(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 1 791 927 B1

(12) **EUROPEAN PATENT SPECIFICATION**

(45) Date of publication and mention
of the grant of the patent:
11.01.2012 Bulletin 2012/02

(21) Application number: **05856354.5**

(22) Date of filing: **23.09.2005**

(51) Int Cl.:
***C09K 11/06** (2006.01)*

(86) International application number:
PCT/KR2005/003169

(87) International publication number:
WO 2006/080637 (03.08.2006 Gazette 2006/31)

(54) NEW COMPOUND AND ORGANIC LIGHT EMITTING DEVICE USING THE SAME(6)

NEUE VERBINDUNG UND ORGANISCHE LICHTEMITTIERENDE VORRICHTUNG DAMIT (6)

NOUVEAU COMPOSE ET DISPOSITIF ELECTROLUMINESCENT ORGANIQUE UTILISANT CE COMPOSE (6)

(84) Designated Contracting States:
DE GB NL

(30) Priority: **24.09.2004 KR 20040077214**

(43) Date of publication of application:
06.06.2007 Bulletin 2007/23

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- **TRITSCHLER W ET AL: "SYNTHESE UND KONFORMATION VON SPIROACRIDANEN// SYNTHESIS AND CONFORMATION OF SPIROACRIDANS" CHEMISCHE BERICHTE, VERLAG CHEMIE GMBH. WEINHEIM, DE, vol. 117, 1 January 1984 (1984-01-01), pages 2703-2713, XP008051454 ISSN: 0009-2940**
- **TRITSCHLER ET AL. CHEMISCHE BERICHTE vol. 117, no. 8, 1984, pages 2703 - 2713, XP008095206**

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Description

Technical Field

[0001] The present invention relates to an organic light emitting device which comprises a fluorene derivative capable of significantly improving a lifespan, efficiency, and electrochemical and thermal stabilities thereof.

Background Art

[0002] An organic light emission phenomenon is an example of a conversion of current into visible rays through an internal process of a specific organic molecule. The organic light emission phenomenon is based on the following mechanism. When organic material layers are interposed between an anode and a cathode, if voltage is applied between the two electrodes, electrons and holes are injected from the cathode and the anode into the organic material layer. The electrons and the holes which are injected into the organic material layer are recombined to form an exciton, and the exciton is reduced to a bottom state to emit light. An organic light emitting device which is based on the above mechanism typically comprises a cathode, an anode, and organic material layer(s), for example, organic material layers including a hole injection layer, a hole transport layer, a light emitting layer, and an electron transports layer; interposed therebetween.

[0003] The materials used in the organic light emitting device are mostly pure organic materials or complexes of organic material and metal. The material used in the organic light emitting device may be classified as a hole injection , material, a hole transport material, a light emitting material, an electron transport material, or an electron injection material, according to its use. In connection with this, an organic material having a p-type property, which is easily oxidized and is electrochemically stable when it is oxidized, is mostly used as the hole injection material or the hole transport material. Meanwhile, an organic material having an n-type property, which is easily reduced and is electrochemically stable when it is reduced, is used as the electron injection material or the electron transport material. As the light emitting layer material, an organic material having both p-type and n-type properties is preferable, which is stable when it is oxidized and when it is reduced. Also a material having high light emission efficiency for conversion of the exciton into light when the exciton is formed is preferable.

[0004] In addition, it is preferable that the material used in the organic light emitting device further have the following properties.

[0005] First, it is preferable that the material used in the organic light emitting device have excellent thermal stability. The reason is that joule heat is generated by movement of electric charges in the organic light emitting device. NPB, which has recently been used as the hole transport layer material, has a glass transition temperature of 100°C or lower, thus it is difficult to apply to an organic light emitting device requiring a high current.

[0006] Second, in order to produce an organic light emitting device that is capable of being actuated at low voltage and has high efficiency, holes and electrons which are injected into the organic light emitting device must be smoothly transported to a light emitting layer, and must not be released out of the light emitting layer. To achieve this, a material used in the organic light emitting device must have a proper band gap and a - proper HOMO and LUMO energy levels. A LUMO energy level of PEDOT:PSS, which is currently used as a hole transport material of an organic light emitting device produced using a solution coating method, is lower than that of an organic material used as a light emitting layer material, thus it is difficult to produce an organic light emitting device having high efficiency and a long lifespan.

[0007] Moreover, the material used in the organic light emitting device must have excellent chemical stability, electric charge mobility, and interfacial characteristic with an electrode or an adjacent layer. That is to say, the material used in the organic light emitting device must be little deformed by moisture or oxygen. Furthermore, proper hole or electron mobility must be assured so as to balance densities of the holes and of the electrons in the light emitting layer of the organic light emitting device to maximize the formation of excitons. Additionally, it has to be able to have a good interface with an electrode including metal or metal oxides so as to assure stability of the device.

[0008] Accordingly, there is a need to develop an organic light emitting device including an organic material having the above-mentioned requirements in the art.

Disclosure of Invention

Technical Problem

[0009] Therefore, the object of the present inventions is to provide an organic light emitting device which is capable of satisfying conditions required of a material usable for an organic light emitting device, for example, a proper energy level, electrochemical stability, and thermal stability, and which includes a fluorene derivative having a chemical structure capable of playing various roles required in the organic light emitting device, depending on a substituent group.

The document WO 2004/020371 discloses fluorene compounds wherein the two benzene rings are substituted with arylamine moities. These compounds are applied in organic light-emitting devices in the light emitting layer or as hole transporting material.
The document-US 2004/021-9386 discloses triarylamine derivatives suitable as hole-transporting materials in organic light-emitting devices.

Technical Solution

[0010] The present invention provides an organic light emitting device which comprises a first electrode, organic material layers comprising a light emitting layer and one of a hole-injection layer, a hole-transport layer and a layer which both injects and transports holer; and a second electrode; wherein the first electrode, the organic material layers and the second electrode form a layered structure and the hole-injection layer, the hole-transport layer or the layer which both injects and transports holes includes a compound according to claim 1.

Brief Description of the Drawings

[0011] FIG. 1 illustrates an organic light emitting device comprising a substrate 1, an anode 2, a light emitting layer 3; and a cathode 4: and

[0012] FIG. 2 illustrates an organic light emitting device comprising a substrate 1, an anode 2, a hole injection layer 5, a hole transport layer 6, a light emitting layer 7, an electron transport layer 8, and a cathode 4.

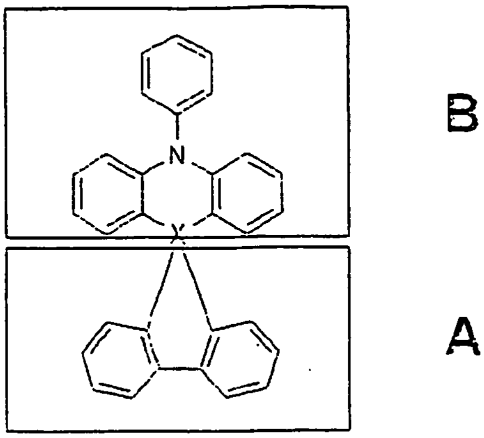
Best Mode for Carrying Out the Invention

[0013] Hereinafter, a detailed description will be given of the present invention.

[0014] Various substituent groups can be introduced into the core structure of the compound contained in an organic material layer. The core structure contains a fluorene group bonded to a combination of an acridine group and a carbazolyl group to form a spiro structure, thereby the compound has characteristics suitable for use in an organic material layer of the organic light emitting device. This will be described in detail, below.

[0015] The steric core structure of the compound can be divided into two portions, A and B; for explanation, as shown in the following figure.

[0016]



[0017] The compound has the steric core structure in which a plane A meets with a plane B at right angles around X (a carbon atom), and conjugation does not occur between the A and B portions around X. Furthermore, since one nitrogen atom is positioned among three aryl groups in the plane B, conjugation is limited in the plane B.

[0018] The conjugation length of the compound has a close relationship with an energy band gap. In detail, the energy band gap is reduced as the conjugation length of the compound increases. As described above, since a conjugation structure is limited in the core structure of the compound, the core structure has a large energy band gap.

[0019] Additionally, by introducing various substituent groups into the core structure, compounds having intrinsic characteristics of the substituent groups can be obtained. Particularly, since the core structure of the compound includes the arylamine structure, it has an energy level suitable for the hole injection and/or hole transport materials in the organic light emitting device. In the present invention, the compound having the proper energy level is selected depending on the substituent groups; thereby it is possible to realize a device having a low actuating voltage and a high light efficiency,

[0020] Furthermore, various substituent groups can be symmetrically introduced into the core structure (the A and B portions are located at both sides of the core structure) so as to precisely control the energy band gap, improve interfacial characteristics with organic materials, and apply the compound to various fields.

[0021] Meanwhile, since the compound has a high glass transition temperature (Tg), it has excellent thermal stability. For example, the glass transition temperature of the compound of Formula 2-4 is 1.48°C, which is still higher than that of conventionally used NPB (Tg: 96°C). Such increase in thermal stability is an important factor providing actuating stability to the device.

[0022] Furthermore, the compound may be used to form the organic material layer using a vacuum deposition process or a solution coating process during the production of the organic light emitting device. In connection with this, illustrative, but non-limiting, examples of the solution coating process include a spin coating process, a dip coating process, an inkjet printing process, a screen printing process, a spray process, and a roll coating process.

[0023] For example, the compound has excellent solubility in a polar solvent, such as xylene, dichloroethane, or NMP, which is used during the production of the device, and forms a thin film very well through the process using a solution, thus the solution coating process may be applied to produce the device.

[0024] Tertiary alcohol, which is produced by a reaction of a lithiated aryl and keto group, is heated in the presence of an acid catalyst to form a hexagonal cyclic structure while water is removed, thereby producing the compound having a spiro structure according to the present invention. The above-mentioned procedure for producing the compound is well known in the art, and those skilled in the art can change the production conditions during the production of the compound. The production will be described in detail in the preparation examples later.

[0025] The organic light emitting device of the present invention can be produced using known materials through a known process, modified only in that the hole-injection layer, the hole-transport layer or the layer which both injects and transports holes includes the compound of the present invention.

[0026] The organic light emitting device of the present invention may comprise a hole injection layer, a hole transport layer, a light emitting layer, an electron transport layer, and an electron injection layer as the organic material layers. However, the structure of the organic light emitting device is not limited to this, but may comprise a smaller number of organic material layers.

[0027] Furthermore, the organic light emitting device of the present invention may be produced, for example, by sequentially layering a first electrode, organic material layers, and a second electrode on a substrate. In connection with this, a physical vapor deposition (PVD) method, such as a sputtering method or an e-beam evaporation method, may be used, but the method is not limited to these

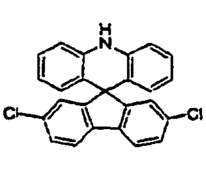
[0028] A method of producing the compound of the invention and the preparation of the organic light emitting device using the same will be described in detail in the following preparation examples and examples. However, the following preparation examples and examples are set forth to illustrate, but are not to be construed to limit the present invention.

Mode for the Invention

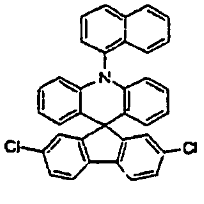
[0029] A better understanding of a method of producing an organic compound according to the invention and the preparation of an organic light emitting device using the same may be obtained in light of the following preparation examples and examples which are set forth to illustrate, but are not to be construed to limit the present invention.

[0030] In order to produce the compound, any one of the compounds of the following Formulae a to g may be used as a starting material.

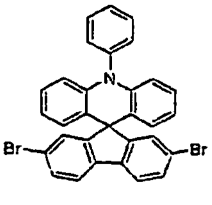
[0031]



[Formula a]



[Formula b]



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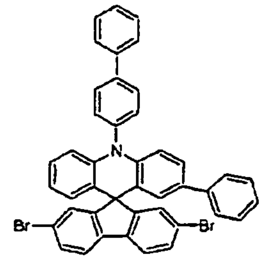
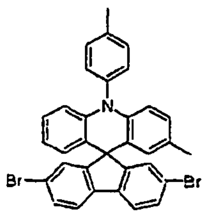
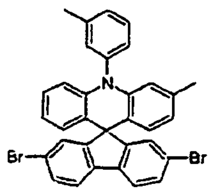


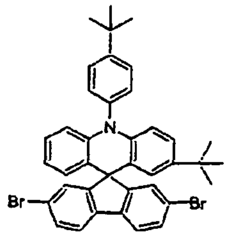
[Formula d]



[Formula e]



[Formula f]



[Formula g]

[0032] PREPARATION EXAMPLE 1: Preparation of a starting material represented by Formula a

[0033] 1) After 10 g of diphenylamine (59 mmol) and 8.04 ml of bromomethyl methyl ether (88.6 mmol) were dissloved in 100 ml of tetrahydrofuran, 12.4 ml of triethylamine (88.6 mmol) were added thereto. Sting was conducted in a nitrogen current for 5 hours, and an organic layer was then extracted using distilled water. The extracted organic layer was subjected to a column separation process at a ratio of n-hexane/tetrahydrofuran of 15:1, and vacuum dried to produce 12 g of tertiary amine (yield 90 %).

[0034] 2) The amine compound produced in 1) (12.0 g, 56.3 mmol) was dissolved in 100 ml of purified THF and cooled to -78°C, and n-BuLi (2.5 M hexane solution, 22.5 ml, 56.3 mmol) was slowly addred thereon. Stirring was conducted at the same temperature for 30 min, and a 2,7-dichloro-9-fluorenone compound (14.0 g, 56.3 mmol) was added thereto. After stirring was conducted at the same temperature for 40 min, the temperature was raised to normal temperature and stirring was carried out for an additional 3 hours. The reaction was completed in an ammonium chloride aqueous solution, and extraction was conducted with ethyl ether. Water was removed from an organic material layer using anhydrous magnesium sulfate, and an organic solvent was then removed therefrom. The produced solid was dispersed in ethanol, stirred for one day, filtered, and vacuum dried. After an intermediate material was dispersed in 100 ml of acetic acid, ten drops of concentrated sulfuric acid were dropped thereto and reflux was conducted for 4 hours. The resulting solid was filtered, washed with ethanol, and vacuum dried to produce 21.8 g of amine (96.8 % yield). MS: [M+H]+ = 401.

[0035] PREPARATION EXAMPLE 2: Preparation of a starting material represented by Formula b

[0036] A compound of Formula a (9.00 g, 22.5 mmol), 1-iodonaphthalene (11.4 g, 45.0 mmol), potassium carbonate (6.22 g, 45.0 mmol), copper iodide (214 mg, 1.13 mmol), and xylene (250 ml) were heated in a nitrogen atmosphere overnight. After cooling to normal temperature, a product was extracted with ethyl acetate, water was removed with anhydrous magnesium sulfate, and the solvent was removed at a reduced pressure. The resulting product was passed through a silica gel column using a hexane solvent to produce a compound, the solvent was removed at a reduced pressure, and vacuum drying was conducted to produce the compound of Formula b (5.0 g, 42 % yield). MS: $[M+H]^+ = 527$.

[0037] PREPARATION EXAMPLE 3: Preparation of a starting material represented by Formula c

[0038] 1) 5.08 ml of diphenylamine (30 mmol), 5.6 ml of 1-bromo-2-iodobenzene (45 mmol), 2.29 g of copper (36 mmol), and 4.98 g of potassium carbonate (36 mmol) were refluxed for 5 days. After cooling, extraction was conducted with water and ethyl acetate, water was removed with anhydrous magnesium sulfate, and a column separation process was conducted using n-hexane. Ethanol was added to achieve solidification, thereby creating 4.4 g of product (45 % yield). MS: $[M+H]^+= 323$.

[0039] 2) 1.36 g of compound produced in 1) (4.19 mmol) were dissolved in 15 ml of THF in a dried flask, and a dry ice/acetone bath was provided therein. 6.18 ml (2.5 eq) of t-butyllithium (1.7 M pentane solution) were dropped thereon for 30 min and stirring was conducted for an additional 1 hour. 1.18 g of 2,7-dibromo-9-fluorenone (3.49 mmol) were dissolved in 20 ml of THF and then injected using a syringe into a reaction solution, and stirring was conducted for an additional 1 hour. After a further stirring at normal temperature for the additional 1 hour, a saturated NH_4Cl aqueous solution was added thereto and then stirred for 30 min. Extraction was conducted with water and ethyl acetate, water was removed with anhydrous magnesium sulfate, and vacuum drying was carried out to create a product.

[0040] 3) The unpurified compound produced in 2) was dissolved in 5 ml of acetic acid, 5 drops of concentrated sulfuric acid were added therein, and stirring was conducted at 60°C for one day. After cooling, with filtration washing was conducted using water and a 5 % $NaHCO_3$ aqueous solution, and then a column separation process was conducted using a solution of n-hexane and ethyl acetate at a ratio of 9:1. Ethanol was added to the resulting substance to achieve solidification, filtration was conducted, and vacuum drying was conducted to create 1.76 g of product (89.2 %). MS: $[M+H]^+ = 566$.

[0041] PREPARATION EXAMPLE 4: Preparation of a starting material represented by Formula d

[0042] 1) 2.0 g of 4-aminobiphenyl (12 mmol) and 2.74 g of 4-bromobiphenyl (12.0 mmol) were dissolved in 50 ml of xylene, 1.72 g of sodium tert-butoxide (18 mmol), 0.1.1 g of tris(dibenzylideneacetone)dipalladium(0) (0.12 mmol), and 0.036 g of tri-tert-butylphosphine (0.18 mmol) were added thereto, and reflux was conducted in a nitrogen current for 3 hours. Distilled water was added in a reaction solution to complete a reaction, an organic layer was extracted, and water was removed with anhydrous magnesium sulfate. The solvent was removed at a reduced pressure, recrystallization was conducted using hexane, and vacuum drying was conducted to create 2.2 g of product (57 % yield). MS: $[M+H]^+$= 322.

[0043] 2) 2.2 g of compound produced in 1) (6.8 mmol), 1.25 ml of 1-bromo-2-iodobenzene (10 mmol), 1.2 g of potassium carbonate (8.8 mmol), and 0.56 g of copper (8.8 mmol) were refluxed in 5 ml of nitrobenzene in a nitrogen atmosphere for 30 hours, and then cooled to normal temperature. THF was added thereto, stirring was conducted for 30 min, an insoluble solid was filtered and thus removed, and the solvent was removed at a reduced pressure. A column separation process was conducted at a ratio of n-hexane/toluene of 3:1, recrystallization was conducted using ethanol, and vacuum drying was conducted to produce 2.4 g of compound (yield 74 %). MS : $[M+H]^+$ = 477.

[0044] 3) 2.30 g of compound produced in 2) (4.83 mmol) were dissolved in 30 ml of THF, and 5.68 ml (9.66 mmol) of t-BuLi (1.7 M pentane solution) were dropped thereon at -78°C for 30 min. After stirring was conducted for 2 hours, 1.36 g of 2,7-dibromo-9-fluorenone compound (4.03 mmol), dissolved in 8 ml of THF, were added thereto. After stirring at the same temperature for 2 hours, a cooling bath (dry ice/acetone) was removed and stirring was conducted at normal temperature for 1 hour. 16 ml of saturated ammonium chloride aqueous solution were added thereto and stirring was conducted for 30 min to complete a reaction. A product was extracted with ethyl acetate and solidification was achieved using ethanol to produce a compound. The compound was vacuum dried and then dissolved in 5 ml of acetic acid, 5 drops of concentrated sulfuric acid solution were added thereto, and stirring was conducted at 60°C overnight. After cooling to normal temperature, a solvent was removed at a reduced pressure and an organic layer was extracted with ethyl acetate. Byproducts contained in the product were removed through a column separation process using n-hexane and tetrahydrofuran at a ratio of 4:1. The product was recrystallized with ethanol and vacuum dried to create 0.77 g of product (27 %). MS: $[M+H]^+$= 718.

[0045] PREPARATION EXAMPLE 5: Preparation of a starting material represented by Formula e

[0046] 1) 1.97 g of 4,4-ditolylamine (10 mmol); 1.87 ml of 1-bromo-2-iodobenzene (15 mmol), 95 mg of copper iodide (0.5 mmol), and 4.15 g of potassium carbonate (30 mmol) were added to 100 ml of xylene and refluxed for 1 week. After cooling, water was added thereto, extraction was conducted with ethyl acetate, water was removed with anhydrous magnesium sulfate, and a column separation process was conducted using only n-hexane to create 2.219 g of product (yield 63 %). MS: $[M]^+$= 353.

[0047] 2) The resulting compound (428 mg, 1.21 mmol) was dissolved in 6 ml of purified THF in a dried flask and a dry ice/acetone bath was provided therein. 1.43 ml of t-BuLi (1.7 M pentane solution, 2.43 mmol) were dropped thereon for 30 min and stirring was conducted at the same temperature for an additional 1 hour. A 2,7-dibromo-9-fluorenone compound (338 mg, 1.00 mmol) was dissolved in 6 ml of THF and then injected using a syringe into a reaction solution, and stirring was conducted for an additional 1 hour. After the stirring at normal temperature for the additional 1 hour, a saturated ammonium chloride aqueous solution was added thereto to complete the reaction, and then stirring was conducted for 30 min. Extraction was conducted with ethyl acetate, water was removed with anhydrous magnesium sulfate, and vacuum drying was carried out.

[0048] 3) The compound produced in 2) was dissolved in 5 ml of acetic acid, 5 drops of concentrated sulfuric acid were loaded thereon, and stirring was conducted at 60°C for one day. After cooling to normal temperature, washing with filtration was conducted using water and a 5 % sodium bicarbonate aqueous solution, and a column separation process was conducted using a solution of n-hexane and tetrahydrofuran at a ratio of 9:1. Ethanol was used to achieve solidification, and filtration and drying were conducted (528 mg, yield 89 %). MS : $[M+H]^+$= 594.

[0049] PREPARATION EXAMPLE 6: Preparation of a starting material represented by Formula f

[0050] 1) 2.28 ml of m,m'-ditolylamine (12 mmol), 2.26 ml of 1-bromo-2-iodobenzene (18.0 mmol), 991 mg of copper (1.3 eq), 2.16 g of potassium carbonate (15.6 mmol), and 1 ml of nitrobenzene were refluxed for 3 days. After cooling, nitrobenzene was distilled at a reduced pressure and thus removed, extraction was conducted with water and ethyl acetate, and water was removed from an organic layer using anhydrous magnesium sulfate. After the organic solvent was removed, a column separation process was conducted using a solvent of n-hexane and ethyl acetate at a ratio of 50:1, and ethanol was used to achieve solidification. MS: $[M+H]^+$= 353 (-Br), 400 (-I). Yield: 68% (2.9 g).

[0051] 2) 779 mg of compound produced in 1) (2.21 mmol) were dissolved in 8 ml of purified THF in a dried flask, and a dry ice/acetone bath was provided therein. 2.6 ml (4.4 mmol) of t-butyllithium (1.7 M pentane solution) were dropped thereon for 30 min and further stirring was conducted for an additional 1 hour. 0.622 g of 2,7-dibromo-9-fluorenone (1.83 mmol) were dissolved in 9 ml of THF and then injected using a syringe into a reaction solution, and stirring was conducted at the same temperature for an additional I hour. After the stirring at normal temperature for the additional 1 hour, a saturated ammonium chloride aqueous solution was added thereto and then stirred for 30 min. Extraction was conducted with water and ethyl acetate, water was removed from an organic layer using anhydrous magnesium sulfate, and vacuum drying was carried out.

[0052] 3) The unpurified compound produced in 2) was dissolved in 5 ml of acetic acid, 5 drops of concentrated sulfuric acid were added thereto, and stirring was conducted at 60°C for one day. After cooling, washing was conducted during filtration using water and a 5 % $NaHCO_3$ aqueous solution, and a column separation process was conducted using a solution of n-hexane and ethyl acetate at a ratio of 9:1. Ethanol was used for solidification, and filtration and vacuum drying were conducted. MS: $[M+H]^+$= 594. Yield: 73.9% (0.807 g).

[0053] PREPARATION EXAMPLE 7: Preparation of a starting material represented by Formula g

[0054] 1) 2-bromoaniline (800 mg, 4,70 mmol), 1-tert-butyl-4-iodobenzene (1 ml, 5.64 mmol), potassium hydroxide (1.06 g, 18.8 mmol), copper chloride (19 mg, 0.16 mmol), 1,10-phenanthroline (34 mg, 0.16 mmol), and xylene (16 ml) were heated in a nitrogen atmosphere overnight. After the resulting reactants were cooled to normal temperature, a product was extracted with ethyl acetate, water was removed with anhydrous magnesium sulfate, and the solvent was removed at a reduced pressure. The resulting product was passed through a silica gel column using a hexane solvent to produce secondary amine and tertiary amine. A mixture of secondary amine and tertiary amine was vacuum dried, added to toluene (15 ml) along with 1-tert-butyl-4-iodobenzene (0.8 mL, 4.42 mmol), copper iodide (35 mg, 0.18 mmol), and potassium carbonate (763 mg, 5.52 mmol), and heated in a nitrogen atmosphere overnight. They were cooled to normal temperature and extracted with ethyl acetate, water was removed with anhydrous magnesium sulfate, and the solvent was removed at a reduced pressure. The product was passed through a silica gel column using a hexane solvent, the solvent was removed at a reduced pressure, and vacuum drying was conducted to produce a desired white solid (0.67 g, 33 %). MS: $[M+H]^+$= 437.

[0055] 2) 0.5 g of compound produced in 1) (1.2 mmol) were dissolved in 6 ml of purified THF in a dried flask, and a dry ice/acetone bath was provided therein. t-BuLi (1.7 M pentane solution, 1.35 ml, 2.3 mmol) was dropped thereon at -78°C for 30 min. After the reactants were stirred for 1 hour, 338 mg of 2,7-dibromo-9-tluorenone (1 mmol) were dissolved in 5 ml of THF and then injected using a syringe into a reaction solution, and stirring was conducted at the same temperature for 1 hour. A dry ice/ acetone bath was removed and stirring was conducted at normal temperature for 1 hour. Saturated ammonium chloride aqueous solution (10 ml) was added thereto and stirring was conducted for 30 min to complete a reaction. The resulting product was extracted with ethyl acetate and a column separation process was conducted using a solvent of n-hexane and ethyl acetate at a ratio of 9:1 to produce a compound.

[0056] 3) The compound produced in 2) was vacuum dried and dissolved in acetic acid (5 ml), a concentrated sulfuric acid solution (3 drops) was added thereto, and stirring was conducted at 60°C for one day. After the reactants were cooled to normal temperature, a product was filtered using a filter paper and washed with water. Byproducts contained in the product were removed through a column separation process using a solvent of n-hexane and ethyl acetate at a ratio of 9:1, and vacuum drying was conducted to create 0.31 g of product (yield 36 %). MS: $[M+H]^+$= 678.

[0057] EXAMPLE 1: Preparation of the compound represented by Formula 2-1

[0058] After 5.02 g of the compound of Formula c (8.88 mmol) and 1.81 g of diphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 2-1 (5.2 g, yield 78.9 %). MS: $[M+H]^+$= 742.

[0059] EXAMPLE 2: Preparation of the compound represented by Formula 3-1

[0060] After 4.68 g of compound of Formula b (8.88 mmol) and 1.81 g of diphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 3-1 (5.1 g, yield 72.5 %). MS: $[M+H]^+$= 793.

[0061] EXAMPLE 3: Preparation of the compound represented by Formula 3-2

[0062] After 4.68 g of the compound of Formula b (8.88 mmol) and 2.34 g of N-phenyl-1-naphthylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone) palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 3-2 (5.6 g, yield 71 %). MS: $[M+H]^+$= 893.

[0063] EXAMPLE 4: Preparation of the compound represented by Formula 26-1

[0064] After 5.28 g of the compound of Formula e (8.88 mmol) and 1.81 g of diphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 26-1 (4.9 g, yield 71.7 %). MS: $[M+H]^+= 771$.

[0065] EXAMPLE 5: Preparation of the compound represented by Formula 27-1

[0066] After 5.27 g of the compound of Formula f (8.88 mmol) and 1.81 g of diphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 27-1 (5.0 g, yield 73 %). MS: $[M+H]^+= 771$.

[0067] EXAMPLE 6: Preparation of the compound represented by Formula 28-1

[0068] After 6.02 g of the compound of Formula g (8.88 mmol) and 1.81 g of diphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 28-1 (5.6 g, yield 73.8 %), MS: $[M+H]^+= 855$.

[0069] EXAMPLE 7: Preparation of the compound represented by Formula 29-1

[0070] After 6.37 g of the compound of Formula d (8.88 mmol) and 1.81 g of diphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.32 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 2 hours. Distilled water was added to the reaction solution to complete the reaction, and the organic layer was extracted. A column separation process was conducted using a solvent of n-hexane and tetrahydrofuran at a ratio of 9:1, stirringstirtingucted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 29-1 (6.2 g, yield 77 %). MS: $[M+H]^+= 895$.

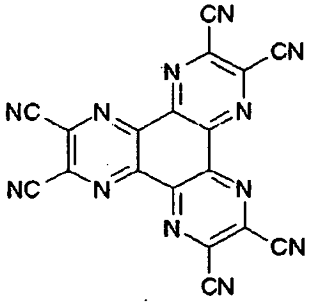
[0071] EXAMPLE 8: Production of an organic light emitting device

[0072] A glass substrate (coming 7059 glass), on which ITO (indium tin oxide) was applied to a thickness of 1000 Å to form a thin film, was put in distilled water, in which a detergent was dissolved, and washed using ultrasonic waves. In connection with this, a product manufactured by Fischer Inc. was used as the detergent, and distilled water was produced by filtering twice using a filter manufactured by Millipore Inc. After ITO was washed for 30 min, ultrasonic washing was conducted twice using distilled water for 10 min. After the washing using distilled water was completed, ultrasonic washing was conducted using isopropyl alcohol, acetone, and methanol solvents, and drying was then conducted. Next, it was transported to a plasma washing machine. The substrate was dry washed using nitrogen plasma under a pressure of 14 mtorr at 85 W for 5 min, and then transported to a vacuum evaporator.

[0073] Hexanitrile hexaazatriphenylene (hereinafter, referred to as "HAT") of the following Formula was vacuum deposited to a thickness of 500 Å by heating on a transparent ITO electrode, which was prepared through the above procedure, so as to form an anode including an ITO conductive layer and an N-type organic material.

[HAT]

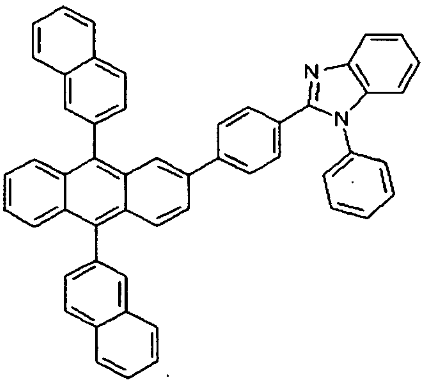
[0074]



[0075] The compound of Formula 28-I (400 Å) was vacuum deposited thereon to form a hole transport layer. Alq3 was vacuum deposited to a thickness of 300 Å on the hole transport layer to form a light emitting layer. An electron transport layer material of the following Formula was deposited to a thickness of 200 Å on the light emitting layer to form an electron transport layer.

[0076] [Electron transport layer material]

[0077]



[0078] Lithium fluoride (LiF) having a thickness of 12 Å and aluminum having a thickness of 2000 Å were sequentially deposited on the electron transport layer to form a cathode.

[0079] In the above procedure, the deposition speed of an organic material was maintained at 0.3 - 0.8 Å/sec. Furthermore, lithium fluoride and aluminum were deposited at speeds of 0.3 Å/sec and 1.5 - 2.5 Å/sec, respectively, on the cathode. During the deposition, a vacuum was maintained at 1 - 3 X 10^{-7}.

[0080] The resulting device had an electric field of 5.47 V at a forward current density of 100 mA/cm^2, and emitted green light at a light efficiency of 2.91 1m/W. The operation and light emission of the device at the above-mentioned actuating voltage mean that the compound of Formula 28-1, which formed the layer between the hole injection layer and the light emitting layer, functions to transport holes.

[0081] EXAMPLE 9: Production of an organic light emitting device

[0082] The procedure of example 1 was repeated to produce an organic light emitting device except that a HAT thin film (80 Å: used to improve interfacial characteristics) was formed on an ITO transparent electrode which was prepared through the same procedure as example 7 and a compound of Formula 28-1 was deposited thereon to a thickness of 1100 Å to form a layer for both injecting and transporting holes.

[0083] The resulting device had an electric field of 6.75 V at a forward current density of 100 mA/cm^2, and emitted green light at a light efficiency of 2.19 lm/W. The operation and light emission of the device at the above-mentioned actuating voltage mean that the compound of Formula 1-4 functions to inject and transport the holes, and it can be seen that, even when the layer for bothe injecting and transporting the holes is formed using the compound of the present invention, it is possible to assure actuation at low voltage and excellent light efficiency.

[0084] EXAMPLE 10: Production of an organic light emitting device

[0085] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 2-1.

[0086] The resulting device had an electric field of 5.63 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 1.83 lm/W.

[0087] EXAMPLE 11: Production of an organic light emitting device

[0088] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 3-1.

[0089] The resulting device had an electric field of 5.77 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 1.66 lm/W.

[0090] EXAMPLE 12: Production of an organic light emitting device

[0091] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 3-2.

[0092] The resulting device had an electric field of 5.72 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 1.78 lm/W.

[0093] EXAMPLE 13: Production of an organic light emitting device

[0094] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 26-1.

[0095] The resulting device had an electric field of 5.70 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 2.1 lm/W.

[0096] EXAMPLE 14: Production of an organic light emitting device

[0097] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 27-1.

[0098] The resulting device had an electric field of 5.63 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 2.0 lm/W.

[0099] EXAMPLE 15: Production of an organic light emitting device

[0100] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 29-1.

[0101] The resulting device had an electric field of 5.80 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 1.82 lm/W.

[0102] EXAMPLE 16: Production of an organic light emitting device

[0103] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 3-1.

[0104] The resulting device had an electric field of 5.77 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 1.66 lm/W.

[0105] EXAMPLE 17: Production of an organic light emitting device

[0106] The procedure of example 9 was repeated to produce a device except that the compound of Formula 28-1 was substituted with the compound of Formula 3-2.

[0107] The resulting device had an electric field of 5.72 V at a forward current density of 100 mA, and emitted green light at a light efficiency of 1.78 lm/W.

Industrial Applicability

[0108] The compound of the present invention can be used as an organic material layer material, particularly, hole injection and/or transport materials in an organic light emitting device, and when applied to an organic light emitting device it is possible to reduce the actuating voltage of the device, to improve the light efficiency thereof, and to improve the lifespan of the device through the thermal stability of the compound.

Claims

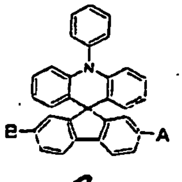
1. An organic light-emitting device comprising:

a first electrode;

organic material layers comprising a light-emitting layer and one of a hole-injection layer, a hole-transport layer and a layer which both injects and transports holes; and

a second electrode;

wherein the first electrode, the organic material layers and the second electrode form a layered structure; and the hole-injection layer, the hole-transport layer or the layer which both injects and transports holes comprises a compound represented by one of the following formulae:



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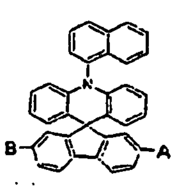

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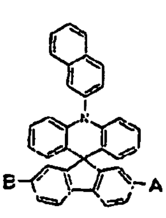
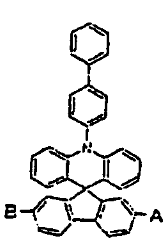
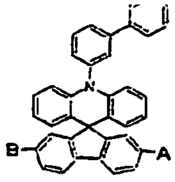
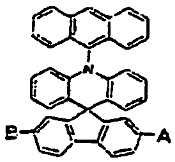
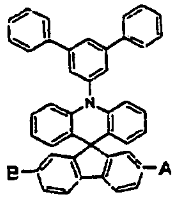
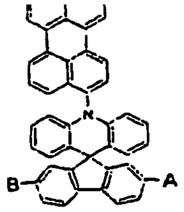


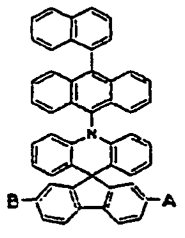


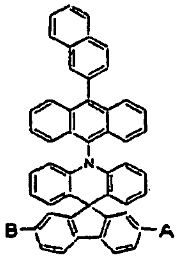


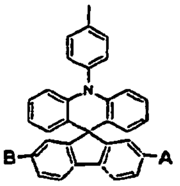


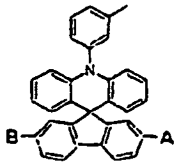


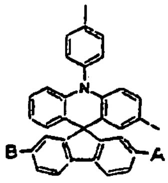


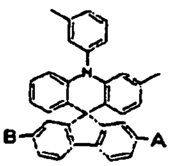


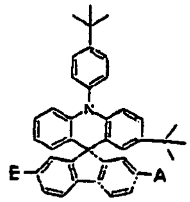


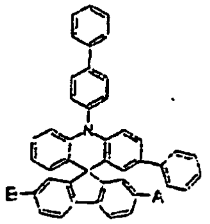




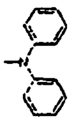


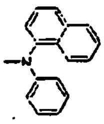


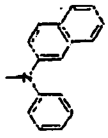


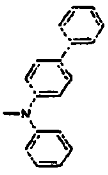


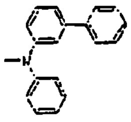
wherein A and B each independently represents one of the following groups:

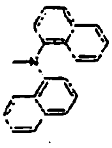


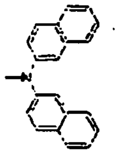


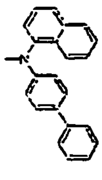


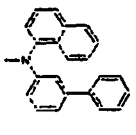


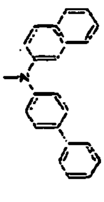


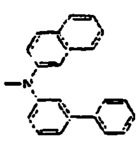


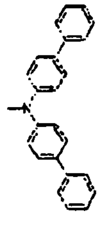


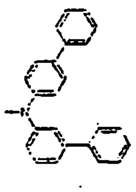


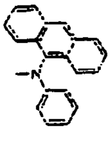


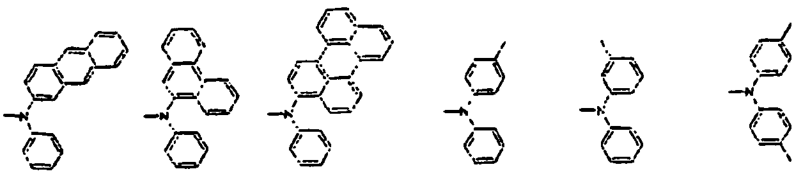


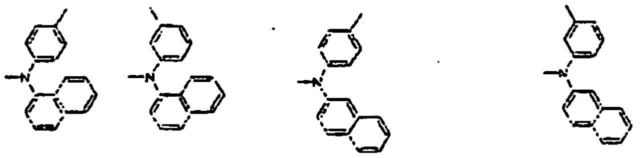








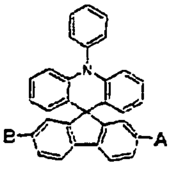




Patentansprüche

1. Organische Licht-emittierende Vorrichtung, umfassend:

   eine erste Elektrode;
   Schichten aus organischem Material, umfassend eine Licht-emittierende Schicht und eines aus einer Loch-Injektionsschicht, einer Loch-Transportschicht und einer Schicht, die sowohl Löcher injiziert als auch transportiert; und
   eine zweite Elektrode;
   wobei die erste Elektrode, die Schichten aus organischem Material und die zweite Elektrode eine Schichtstruktur bilden; und
   die Loch-Injektionsschicht, die Loch-Transportschicht oder die Schicht, die sowohl Löcher injiziert als auch transportiert, eine Verbindung umfasst, die durch eine der nachstehenden Formeln dargestellt ist:



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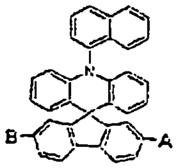

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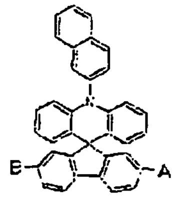
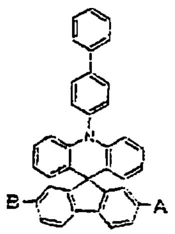
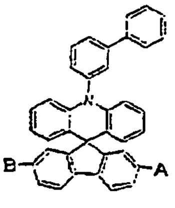
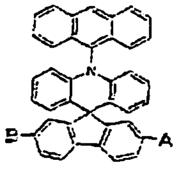
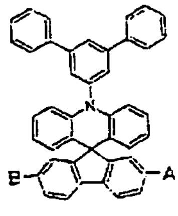
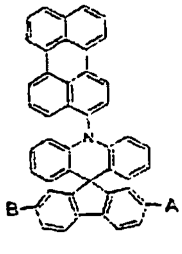


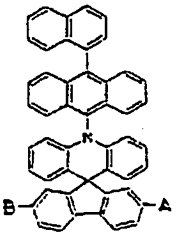


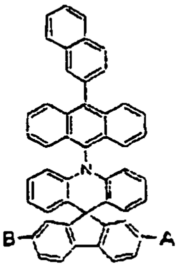


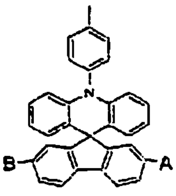


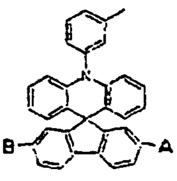


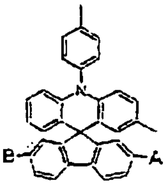


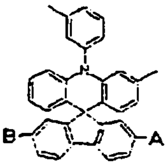


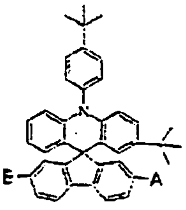


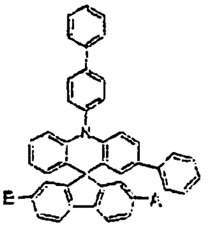




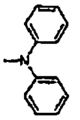


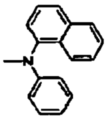


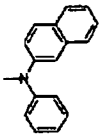


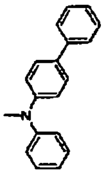


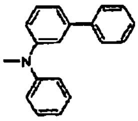
worin A und B jeweils unabhängig voneinander eine der nachstehenden Gruppen darstellen:

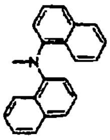


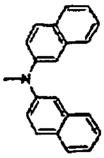


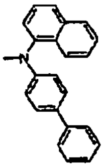


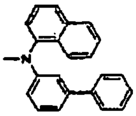


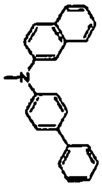


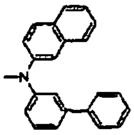


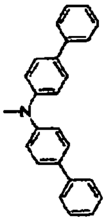


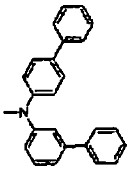


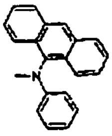


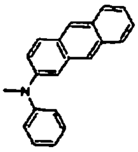


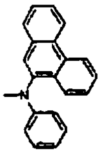


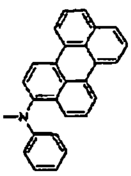


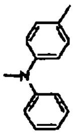


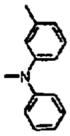


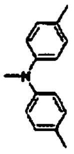


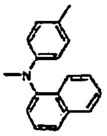


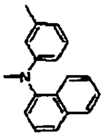


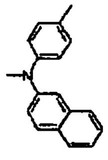


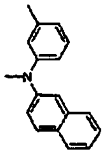








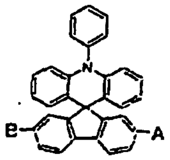




Revendications

1. Dispositif électroluminescent organique comprenant :

une première électrode ;
des couches de matière organique comprenant une couche électroluminescente et l'une d'une couche d'injection de trous, d'une couche de transport de trous et d'une couche qui à la fois injecte et transporte des trous ; et
une seconde électrode ;
dans lequel la première électrode, les couches de matière organique et la seconde électrode forment une structure stratifiée ; et
la couche d'injection de trous, la couche de transport de trous ou la couche qui à la fois injecte et transporte des trous comprend un composé représenté par une des formules suivantes :



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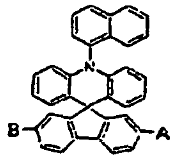

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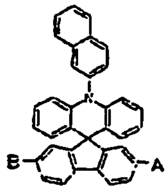
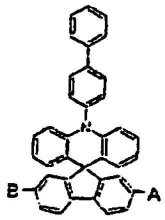
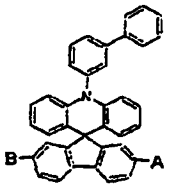
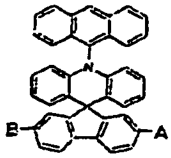
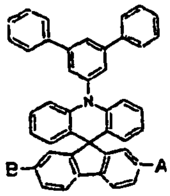
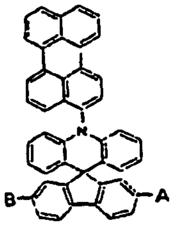


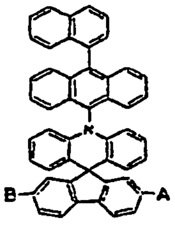


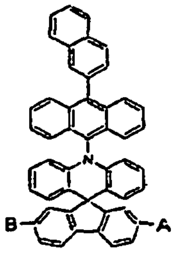


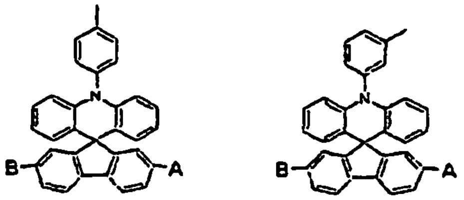


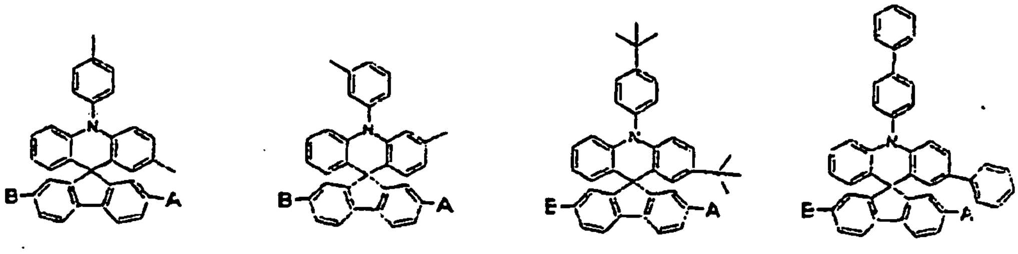




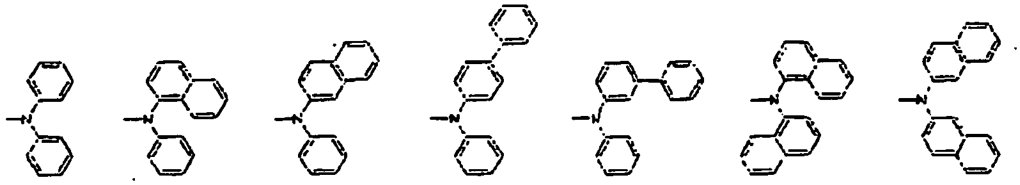


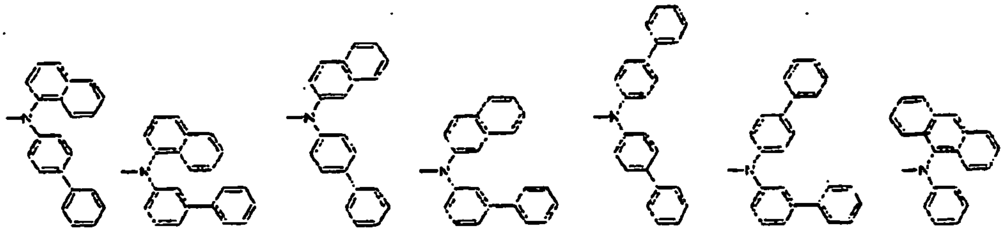


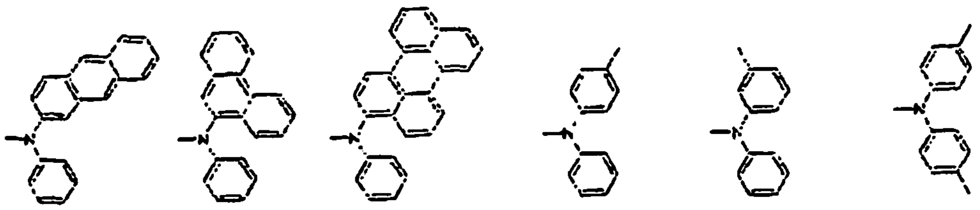


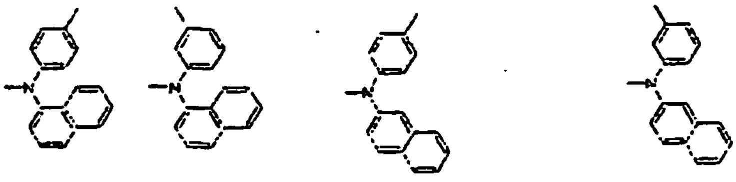


dans lesquelles A et B représentent chacun indépendamment un des groupes suivants :

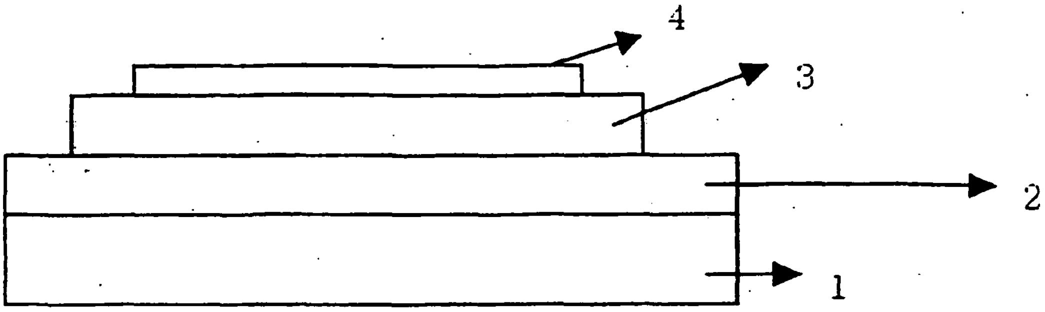




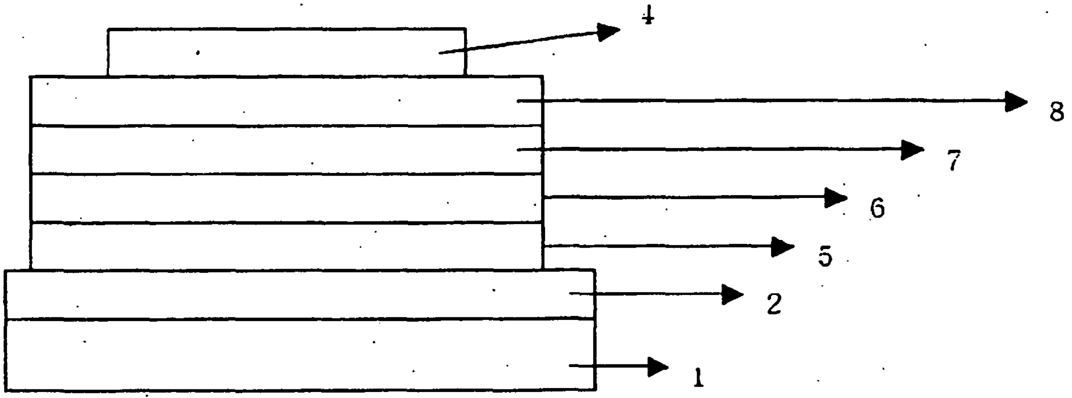




[Fig. 1]



[Fig. 2]



REFERENCES CITED IN THE DESCRIPTION

Patent documents cited in the description

- WO 2004020371 A **[0009]**
- US 20040219386 A **[0009]**

专利名称(译)	新型化合物及使用其的有机发光装置（6）		
公开(公告)号	EP1791927B1	公开(公告)日	2012-01-11
申请号	EP2005856354	申请日	2005-09-23
[标]申请(专利权)人(译)	乐金化学股份有限公司		
申请(专利权)人(译)	LG化学公司		
当前申请(专利权)人(译)	LG化学公司		
[标]发明人	YOON SEOK HEE 302 904 SONGGANG GREEN APT MOON JAE MIN 3 507 LG CHEM DORMITORY HWANG IN HO LEE MIN JEONG CHO WOOK DONG 107 1006 EXPO APT 15 3 464 1 KIM JI EUN 7 403 LG CHEM APT 381 42 JEON BYUNG SUN 1006 TAEYOUNG APT SILLIM 5 DONG		
发明人	YOON, SEOK HEE 302-904 SONGGANG GREEN APT. MOON, JAE MIN 3-507 LG CHEMISTRY DORMITORY HWANG, IN HO LEE, MIN JEONG CHO, WOOK DONG 107-1006 EXPO APT., 15/3 464-1 KIM, JI EUN 7-403 LG CHEMISTRY APT., 381-42 JEON, BYUNG SUN 1006, TAEYOUNG APT. SILLIM 5-DONG		
IPC分类号	C09K11/06		
CPC分类号	C09K11/06 C07D221/20 C09K2211/1011 C09K2211/1029 H01L51/0039 H01L51/006 H01L51/5048 H01L51/5088 H05B33/14 Y10S428/917		
优先权	1020040077214 2004-09-24 KR		
其他公开文献	EP1791927A4 EP1791927A1		
外部链接	Espacenet		

摘要(译)

公开了一种有机发光器件。 有机发光器件包括第一电极，包括发光层的有机材料层和第二电极。 第一电极，有机材料层和第二电极形成层状结构，并且有机材料层的至少一层包括其中热固性或光致的式1的化合物或式1的化合物 -介绍了可交联的官能团。