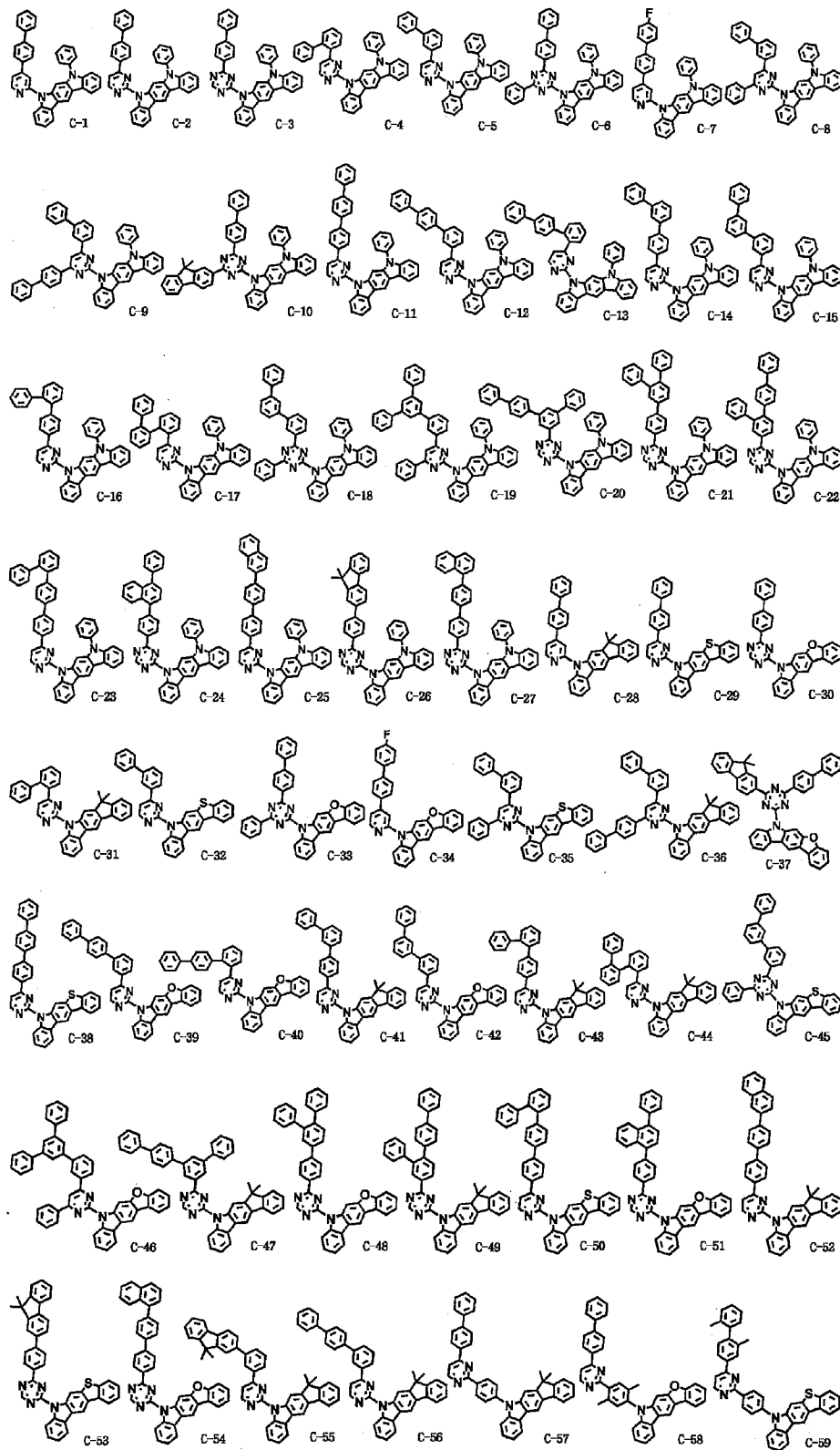
R_1 to R_4 each independently represent hydrogen, deuterium, a halogen, a (C1-C30)alkyl group, a (C6-C30)aryl group, a 3- to 30-membered heteroaryl group, a carbazolyl group, or -SiR₁₃R₁₄R₁₅;

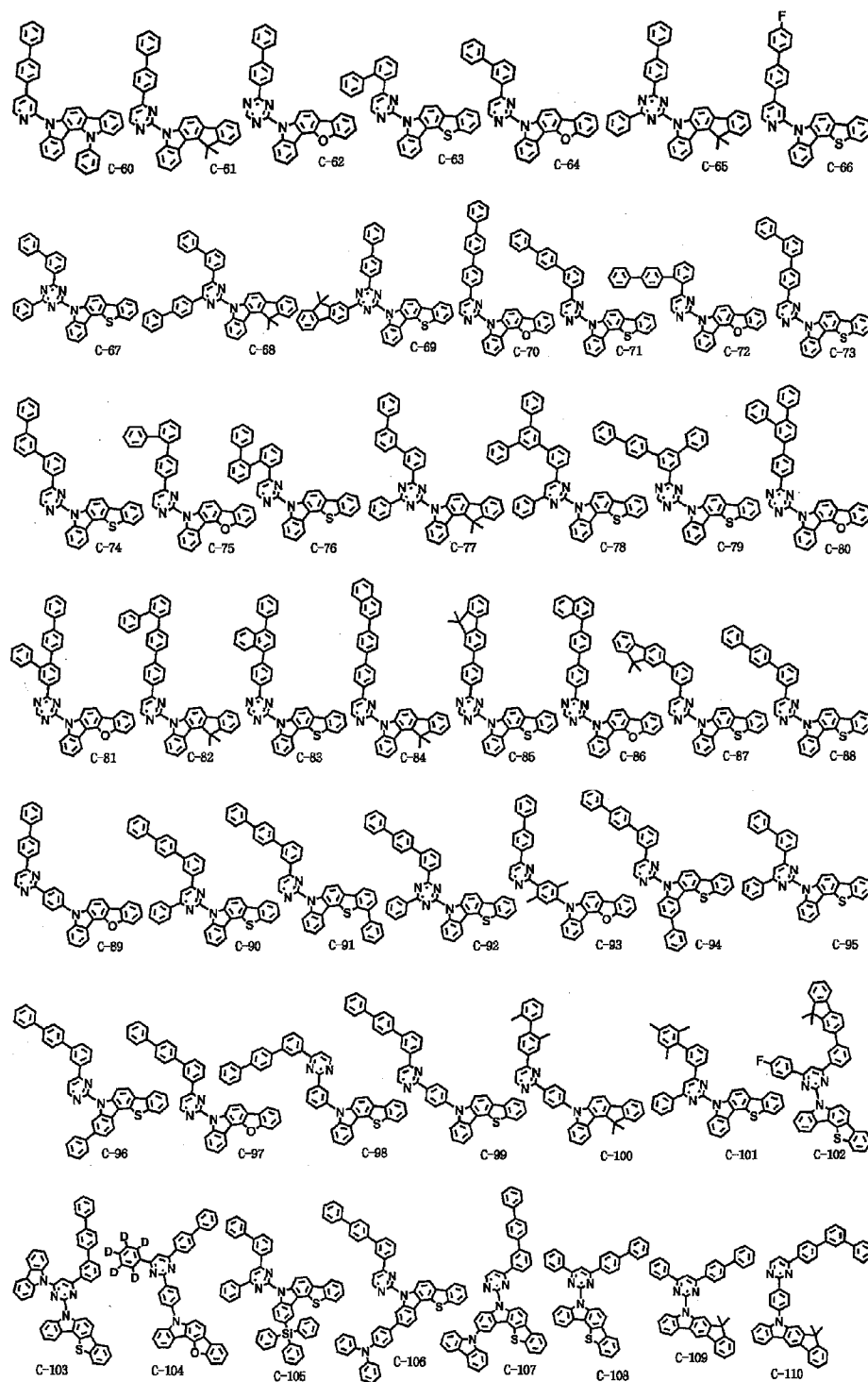
R_5 to R_7 each independently represent a (C1-C30)alkyl group, a (C6-C30)aryl group, or a 3- to 30-membered heteroaryl group; or R_5 and R_6 are linked to each other to form a mono- or polycyclic, (C5-C30)alicyclic or aromatic ring whose carbon atom(s) may be replaced with at least one hetero atom selected from nitrogen, oxygen and sulfur;

R_{13} 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 and heteroarylene groups in L_1 , the alkyl, aryl, heteroaryl and carbazolyl groups in Ar_1 and Ar_2 , and the alkyl, aryl and heteroaryl groups in R_1 to R_7 and R_{13} 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 3- to 30-membered heteroaryl group substituted or unsubstituted with a (C1-C30)alkyl or a (C6-C30)aryl; a (C3-C30)cycloalkyl group; a 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 carbazolyl group; a di(C1-C30)alkylamino group; a di(C6-C30)arylamino group; a (C1-C30)alkyl(C6-C30)arylamino group; a (C6-C30)aryl(C1-C30)alkyl group; and a (C1-C30)alkyl(C6-C30)aryl group.

[Claim 5]

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





[Claim 6]

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

INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2012/005839

A. CLASSIFICATION OF SUBJECT MATTER

*C07D 487/04 (2006.01) C07D 495/04 (2006.01) C07D 491/048 (2006.01) C07D 403/04 (2006.01) C07D 405/04 (2006.01)
C07D 409/04 (2006.01) C07F 7/02 (2006.01) C09K 11/06 (2006.01) H01L 51/54 (2006.01) H01L 27/32 (2006.01) H05B
33/14 (2006.01) H01L 51/50 (2006.01) C07D 403/10 (2006.01) C07D 405/10 (2006.01) C07D 409/10 (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)

CAS Registry and CAPlus via STN: substructure search in accordance with Formula (I)

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 8 November 2012	Date of mailing of the international search report 08 November 2012
Name and mailing address of the ISA/AU AUSTRALIAN PATENT OFFICE PO BOX 200, WODEN ACT 2606, AUSTRALIA Email address: pct@ipaustalia.gov.au Facsimile No.: +61 2 6283 7999	Authorised officer Marie-Luise Huber AUSTRALIAN PATENT OFFICE (ISO 9001 Quality Certified Service) Telephone No. 0262832788

INTERNATIONAL SEARCH REPORT		International application No.
C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		PCT/KR2012/005839
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2011060867 A1 (Merck Patent GmbH) 26 May 2011 & CAPlus Online Abstract Accession No. 2011:651168 CAPlus Abstract: RN 1257247-96-3; Table 6, Compound IC1, page 116;	1-4 and 6
X	WO 2011032686 A1 (Merck Patent GmbH) 24 March 2011 & CAPlus Online Abstract Accession No. 2011:372843 CAPlus Abstract: RN's 1257247-96-3, 1257248-10-4, 1257248-42-2; Compounds 14-16, pages 96-98; Several compounds from line 10, page 63 to line 15, page 69;	1-6
X	Endo, Ayataka et al., "Efficient up-conversion of triplet excitons into a singlet state and its application for organic light emitting diodes", Applied Physics Letters (2011), 98(8), 083302/1-083302/3, ISSN: 0003-6951 & CAPlus Online Abstract Accession No. 2011:242340 CAPlus Abstract: RN 1286756-79-3	1-4 and 6
X	WO 2010136109 A1 (Merck Patent GmbH) 02 December 2010 & CAPlus Online Abstract Accession No. 2010:1498208 CAPlus Abstract: RN's 1257247-96-3, 1257248-10-4, 1257248-42-2; page 39, column 1, compounds 2, 4; page 42, column 1, compound 1; page 44, column 1, compounds 5-7 and column 2, compounds 5-7; all compounds on pages 45-46; Examples 13, 20 and 37; Table 4: Examples H22 - H23; Table 5: Example SH1; page 72, line 32 to page 73, line 26	1-6
P,X	KR 2012078301 A1 (Cheil Industries, Inc.) 10 July 2012 CAPlus Online Abstract Accession No. 2012:1014525: RN's 1391729-50-2, 1391729-52-4, 1391729-53-5 and 1391729-54-6; Examples E-2, E-4, E-6 and E-8 on pages 18, 37 and 58;	1-4 and 6
P,X	US 20120126208 A1 (Kawamura et al.) 24 May 2012 CAPlus Online Abstract Accession No. 2012:739172: RN 1377150-38-3; Compound TB-26, paragraph [0298], page 28; Example 30, Table 7, paragraph [0427], page 34;	1-4 and 6
P,X	WO 2012039561 A1 (Rohm and Haas Electronic Materials Korea Ltd.) 29 March 2012 CAPlus Online Abstract Accession No. 2012:458227: RN 1369586-62-8; pages 9-12: Examples 2-3, 6, 14, 28, 32-33, 35-36 and 46-48; page 31, paragraphs [222] - [223]; Claims 4-10;	1-4 and 6
P,X	WO 2012013271 A1 (Merck Patent GmbH) 02 February 2012 CAPlus Online Abstract Accession No. 2012:157796: RN 1356920-54-1; page 79: Example M2	1-4 and 6
P,X	WO 2011137922 A1 (Merck Patent GmbH) 10 November 2011 CAPlus Online Abstract Accession No. 2011:1438715: RN 1257248-42-2; Several compounds on pages 60-63 and 66; Example 7, compound 10, page 96; Figure 1;	1-4 and 6
P,X	WO 2011108902 A2 (Duksan High Metal Co., Ltd.) 09 September 2011 CAPlus Online Abstract Accession No. 2011:1136802: RN's 1333333-75-7, 1333333-76-8, 1333333-80-4, 1333333-81-5, 1333333-82-6, 1333333-83-7, 1333333-84-8, 1333333-85-9, 1333333-86-0, 1333333-87-1 and 1333333-88-2. and Figure 1; paragraphs [9], [19], [33] and [44]-[45]; compounds 39-41 and 44-52, pages 7-8; Claims and Figures;	1-4 and 6
Form PCT/ISA/210 (fifth sheet) (July 2009)		

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

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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)			