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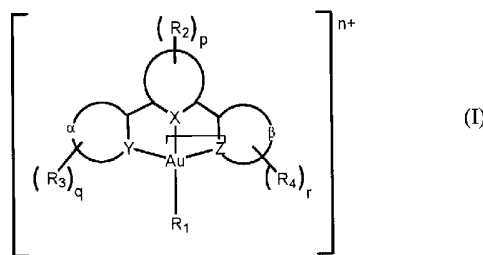
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[Continued on next page]

(54) Title: LUMINESCENT GOLD(III) COMPOUNDS, THEIR PREPARATION, AND LIGHT-EMITTING DEVICES CONTAINING SAME



(I)

(57) Abstract: A class of luminescent gold(III) compounds with a tridentate ligand and at least one strong σ -donating group having the chemical structure represented by the general formula (I): wherein R_1 - R_4 each independently represent the group containing hydrogen, halogen, alkynyl, substituted alkynyl, alkyl, substituted alkyl, aryl, substituted aryl, heteroaryl, substituted heteroaryl, alkoxy, substituted alkoxy, amino, substituted amino, cyano, nitro, alkylcarbonyl, alkoxy carbonyl, arylcarbonyl, aryloxy carbonyl, mono- or dialkylaminocarbonyl, alkylcarbonyloxy, arylcarbonyloxy, aryloxy, alkoxy carbonyl, aryloxy carbonyloxy group, and the like; X, Y and Z each independently represent a heteroatom or a carbon; C-X-C represents an aromatic or heterocyclic 5- or 6-membered ring; α and β each independently represent a bridge for an aromatic or heterocyclic 5- or 6-membered ring or represent a break for non-cyclic moiety; C-X, C-Y and C-Z each independently represent a single bond or double bond; n represents a zero or an integer; p, q and r represent positive integers. These novel compounds are synthesized and found to exhibit photoluminescence and electroluminescence. Some of them are thermally stable for vacuum deposition and can be used as new light-emitting material, both in the role of emitter and dopant, of organic light-emitting devices (OLEDs) to give high brightness and efficiency. Organic light-emitting devices (OLEDs) employing such luminescent compounds fabricated by vacuum deposition, spin coating or other device fabrication methods are described. The color of the electroluminescence changes upon variation of applied DC voltage, and the electroluminescence wavelength shifts to the red upon an increase in the dopant concentration.



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(A) TITLE OF THE INVENTION.

5 LUMINESCENT GOLD(III) COMPOUNDS, THEIR
PREPARATION, AND LIGHT-EMITTING DEVICES
CONTAINING SAME

(B) CROSS-REFERENCES TO RELATED APPLICATIONS.

Not Applicable.

10 (C) STATEMENT REGARDING FEDERALLY SPONSORED
RESEARCH OR DEVELOPMENT.

Not Applicable.

15 (D) INCORPORATION-BY-REFERENCE OF MATERIAL SUBMITTED ON A
COMPACT DISK.

Not Applicable.

20 (E) BACKGROUND OF THE INVENTION.

(1) Field of the Invention.

The present invention is directed to a new class of luminescent gold(III) compounds containing a tridentate ligand and having at least one strong σ -donating group. Such compounds can be used as light-emitting material in phosphorescence based organic light-emitting devices (OLEDs), wherein different colors can be obtained by varying the applied DC voltage or the dopant concentration of said compounds.

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(2) Description of Related Art Including Information Disclosed Under 37 C.F.R. 1.97 and 1.98.

The market for flat-panel displays has attracted considerable attention in connection with the development of electroluminescent materials. The electroluminescence materials used are generally categorized into conjugated polymers or low-molar-mass small molecules. OLEDs are among the most important candidates for the use of electroluminescent materials in commercial flat-panel displays because OLEDs possess the advantages of robustness, ease of fabrication and color tuning, wide viewing angle, high brightness and contrast ratios, low turn-on voltage and low energy consumption.

A typical OLED structure is composed of a thin film of organic material sandwiched between a transparent conductor such as indium tin oxide (ITO), and a vapor deposited metal cathode. Upon applying an electrical potential, excitons are formed by the recombination of the holes and electrons, injected from the ITO electrode and metal cathode, respectively. Electroluminescence is generated in the organic material from the radiative relaxation of excitons. Higher performance of the device can be achieved by using multiple organic layers for separation of hole and electron transporting layers.

Although electroluminescence from organic polymers was initially reported years ago [Kaneto, K.; Yoshino, K.; Koa, K.; Inuishi, Y. *Jpn. J. Appl. Phys.* 18, 1023 (1974)], it was only after the report on yellow-green electroluminescence from poly(*p*-phenylenevinylene) (PPV) that light-emitting polymers and OLEDs have received much attention [Burroughs, J.H.; Bradley, D.D.C.; Brown, A.R.; Marks, N.; Friend, R.H.; Burn, P.L.; Holmes, A.B., *Nature* 347, 539 (1990)]. Later on, similar studies were reported by using PPV derivatives as the light-emitting polymers [Braun, D.; Heeger, A.J., *Appl. Phys. Lett.* 58, 1982 (1991)]. Since then a number of new electroluminescent polymers have been investigated for improved properties.

Electroluminescence of organic materials was discovered in anthracene crystals immersed in liquid electrolyte in 1965 [Helfrich, W.; Schneider, W.G. *Phys. Rev. Lett.* 14, 229 (1965)]. Although lower operating voltages could be achieved by using a thin film of anthracene as well as solid electrodes, very low efficiency of such a single-layer device was encountered. High-performance green electroluminescence from an organic small molecule, aluminum tris(quinolate) (Alq₃), was first reported in

1987 [Tang, C.W.; VanSlyke, S.A. *Appl. Phys. Lett.* 51, 913 (1987)]. A double-layer OLED with high efficiency and low operating voltage was described, in which Alq₃ was utilized both as emitting layer and electron transporting layer. Subsequent modifications of the device with triple-layer structure gave better performance with higher efficiency.

Some improvements in OLED efficiencies have been achieved by using phosphorescent material to generate light emission from both singlet and triplet excitons. One approach, particularly for small-molecule OLEDs, is to harvest triplet excitons efficiently through incorporation of heavy metal centers, which would increase spin-orbit coupling and hence intersystem crossing into the triplet state. In 1998, Baldo et al. demonstrated a phosphorescence electroluminescence device with high quantum efficiency by using platinum(II) 2,3,7,8,12,13,17,18-octaethyl-21H,23H-porphine (PtOEP) as a dye [Baldo, M.A.; O'Brien, D.F.; You, Y.; Shoustikow, A.; Sibley, S.; Thompson, M.E.; Forrest, S.R. *Nature* 395, 151 (1998)]. A multilayer device in which the emitting layer of Alq₃ is doped with PtOEP showed a strong emission at 650 nm attributed to the triplet excitons of PtOEP. Cyclometalated iridium(III) is known to show phosphorescence and is another class of materials used for high efficiency OLEDs. Baldo et al. reported the use of *fac*-tri(2-phenylpyridine)iridium(III) [Ir(ppy)₃] as phosphorescence emitting material which was doped in 4,4'-N,N'-dicarbazole-biphenyl (CBP) as a host in an OLED to give high quantum efficiency [Baldo, M.A.; Lamansky, S.; Burrows, P.E.; Thompson, M.E.; Forrest, S.R. *Appl. Phys. Lett.* 75, 4 (1999)]. In addition, *fac*-tri(2-phenylpyridine)iridium(III) [Ir(ppy)₃] was used as phosphorescence sensitizer for high efficiency fluorescent OLED [Baldo, M.A.; Thompson, M.E.; Forrest, S.R. *Nature*, 403, 750 (2000)]. Using the concept of a phosphorescence emitter with a higher population of excitations, very high efficiency red fluorescence from [2-methyl-6-[2-(2,3,6,7-tetrahydro-1H,5H-benzo[II]quinolizin-9-yl)-ethenyl]-4H-pyran-4-ylidene]propanedinitrile (DCM2) was found in a multilayer OLED composed of Ir(ppy)₃ and DCM2 dopant layers. In a sensitization process, energy is transferred from Ir(ppy)₃ to DCM2 to give such high efficiency fluorescence.

Apart from the enhancement of the emission efficiency, the ability to bring about a variation in the emission color would be important. Most of the common approaches involve the use of different light-emitting materials or multi-component

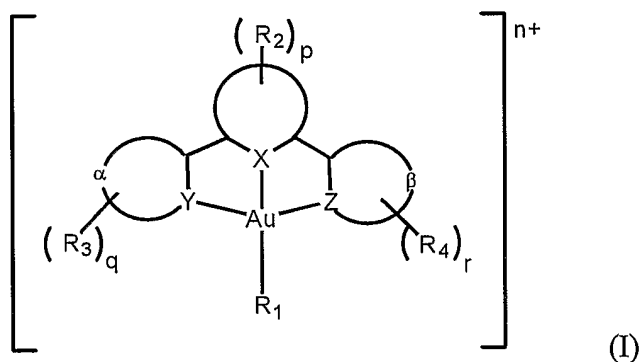
blended mixtures of light-emitting materials with different emission characteristics for color tuning. Examples that employ a single light-emitting material as dopant to generate more than one emission color have been rare. Recent studies have shown that different emission colors from a single emissive dopant could be generated by using phosphorescent material through a change in the direction of the bias or in the dopant concentration. Welter et al. reported the fabrication of a simple OLED consisting of semiconducting polymer PPV and phosphorescent ruthenium polypyridine dopant [Welter, S.; Krunner, K.; Hofstraat, J.W.; De Cola, D. *Nature*, 421, 54 (2003)]. At forward bias, red emission from the excited state of the phosphorescent ruthenium polypyridine dopant was observed, while the OLED emitted a green emission at reverse bias in that the lowest excited singlet state of PPV was populated. Adamovich et al. reported the use of a series of phosphorescent platinum(II) [2-(4,6-difluorophenyl)pyridinato-N,C^{2'}] β -diketonates as single emissive dopant in OLED [Adamovich, V.; Brooks, J.; Tamayo, A.; Alexander, A.M.; Djurovich, P.R.; D'Andrade, B.W.; Adachi, C.; Forrest, S.R.; Thompson, M.E. *New. J. Chem.* 26, 1171 (2002)]. Both blue emission from the monomeric species and orange emission from the aggregates were observed in such OLED and the relative intensity of the orange emission increases as the doping level is increased. As a result, the electroluminescence color can be tuned by changing the dopant concentration, and white illumination sources of an OLED can be obtained in a doping concentration with equal intensities of the monomeric and aggregate bands. In both cases, the change of electroluminescence color in OLED can be accomplished upon a variation of the external stimulus or fabrication conditions while keeping the light-emitting material the same.

Despite recent interest in electrophosphorescent materials, in particular metal complexes with heavy metal centers, most of the work has been focused on the use of iridium(III), platinum(II) and ruthenium(II), while other metal centers have been relatively less extensively explored. In contrast to the isoelectronic platinum(II) compounds which are known to show rich luminescence properties, very few examples of luminescent gold(III) compounds have been reported, probably due to the presence of low-energy d-d ligand field (LF) states and the electrophilicity of the gold(III) metal center. The introduction of strong σ -donating ligands into gold(III) compounds to

enhance the luminescence properties as a result of the enlargement of d-d splitting has been considered. Yam et al. first demonstrated that gold(III) aryl compounds are photo-stable and are capable of displaying interesting photoluminescence properties which occur even at room temperature [Yam, V.W.W.; Choi, S.W.K.; Lai, T.F.; Lee, W.K. *J. Chem. Soc., Dalton Trans.* 1001(1993)]. Another interesting donor ligand is the alkynyl group. But despite the fact that a number of gold(I) alkynyls are known and have been shown to exhibit interesting luminescence properties, the chemistry of gold(III) alkynyls has been essentially ignored, except for a brief report on the synthesis of an alkynylgold(III) compound of 6-benzyl-2,2'-bipyridine in the literature [Cinellu, M.A.; Minghetti, G.; Pinna, M.V.; Stoccoro, S.; Zucca, A.; Manassero, M. *J. Chem. Soc. Dalton Trans.* 2823 (1999)], but their luminescence behaviour has remained totally unexplored. The present inventors have described herein the design, synthesis and photoluminescence behaviors of luminescent gold(III) compounds with a tridentate ligand and at least one strong σ -donating group, and the use of such compounds as electrophosphorescent material in OLEDs to give strong electroluminescence with high efficiency.

(F) BRIEF SUMMARY OF THE INVENTION

The present invention is directed to novel luminescent gold(III) compounds, their preparation, and OLEDs containing them. The compounds have the chemical structure shown in generic formula (I):



wherein R_1 , R_2 , R_3 and R_4 each independently represents a substituent selected from the group consisting of hydrogen, halogen, alkynyl, substituted alkynyl, alkyl, substituted alkyl, aryl, substituted aryl, heteroaryl, substituted heteroaryl, alkoxy, substituted

alkoxyl, amino, substituted amino, cyano, nitro, alkylcarbonyl, alkoxy carbonyl, arylcarbonyl, aryloxy carbonyl, mono- or dialkylaminocarbonyl, alkylcarbonyloxy, arylcarbonyloxy, aryloxy, alkoxy carbonyl, aryloxy carbonyloxy group, and the like; X, Y and Z each independently represent a heteroatom or a carbon; $\overline{\text{C-X-C}}$ represents an aromatic or heterocyclic 5- or 6-membered ring; α and β each independently represents a bridge for an aromatic or heterocyclic 5- or 6-membered ring, or a break for a non-cyclic moiety; C-X, C-Y and C-Z each independently represents a single bond or double bond; n is a zero or an integer; and p, q and r are positive integers. The compounds of the present invention each contains one tridentate ligand and at least one strong σ -donating group coordinating to a gold(III) metal center.

The luminescent gold(III) compounds of the present invention show photoluminescence *via* triplet excited state upon photo-excitation, or electroluminescence *via* triplet exciton upon applying a DC voltage. Preferred compounds of the invention are thermally stable and volatile enough to be able to form a thin layer by sublimation or vacuum deposition.

The present invention is also directed to the use of luminescent compounds of general formula (I) as phosphorescent emitters or dopants fabricated into OLEDs to generate electroluminescence. In one embodiment, the light-emitting material used as a phosphorescent emitter or dopant in an OLED can comprise a gold(III) compound coordinated with one tridentate ligand and at least one strong σ -donating ligand.

In an OLED according to the present invention, the luminescent compound is included in a light-emitting layer or as a dopant arranged between a pair of electrodes. The typical structure of an OLED using the luminescent compound of the present invention as a light-emitting layer without an electron blocking layer is in the order shown in FIG. 1: anode/hole transporting layer/luminescent gold(III) compound as a light-emitting layer/electron injection layer/cathode.

The typical structure of an OLED using the luminescent compound of the present invention as a light-emitting layer with an electron blocking layer is in the order shown in FIG. 2: anode/hole transporting layer/luminescent gold(III) compound as light-emitting layer/hole blocking layer/electron injection layer/cathode.

The typical structure of an OLED using the luminescent compound of the present invention as a dopant with an electron blocking layer is in the order shown in

FIG. 3: anode/hole transporting layer/luminescent gold(III) compound doped in host/hole blocking layer/electron transporting layer/cathode.

(G) BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows the structure of an OLED using the luminescent compound of the present invention as the light-emitting layer without an electron blocking layer.

FIG. 2 shows the structure of an OLED using the luminescent compound of the present invention as the light-emitting layer with an electron blocking layer.

FIG. 3 shows the structure of an OLED using the luminescent copolymer of the present invention as a dopant with an electron blocking layer.

FIG. 4 shows the UV-vis absorption and emission spectra of compound 1 in dichloromethane at 298° K. No instrumental correction was applied for the emission wavelength.

FIG. 5 shows the UV-vis absorption and emission spectra of compound 4 in dichloromethane at 298° K. No instrumental correction was applied for the emission wavelength.

FIG. 6 shows the UV-vis absorption and emission spectra of compound 6 in dichloromethane at 298° K. No instrumental correction was applied for the emission wavelength.

FIG. 7 UV-vis absorption and emission spectra of compound 8 in dichloromethane at 298° K. No instrumental correction was applied for the emission wavelength.

FIG. 8 shows the solid state (thin film) emission spectrum of compound 1 at 298° K. No instrumental correction was applied for the emission wavelength.

FIG. 9 shows the solid state (thin film) emission spectrum of compound 8 at 298° K. No instrumental correction was applied for the emission wavelength.

FIG. 10A shows the multilayer OLED with the structure of ITO/NPB (60 nm)/ compound 1 (30 nm)/Alq₃ (10 nm)/LiF (1 nm)/Al (90 nm).

FIG. 10B shows the electroluminescence spectra of device 1 (see Fig. 1) at different applied DC voltages; the emission intensities are normalized in the range of 450 - 480 nm.

FIG. 11A shows the multilayer OLED of device 2 (see Fig. 2) with the structure of ITO/TPD (70 nm)/ compound 1 (10 nm)/Alq₃ (40 nm)/LiF (0.6 nm)/Al (150 nm).

FIG. 11B shows the electroluminescence spectrum of device 2 (see Fig. 2) upon applying 7 V DC voltage.

FIG. 11C shows the current density and luminance *versus* voltage characteristics of device 2 (see Fig. 2).

5 FIG. 11D shows the current and power efficiency *versus* current density characteristics of device 2 (see Fig. 2).

FIG. 12A shows the multilayer OLED with the structure of device 3 (see Fig. 3) ITO/TPD (70 nm)/ compound 8 (30 nm)/BCP (30 nm)/Alq₃ (10 nm)/LiF (0.6 nm)/Al (150 nm).

10 FIG. 12B shows the electroluminescence spectrum of device 3 (see Fig. 3) upon applying 9 V DC voltage.

FIG. 12C shows the current density and luminance *versus* voltage characteristics of device 3 (see Fig. 3).

15 FIG. 12D shows the current and power efficiency *versus* current density characteristics of device 3 (see Fig. 3).

FIG. 13A shows the multilayer OLED of device 4 with the structure: ITO/TPD (40 nm)/CBP doped with (X %) compound 8 (30 nm)/BCP (20 nm)/Alq₃ (10 nm)/LiF (0.6 nm)/Al (150 nm) [X = 1 (device 4a), 3 (device 4b), 6 (device 4c), 12 (device 4d), 18 (device 4e), 100 (device 4f)].

20 FIG. 13B shows normalized electroluminescence spectra of devices 4a-f with different concentrations of compound 8 1% (device 4a), 3 (device 4b), 6 (device 4c), 12 (device 4d), 18 (device 4e), 100 (device 4f) as dopant upon applying 12 V DC voltage.

25 FIG. 14A shows device 5: ITO/TPD (70 nm)/CBP (compound 8, 6 wt%) (30 nm)/TPBi (60nm)/LiF (1 nm)/Al (150 nm).

FIG. 14B shows a plot of current density and luminance *versus* voltage for device 5.

FIG. 14C shows a plot of current and power efficiency *versus* current density for device 5.

30 FIG. 14D plot of quantum efficiency *versus* current density. Inset: spectra characteristics of device 5.

(H) DETAILED DESCRIPTION OF THE INVENTION

The present invention is related to the syntheses, spectral characterization, and luminescence properties of a class of luminescent gold(III) compounds with one tridentate ligand and at least one strong σ -donating group; and the use of such compounds as light-emitting material in OLEDs to provide electroluminescence with high efficiency and brightness. The compounds have the following structural characteristics:

- (1) at least one gold metal center at an oxidation state of +3;
- (2) said one gold metal center having four coordination sites;
- (3) one tridentate ligand with one to three aromatic or heterocyclic ring(s) coordinating to the gold metal center;
- (4) one monodentate ligand coordinating to the gold metal center;
- (5) at least one strong σ -donating ligand coordinating to the gold metal center; and
- (6) the compounds being charged or neutral.

Gold(III) compounds have been rarely observed to emit, in contrast to their isoelectronic platinum(II) compounds which are known to display rich luminescence properties. The lack of luminescence behavior in gold(III) compounds may be due to the presence of low-lying d-d ligand field (LF) states and the electrophilicity of the gold(III) metal center. Gold(III) aryl compounds [Yam, V.W.W.; Choi, S.W.K.; Lai, T.F.; Lee, W.K. *J. Chem. Soc., Dalton Trans.* 1001(1993)] are exceptions in that they show interesting luminescence properties even at room temperature and possess photostability upon light irradiation. Without wishing to be bound by theory, it is believed that the coupling of strong σ -donating ligands to gold(III) renders the metal centre more electron-rich, thereby raising the energy of the d-d states, which results in an improvement or enhancement of the luminescence by increasing the chances for population of the emissive state. A class of luminescent gold(III) compounds with one tridentate ligand and at least one strong σ -donating group will be described in detail hereinbelow.

The luminescent gold(III) compounds of the present invention can be formed into thin films by vacuum deposition, spin-coating or other known fabrication methods. Different multilayer OLEDs have been fabricated using the compounds of

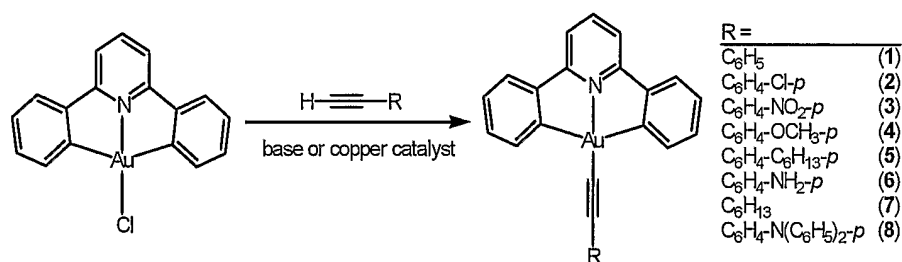
the present invention as light-emitting material or as dopant in the emitting layer. In general, the OLEDs consist of one anode and one cathode, between which are the hole transporting layer, light-emitting layer, and electron transporting or injection layer.

The present invention will be illustrated more specifically by the following non-limiting examples, it being understood that changes and variations can be made therein without deviating from the scope and the spirit of the invention as hereinafter claimed.

EXAMPLE 1

Synthesis and characterization

Compounds 1-8 were synthesized according to the following methodology. The precursor compound, $[\text{Au}(\text{C}^{\wedge}\text{N}^{\wedge}\text{C})\text{Cl}]$, was prepared according to the modification of a procedure reported in the literature [Wong, K.H.; Cheung, K.K.; Chan, M.C.W.; Che, C.M. *Organometallics*, 17, 5305(1998)]. The desired compounds were synthesized by the reaction of $[\text{Au}(\text{C}^{\wedge}\text{N}^{\wedge}\text{C})\text{Cl}]$ with various alkynes in the presence of a base or copper catalyst in an organic solvent. For example, to a mixture of $[\text{Au}(\text{C}^{\wedge}\text{N}^{\wedge}\text{C})\text{Cl}]$, terminal alkyne and Et_3N in degassed dichloromethane solution was added CuI . The reaction mixture was stirred for 6 hours under a nitrogen atmosphere at room temperature. The crude product was purified by column chromatography on silica gel using dichloromethane as eluent. Pale yellow crystals were obtained from slow diffusion of diethyl ether into the dichloromethane solution of the compounds.



The characteristic spectral properties of compounds 1-8 are as follows:

$[\text{Au}(\text{C}^{\wedge}\text{N}^{\wedge}\text{C})\text{C}\equiv\text{C-C}_6\text{H}_5]$ (Compound 1)

Yield: 88 %. ^1H NMR (300 MHz, CH_2Cl_2 , 298° K, relative to Me_4Si): δ 8.04 (dd, 7.4 and 1.0 Hz, 2 H, $\text{C}^{\wedge}\text{N}^{\wedge}\text{C}$), 7.92 (t, 8.0 Hz, 1 H, $\text{C}^{\wedge}\text{N}^{\wedge}\text{C}$), 7.62 (m, 4 H, $\text{C}^{\wedge}\text{N}^{\wedge}\text{C}$ and C_6H_5), 7.54 (d, 8.0 Hz, 2 H, $\text{C}^{\wedge}\text{N}^{\wedge}\text{C}$), 7.26 – 7.44 (m, 7 H, $\text{C}^{\wedge}\text{N}^{\wedge}\text{C}$ and

C₆H₅); positive EI-MS: m/z 527 [M]⁺; IR (KBr): 2147 cm⁻¹ ν (C≡C); elemental analyses calc'd for C₂₅H₁₆NAu (found): C 56.93 (56.57), H 3.04 (3.05), N 2.66 (2.66).

[Au(C^{^N^C})C≡C-C₆H₄-Cl-*p*] (Compound 2)

5 Yield: 85 %. ¹H NMR (300 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.00 (dd, 7.2 and 1.0 Hz, 2 H, C^{^N^C}), 7.90 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.50 – 7.60 (m, 6 H, C^{^N^C} and C₆H₄), 7.25 – 7.42 (m, 6 H, C^{^N^C} and C₆H₄); positive EI-MS: m/z 562 [M]⁺; IR (KBr): 2157 cm⁻¹ ν (C≡C); elemental analyses calc'd for C₂₅H₁₅NCIAu·1/2H₂O (found): C 52.59 (52.85), H 2.80 (2.66), N 2.45 (2.40).

10

[Au(C^{^N^C})C≡C-C₆H₄-NO₂-*p*] (Compound 3)

Yield: 80 %. ¹H NMR (400 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.22 (d, 9.0 Hz, 2 H, C₆H₄), 8.00 (dd, 7.6 and 1.2 Hz, 2 H, C^{^N^C}), 7.94 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.73 (d, 9.0 Hz, 2 H, C₆H₄), 7.64 (dd, 7.6 and 1.2 Hz, 2 H, C^{^N^C}), 7.55 (d, 8.0 Hz, 2 H, C^{^N^C}), 7.41 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 7.32 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}); positive EI-MS: m/z 572 [M]⁺; IR (KBr): 2146 cm⁻¹ ν (C≡C); elemental analyses calc'd for C₂₅H₁₅N₂O₂Au (found): C 51.64 (51.62), H 2.75 (2.69), N 4.82 (4.75).

20

[Au(C^{^N^C})C≡C-C₆H₄-OCH₃-*p*] (Compound 4)

Yield: 86 %. ¹H NMR (400 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.02 (dd, 7.6 and 1.0 Hz, 2 H, C^{^N^C}), 7.90 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.60 (dd, 7.6 and 1.0 Hz, 2 H, C^{^N^C}), 7.50 – 7.56 (m, 4 H, C^{^N^C} and C₆H₄), 7.40 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 7.27 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 6.91 (d, 8.9 Hz, 2 H, C₆H₄), 3.88 (s, 3 H, OCH₃); positive EI-MS: m/z 557 [M]⁺; IR (KBr): 2157 cm⁻¹ ν (C≡C); elemental analyses calc'd for C₂₆H₁₈NOAu·1/2H₂O (found): C 55.12 (55.15), H 3.36 (3.28), N 2.47 (2.48).

30

[Au(C^{^N^C})C≡C-C₆H₄-C₆H₁₃-*p*] (Compound 5)

Yield: 75 %. ¹H NMR (300 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.00 (dd, 7.4 and 1.0 Hz, 2 H, C^{^N^C}), 7.87 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.57 (dd, 7.4 and

1.0 Hz, 2 H, C^{^N^C}), 7.47 – 7.51 (m, 4 H, C^{^N^C} and C₆H₄), 7.37 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 7.24 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 7.18 (d, 8.3 Hz, 2 H, C₆H₄), 2.64 (t, 7.7 Hz, 2 H, CH₂-CH₂-(CH₂)₃-CH₃), 1.64 (m, 2 H, CH₂-CH₂-(CH₂)₃-CH₃), 1.34 (m, 6 H, CH₂-CH₂-(CH₂)₃-CH₃), 0.90 (t, 7.0 Hz, 3 H, CH₂-CH₂-(CH₂)₃-CH₃); positive EI-MS: *m/z* 611 [M]⁺; IR (KBr): 2149 cm⁻¹ ν(C≡C); elemental analyses calc'd for C₃₁H₂₈NAu·1/2H₂O (found): C 60.00 (59.91), H 4.68 (4.60), N 2.26 (2.25).

Au(C^{^N^C})C≡C-C₆H₄-NH₂-*p*] (Compound 6)

Yield: 80 %. ¹H NMR (300 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.07 (dd, 7.4 and 1.0 Hz, 2 H, C^{^N^C}), 7.92 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.65 (dd, 7.4 and 1.0 Hz, 2 H, C^{^N^C}), 7.56 (d, 8.0 Hz, 2 H, C^{^N^C}), 7.39 – 7.45 (m, 4 H, C^{^N^C} and C₆H₄), 7.30 (dt, 7.5 and 1.3 Hz, 2 H, C^{^N^C}), 6.67 (d, 8.6 Hz, 2 H, C₆H₄), 3.84 (s, 2 H, NH₂); positive EI-MS: *m/z* 542 [M]⁺; IR (KBr): 2143 cm⁻¹ ν(C≡C); elemental analyses calc'd for C₂₅H₁₇N₂Au·1/2H₂O (found): C 54.45 (54.59), H 3.27 (3.13), N 5.08 (5.04).

[Au(C^{^N^C})C≡C-C₆H₁₃] (Compound 7)

Yield: 85 %. ¹H NMR (300 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.00 (dd, 7.2 and 1.0 Hz, 2 H, C^{^N^C}), 7.90 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.62 (dd, 7.2 and 1.0 Hz, 2 H, C^{^N^C}), 7.53 (d, 8.0 Hz, 4 H, C^{^N^C}), 7.40 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 7.28 (dt, 7.3 and 1.3 Hz, 2 H, C^{^N^C}), 2.49 (t, 6.9 Hz, 2 H, CH₂-(CH₂)₂-(CH₂)₂-CH₃), 1.63 – 1.71 (m, 4 H, CH₂-(CH₂)₂-(CH₂)₂-CH₃), 1.40 (m, 4 H, CH₂-(CH₂)₂-(CH₂)₂-CH₃), 0.95 (t, 7.0 Hz, 3 H, CH₂-(CH₂)₂-(CH₂)₂-CH₃); positive EI-MS: *m/z* 536 [M]⁺; IR (KBr): 2155 cm⁻¹ ν(C≡C); elemental analyses calc'd for C₂₅H₂₄NAu (found): C 56.08 (55.96), H 4.52 (4.60), N 2.62 (2.53).

[Au(C^{^N^C})C≡C-C₆H₄-N(C₆H₅)₂-*p*] (Compound 8)

Yield: 72 %. Yield: 80 %. ¹H NMR (300 MHz, CH₂Cl₂, 298° K, relative to Me₄Si): δ 8.05 (dd, 7.2 and 1.2 Hz, 2 H, C^{^N^C}), 7.92 (t, 8.0 Hz, 1 H, C^{^N^C}), 7.64 (dd, 7.2 and 1.2 Hz, 2 H, C^{^N^C}), 7.56 (d, 8.0 Hz, 2 H, C^{^N^C}), 7.48 (d, 8.8 Hz, 2 H, C^{^N^C}), 7.42 (t, 7.2 Hz, 2 H, C^{^N^C} H's), 7.32 – 7.26 (m, 6H, C^{^N^C}, C₆H₄ and N-C₆H₅), 7.14 – 7.02 (m, 8H, C^{^N^C}, C₆H₄ and N-C₆H₅); positive EI-MS: *m/z* 694 [M]⁺;

IR (KBr): 2143 cm^{-1} $\nu(\text{C}\equiv\text{C})$; elemental analyses calc'd for $\text{C}_{37}\text{H}_{25}\text{N}_2\text{Au}\cdot 1/2\text{H}_2\text{O}$ (found): C 60.85 (61.10), H 3.67 (3.53), N 3.78 (3.80).

UV-vis absorption properties

5 All the luminescent gold(III) compounds exhibit an intense absorption band at 312 – 327 nm and a moderately intense vibronic-structured absorption band at 362 – 426 nm in dichloromethane at 298° K. The photophysical data of 1 – 8 are summarized in Table 1. In general, the electronic absorption energies are insensitive to the nature of the alkynyl ligands. The low-energy vibronic-structured absorption band show
10 vibrational progression spacings of 1310-1380 cm^{-1} , corresponding to the skeletal vibrational frequency of the $\text{C}\equiv\text{N}\text{C}$ ligand. Such low-energy absorptions are assigned as intraligand (IL) $\pi\text{-}\pi^*$ transition. An additional shoulder appeared in each of the electronic absorption spectra of compounds 6 and 8 at *ca.* 415 and 426 nm, respectively (see Figures 6 and 7). Since the alkynyl ligand with electron-rich amino substituent has
15 better electron-donating property, the presence of a low-lying alkynyl-to-diarylpyridine ligand-to-ligand charge transfer (LLCT) transition is possible. Thus the low-energy absorptions in compounds 6 and 8 are assigned as an admixture of intraligand (IL) $\pi\text{-}\pi^*(\text{C}\equiv\text{N}\text{C})/\text{LLCT } \pi(\text{C}\equiv\text{C}-\text{C}_6\text{H}_4-\text{NR}_2\text{-}p) \rightarrow \pi^*(\text{C}\equiv\text{N}\text{C})$ transition.

20 Photoluminescence properties

Unlike most other Au(III) compounds which are non-emissive or only show luminescence at low temperature compounds, 1 – 8 display intense luminescence at 468 – 625 nm in the solution state at room temperature (Table 1). In general, the emission energies of the compounds were found to be insensitive to the nature of the alkynyl
25 ligands (Figures 1 and 2). A vibronic-structured emission band with band maximum at 473 nm is observed for 1 – 5 and 7 in dichloromethane at room temperature. The vibrational progression spacings of *ca.* 1300 cm^{-1} are in line with the $\text{C}=\text{C}$ and $\text{C}=\text{N}$ stretching frequency of the tridentate ligand, indicative of the involvement of tridentate ligand in the excited state origin. Similar to the low-energy absorption band in the
30 electronic absorption studies, the luminescence is assigned as originated from metal-perturbed intra-ligand $^3[\pi\text{-}\pi^*]$ state of the tridentate $\text{C}\equiv\text{N}\text{C}$ ligand. Compounds 6 and 8 exhibit a structureless emission band at lower energy in dichloromethane at room

temperature (Figures 3 and 4). The emission spectra of compounds 1 - 8 in the solid state show a low-energy structureless band at around 570 nm (Figures 4 and 5). The red shift of the solid-state emission relative to that in the solution state is attributed to the excimeric intraligand emission arising from the π stacking of the C^NC ligand, probably due to the ordered packing of the molecules in the solid state.

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Table 1 Photophysical data for Compounds 1 through 8.

Compound	Medium (T[K])	Absorption	Emission
		λ_{max} [nm] (ϵ_{max} [dm ³ mol ⁻¹ cm ⁻¹])	λ_{max} [nm]
1	CH ₂ Cl ₂ (298)	312 (19890), 322 (19980), 364 (5050), 381 (5870), 402 (4870)	476, 506, 541, 582
	Solid (298)		588
	Thin film (298) ^a		568
2	CH ₂ Cl ₂ (298)	312 (19400), 322 (19640), 365(4640), 382(5170), 402(4305)	476, 506, 539, 584
	Solid (298)		558
3	CH ₂ Cl ₂ (298)	312 sh (27160), 327 (36005), 364 sh (17995), 382 sh (10170), 403 (5630)	477, 508, 546, 593
	Solid (298)		563
4	CH ₂ Cl ₂ (298)	312 (13820), 322 (13455), 362 (6400), 380 (6245), 400 (4190)	474, 505, 539, 584
	Solid (298)		555
5	CH ₂ Cl ₂ (298)	312 (17855), 322 (18100), 363 (5785), 381 (5945), 401 (4455)	475, 505, 538, 583
	Solid (298)		556
6	CH ₂ Cl ₂ (298)	310 (19195), 322 sh (15680), 365 (8855), 381 (10100), 399 (8300), 415 sh (3410)	611
	Solid (298)		585
7	CH ₂ Cl ₂ (298)	311 (14775), 320 (13925), 364 (3810), 380 (4900), 400 (4280)	473, 505, 537, 583
	Solid (298)		555
8	CH ₂ Cl ₂ (298)	312 (37090), 322 (38325), 364 (8525), 384 (10040), 400 (10035), 426 sh (4145)	625
	Thin film (298) ^a		564

^a prepared by vacuum deposition

EXAMPLE 2

Figure 10A shows an illustrative OLED structure of device 1: ITO/4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]biphenyl (NPB) (60 nm)/compound 1 (30 nm)/aluminum tris(8-hydroxyquinoline) (Alq_3) (10 nm)/LiF (1 nm)/Al (90 nm). NPB and Alq_3 act as the hole transporting material and the electron transporting or injection material, respectively. Electroluminescence spectra of device 1 at different DC voltages applied are shown in Figure 10B. In general, both the emission band of NPB in the range of 450 – 520 nm and the emission band of compound 1 at about 585 nm are observed upon application of DC voltage. At a lower DC voltage, the emission arising from compound 1 is more intense relative to the emission band of NPB. Upon increasing the DC voltage, the relative emission intensity ratio of compound 1: NPB decreases gradually. Therefore, the emission color of device 1 can be tuned from orange to green by applying different DC voltages.

EXAMPLE 3

Figure 11A shows an illustrative OLED structure of device 2: ITO/ $\text{N,N}'$ -diphenyl- $\text{N,N}'$ -bis(3-methylphenyl)-[1,9-biphenyl]-4,4'-diamine (TPD) (70 nm)/compound 1 (10 nm)/ Alq_3 (40 nm)/LiF (0.6 nm)/Al (150 nm). TPD acts as the hole transporting material. The electroluminescence spectrum of device 2 is depicted in Figure 11B. The EL spectrum shows only one band at around 585 nm. The EL spectrum compares well with the photoluminescence (PL) spectrum of compound 1 (Figure 7), indicating that both EL and PL arise from the same excited state or the same type of exciton, which is attributed to the excimeric intraligand emission resulting from the π stacking of the $\text{C}^{\wedge}\text{N}^{\wedge}\text{C}$ ligand. Figure 11C and 11D show the characteristics of device 2 with the relationship between current density, luminance and voltage; and between current, power efficiency and current density. The turn-on voltage is about 5 V.

EXAMPLE 4

Figure 12A shows an illustrative OLED structure of device 3: ITO/TPD (70 nm)/ compound 8 (30 nm)/2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) (30 nm)/ Alq_3 (10 nm)/LiF (0.6 nm)/Al (150 nm). BCP acts as the hole blocking material.

The electroluminescence spectrum of device 3 is depicted in Figure 12B. The EL spectrum shows only one band at around 580 nm. The EL spectrum compares well with the photoluminescence (PL) spectrum of compound 8 (shown in Figure 5), indicating that both EL and PL arise from the same excited state or the same type of exciton, which is attributed to the excimeric intraligand emission resulting from the π stacking of the C[^]N[^]C ligand. Figures 12C and 12D show the characteristics of device 3 with the relationship between current density, luminance and voltage; and between current, power efficiency and current density. The turn-on voltage is about 6.5 V. The relatively higher turn on voltage is due to the increase in emitting layer thickness (30 nm) and the introduction of an additional hole blocking layer.

EXAMPLE 5

Figure 13A shows the general OLED structure: ITO/TPD (70 nm)/4,4'-N,N'-dicarbazole-biphenyl (CBP) doped with (X %) compound 8 (30 nm)/BCP (30 nm)/Alq₃ (10 nm)/LiF (0.6 nm)/Al (150 nm) [X = 1 (device 4a), 3 (device 4b), 6 (device 4c), 12 (device 4d), 18 (device 4e), 100 (device 4f)]. CBP acts as the host material. Figure 13B displays the normalized EL spectra of devices 4a-f with different concentration of compound 8 as dopant upon applying 12 V DC voltage. It is clear that the EL band shifts to red from 500 nm to 580 nm upon increasing the dopant concentration. Since the emitting layer in device 4 is fabricated by the simultaneous vacuum deposition of compound 8 and the host, a higher dopant concentration of compound 8 may give rise to a higher order and better packing of the molecules, leading to stronger π stacking of the C[^]N[^]C ligand, and hence a lower energy excimeric intraligand emission. Therefore, a dependence of the EL color on the dopant concentration of the luminescent gold(III) compound, leading to concentration-dependent color tuning, can be accomplished in the present invention.

EXAMPLE 6

Device 5 with the following OLED structure: ITO/TPD (70 nm)/CBP (compound 8, 6wt%) (30 nm)/1,3,5-tris(2'-(1'-phenyl-1'-H-benzimidazole)benzene (TPBi) (60nm)/LiF (1 nm)/Al (150 nm) is fabricated (Figure 13). Higher efficiency is obtained in device 5 by using TPBi as electron transporting material which has higher

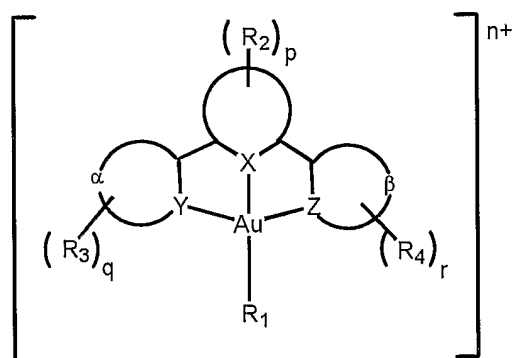
mobility than Alq_3 . The characteristics of device 5 are illustrated in Figure 13B which shows (a) plot of current density and luminance *versus* voltage; (b) plot of current and power efficiency *versus* current density; and (c) plot of quantum efficiency *versus* current density. Inset: spectra characteristics of device 5.

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(I) CLAIMS:

1. A luminescent compound comprising a tridentate ligand and at least one strong σ -donating group coordinating to a gold(III) metal center.
2. A luminescent compound having the chemical structure represented by any of the following general formula (I), wherein R_1 - R_4 each independently represent the group containing hydrogen, halogen, alkynyl, substituted alkynyl, alkyl, substituted alkyl, aryl, substituted aryl, heteroaryl, substituted heteroaryl, alkoxy, substituted alkoxy, amino, substituted amino, cyano, nitro, alkylcarbonyl, alkoxy carbonyl, arylcarbonyl, aryloxy carbonyl, mono- or dialkylaminocarbonyl, alkylcarbonyloxy, arylcarbonyloxy, aryloxy, alkoxy carbonyl, aryloxy carbonyloxy group, and the like; X, Y and Z each independently represent a heteroatom or a carbon; $\overline{C-X-C}$ represents an aromatic or heterocyclic 5- or 6-membered ring; α and β each independently represent a bridge for an aromatic or heterocyclic 5- or 6-membered ring or represent a break for non-cyclic moiety; C-X, C-Y and C-Z each independently represent a single bond or double bond; n represents a zero or an integer; p, q and r represent positive integers.

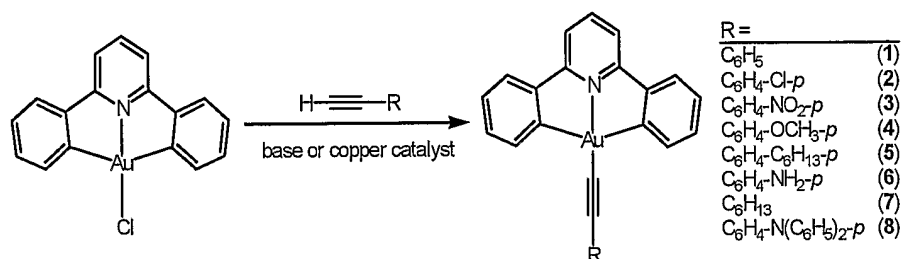


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3. A compound according to claim 2 wherein said compound can be deposited as a thin film.
4. A compound according to claim 3 wherein said thin layer can be deposited by sublimation, vacuum deposition, or spin-coating.

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5. A compound according to claim 2 wherein said compound has photoluminescence properties.
6. A compound according to claim 5 wherein said compound has electroluminescence properties.
7. A compound of claim 2 wherein said compound can be fabricated into a light-emitting device.
8. A light-emitting device wherein the compound of claim 2 is used as light-emitting layer.
9. A light-emitting device according to claim 8 wherein the compound of claim 2 is used as a dopant in the light-emitting layer.
10. A light-emitting device according to claim 8 wherein the luminescence color of the compound varies with the applied voltage.
11. A light-emitting device according to claim 9 wherein the luminescence band maximum of the compound varies with the concentration of the dopant.
12. A method for preparing a luminescent compound with a tridentate ligand and at least one strong σ -donating group coordinating to a gold(III) metal center, comprising the following reaction:



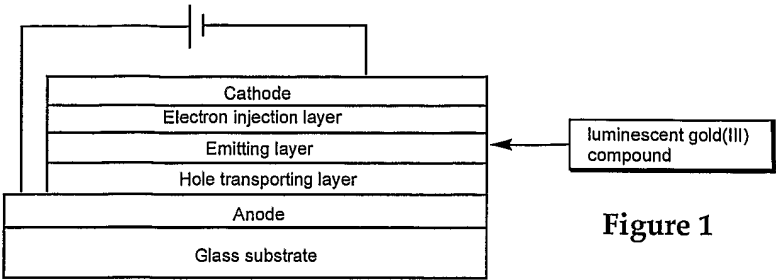


Figure 1

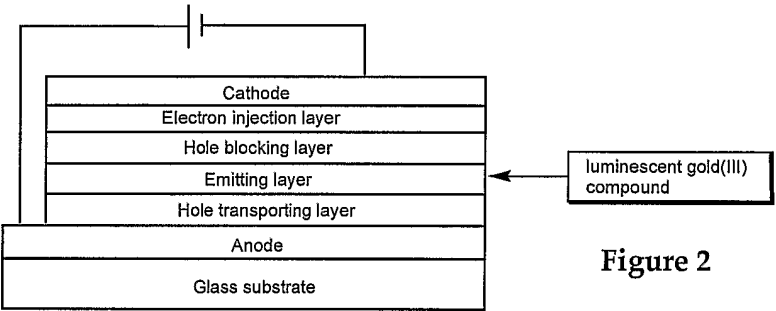


Figure 2

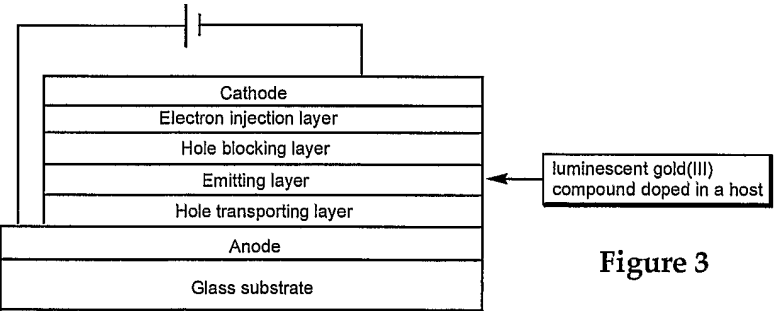


Figure 3

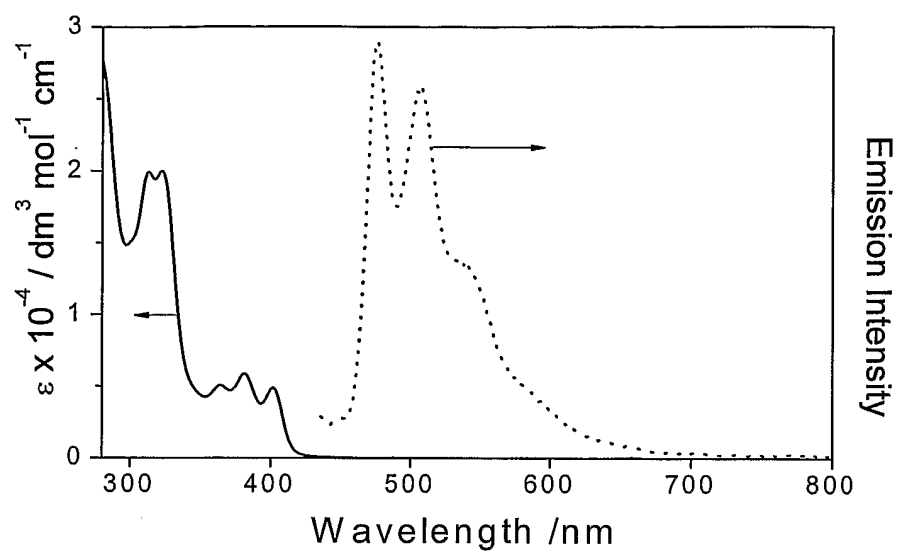


Figure 4

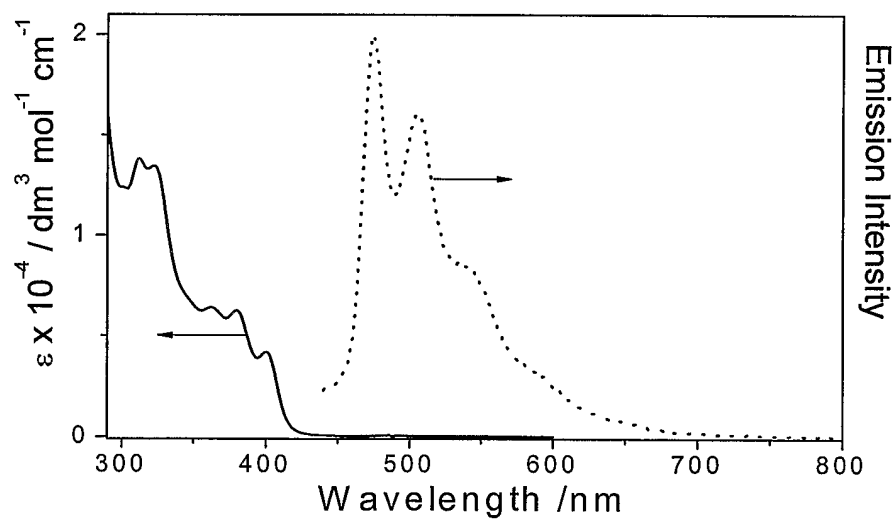


Figure 5

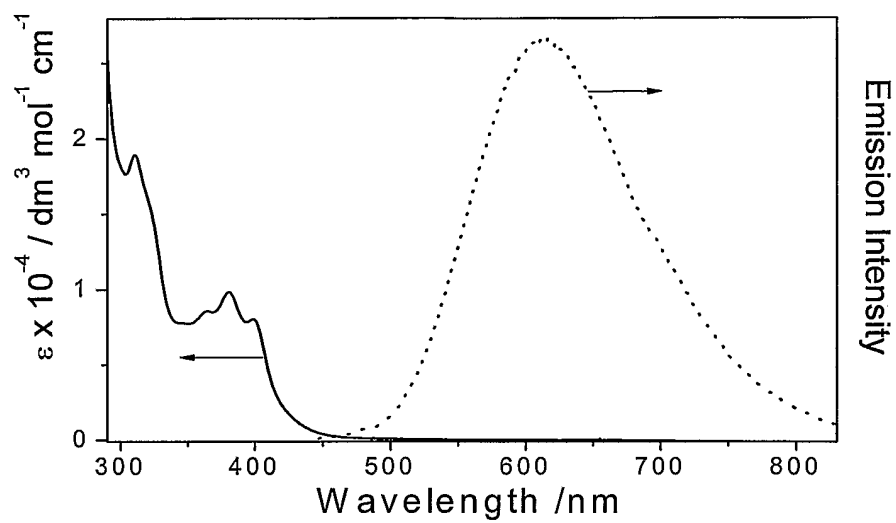


Figure 6

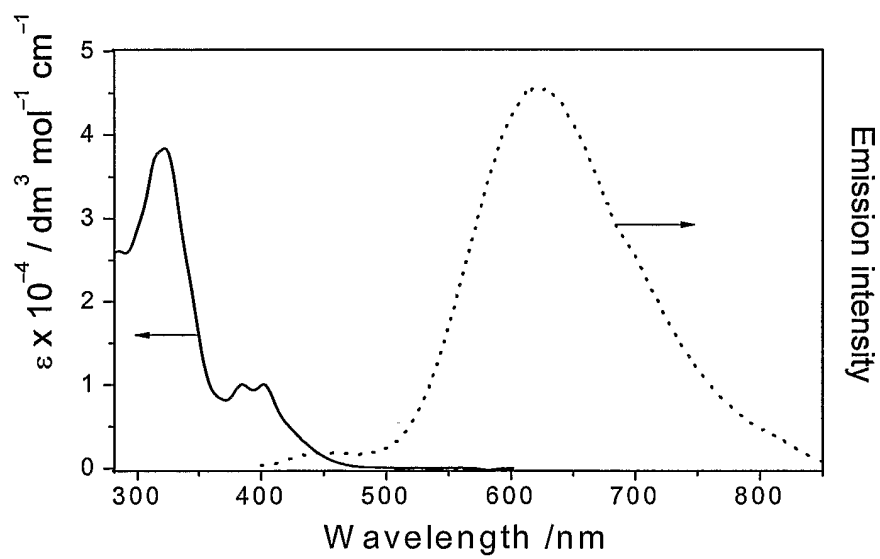


Figure 7

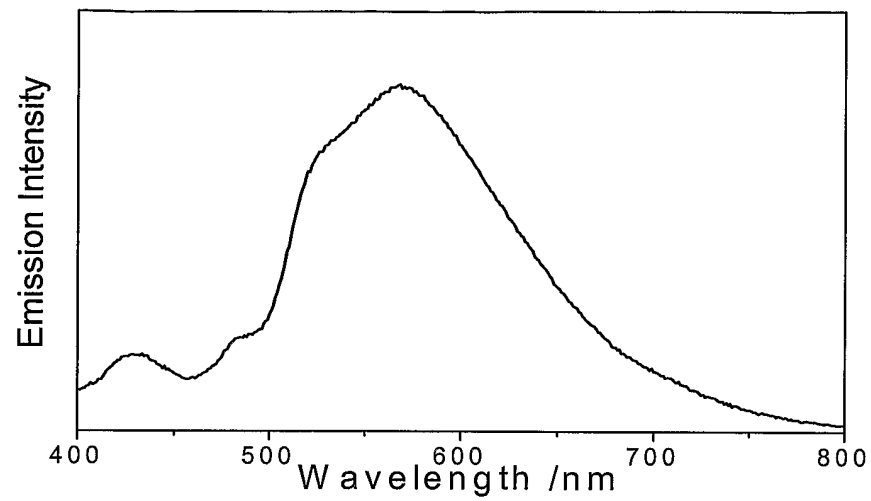


Figure 8

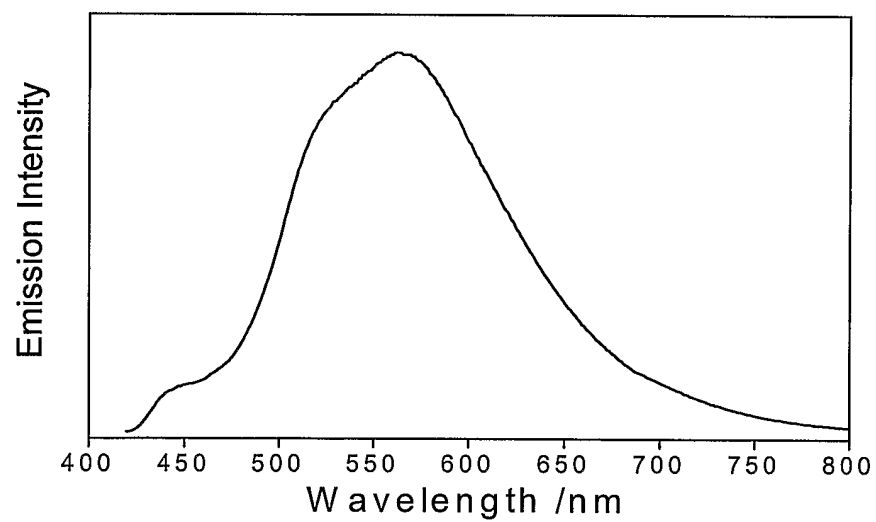


Figure 9

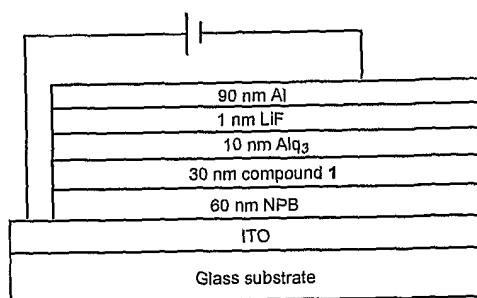


Figure 10A

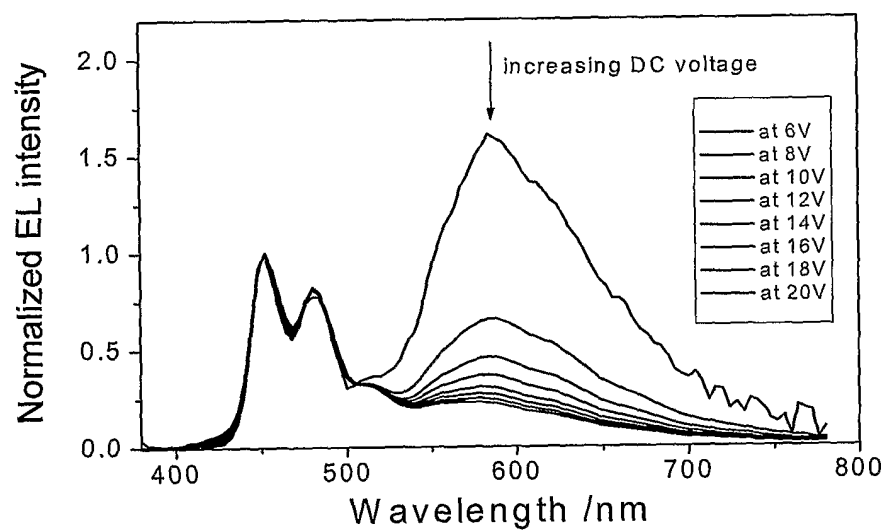


Figure 10B

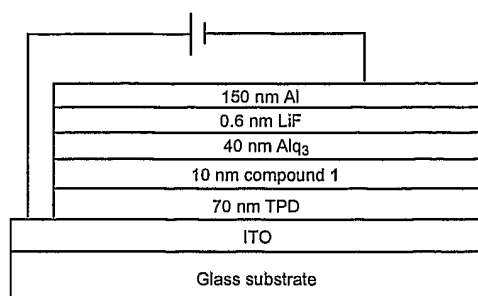


Figure 11A

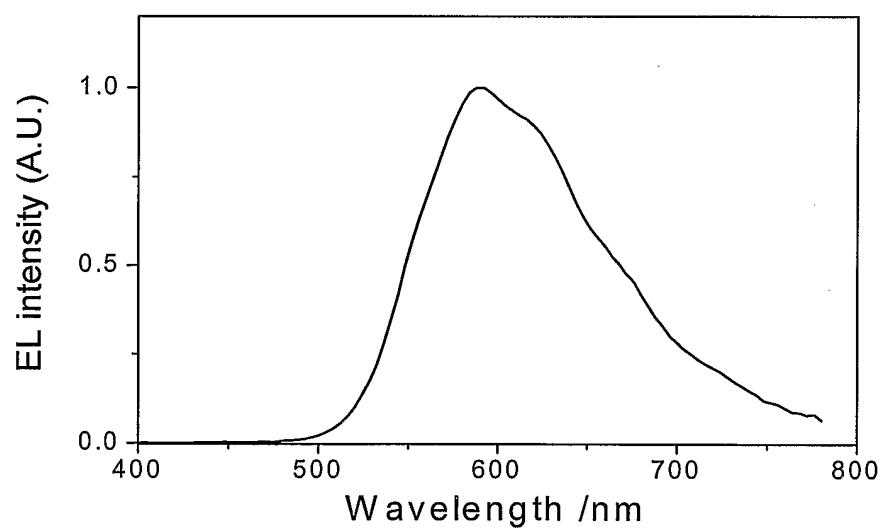


Figure 11B

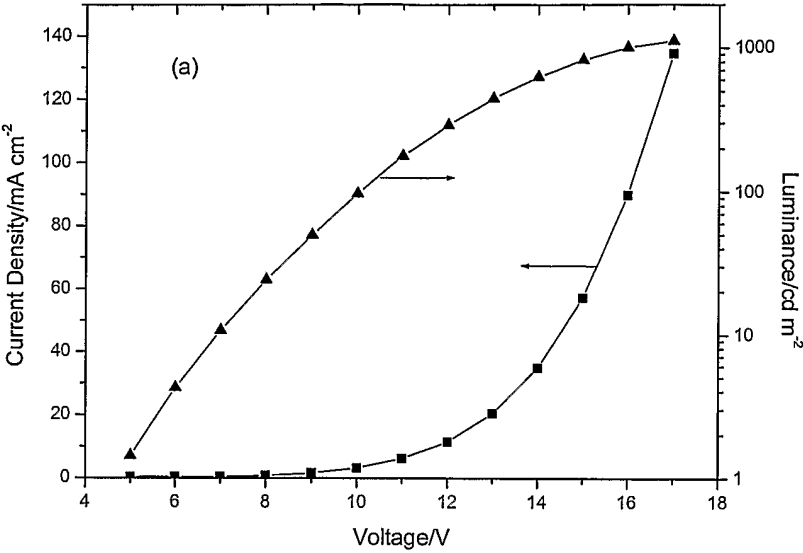


Figure 11C

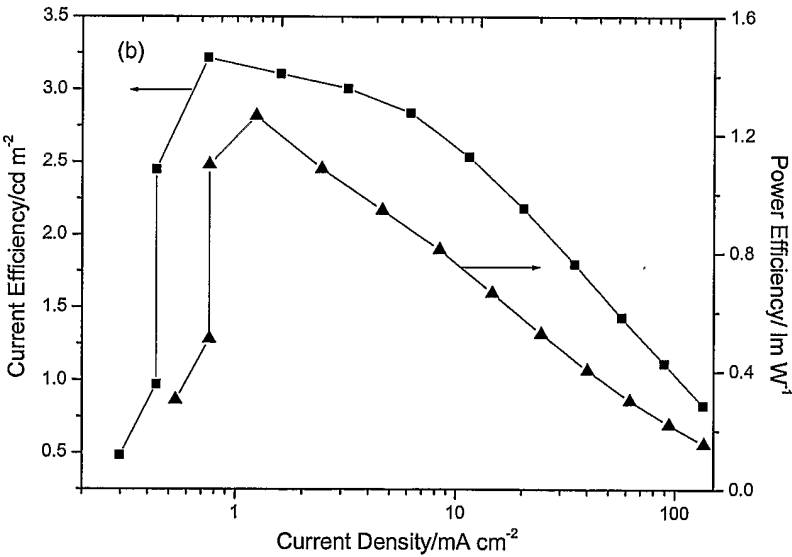


Figure 11D

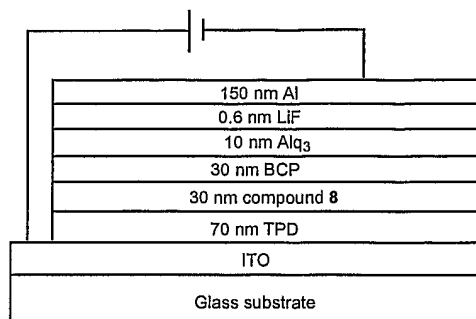


Figure 12A

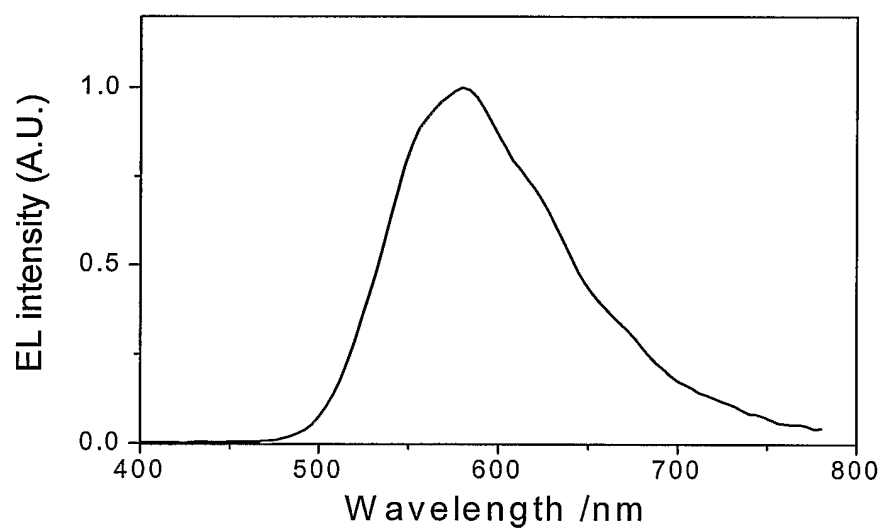


Figure 12B

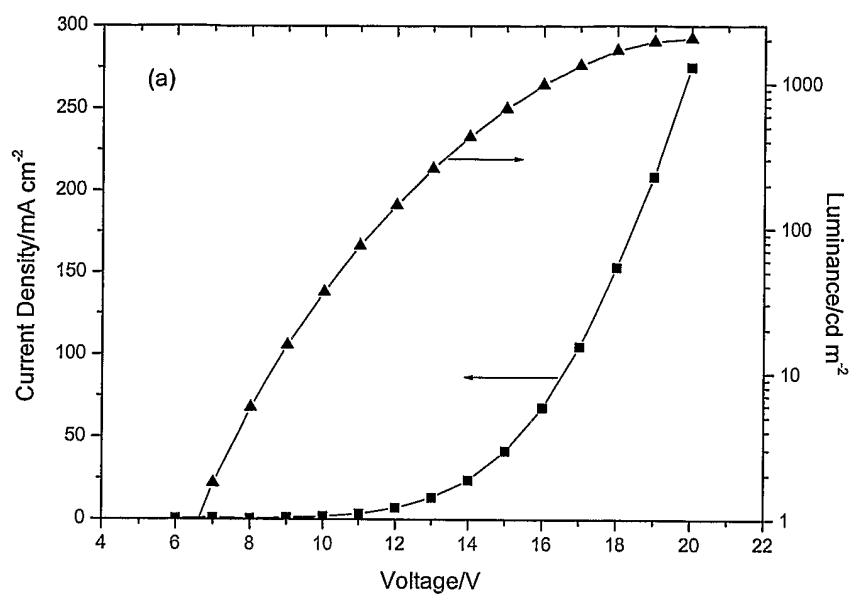


Figure 12C

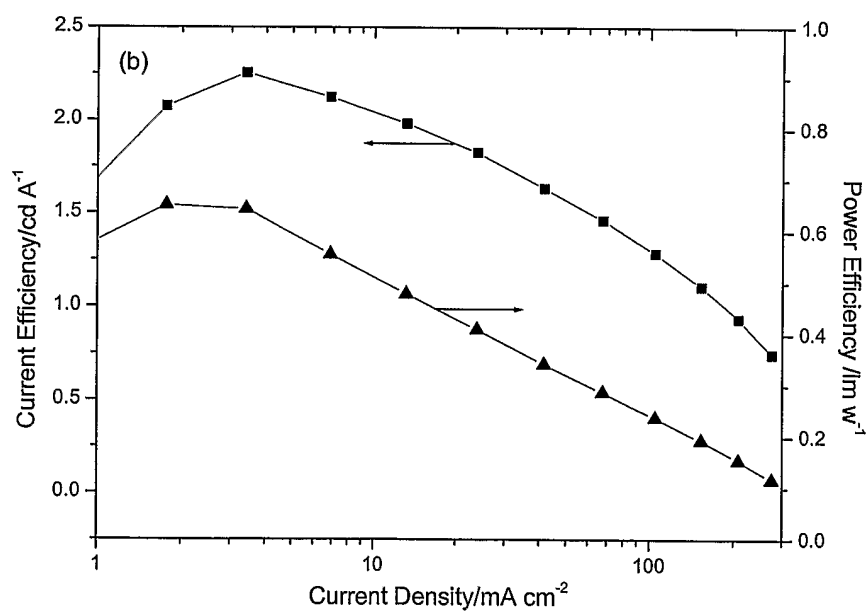


Figure 12D

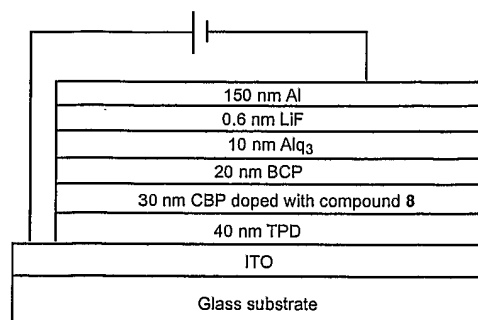


Figure 13A

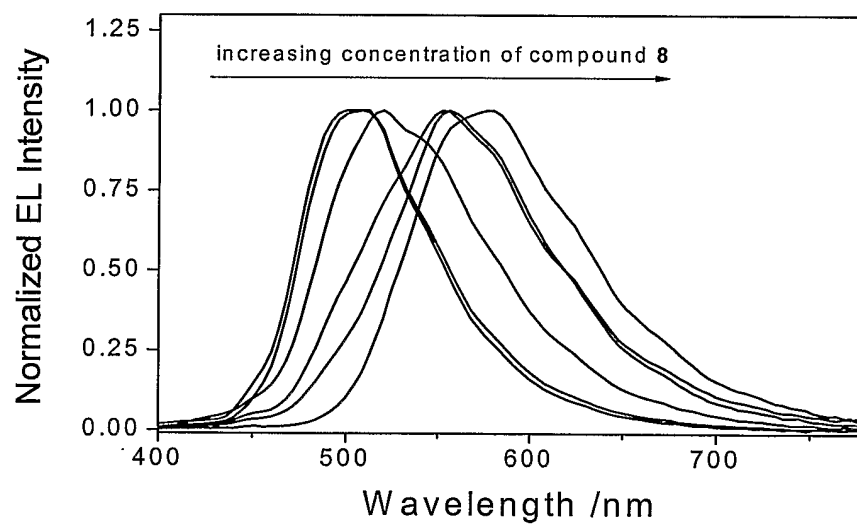


Figure 13B

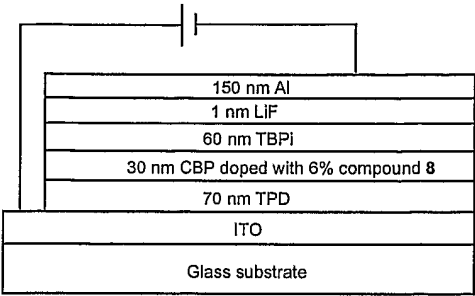


Figure 14A

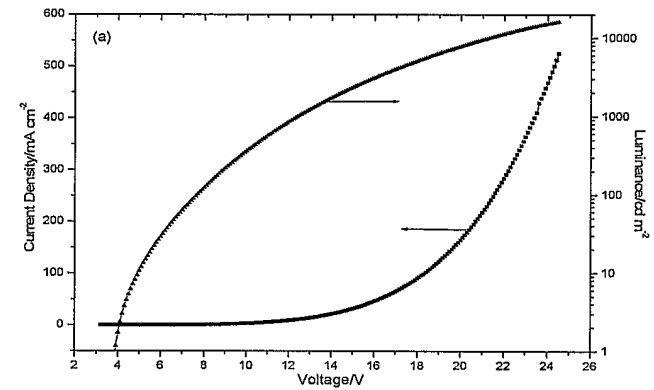


Figure 14B

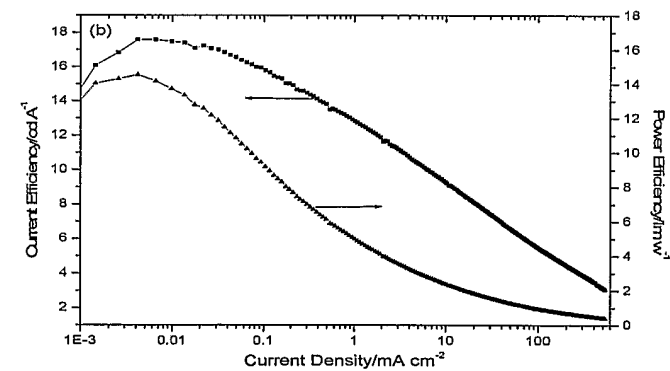


Figure 14C

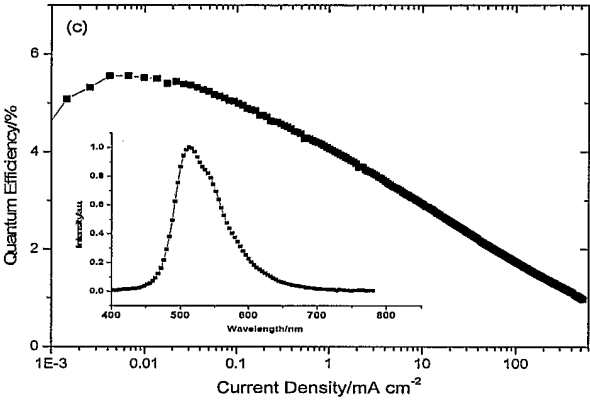


Figure 14D

INTERNATIONAL SEARCH REPORT

International application No.
PCT/CN2005/0001589

A. CLASSIFICATION OF SUBJECT MATTER

IPC7: C09K11/06, H05B33/14

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)

IPC7: C09K11/06, H05B33/14, H01L33/00, C07D213/02, C07F15/00.

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

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

STN, WPI, EPODOC, PAJ, CPRS: Au, gold, organogold, tridentate, OLEDs

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	Application of 2,6-Diphenylpyridine as a Tridentate [L] Dianionic Ligand in Organogold(III) Chemistry. Structural and Spectroscopic Properties of Mono- and Binuclear Transmetalated Gold(III) Complexes; Wong, Kar-Ho et al; Organometallics (1998), 17(16), 3505-3511.	1-7
A	CN1381545A, 27. Nov. 2002(27.11.2002), the university of Hong Kong, see whole document.	1-12
PX	A novel class of phosphorescent gold(III) alkynyl-based organic light-emitting devices with Tunable colour; Keith man-Chung Wong et al; <i>Chemical Communication</i> ; 2005, 2906-2908	1-12
PX	Luminescent Fold(III) Alkynyl Complexes: Synthesis, Structural Characterization, and Luminescence Properties; Vivial Wing-Wuh Yam et al; <i>Angew. Chem. Int. Ed</i> 2005,44,3107-3110	1-12

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents:

“A” document defining the general state of the art which is not considered to be of particular relevance

“E” earlier application or patent but published on or after the international filing date

“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)

“O” document referring to an oral disclosure, use, exhibition or other means

“P” document published prior to the international filing date but later than the priority date claimed

“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

“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

“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

“&” document member of the same patent family

Date of the actual completion of the international search
27.Dec.2005 (27.12.2005)

Date of mailing of the international search report

19 · JAN 2006 (19 · 01 · 2006)

Name and mailing address of the ISA/CN
The State Intellectual Property Office, the P.R.China
6 Xitucheng Rd., Jimen Bridge, Haidian District, Beijing, China
100088
Facsimile No. 86-10-62019451

Authorized officer

Wu, Hongxiu

Telephone No. 010-62085652



INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2005/001589

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
Claims_____relate to subject-matter considered by this Authority to be covered by the provisions of Rule 67.1 (iv) PCT.
consequently, no opinion will be formulated with respect to the industrial applicability of the subject-matter of these claims
(Art. 34(4)(a)(i) PCT)
2. ☒ Claims Nos.: 1-11 (in part)
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

see further Information sheet PCT/ISA/210
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fees, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on protest

- ☐ The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.
- ☐ The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.
- ☐ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2005/001589

Continuation of Box II.2

Present Claims 1-11 relate to an extremely large number of possible compounds. Support within the meaning of Article 6 PCT and/or disclosure within the meaning of Article 5 PCT is to be found, however, for only a very small proportion of the compounds. In present case, the claims so lack support, and the application so lacks disclosure, that a meaningful search over the whole of the claimed scope is impossible. Consequently, the search has been carried out for those parts of the claims which appear to be supported and disclosed, namely part of claims 1-11 and claim 12.

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/CN2005/001589

Patent Documents referred in the Report	Publication Date	Patent Family	Publication Date
CN1381545A	27.11.2005	US2002179885A1	05.12.2002
		JP2002363552A	18.12.2002