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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. The organic electroluminescent compound according to the present invention is represented by Chemical Formula 1:

[2] [Chemical Formula 1]

[3]
$$A_{18}^{A_{17},A_{16}}$$
 $A_{19}^{A_{15}}$ $A_{19}^{A_{15}}$

Background Art

- [4] In general, the organic EL device commonly has a configuration of anode/hole injection layer (HIL)/hole transport layer (HTL)/emission material layer (EML)/electron transport layer (ETL)/electron injection layer (EIL)/cathode. Organic electroluminescent devices emitting blue, green or red light may be created depending on how to form the emission material layer.
- [5] At present, 4,4'-bis(carbazol-9-yl) biphenyl (CBP) is the most widely known as a host material for a phosphorescent material. Highefficiency OLEDs using a hole blocking layer comprising 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP), Bis(2-methyl-8-quinolinato)(ρ-phenyl-phenolato)aluminum(III) (BAlq), etc. are reported. Highperformance OLEDs using BAlq derivatives as a host were reported by Pioneer (Japan) and others.
- Although these materials provide good electroluminescence characteristics, they are disadvantageous in that degradation may occur during the hightemperature deposition process in vacuum because of low glass transition temperature and poor thermal stability. Since the power efficiency of an OLED is given by $(\pi / \text{voltage}) \times \text{current}$ efficiency, the power efficiency is inversely proportional to the voltage. High power efficiency is required to reduce the power consumption of an OLED. Actually, OLEDs using phosphorescent materials provide much better current efficiency (cd/A) than

those using fluorescent materials. However, when the existing materials such as BAlq, CBP, etc. are used as a host of the phosphorescent material, there is no significant advantage in power efficiency (lm/W) over the OLEDs using fluorescent materials because of high driving voltage.

[7] Further, the OLED devices do not have satisfactory operation life. Therefore, development of more stable, higherperformance host materials is required.

Disclosure of Invention

Technical Problem

[8] Accordingly, an object of the present invention is to provide an organic electroluminescent compound having luminescence efficiency and device operation life improved over existing host materials and having superior backbone with appropriate color coordinates in order to solve the aforesaid problems. Another object of the present invention is to provide a highly efficient and longlife organic electroluminescent device employing the organic electroluminescent compound as an electroluminescent material.

Solution to Problem

[9] The present invention relates to organic electroluminescent compounds represented by Chemical Formula 1, and an organic electroluminescent device using the same. The organic electroluminescent compounds according to the present invention exhibit high luminous efficiency, excellent color purity and life property of the material, so that OLED's with very excellent operation life can be manufactured therefrom.

[10] [Chemical Formula 1]

[11]
$$A_{18}^{A_{17},A_{16}}$$
 $A_{19}^{A_{15}}$ $A_{19}^{A_{15}}$

- [12] wherein
- [13] A₁ through A₁₉ independently represent CR_1 or N, X represents - $(CR_2R_3)_{1}$ -, -N(R₄)-, -S-, -O-, -Si(R₅)(R₆)-, -P(R₇)-, -P(=O)(R₈)- or -B(R₉)-, and Ar₁ represents (C6-C40)arylene with or without substituent(s) or (C3-C40)heteroarylene with or without substituent(s), except for the case where m is 0, and A₁₅ through A₁₉ are CR_1 at the same time;
- [14] R₁ through R₉ independently represent hydrogen, deuterium, halogen, (C1-C30)alkyl

with or without substituent(s), (C6-C30)aryl with or without substituent(s), substituted or unsubstituted (C6-C30)aryl fused with one or more (C3-C30)cycloalkyl(s) with or without substituent(s), (C3-C30)heteroaryl with or without substituent(s), 5- to 7-membered heterocycloalkyl with or without substituent(s), 5- to 7-membered heterocycloalkyl fused with one or more aromatic ring(s) with or without substituent(s), (C3-C30)cycloalkyl fused with one or more aromatic ring(s) with or without substituent(s), (C3-C30)cycloalkyl fused with one or more aromatic ring(s) with or without substituent(s), cyano, trifluoromethyl, NR₂₁R ₂₂, BR₂₃R₂₄, PR₂₅R₂₆, P(=O)R₂₇R₂₈, R^aR^bR^cSi-, R^dY-, R^cC(=O)-, R^fC(=O)O-, (C6-C30)ar(C1-C30)alkyl with or without substituent(s), (C2-C30)alkenyl with or without substituent(s), carboxyl, nitro,

$$R_{61}$$
, R_{62} , or hydroxyl, or each of them may be linked to an R_{61} R_{62} R_{62} R_{62} R_{61}

adjacent substituent via (C3-C30)alkylene or (C3-C30)alkenylene with or without a fused ring to form an alicylic ring, a mono or polycyclic aromatic ring or a mono or polycyclic heteroaromatic ring;

- [15] W represents $-(CR_{51}R_{52})_{n-}$, $-(R_{51})C=C(R_{52})-$, $-N(R_{53})-$, -S-, -O-, $-Si(R_{54})(R_{55})-$, $-P(R_{56})-$, $-P(=O)(R_{57})-$, -C(=O)- or $-B(R_{58})-$, and R_{51} through R_{58} and R_{61} through R_{63} are the same as R_1 through R_9 ;
- [16] the heterocycloalkyl or heteroaryl may contain one or more heteroatom(s) selected from B, N, O, S, P(=O), Si and P;
- R₂₁ through R₂₈ independently represent (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s) or (C3-C30)heteroaryl with or without substituent(s), R^a, R^b, and R^c independently represent (C1-C30)alkyl with or without substituent(s) or (C6-C30)aryl with or without substituent(s), Y represents S or O, R^d represent (C1-C30)alkyl with or without substituent(s) or (C6-C30)aryl with or without substituent(s), (C1-C30)alkoxy with or without substituent(s), (C6-C30)aryl with or without substituent(s) or (C6-C30)aryloxy with or without substituent(s), R^f represent (C1-C30)alkyl with or without substituent(s), (C1-C30)alkoxy with or without substituent(s), (C1-C30)alkoxy with or without substituent(s) or (C6-C30)aryloxy with or without substituent(s)
- [18] m represents an integer 0 to 2; and
- [19] 1 and n represent an integer 1 or 2.

[20]

[21] In the present invention, "alkyl", "alkoxy" and other substituents containing "alkyl"

moiety include both linear and branched species. In the present invention, "cycloalkyl"

includes both adamantyl with or without substituent(s) and (C7-C30)bicycloalkyl with or without substituent(s).

- In the present invention, "aryl" means an organic radical derived from an aromatic hydrocarbon by the removal of one hydrogen atom, and may include a 4- to 7-membered, particularly 5- or 6-membered, single ring or fused ring, including a plurality of aryls linked by chemical bond(s). Specific examples include phenyl, naphthyl, biphenyl, anthryl, indenyl, fluorenyl, phenanthryl, triphenylenyl, pyrenyl, perylenyl, chrysenyl, naphthacenyl, fluoranthenyl, etc., but are not limited thereto. "heteroaryl" means an aryl group containing 1 to 4 heteroatom(s) selected from B, N, O, S, P(=O), Si and P as aromatic ring backbone atom(s), other remaining aromatic ring backbone atoms being carbon. It may be 5- or 6-membered monocyclic heteroaryl or polycyclic heteroaryl resulting from condensation with a benzene ring, and may be partially saturated. Further, the heteroaryl includes more than one heteroaryls linked by chemical bond(s). The heteroaryl includes a divalent aryl group wherein the heteroatom(s) in the ring may be oxidized or quaternized to form, for example, an Noxide or a quaternary salt.
- Specific examples include monocyclic heteroaryl such as furyl, thienyl, pyrrolyl, imidazolyl, pyrazolyl, thiadiazolyl, isothiazolyl, isoxazolyl, oxazolyl, oxadiazolyl, triazinyl, tetrazinyl, triazolyl, tetrazolyl, furazanyl, pyridyl, pyrazinyl, pyrimidinyl, pyridazinyl, etc., polycyclic heteroaryl such as benzofuryl, benzothienyl, isobenzofuryl, benzimidazolyl, benzothiazolyl, benzoisoxazolyl, benzoisoxazolyl, benzoisoxazolyl, isoindolyl, indolyl, indazolyl, benzothiadiazolyl, quinolyl, isoquinolyl, cinnolinyl, quinazolinyl, quinolizinyl, quinoxalinyl, carbazolyl, phenanthridinyl, benzodioxolyl, etc., an Noxide thereof (e.g., pyridyl Noxide, quinolyl Noxide, etc.), a quaternary salt thereof, etc., but are not limited thereto.
- The "(C1-C30)alkyl" groups described herein may include (C1-C20)alkyl or (C1-C10)alkyl and the "(C6-C30)aryl" groups include (C6-C20)aryl or (C6-C12)aryl. The "(C3-C30)heteroaryl" groups include (C3-C20)heteroaryl or (C3-C12)heteroaryl and the "(C3-C30)cycloalkyl" groups include (C3-C20)cycloalkyl or (C3-C7)cycloalkyl. The "(C2-C30)alkenyl or alkynyl" group include (C2-C20)alkenyl or alkynyl, (C2-C10)alkenyl or alkynyl.
- In "with or without substituent(s)", the substituent is further substituted by one or more substituent(s) selected from the group consisting of deuterium, halogen, (C1-C30)alkyl with or without halogen substituent(s), (C6-C30)aryl, (C3-C30)heteroaryl with or without (C6-C30)aryl substituent(s), 5- to 7-membered heterocycloalkyl, 5- to 7-membered heterocycloalkyl fused with one or more aromatic ring(s), (C3-C30)cycloalkyl, (C3-C30)cycloalkyl fused with one or more aromatic ring(s), tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl,

(C2-C30)alkenyl, (C2-C30)alkynyl, cyano, carbazolyl, NR₃₁R₃₂, BR₃₃R₃₄, PR₃₅R₃₆, P(=O)R₃₇R₃₈, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyl(C6-C30)aryl, (C1-C30)alkyloxy, (C1-C30)alkylthio, (C6-C30)aryloxy, (C6-C30)arylthio, (C1-C30)alkoxycarbonyl, (C1-C30)alkylcarbonyl, (C6-C30)arylcarbonyl, (C6-C30)aryloxycarbonyl, (C1-C30)alkoxycarbonyloxy, (C1-C30)alkylcarbonyloxy, (C6-C30)arylcarbonyloxy, (C6-C30)arylcarbonyloxy, (C6-C30)arylcarbonyloxy, arboxyl, nitro and hydroxyl, or is linked to an adjacent substituent to form a ring, wherein R₃₁ through R₃₈ independently represent (C1-C30)alkyl, (C6-C30)aryl or (C3-C30)heteroaryl.

[26] The R₁ through R₉,R₂₁ through R₂₈, R₅₁ through R₅₈ and R₆₁ through R₆₃ are independently selected from hydrogen, deuterium, halogen, alkyl such as methyl, ethyl, propyl, butyl, pentyl, hexyl, ethylhexyl, heptyl, octyl, etc., aryl such as phenyl, naphthyl, fluorenyl, biphenyl, phenanthryl, terphenyl, pyrenyl, perylenyl, spirobifluorenyl, fluoranthenyl, chrysenyl, triphenylenyl, etc., aryl fused with one or more cycloalkyl such as 1,2-dihydroacenaphthyl, heteroaryl such as dibenzothiophenyl, dibenzofuryl, carbazolyl, pyridyl, furyl, thienyl, quinolyl, triazinyl, pyrimidinyl, pyridazinyl, quinoxalinyl, phenanthrolinyl, etc., heterocycloalkyl fused with one or more aromatic ring such as benzopyrrolidino, benzopiperidino, dibenzomorpholino, dibenzoazepino, etc., amino substituted by aryl such as phenyl, naphthyl, fluorenyl, biphenyl, phenanthryl, terphenyl, pyrenyl, perylenyl, spirobifluorenyl, fluoranthenyl, chrysenyl, triphenylenyl, etc. or heteroaryl such as dibenzothiophenyl, dibenzofuryl, carbazolyl, pyridyl, furyl, thienyl, quinolyl, triazinyl, pyrimidinyl, pyridazinyl, quinoxalinyl, phenanthrolinyl, etc., aryloxy such as biphenyloxy, etc., arylthio such as biphenylthio, etc., aralkyl such as biphenylmethyl, triphenylmethyl, etc.,

$$R_{61}$$
 or R_{62} , but are not limited thereto, and may be further sub-

stituted as shown in Chemical Formula 1.

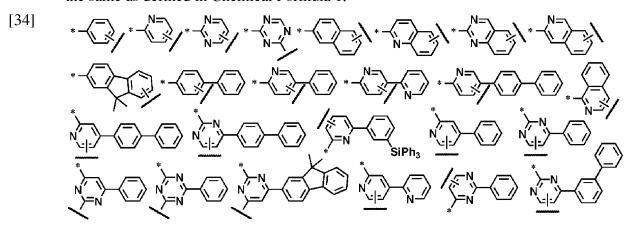
[27] More specifically, the R_1 through R_9 may be exemplified as following structures but are not limited thereto.

$$[28] \\ R_{71}^{-} \mid \underset{R_{73}}{\overset{R_{72}}{\nearrow}} N^{-} \mid \underset{R_{76}}{\overset{R_{75}}{\nearrow}} \underset{R_{74}}{\overset{R_{74}}{\nearrow}} \mid \underset{R_{80}}{\overset{R_{75}}{\nearrow}} \underset{R_{81}}{\overset{R_{71}}{\nearrow}} \mid \underset{R_{82}}{\overset{R_{83}}{\nearrow}} \mid \underset{R_{82}}{\overset{N}{\nearrow}} \mid \underset{R_{84}}{\overset{N}{\nearrow}} \mid \underset{R_{84}}{\overset{N}{\nearrow}} \mid \underset{R_{85}}{\overset{N}{\nearrow}} \mid \underset{R_{87}}{\overset{N}{\nearrow}} \mid \underset{R_{87}}{\overset{N}{\nearrow}} \mid \underset{R_{87}}{\overset{N}{\nearrow}} \mid \underset{R_{87}}{\overset{N}{\nearrow}} \mid \underset{R_{87}}{\overset{N}{\nearrow}} \mid \underset{R_{96}}{\overset{N}{\nearrow}} \mid \underset{N}{\overset{N}{\nearrow}} \mid \underset{N}{\overset{N}{\nearrow$$

[29]

$$\begin{bmatrix} 30 \end{bmatrix} \qquad \begin{matrix} R_{103} - S_{-} \\ R_{104} - O_{-} \end{matrix} \qquad \begin{matrix} S \\ R_{106} \end{matrix} \qquad \begin{matrix} R_{105} \end{matrix} \qquad \begin{matrix} R_{107} \end{matrix} \qquad \begin{matrix} R_{109} \end{matrix} \qquad \begin{matrix} R_{109} \end{matrix} \qquad \begin{matrix} R_{111} \\ R_{110} \end{matrix} \qquad \begin{matrix} R_{111} \\ R_{112} \end{matrix} \qquad \begin{matrix} R_{111} \\ R_{121} \end{matrix} \qquad \begin{matrix} R_{121} \end{matrix} \qquad \begin{matrix} R_{127} \\ R_{128} \end{matrix} \qquad \begin{matrix} R_{130} \end{matrix} \qquad \begin{matrix} R_{134} \\ R_{133} \end{matrix} \qquad \begin{matrix} R_{134} \\ R_{135} \end{matrix} \qquad \begin{matrix} R_{137} - S_{-} \\ R_{138} \end{matrix} \qquad \begin{matrix} R_{137} - S_{-} \\ R_{138} \end{matrix} \qquad \begin{matrix} R_{138} \\ R_{138} \end{matrix} \qquad \begin{matrix} R_{134} \\ R_{135} \end{matrix} \qquad \begin{matrix} R_{137} - S_{-} \\ R_{138} \end{matrix} \qquad \begin{matrix} R_{138} \\ R_{138} \end{matrix}$$

- [31] wherein
- R₇₁ through R₁₃₈ independently represent hydrogen, deuterium, halogen, [32] (C1-C30)alkyl, (C6-C30)aryl, (C6-C30)aryl fused with one or more (C3-C30)cycloalkyl(s), (C3-C30)heteroaryl, 5- to 7-membered heterocycloalkyl, 5- to 7-membered heterocycloalkyl fused with one or more aromatic ring(s), (C3-C30)cycloalkyl, (C3-C30)cycloalkyl fused with one or more aromatic ring(s), cyano, amino, (C1-C30) alkylamino, (C6-C30) arylamino, NR₄₁R₄₂, BR₄₃R₄₄, PR₄₅R₄₆, $P(=O)R_{47}R_{48}$, tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyloxy, (C1-C30)alkylthio, (C6-C30) aryloxy, (C6-C30) arylthio, (C1-C30) alkoxycarbonyl, (C1-C30) alkylcarbonyl, (C6-C30)arylcarbonyl, (C2-C30)alkenyl, (C2-C30)alkynyl, (C6-C30)aryloxycarbonyl, (C1-C30)alkoxycarbonyloxy, (C1-C30)alkylcarbonyloxy, (C6-C30)arylcarbonyloxy, (C6-C30)aryloxycarbonyloxy, carboxyl, nitro or hydroxyl, or each of them may be linked to an adjacent substituent via (C3-C30)alkylene or (C3-C30)alkenylene with or without a fused ring to form an alicylic ring or a mono or polycyclic aromatic ring, wherein R₄₁ through R₄₈ independently represent (C1-C30)alkyl, (C6-C30)aryl or (C3-C30)heteroaryl.
- [33] The \star (Ar₁) is exemplified as following structures but not limited thereto. m is the same as defined in Chemical Formula 1.



[35]

[40]

[36] The organic electroluminescent compound according to the present invention may be specifically exemplified as following compounds but is not limited thereto.

[37] [38] [39]

[43]

[47] The organic electroluminescent compound according to the present invention may be

prepared as shown in following Reaction Scheme 1.

[48] [Reaction Scheme 1]

$$\begin{bmatrix} 49 \end{bmatrix} \xrightarrow{A_{17}A_{16}} \xrightarrow{A_{18},A_{15}} \xrightarrow{A_{18},A_{15}} \xrightarrow{A_{18},A_{15}} \xrightarrow{A_{18},A_{15}} \xrightarrow{A_{18},A_{15}} \xrightarrow{A_{18},A_{15}} \xrightarrow{A_{19},A_{15}} \xrightarrow{A_{19},A_{15}$$

- [50] wherein
- [51] A_1 through A_{19} , X, Ar_1 and m are the same as defined in Chemical Formula 1.
- Provided is an organic electroluminescent device, which comprises a first electrode; a second electrode; and one or more organic layer(s) interposed between the first electrode and the second electrode, wherein the organic layer comprises one or more organic electroluminescent compound(s) represented by Chemical Formula 1. The organic electroluminescent compound is used as a host material of the electroluminescent layer.
- [53] In addition, the organic layer may include the electroluminescent layer, and the electroluminescent layer may further include one or more dopants besides one or more organic electroluminescent compounds of Chemical Formula 1. The dopant applied to the organic electroluminescent device of the present invention is not specifically limited.
- [54] Preferably, the dopant applied to the organic electroluminescent device of the present invention is selected from following Chemical Formula 2.
- [55] [Chemical Formula 2]
- [56] $M^1L^{101}L^{102}L^{103}$
- [57] wherein
- [58] M¹ is a metal selected from a group consisting of Group 7, Group 8, Group 9, Group 10, Group 11, Group 13, Group 14, Group 15 and Group 16 metals, and ligand L¹0¹, L¹0² and L¹0³ are independently selected from the following structures;

- [63] wherein
- [64] R₂₀₁ through R₂₀₃ independently represent hydrogen, (C1-C30)alkyl with or without halogen substituent(s), (C6-C30)aryl with or without (C1-C30)alkyl substituent(s) or halogen;
- [65] R_{204} through R_{219} independently represent hydrogen, (C1-C30)alkyl with or without substituent(s), (C1-C30)alkoxy with or without substituent(s), (C3-C30)cycloalkyl with or without substituent(s), (C2-C30)alkenyl with or without substituent(s), (C6-C30)aryl with or without substituent(s), mono- or di-(C1-C30)alkylamino with or without substituent(s), SF_5 , tri(C1-C30)alkylsilyl with or without substituent(s), SF_5 , tri(C1-C30)alkylsilyl with or without substituent(s), SF_5 , tri(C6-C30)arylsilyl with or without substituent(s), tri(C6-C30)arylsilyl with or without substituent(s), cyano or halogen;
- [66] R₂₂₀ through R₂₂₃ independently represent hydrogen, (C1-C30)alkyl with or without halogen substituent(s) or (C6-C30)aryl with or without (C1-C30)alkyl substituent(s);
- [67] R₂₂₄ and R₂₂₅ independently represent hydrogen, (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s) or halogen, or R₂₂₄ and R₂₂₅ may be linked via (C3-C12)alkylene or (C3-C12)alkenylene with or without a fused ring to form an alicylic ring or a mono or polycyclic aromatic ring;
- [68] R₂₂₆ represents (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or

without substituent(s), (C5-C30)heteroaryl with or without substituent(s) or halogen; R₂₂₇ through R₂₂₉ independently represent hydrogen, (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s) or halogen; and

[70] Q represents $R_{231}R_{232}$, $R_{233}R_{234}$ or $R_{237}R_{238}R_{241}$, wherein R_{231} through R_{242} $R_{239}R_{240}$

independently represent hydrogen, (C1-C30)alkyl with or without halogen substituent(s), (C1-C30)alkoxy, halogen, (C6-C30)aryl with or without substituent(s), cyano or (C5-C30)cycloalkyl with or without substituent(s), or each of them may be linked to an adjacent substituent via alkylene or alkenylene to form a spiro ring or a fused ring, or may be linked to R_{207} or R_{208} via alkylene or alkenylene to form a saturated or unsaturated fused ring.

[71]

[69]

- The meaning of the electroluminescent layer may be a single layer as a layer where the light is emitted or may be a multiple layer where two or more layers are laminated. In the configuration of the present invention, when hostdopant are used in mixture, it is confirmed that the luminous efficiency are remarkably improved by the electroluminescent host of the present invention. It may be configured at doping concentration of 0.5 to 10wt%. Compared to other host materials, the electroluminescent host of the present invention has superior conductivity with respect to the hole and electron and excellent stability in material, thereby showing a characteristic of remarkably increasing its life span as well as improving the luminous efficiency.
- [73] The M¹ is selected from a group consisting of Ir, Pt, Pd, Rh, Re, Os, Tl, Pb, Bi, In, Sn, Sb, Te, Au and Ag. The dopant compounds of Chemical Formula 2 are exemplified by the compounds described in Korean Patent Application No. 10-2008-0112855, but are not limited thereto.
- In the organic electronic device of the present invention, the organic layer may further include, in addition to the organic electroluminescent compound represented by Chemical Formula 1, one or more compound(s) selected from a group consisting of arylamine compounds and styrylarylamine compounds, at the same time. The arylamine compounds or styrylarylamine compounds are exemplified in Korean Patent Application No. 10-2008-0123276, 10-2008-0107606 or 10-2008-0118428, but are not limited thereto.
- [75] Further, in the organic electroluminescent device of the present invention, the organic layer may further include, in addition to the organic electroluminescent compound represented by Chemical Formula 1, one or more metal(s) selected from a group consisting of organic metals of Group 1, Group 2, 4th period and 5th period transition metals, lanthanide metals and dtransition elements or complex compound(s). The

organic layer may include an electroluminescent layer and a charge generating layer.

Further, the organic layer may include, in addition to the organic electroluminescent compound of Chemical Formula 1, one or more organic electroluminescent layer(s) emitting blue, green or red light at the same time in order to embody a whiteemitting organic electroluminescent device. The compound emitting blue, green or red light may be exemplified by the compounds described in Korean Patent Application No. 10-2008-0123276, 10-2008-0107606 or 10-2008-0118428, but are not limited thereto.

- In the organic electroluminescent device of the present invention, a layer (hereinafter referred to as "surface layer") selected from a chalcogenide layer, a metal halide layer and a metal oxide layer may be placed on the inner surface of one or both electrode(s) among the pair of electrodes. More specifically, a metal chalcogenide (including oxide) layer of silicon or aluminum may be placed on the anode surface of the electroluminescent medium layer, and a metal halide layer or metal oxide layer may be placed on the cathode surface of the electroluminescent medium layer. Operation stability may be attained therefrom.
- [78] The chalcogenide may be, for example, SiO_x ($1 \le x \le 2$), AlO_x ($1 \le x \le 1.5$), SiON, SiAlON, etc. The metal halide may be, for example, LiF, MgF_2 , CaF_2 , a rare earth metal fluoride, etc. The metal oxide may be, for example, Cs_2O , Li_2O , MgO, SrO, BaO, CaO, etc.
- [79] In the organic electroluminescent device according to the present invention, it is also preferable to arrange on at least one surface of the pair of electrodes thus manufactured a mixed region of an electron transport compound and a reductive dopant, or a mixed region of a hole transport compound and an oxidative dopant. In that case, since the electron transport compound is reduced to an anion, injection and transport of electrons from the mixed region to an electroluminescent medium are facilitated. In addition, since the hole transport compound is oxidized to a cation, injection and transport of holes from the mixed region to an electroluminescent medium are facilitated. Preferable oxidative dopants include various Lewis acids and acceptor compounds. Preferable reductive dopants include alkali metals, alkali metal compounds, alkaline earth metals, rareearth metals, and mixtures thereof.
- [80] Further, a white-emitting electroluminescent device having two or more electroluminescent layers may be manufactured by employing a reductive dopant layer as a charge generating layer.

Advantageous Effects of Invention

[81] Since the organic electroluminescent compound according to the present invention exhibits good luminous efficiency and excellent life property, it may be used to manufacture OLED devices having very superior operation life.

Mode for the Invention

[82] The present invention is further described with respect to organic electroluminescent compounds according to the present invention, processes for preparing the same, and luminescence properties of devices employing the same. However, the following examples are provided for illustrative purposes only and they are not intended to limit the scope of the present invention.

[83] [84]

[Preparation Example 1] Preparation of Compound 1

[86] Preparation of Compound 1-1

9H-carbazole (10 g, 41.10 mmol), 2-chloropyridine (5.60 g, 49.32 mmol), Pd(OAc)₂ (0.46 g), NaOt-bu (7.9 g, 82.20 mmol), toluene (100 mL), P(t-bu)₃ (2 mL, 4.11 mmol, 50% in toluene) were added and stirred under reflux. 10 hours later, the mixture was cooled to room temperature and distilled water was added. Extracting with EA and drying with MgSO₄, drying under reduce pressure was performed. Compound **1-1** (8.3 g, 33.98 mmol, 83%) was obtained via column separation.

[88] <u>Preparation of Compound 1-2</u>

1-neck flask was filled with Compound **1-1** (8.3 g, 33.98 mmol), formed a vacuum and was filled with argon. After tetrahydrofuran (500 mL) was added, the mixture was stirred at 0°C for 10 minutes. NBS (7.35 g, 40.78 mmol) was added thereto and stirred at room temperature for one day. Upon completion of the reaction, the product was extracted with distilled water and EA. After drying an organic layer with MgSO₄ and removing solvent by a rotary type evaporator, Compound **1-2** (8.5 g, 26.30 mmol, 77%) was obtained via column chromatography using hexan and EA as developing solvent.

[90] <u>Preparation of Compound 1-3</u>

The 1-neck flask was filled with Compound **1-2** (8.5 g, 26.30 mmol), formed a vacuum and was filled with argon. After tetrahydrofuran (500 mL) was added, the mixture was stirred at 78°C for 10 minutes. n-BuLi(2.5M in hexane) (15.8 mL, 39.45 mmol) was added dropwise and stirred at -78°C for 1 hour and a half. Trimethylborate (4.85 mL, 39.45 mmol) was added at -78°C. The mixture was stirred at -78°C for 30 minutes and stirred at room temperature for 4 hours. Upon completion of the reaction, the product was extracted with distilled water and EA. After drying an organic layer with MgSO₄ and removing solvent by a rotary type evaporator, Compound **1-3** (5.2 g,

18.05 mmol, 68.6%) was obtained via column chromatography using hexan and EA as developing solvent.

[92] Preparation of Compound 1

[93] Compound **1-3** (5.0 g, 17.4 mmol), 2-bromodibenzo[b,d]furan (5.2 g, 20.88 mmol), Pd(PPh₃)₄ (0.8 g, 0.7 mmol), 2M K₂CO₃ aqueous solution (20 mL), toluene (100 mL), and ethanol (50 mL) were added and stirred under reflux for 12 hours. After washing with distilled water, extracting with EA, and drying with MgSO₄, distillation under reduced pressure followed by column separation yielded Compound **1** (4.3 g, 10.48 mmol, 60 %).

[94] [Preparation Example 2] Preparation of Compound 49

[96] Preparation of Compound 2-1

[97] 2,4,6-trichloropyrimidine (10 g, 54.51 mmol), phenylboronic acid (16.6 g, 136,29 mmol), Pd(PPh₃)₄ (3.15 g, 2.72 mmol), 2M K₂CO₃ (50 mL), toluene (100 mL), and ethanol (30 mL) were added and stirred under reflux. 4 hours later, the mixture was cooled to room temperature and distilled water was added thereto. After extracting with EA and drying with MgSO₄, distillation under reduced pressure followed by column separation yielded Compound **2-1** (7 g, 26.24 mmol, 48.14%).

[98] Preparation of Compound 2-2

[99] NaH (1.57 g, 39.36 mmol, 60% in mineral oil) was mixed with DMF (70 mL) and Compound **2-1** (7 g, 26.24 mmol) was dissolved in DMF (60 mL). 1 hour later, Compound 9H-carbazole was dissolved in DMF (70 mL). The mixture was stirred for 10 hours. After adding distilled water, extracting with EA, and drying with MgSO₄, distillation under reduced pressure followed by column separation yielded Compound **2-2** (7 g, 14.78 mmol, 56.33%).

[100] Preparation of Compound 2-3

[101] Compound **2-3** (5.7 g, 11.97 mmol, 80.9%) was obtained by combining Compound **2-2** (7 g, 14.78 mmol) in Preparation Example 1 according to the same method as the preparation of Compound 1-2.

[102] <u>Preparation of Compound 2-4</u>

[103] Compound **2-4** (3.4 g, 7.70 mmol, 64.4%) was obtained by combining Compound **2-3** (5.7 g, 11.97 mmol) in Preparation Example 1 according to the same method as the preparation of Compound 1-3.

[104] Preparation of Compound **49**

[105] Compound **49** (3.2 g, 5.52 mmol, 72%) was obtained by using Compound **2-4** (3.4 g, 7.70 mmol)and 2-bromodibenzo[b,d]thiophene inPreparation Example 1 according to the same method as the preparation of Compound 1.

[106] [Preparation Example 3] Preparation of Compound 51

- [108] Preparation of Compound 3-1
- [109] Compound 3-1(13.2 g, 47.7 mmol, 87.5%) was obtained by combining 2,4,6-trichlorotriazine (10 g, 54.51 mmol) in Preparation Example 2 according to the same method as the preparation of Compound 2-1.
- [110] Preparation of Compound 3-2
- [111] Compound 3-2(14.5 g, 36.39 mmol, 76.3%) was obtained by combining Compound **3-1** (13.2 g, 47.7 mmol) in Preparation Example 2 according to the same method as the preparation of Compound 2-2.
- [112] Preparation of Compound 3-3
- [113] Compound **3-3** (14.6 g, 30.59 mmol, 84%) was obtained by combining Compound **3-2** (14.5 g, 36.39 mmol) in Preparation Example 2 according to the same method as the preparation of Compound 2-3.
- [114] Preparation of Compound 3-4
- [115] Compound **3-4** (7.2 g, 16.28 mmol, 53.2%) was obtained by combining Compound **3-3** (14.6 g, 30.59 mmol) in Preparation Example 2 according to the same method as the preparation of Compound 2-4.
- [116] Preparation of Compound 51
- [117] Compound **51** (5.1 g, 8.63 mmol, 53%) was obtained by using Compound **3-4** (7.2 g, 16.28 mmol) and 2-bromo-9,9-dimethyl-9H-fluorene in Preparation Example 2 according to the same method as the preparation of Compound 49.

[118] [Preparation Example 4] Preparation of Compound **62**

- [121] Preparation of Compound 4-1
- [122] 1,3-dibromobenzene (28 g, 0.119 mol) was dissolved in THF (600 mL) and n-BuLi (47.5 mL) was slowly added dropwise at -78°C. After reacting and stirring for 1 hour, Compound 3-1 (47.5 mL) was slowly added dropwise and slowly heated. The mixture was stirred at room temperature for 5 hours. Upon completion of the reaction, the product was extracted with EA and distilled water. Compound **4-1** (15.7 g, 40.43 mmol, 40.4%) was obtained via column separation.
- [123] Preparation of Compound 4-2
- [124] Compound **4-2** (12.5 g, 26.34 mmol, 65.2%) was obtained by combining Compound **4-1** (15.7 g, 40.43 mmol) in Preparation Example 1 according to the same method as the preparation of Compound 1-1.
- [125] Preparation of Compound **4-3**
- [126] Compound **4-3** (9.8 g, 17.71 mmol, 67.3%) was obtained by combining Compound **4-2** (12.5 g, 26.34 mmol) in Preparation Example 1 according to the same method as the preparation of Compound 1-2.
- [127] Preparation of Compound 4-4
- 9H-carbazole (70 g, 0.42 mmol), Iodobenzene (46 mL), cooper (40 g), potassium-carbonate (174 g), 18-crown-6 (9 g), and 1,2-Dichlorobenzene (2 L) were added and stirred under reflux for 12 hours. Upon completion of the reaction, the product was extracted with EA and dried with MgSO₄. Distillation under reduced pressure followed by column separation yielded Compound **4-4** (63.4 g, 260.58 mmol, 62%).

- [129] Preparation of Compound 4-5
- [130] Compound **4-5** (52.4 g, 162.63 mmol, 62.4%) was obtained by combining Compound **4-4** (63.4 g, 260.58 mmol) in Preparation Example 1 according to the same method as the preparation of Compound 1-2.
- [131] Preparation of Compound 4-6
- [132] Compound **4-6** (20.3 g, 70.70 mmol, 43%) was obtained by combining Compound **4-5** (52.4 g, 162.63 mmol) in Preparation Example 1 according to the same method as the preparation of Compound 1-3.
- [133] Preparation of Compound **62**
- [134] Compound **62** (5.7 g, 7.96 mmol, 50%) was obtained by using Compound **4-3** (9.8 g, 17.71 mmol) and Compound **4-6** in Preparation Example 1 according to the same method as the preparation of Compound 1.

[135]

- Organic electroluminescent Compounds 1 to 68 were prepared according to Preparation Examples 1 to 4 and Table 1 shows ¹H NMR and MS/FAB of the prepared organic electroluminescent compounds.
- [137] [Table 1]

[138]

Cmpd.	¹H NMR(CDCl3, 200 MHz)	M	S/FAB
ипра.	·	found	calculated
1	$\delta = 7.25(1H, m), 7.32~7.4(4H, m), 7.66~7.81(6H, m), 7.87~7.94(4H, m), 8.01(1H, m), 8.41(1H, m), 8.55(1H, m)$	410.47	410.14
4	$\delta = 7.25(1H, m), 7.32 \sim 7.41(4H, m), 7.51 \sim 7.52(5H, m), 7.66 \sim 7.81(6H, m), 7.87 \sim 7.94(3H, m), 8.4(1H, m), 8.47(1H, m), 8.55(1H, m)$	486.56	486.17
5	$ 8 = 7.11(1H, m), 7.25(1H, m), 7.32~7.41(11H, m), \\ 7.66~7.81(6H, m), 7.87~7.94(3H, m), 8.3(2H, m), \\ 8.55~8.6(2H, m) $	562.66	562.20
7	δ = 7.25(11I, m), 7.32~7.42(51I, m), 7.49~7.52(51I, m), 7.66~7.81(7H, m), 7.87~7.94(4H, m), 8.43(1H, s), 8.55(11I, m)	536.62	536.19
8	$ \begin{array}{llllllllllllllllllllllllllllllllllll$	616.87	616.29
9	δ = 7.11(1H, m), 7.25(1H, m), 7.32(1H, m), 7.33(1H, m), 7.37~7.46(22H, m), 7.64~7.81(7H, m), 7.87~7.94(3H, m), 8.27(1H, m), 8.4(1H, m), 8.55~8.6(2H, m)	821.05	820.29
10	δ = 7.14(1H, m), 7.25(1H, m), 7.32~7.38(5H, m), 7.66(1H, m), 7.69(1H, m), 7.7(1H, m), 7.71(1H, m), 7.72~7.81(8H, m), 8.53~8.59(3H, m), 9.3(1H, m), 9.92(1H, m)	564.63	564.20
11	8 = 7.14(1H, m), 7.25(2H, m), 7.32~7.41(4H, m), 7.51~7.52(4H, m), 7.66~7.81(7H, m), 7.87~7.94(3H, m), 8.53~8.55(2H, m), 9.3(1H, m), 9.41(1H, m)	563.65	563.20
12	$\delta = 7.25(1 \text{III, m}), 7.32 \sim 7.41(4 \text{II, m}), 7.51 \sim 7.52(4 \text{II, m}), 7.66 \sim 7.81(8 \text{II, m}), 7.87 \sim 8(4 \text{II, m}), 8.3(2 \text{II, m}), 8.44(1 \text{III, m}), 8.55 \sim 8.6(2 \text{II, m})$	562.66	562.20
13	$\delta = 7.25(1II, m), 7.32~7.41(11II, m), 7.66~7.81(8II, m), 7.87~7.94(3II, m), 8.2(2II, m), 8.3(4II, m), 8.55(1II, m)$	638.75	638.24
14	δ = 7.25(1H, m), 7.32~7.41(11H, m), 7.66~7.81(6H, m), 7.87~7.94(3H, m), 8.26~8.3(3H, m), 8.55~8.56(2H, m), 9.93(2H, m)	640.73	640.23
15	8 = 7.25(1H, m), 7.32~7.51(11H, m), 7.66~7.81(6H, m), 7.87~7.94(3H, m), 8.09(1H, m), 8.28(5H, m), 8.55(1H, m)	640.73	640.23
16	$\delta = 7.25(1H, m), 7.32~7.46(16H, m), 7.55(3H, m), 7.61(1H, m), 7.62~7.71(10H, m), 7.87~8(4H, m), 8.3(2H, m), 8.44(1H, m), 8.55~8.6(2H, m)$	821.05	820.29
17	$\delta = 1.72(6\text{II}, \text{ s}), 7(1\text{II}, \text{ m}), 7.17(1\text{II}, \text{ m}), 7.25~7.26(2\text{II}, \text{ m}), 7.32~7.38(4\text{II}, \text{ m}), 7.51(1\text{II}, \text{ m}), 7.66~7.81(6\text{II}, \text{ m}), 7.87~7.96(5\text{II}, \text{ m}), 8.07(1\text{II}, \text{ m}), 8.14(1\text{II}, \text{ m}), 8.5~8.55(2\text{II}, \text{ m})$	602.72	602.24
18	$\delta = 7.32 \sim 7.4(3H, m), 7.66 \sim 7.81(8H, m), 7.87 \sim 8.01(6H, m)$	460.52	460.16

m), 8.16(1II, m), 8.41(1II, m), 8.54(1II, m)			
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	(2H,	536.62	536.19
δ = 7.32~7.41(3II, m), 7.51~7.52(5II, m), 7.66~7.81 m), 7.87~7.96(4II, m), 8.16(1II, m), 8.4(1II, m), 8.47 m), 8.54(1II, m)	(Ш,	536.62	536.19
8 = 7.11(1H, m), 7.32~7.54(10H, m), 7.66~7.81(8H, 7.87~7.96(4H, m), 8.16(1H, m), 8.3(2H, m), 8.54(1H, 8.6(1H, m)		612.72	612.22
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	т), т),	764.91	764.28
δ = 7.32~7.46(15H, m), 7.55(3H, m), 7.61(1H, 7.62(1H, m), 7.66~7.71(11H, m), 7.87~8(5H, m), 8.16 m), 8.3(2H, m), 8.44(1H, m), 8.54(1H, m), 8.6(1H, m)		871.11	870.31
8 = 7.32~7.54(10H, m), 7.66~7.81(8H, m), 7.87~7.96 32 m), 8.16(1H, m), 8.26~8.3(3H, m), 8.54~8.56(2H, 9.93(2H, m)	m),	690.79	690.24
8 = 1.72(6H, s), 7(1H, m), 7.17(1H, m), 7.26(1H, 7.32~7.38(3H, m), 7.51(1H, m), 7.66~7.81(8H, 7.87~7.96(6H, m), 8.07(1H, m), 8.14~8.16(2H, 8.5~8.54(2H, m)	m), m), m),	652.78	652.25
34 $\delta = 7.32 \sim 7.51(10\text{II}, \text{ m}), 7.66 \sim 7.81(8\text{II}, \text{ m}), 7.87 \sim 7.96$ m), $8.09(1\text{II}, \text{ m}), 8.16(1\text{II}, \text{ m}), 8.28(5\text{II}, \text{ m}), 8.54(1\text{II}, \text{ m})$		690.79	690.24
35 $\delta = 7.41 \sim 7.54(18\text{II}, \text{ m}), 7.69 \sim 7.81(11\text{II}, \text{ m}), 7.87(11\text{II}, \text{ m}), 8.18 \sim 8.2(3\text{II}, \text{ m}), 8.3(4\text{II}, \text{ m})$		790.95	790.30
δ = 7.11(1H, m), 7.22~7.25(5H, m), 7.32~7.41(4H, 7.51~7.52(8H, m), 7.66~7.81(5H, m), 7.88~7.89(3H, 7.97~8(2H, m), 8.18(1H, m), 8.43(1H, m), 8.6(1H, 8.81(2H, m))	m), m),	715.84	715.26
δ = 7.11(111, m), 7.32~7.54(1111, m), 7.66~7.81(511, 7.89(111, m), 8(111, m), 8.18(111, m), 8.3(211, m), 8.43 m), 8.6(111, m), 9.34(111, m)		563.65	563.20
δ = 7.11(1Π, m), 7.32~7.41(1Π, m), 7.66~7.81(5Π, 7.89(1Η, m), 8(1Η, m), 8.18(1Η, m), 8.3(2Η, m), 8.43 m), 8.51(1Η, m), 8.6(1Η, m)		563.65	563.20
δ = 7.11(1H, m), 7.32(1H, m), 7.38~7.47(11H, 39 7.66~7.81(6H, m), 7.89~7.92(2H, m), 8(1H, m), 8.18 m), 8.3(2H, m), 8.6(1H, m), 8.91(1H, m)		613.70	613.22
δ = 7.11(1H, m), 7.32~7.54(10H, m), 7.66~7.81(6H, 40 7.89(Ш, m), 8(Ш, m), 8.18(Ш, m), 8.3(2H, m), 8.75(1H, m), 9.39(2H, m)		614.69	614.21
41 $\delta = 7.11(111, m), 7.25~7.33(411, m), 7.41~7.54(1011, m)$	m),	779.88	779.27

	7.63(1H, m), 7.66~7.71(8H, m), 7.87(1H, m), 7.94(1H, m), 8.12(1H, m), 8.3(2H, m), 8.55~8.6(2H, m), 8.74(2H, m)		
42	$\delta = 7.25(1H, m), 7.32~7.41(5H, m), 7.51(4H, m), 7.66~7.81(10H, m), 7.87~7.94(3H, m), 8.55(1H, m), 8.63(1H, s), (H,)$	563.65	563.20
43	$ \delta = 7.25(1 \text{H}, \text{m}), 7.32 \sim 7.41(5 \text{H}, \text{m}), 7.51(4 \text{H}, \text{m}), \\ 7.66 \sim 7.81(6 \text{H}, \text{m}), 7.87 \sim 7.94(3 \text{H}, \text{m}), 8.28(4 \text{H}, \text{m}), \\ 8.55(1 \text{H}, \text{m}) $	564.63	564.20
44	$ \delta = 7.25(11I, m), 7.32(21I, s), 7.32~7.41(41I, m), 7.51(41I, m), 7.66~7.81(81I, m), 7.87~7.94(31I, m), 8.28(21I, m), 8.55(11I, m) $	563.65	563.20
45	$ \delta = 1.72(12\text{H}, \text{ s}), 7.25~7.38(8\text{H}, \text{ m}), 7.55(2\text{H}, \text{ m}), 7.63~7.81(10\text{H}, \text{ m}), 7.87~7.94(7\text{H}, \text{ m}), 8.55(1\text{H}, \text{ m}), 8.63(1\text{H}, \text{s}), (1\text{H},) $	795.97	795.32
46	δ = 7.25(1H, m), 7.33(1H, m), 7.41(2H, m), 7.5-7.52(6H, m), 7.69(1H, m), 7.77(1H, m), 7.86~7.87(2H, m), 7.94~8(4H, m), 8.28(4H, m), 8.45(1H, m), 8.55(1H, m)	580.70	580.17
47	8 = 7.25(1H, m), 7.33(1H, m), 7.41(2H, m), 7.5~7.52(10H, m), 7.69(1H, m), 7.77(1H, m), 7.85~7.87(6H, m), 7.94~8(4H, m), 8.3(4H, m), 8.45(1H, m), 8.55(1H, m), 8.63(1H, s), (H,)	731.90	731.24
48	$ \begin{array}{llllllllllllllllllllllllllllllllllll$	578.72	578.18
49	δ = 7.25(1II, m), 7.33(1II, m), 7.41(2II, m), 7.5~7.52(6II, m), 7.69(1H, m), 7.77~7.79(5H, m), 7.86~7.87(2H, m), 7.94~8(4H, m), 8.45(1H, m), 8.55(1H, m), 8.63(1H, s), (H,)	579.71	579.18
50	$ \delta = 1.72(6H, s), 7.25~7.41(6H, m), 7.51~7.55(6H, m), 7.61(1H, m), 7.69(1H, m), 7.77~7.79(5H, m), 7.87(2H, m), 7.94(1H, m), 8.06(1H, m), 8.55(1H, m), 8.63(1H, s), (H,) $	589.73	589.25
51	$ \begin{array}{llllllllllllllllllllllllllllllllllll$	590.71	590.25
52	$\delta = 1.72(6\text{II}, \text{ s}), 7.11(1\text{II}, \text{ m}), 7.25(1\text{II}, \text{ m}), 7.28(1\text{II}, \text{ m}), 7.33(1\text{II}, \text{ m}), 7.38~7.51(11\text{II}, \text{ m}), 7.61(1\text{II}, \text{ m}), 7.69(1\text{II}, \text{ m}), 7.77(1\text{II}, \text{ m}), 7.87(2\text{II}, \text{ m}), 7.94(1\text{II}, \text{ m}), 8.06(1\text{II}, \text{ m}), 8.3(2\text{H}, \text{ m}), 8.55~8.6(2\text{H}, \text{ m})$	588.74	588.26
53	$\delta = 1.72(6\text{H, s}), 7.25 \sim 7.41(6\text{H, m}), 7.48 \sim 7.61(15\text{H, m}), 7.69 \sim 7.77(6\text{H, m}), 7.87(2\text{H, m}), 7.94(1\text{H, m}), 8.06(1\text{H, m}), 8.55(1\text{H, m}), 8.63(1\text{H, s}), (\text{H, })$	741.92	741.31
54	$\delta = 0.66(6H, s), 7.25(1H, m), 7.33(2H, m), 7.41(2H, m),$	605.80	605.23

	E = 1 E = 0 (EH		
	7.51~7.52(5II, m), 7.58~7.61(2II, m), 7.69(1II, m),		
<u> </u>	7.77~7.94(10H, m), 8.55(1H, m), 8.63(1H, s), (H,) 8 = 1.3(4H, m), 1.45(4H, m), 7.25(1H, m), 7.33(2H, m),		
55	7.41(2H, m), 7.51~7.52(5H, m), 7.58~7.61(2H, m),	632.83	632.24
33	7.69(III, m), 7.77~7.94(6II, m), 8.28(4II, m), 8.55(III, m)	052.65	0.02.24
	$\delta = 7.25(\text{III}, \text{ m}), 7.33 \sim 7.42(\text{6II}, \text{ m}), 7.48 \sim 7.51(\text{5II}, \text{ m}),$		
56	7.69~7.87(12H, m), 7.94(1H, m), 8.03~8.12(3H, m),	655.72	655.22
30	8.55(III, m), 8.63(III, s), (II,)	000.72	000.22
-	$\delta = 7.25(1H, m), 7.33 \sim 7.55(26H, m), 7.69(1H, m),$		
57	7.72~7.79(13H, m), 7.94(1H, m), 8.55(1H, m), 8.63(1H,	893.95	893.34
()1	s), (H,)	0.001.00	0.50.101
	$\delta = 7.25(\text{III}, \text{m}), 7.33(\text{III}, \text{m}), 7.41 \sim 7.51(\text{1III}, \text{m}),$		
58	7.69~7.81(11H, m), 7.87~7.88(3H, m), 7.94(1H, m),	635.56	635.25
00	8.55(1H, m), 8.63(1H, s), (H,)	000.00	(0.0.20)
	$\delta = 7.25(\text{III}, \text{ m}), 7.33(\text{III}, \text{ m}), 7.41~7.51(\text{1III}, \text{ m}),$		
59	7.69(1H, m), 7.75(1H, m), 7.77~7.87(13H, m), 8.55(1H,	671.72	671.21
	m), 8.63(1H, s), (H,)		
	$\delta = 7.25 - 7.33(3H, m), 7.41 - 7.51(10H, m), 7.58 - 7.63(3H, m)$		
	m), 7.69(III, m), 7.77~7.79(6II, m), 7.87(III, m),	200 =2	
60	7.94~8(2H, m), 8.12(1H, m), 8.18(1H, m), 8.55(1H, m),	638.76	638.25
	8.63(III, s), (II,)		
	$\delta = 7.25(2II, m), 7.33(2II, m), 7.41~7.51(9II, m),$		
61	7.58(2H, m), 7.69(2H, m), 7.77(2H, m), 7.87(2H, m),	639.75	639.24
	7.94(2H, m), 8.28(4H, m), 8.55(2H, m)		
	$\delta = 7.25(2H, m), 7.33(2H, m), 7.41~7.51(11H, m),$		
62	7.58(2II, m), 7.69(2II, m), 7.77(2II, m), 7.87(2II, m),	715.84	715.27
	7.94(2H, m), 8.09(1H, m), 8.28(5H, m), 8.55(2H, m)		
	$\delta = 7.25(1H, m), 7.33~7.51(11H, m), 7.58(2H, m),$		
63	7.69(III, m), 7.77(2II, m), 7.87(III, m), 7.94~8(2II, m),	640.73	640.24
	8.18(III, m), 8.28(4II, m), 8.43(III, m), 8.51~8.55(2II, m)		
	$\delta = 7.25(1H, m), 7.33(1H, m), 7.41~7.51(11H, m),$		
64	7.58(2H, m), 7.69(1H, m), 7.76~7.77(3H, m), 7.87~8(4H,	690.79	690.25
	m), 8.18(111, m), 8.28(411, m), 8.55(111, m), 8.91(111, m)		
	$\delta = 7.25(1H, m), 7.33(1H, m), 7.41~7.56(8H, m),$		
65	7.69(III, m), 7.77~7.79(3II, m), 7.86~7.87(2II, m),	579.71	579.18
	7.94~8(4H, m), 8.09(1H, m), 8.28(1H, m), 8.45(1H, m),		
	8.54~8.55(2H, m) 8 = 7.95.7.99(9H, m) 7.41(9H, m) 7.5.7.51(5H, m)		
	8 = 7.25 - 7.33(3H, m), 7.41(2H, m), 7.5 - 7.51(5H, m),		
66	7.63(III, m), 7.69(III, m), 7.77~7.79(6II, m), 7.87(III, m),	643.79	643.28
	7.94~8(2H, m), 8.12(1H, m), 8.18(1H, m), 8.55(1H, m),		
	8.63(1H, s), (H,) $\delta = 7.25 \sim 7.33(3II, m)$, $7.41(4II, m)$, $7.5 \sim 7.51(9II, m)$,		
67	$0 = 7.25 \times 7.33(311, m), 7.41(411, m), 7.5 \times 7.51(911, m), 7.63(111, m), 7.69(111, m), 7.77 \times 7.79(611, m), 7.87(111, m), 7.63(111, m), 7.87(111, m), 7.71 \times 1.01 \times 1.01$	702 01	702 20
67		793.91	793.30
	$7.94 \sim 8(2 \text{II}, \text{ m}), 8.12(1 \text{II}, \text{ m}), 8.18(1 \text{II}, \text{ m}), 8.28(4 \text{II}, \text{ m}),$		

[142]		8.55(1H, m), 8.63(1H, s), (H,)	
	68	$ \delta = 7.25(1\text{H}, \text{ m}), \ 7.33\text{-}7.42(26\text{H}, \text{ m}), \ 7.61\text{-}7.69(7\text{H}, \text{ m}), \ 7.76\text{-}7.77(3\text{H}, \text{ m}), \ 7.87(1\text{H}, \text{ m}), \ 7.94\text{-}8(2\text{H}, \text{ m}), \ 8.09(1\text{H}, \text{ m}), \ 8.16\text{-}8.18(2\text{H}, \text{ m}), \ 8.28(4\text{H}, \text{ m}), \ 8.54\text{-}8.55(2\text{H}, \text{ m}) $	1023.38

[143] [Example 1] Manufacture of OLED device using the organic electroluminescent

compound according to the present invention

[144] An OLED device was manufactured using the electroluminescent material according to the present invention. First, a transparent electrode ITO thin film (15 Ω/\Box) obtained from a glass for OLED (produced by Samsung Corning) was subjected to ultrasonic washing with trichloroethylene, acetone, ethanol and distilled water, sequentially, and stored in isopropanol before use.

- Then, an ITO substrate was equipped in a substrate folder of a vacuum vapor deposition apparatus, and 4,4',4"-tris(N,N-(2-naphthyl)-phenylamino)triphenylamine (2-TNATA) was placed in a cell of the vacuum vapor deposition apparatus, which was then ventilated up to 10-6 torr of vacuum in the chamber. Then, electric current was applied to the cell to evaporate 2-TNATA, thereby forming a hole injection layer having a thickness of 60 nm on the ITO substrate.
- Then, N,N'-bis(α -naphthyl)-N,N'-diphenyl-4,4'-diamine (NPB) was placed in another cell of the vacuum vapor deposition apparatus, and electric current was applied to the cell to evaporate NPB, thereby forming a hole transport layer having a thickness of 20 nm on the hole injection layer.
- [147] After forming the hole injection layer and the hole transport layer, an electroluminescent layer was formed thereon as follows. Compound **49** was placed in a cell of a vacuum vapor deposition apparatus as host, and Ir(ppy)₃[tris(2-phenylpyridine)iridium] was placed in another cell as a dopant. The two materials were evaporated at different rates such that an electroluminescent layer having a thickness of 30 nm was vapordeposited on the hole transport layer at 4 to 10 wt%.
- Subsequently, tris(8-hydroxyquinoline)-aluminum(III) (Alq) was vapor-deposited with a thickness of 20 nm as an electron transport layer. Then, after vapor-depositing lithium quinolate (Liq) of a following structure with a thickness of 1 to 2 nm as an electron injection layer, an Al cathode having a thickness of 150 nm was formed using another vacuum vapor deposition apparatus to manufacture an OLED. Each compound used in the OLED was purified by vacuum sublimation at 10-6 torr.
- [149] [Example 2] Manufacture of OLED device using the organic electroluminescent compounds of the present invention
- [150] An OLED was manufactured as in Example 1 except that Compound **23** according to the present invention is used as host material on the electroluminescent layer and an organic iridium complex (piq)₂

 Ir(acac)[bis-(1-phenylisoquinolyl)iridium(III)acetylacetonate] is used as electroluminescent dopant.
- [151] [Comparative Examples 1 and 2] Electroluminescent properties of the OLED device using conventional electroluminescent material
- [152] An OLED device was manufactured in the same manner as Examples 1 and 3 except

that 4,4'-Bis(carbazol-9-yl)-biphenyl (CBP) instead of the compounds of the present invention was used as host material in a cell of the vacuum vapor deposition apparatus.

[153]

[154] Luminous efficiency of the OLED devices including the organic electroluminescent compound according to the present invention manufactured in Examples 1 to 2 and Comparative Examples 1 and 2 and the conventional electroluminescent compound was measured at 1,000 cd/m². The result is given in Table 2.

[155] [Table 2]

[156]

No.		Host	Dopant	Hole blocking layer	Driving voltage(V) @1,000 cd/m ²	Power efficiency(cd/A) @1,000 cd/m²	Color
	6	49	Ir(ppy)₃	-	6.7	30.1	Green
	7	59	Ir(ppy)₃	-	6.5	28.1	Green
Example 1	8	60	Ir(ppy)₃	1	6.7	30.3	Green
	9	62	Ir(ppy)₃	i	6.4	28.3	Green
	10	67	Ir(ppy)₃	-	6.3	29.0	Green
	16	23	(piq) ₂ Ir(acac)	-	6.2	7.1	Red
E1-	17	30	(piq) ₂ Ir(acac)	-	6.1	7.6	Red
Example 2	18	34	(piq) ₂ Ir(acac)	-	6.0	7.8	Red
	19	53	(piq) ₂ Ir(acac)	-	6.3	7.6	Red
	20	55	(piq) ₂ lr(acac)	1	6.4	7.3	Red
Comparati Example		CBP	Ir(ppy)₃	ВА1 а	7.5	25.1	Green
	Comparative Example 2		(piq) ₂ Ir(acac)	BAIq	7.5	6.5	Red

[157] As shown in Table 2, the organic electroluminescent compounds according to the present invention have excellent properties compared with the conventional material. In addition, the device using the organic electroluminescent compound according to the present invention as host material for emitting red or green light has excellent electroluminescent properties and drops driving voltage, thereby increasing power efficiency and improving power consumption.

Claims

[Claim 1]

An organic electroluminescent compound represented by Chemical Formula 1:

[Chemical Formula 1]

$$A_{18}$$
 A_{19}
 A_{15}
 A_{19}
 A_{19}
 A_{15}
 A_{19}
 A

wherein

A₁ through A₁₉ independently represent CR₁ or N, X represents -(CR₂R₃ $)_{1}$, $-N(R_4)$ -, -S-, -O-, $-Si(R_5)(R_6)$ -, $-P(R_7)$ -, $-P(=O)(R_8)$ - or $-B(R_9)$ -, and Ar₁ represents (C6-C40) arylene with or without substituent(s) or (C3-C40)heteroarylene with or without substituent(s), except for the case where m is 0, and A_{15} through A_{19} are CR_1 at the same time; R₁ through R₉ independently represent hydrogen, deuterium, halogen, (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s), substituted or unsubstituted (C6-C30)aryl fused with one or more (C3-C30)cycloalkyl(s) with or without substituent(s), (C3-C30)heteroaryl with or without substituent(s), 5- to 7-membered heterocycloalkyl with or without substituent(s), 5- to 7-membered heterocycloalkyl fused with one or more aromatic ring(s) with or without substituent(s),(C3-C30)cycloalkyl with or without substituent(s), (C3-C30)cycloalkyl fused with one or more aromatic ring(s) with or without substituent(s), cyano, trifluoromethyl, NR₂₁R₂₂, BR₂₃R₂₄, PR₂₅R $_{26}$, $P(=O)R_{27}R_{28}$, $R^aR^bR^cSi$, R^dY , $R^eC(=O)$, $R^fC(=O)O$, (C6-C30)ar(C1-C30)alkyl with or without substituent(s), (C2-C30)alkenyl with or without substituent(s), (C2-C30)alkynyl with or without substituent(s), carboxyl, nitro,

$$R_{62}$$
, or hydroxyl, or each of them may be linked to an N_{61}

adjacent substituent via (C3-C30)alkylene or (C3-C30)alkenylene with or without a fused ring to form an alicylic ring, a mono or polycyclic aromatic ring or a mono or polycyclic heteroaromatic ring; W represents $-(CR_{51}R_{52})_n$, $-(R_{51})C=C(R_{52})$, $-N(R_{53})$, -S, -S, -O, $-Si(R_{54})$ (R_{55}) -, $-P(R_{56})$ -, $-P(=O)(R_{57})$ -, -C(=O)- or $-B(R_{58})$ -, and R_{51} through R_{58} and R_{61} through R_{63} are the same as R_1 through R_9 ; the heterocycloalkyl or heteroaryl may contain one or more heteroatom(s) selected from B, N, O, S, P(=O), Si and P; R₂₁ through R₂₈ independently represent (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s) or (C3-C30)heteroaryl with or without substituent(s), Ra, Rb, and Rc independently represent (C1-C30)alkyl with or without substituent(s) or (C6-C30)aryl with or without substituent(s), Y represents S or O, R^d represent (C1-C30)alkyl with or without substituent(s) or (C6-C30)aryl with or without substituent(s), Re represent (C1-C30)alkyl with or without substituent(s), (C1-C30)alkoxy with or without substituent(s), (C6-C30) aryl with or without substituent(s) or (C6-C30) aryloxy with or without substituent(s), Rf represent (C1-C30)alkyl with or without substituent(s), (C1-C30)alkoxy with or without substituent(s), (C6-C30)aryl with or without substituent(s) or (C6-C30)aryloxy with or without substituent(s);

m represents an integer 0 to 2; and 1 and n represent an integer 1 or 2.

[Claim 2]

The organic electroluminescent compound according to claim 1, wherein the substituent of R_1 through R_9 , R_{21} through R_{28} , R_{51} through R_{58} and R_{61} through R_{63} is further substituted by one or more substituent(s) selected from a group consisting of deuterium, halogen, (C1-C30)alkyl with or without halogen substituent(s), (C6-C30)aryl, (C3-C30)heteroaryl with or without (C6-C30)aryl substituent(s), 5- to 7-membered heterocycloalkyl, 5- to 7-membered heterocycloalkyl fused with one or more aromatic ring(s), (C3-C30)cycloalkyl, (C3-C30)cycloalkyl fused with one or more aromatic ring(s), tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl, adamantyl, (C7-C30)bicycloalkyl, (C2-C30)alkenyl, (C2-C30)alkynyl, cyano, carbazolyl, NR₃₁R₃₂, BR₃₃R ₃₄, PR₃₅R₃₆, P(=O)R₃₇R₃₈, (C6-C30)arylc1-C30)alkyl, (C1-C30)alkyl(C6-C30)arylc2-C30)alkylthio, (C6-C30)aryloxy, (C6-C30)arylthio, (C1-C30)alkoxycarbonyl,

 $(C1\hbox{-}C30) alkyl carbonyl, (C6\hbox{-}C30) aryl carbonyl,\\$

(C6-C30)aryloxycarbonyl, (C1-C30)alkoxycarbonyloxy,

(C1-C30)alkylcarbonyloxy, (C6-C30)arylcarbonyloxy,

(C6-C30)aryloxycarbonyloxy, carboxyl, nitro and hydroxyl, or is linked to an adjacent substituent to form a ring, wherein R_{31} through R_{38} independently represent (C1-C30)alkyl, (C6-C30)aryl or (C3-C30)heteroaryl.

[Claim 3]

The organic electroluminescent compound according to claim 1, wherein R_1 through R_9 are selected from the following structures:

wherein

R₇₁ through R₁₃₈ independently represent hydrogen, deuterium, halogen, (C1-C30)alkyl, (C6-C30)aryl, (C6-C30)aryl fused with one or more (C3-C30)cycloalkyl(s), (C3-C30)heteroaryl, 5- to 7-membered heterocycloalkyl, 5- to 7-membered heterocycloalkyl fused with one or more aromatic ring(s),(C3-C30)cycloalkyl, (C3-C30)cycloalkyl fused with one or more aromatic ring(s), cyano, amino, (C1-C30)alkylamino, (C6-C30)arylamino, NR₄₁R₄₂, BR₄₃R₄₄, PR₄₅R₄₆, P(=O)R₄₇R₄₈, tri(C1-C30)alkylsilyl, di(C1-C30)alkyl(C6-C30)arylsilyl, tri(C6-C30)arylsilyl, (C6-C30)ar(C1-C30)alkyl, (C1-C30)alkyloxy, (C1-C30)alkylthio, (C6-C30)aryloxy, (C6-C30)arylthio, (C1-C30)alkoxycarbonyl, (C1-C30)alkylcarbonyl, (C2-C30)alkynyl, (C6-C30)aryloxycarbonyl, (C1-C30)alkoxycarbonyloxy, (C1-C30)alkoxycarbonyloxy, (C1-C30)alkylcarbonyloxy, (C1-C30)alkylcarbonyloxy, (C1-C30)alkylcarbonyloxy, (C1-C30)alkylcarbonyloxy, (C1-C30)alkylcarbonyloxy, (C1-C30)alkylcarbonyloxy,

(C6-C30)aryloxycarbonyloxy, carboxyl, nitro or hydroxyl, or each of them may be linked to an adjacent substituent via (C3-C30)alkylene or (C3-C30)alkenylene with or without a fused ring to form an alicylic ring or a mono or polycyclic aromatic ring, wherein R_{41} through R_{48} independently represent (C1-C30)alkyl, (C6-C30)aryl or (C3-C30)heteroaryl.

[Claim 4]

The organic electroluminescent compound according to claim 1, wherein $*-(Ar_1)_m$ is selected from the following structures:

[Claim 5]

The organic electroluminescent compound according to claim 1, which is selected from the following structures:

[Claim 6]

An organic electroluminescent device comprising the organic electroluminescent compound according any of claims 1 to 5.

[Claim 7]

The organic electroluminescent device according to claim 6, which comprises a first electrode; a second electrode; and one or more organic layer(s) interposed between the first electrode and the second electrode, wherein the organic layer comprises one or more organic electroluminescent compound(s) according to any of claims 1 to 5 and one or

more dopant(s) represented by Chemical Formula 2:

[Chemical Formula 2]

 $M^{1}L^{101}L^{102}L^{103}$

wherein

M¹ is a metal selected from a group consisting of Group 7, Group 8, Group 9, Group 10, Group 11, Group 13, Group 14, Group 15 and Group 16 metals, and ligand L¹⁰¹, L¹⁰² and L¹⁰³ are independently selected from the following structures:

wherein

 R_{201} through R_{203} independently represent hydrogen, (C1-C30)alkyl with or without halogen substituent(s), (C6-C30)aryl with or without (C1-C30)alkyl substituent(s) or halogen;

 R_{204} through R_{219} independently represent hydrogen, (C1-C30)alkyl with or without substituent(s), (C1-C30)alkoxy with or without substituent(s), (C3-C30)cycloalkyl with or without substituent(s), (C2-C30)alkenyl with or without substituent(s), (C6-C30)aryl with or without substituent(s), mono- or di-(C1-C30)alkylamino with or without substituent(s), mono- or di-(C6-C30)arylamino with or without

substituent(s), SF_5 , tri(C1-C30)alkylsilyl with or without substituent(s), di(C1-C30)alkyl(C6-C30)arylsilyl with or without substituent(s), tri(C6-C30)arylsilyl with or without substituent(s), cyano or halogen; R_{220} through R_{223} independently represent hydrogen, (C1-C30)alkyl with or without halogen substituent(s) or (C6-C30)aryl with or without (C1-C30)alkyl substituent(s);

 R_{224} and R_{225} independently represent hydrogen, (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s) or halogen, or R_{224} and R_{225} may be linked via (C3-C12)alkylene or (C3-C12)alkenylene with or without a fused ring to form an alicylic ring or a mono or polycyclic aromatic ring;

R₂₂₆ represents (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s), (C5-C30)heteroaryl with or without substituent(s) or halogen;

 R_{227} through R_{229} independently represent hydrogen, (C1-C30)alkyl with or without substituent(s), (C6-C30)aryl with or without substituent(s) or halogen; and

$$Q \text{ represents } \underset{R_{231}}{R_{232}}, \underset{R_{233}}{R_{233}} \underset{R_{234}}{R_{234}} \text{ or } \underset{R_{239}}{R_{238}} \underset{R_{240}}{R_{242}}, \text{ wherein } R_{231}$$

through R_{242} independently represent hydrogen, (C1-C30)alkyl with or without halogen substituent(s), (C1-C30)alkoxy, halogen, (C6-C30)aryl with or without substituent(s), cyano or (C5-C30)cycloalkyl with or without substituent(s), or each of them may be linked to an adjacent substituent via alkylene or alkenylene to form a spiro ring or a fused ring, or may be linked to R_{207} or R_{208} via alkylene or alkenylene to form a saturated or unsaturated fused ring.

[Claim 8]

The organic electroluminescent device according to claim 7, wherein the organic layer further comprises one or more amine compound(s) selected from a group consisting of arylamine compounds and styry-larylamine compounds, or one or more metal(s) selected from a group consisting of organic metals of Group 1, Group 2, 4th period and 5th period transition metals, lanthanide metals and dtransition elements or complex compound(s).

[Claim 9]

The organic electroluminescent device according to claim 7, wherein the organic layer comprises an electroluminescent layer and a charge generating layer.

[Claim 10]

The organic electroluminescent device according to claim 7, which is a

white lightemitting organic electroluminescent device wherein the organic layer comprises one or more organic compound layer(s) emitting red, green or blue light at the same time.

INTERNATIONAL SEARCH REPORT

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A.		CLASSIFICATION OF SUBJECT MATTER
	r	Int. Cl.

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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, Chemical Abstracts- accessed via STN (substructure search covering full scope of all claims)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
х	US 2009/0085479 A1 (USHIKUBO, T.) 2 April 2009 See [0100]], page 7; [0111], column 2, page 8; [0128]-[0130], pages 10 and 11; light-emitting elements 4-6, [0199]-[0230], pages 16-18; the product of scheme B-1, page 25; and PCCPA, structural formula 2, [0274], page 24, and the product of scheme B-2, page 25	1-4 & 6-10
X	WO 2008/090912 A1 (KONICA MINOLTA HOLDINGS, INC.) 31 July 2008 See abstract; OC-31 and OC-32, page 24; and OC-33 and OC-35, page 25; and PD-1 and PD-6, page 32.	1-3 & 6-10
x	JP 11-329737 A (TAIHO IND. CO. LTD. ET AL.) 30 November 1999 & CAPLUS Accession Number 1999:756830 See CAPLUS abstract; and formula 39, page 20, and structures page 21. JP 11-144867 A (TORAY IND. INC.) 28 May 1999	1-4 & 6
x	& machine translation from http://www19.ipdl.inpit.go.jp/PA1/cgi-bin/PA1INDEX See abstract; working example, [0031]-[0035], chemical formula 7, HTL1 and chemical formula 9, HTL3; and structures pages 7 and 8	1-3 & 6

	o, in bit detailed pages , and o					
	X Further documents are listed in the co	ntinuati	on of Box C X 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"	"E" earlier application or patent but published on or after the "X" document of particular relevance; the claimed invention cannot be considered international filing date or cannot be considered to involve an inventive step when the document is alone					
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	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					
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/KR2010/005092

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	ZHANG, K. et al., "Synthesis and Properties of Carbazole Main Chain Copolymers with Oxadiazole Pendant toward Bipolar Polymer Host: Tuning the HOMO/LUMO Level and Triplet Energy", Chemistry of Materials, 2008, vol 20(23), pages 7324-7331. See P1, scheme 1, page 7327	Junii 11U.
A	KR 10-2010-0079458 A (DUKSAN HIGH METAL CO., LTD.) 08 July 2010 & STN File CAPLUS Accession Number 2010:870481	
P,X	See abstract; examples 106 and 111, page 21, and example 2 on page 24.	1-3 & 6
P,X	JP 2010-114180 A (KONICA MINOLTA HOLDINGS, INC., JAPAN) 20 May 2010 & STN File CAPLUS Accession Number 2010:628012 See abstract; and PH-11 and PH-12, page 14; and PH-21, page 15.	1,3&6
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR2010/005092

This Annex lists the known "A" publication level 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.

	t Document Cited in Search Report	Patent Family Member					
US	2009085479	CN	101399315	JР	2009099966	KR	20090033083
WQ	2008090912	NONE					
JР	11329737	NONE					_
JP	11144867	NONE		•			
KR	102010079458	NONE	,				
JР	2010114180	NONE			·		

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

END OF ANNEX