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(54) **NEW COMPOUND AND ORGANIC LIGHT EMITTING DEVICE USING THE SAME(10)**

NEUE VERBINDUNG UND DIESE VERWENDEnde ORGANISCHE LICHEMITTIERENDE VORRICHTUNG (10)

COMPOSE ET DISPOSITIF D'EMISSION LUMINEUSE ORGANIQUE LE COMPORTANT

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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 ETAL. CHEMISCHE BERICHTE vol. 117, no. 8, 1984, pages 2703 - 2713, XP008051454**

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Description**Technical Field**

5 [0001] The present invention relates to a novel compound which is capable of significantly improving a lifespan, efficiency, and electrochemical and thermal stabilities of an organic light emitting device, the production of the compound, and an organic light emitting device in which the compound is contained in an organic compound layer.

Background Art

10 [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 transport layer, interposed therebetween.

15 [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.

20 [0004] In addition, it is preferable that the material used in the organic light emitting device further have the following properties.

25 [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.

30 [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 or 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.

35 [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.

40 [0008] WO 2004/020371 describing organic light-emitting devices comprising in the light-emitting layer a condensed polycyclic compound in combination with arylamine compounds.

Disclosure of Invention**Technical Problem**

45 [0009] Accordingly, there is a need to develop an organic material having the above-mentioned requirements in the art.

[0010] Therefore, the present inventors provide an organic material which is capable of satisfying conditions required of a material which can be used for an organic light emitting device, for example, a proper energy level, electrochemical stability, and thermal stability, and which has a chemical structure capable of playing various roles required for the

organic light emitting device, depending on a substituent group.

[0011] Furthermore, the present invention aims to provide the production of a novel organic light emitting material found by the present inventors, and an organic light emitting device using the same.

5 Technical Solution

[0012] The present invention provides an organic light emitting device as defined in Claim 1. Preferred embodiments are defined in Claims 2-4.

[0013] A detailed description will be given of the substituent groups of Formula 1.

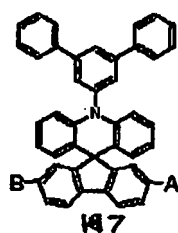
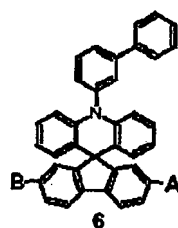
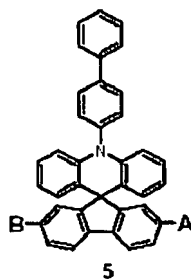
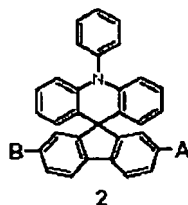
10 [0014] The carbon number of the alkyl, alkoxy, and alkenyl groups of Formula 1 is preferably 1-20.

[0015] Illustrative, but non-limiting, examples of the aryl group of R9 of Formula 1 include monocyclic aromatic rings, such as a phenyl group, a biphenyl group, a terphenyl group, and a stilbene group, and multicyclic aromatic rings, such as a naphthyl group, an anthracenyl group, a phenanthrene group, a pyrenyl group, and a perylenyl group.

15 [0016] Illustrative, but non-limiting, examples of the arylamine group of R3, R9 and R15 of Formula 1 include a diphenylamine group, a dinaphthylamine group, a dibiphenylamine group, a phenylnaphthylamine group, a phenyldiphenylamine group, a ditolylamine group, a phenyltolylamine group, a carbazolyl group, and a triphenylamine group.

[0017] Illustrative, but non-limiting, examples of the heterocyclic group of R9 of Formula 1 include a thiophenyl group, a furan group, a pyrrolyl group, an imidazolyl group, a thiazolyl group, an oxazolyl group, an oxadiazolyl group, a triazolyl group, a pyridyl group, a pyradazine group, a quinolinyl group, an isoquinoline group, and an acridyl group.

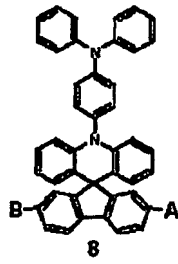
20 [0018] According to the preferred embodiment of the present invention, illustrative, but non-limiting, examples of the compound of Formula 1 include compounds of the following Formulae.



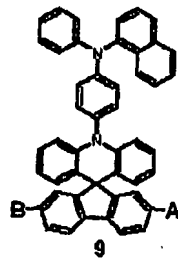
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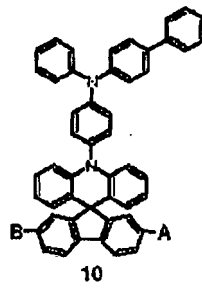


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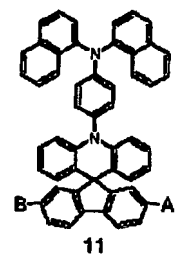
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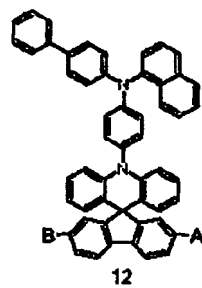
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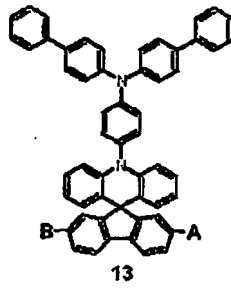


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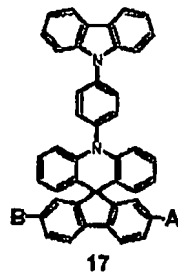
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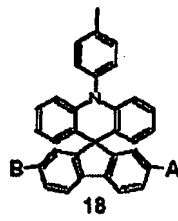
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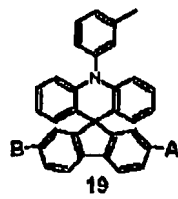
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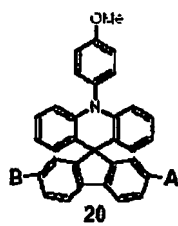
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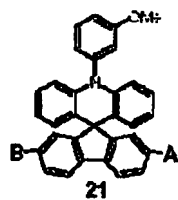


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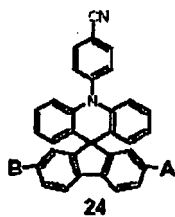


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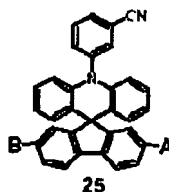


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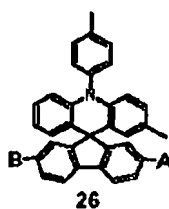


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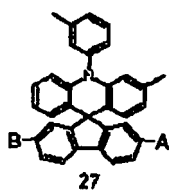
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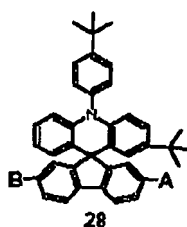
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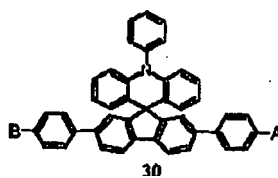


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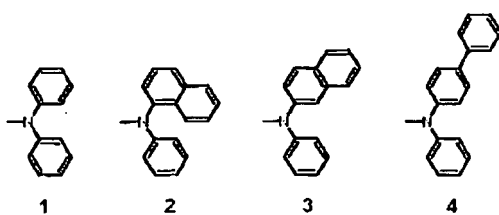
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[0019] Illustrative, but on-limiting, examples of A or B are as follows. Combination of the compounds of the previously discussed Formulae and the following substituent groups A or B can form various derivative compounds. For example, if the compound of Formula 2 is combined with the substituent group 1, the resulting product will be designated by the compound of Formula 2-1.

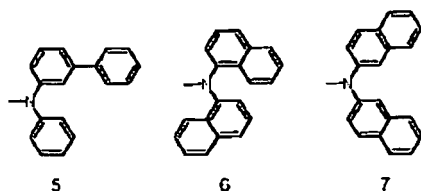
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[0020] A and B

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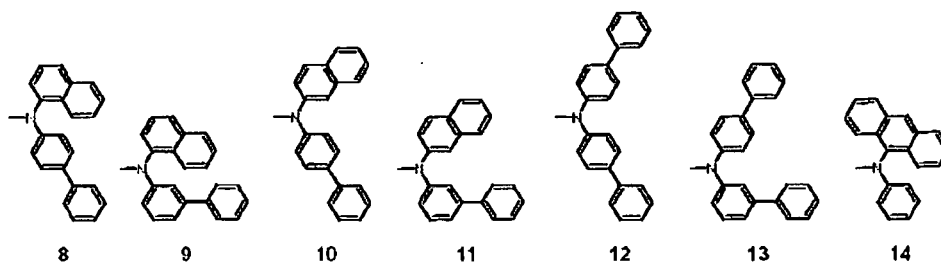


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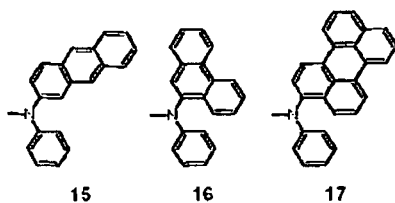
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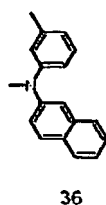
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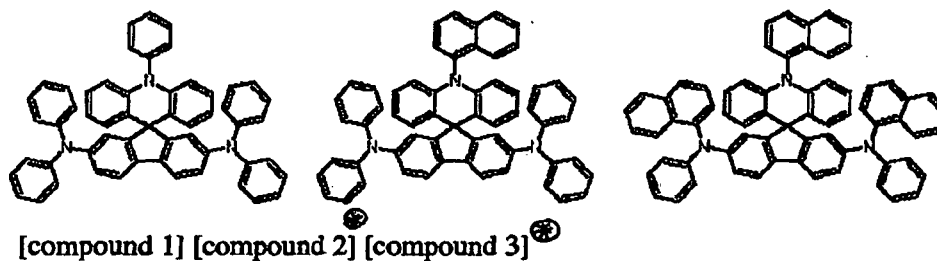
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[0021] Illustrative, but non-limiting, examples of the compound of Formula 1 include the following compounds.

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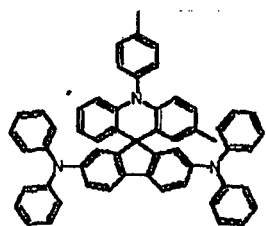


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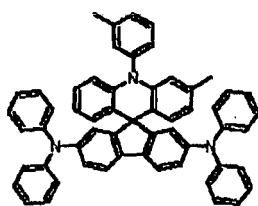
[compound 1] [compound 2] [compound 3]⁺

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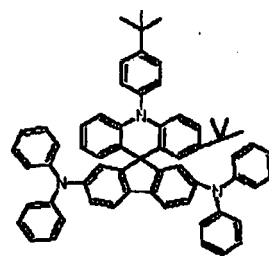
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[compound 4]



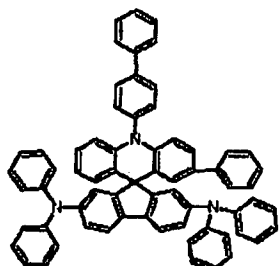
[compound 5]



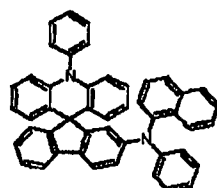
[compound 6]

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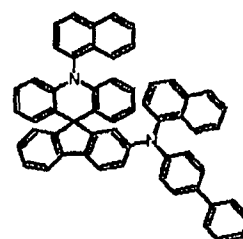
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[compound 7] *



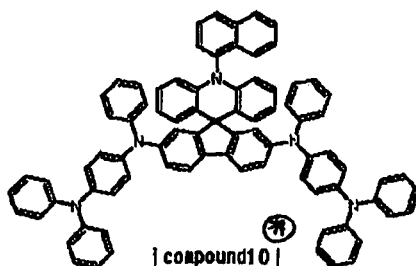
[compound 8]



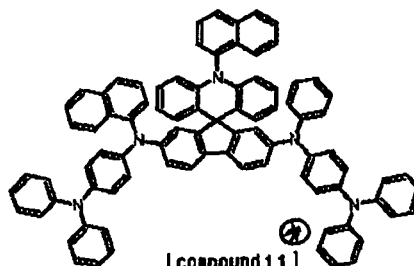
[compound 9] *

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[compound 10] *



[compound 11] *

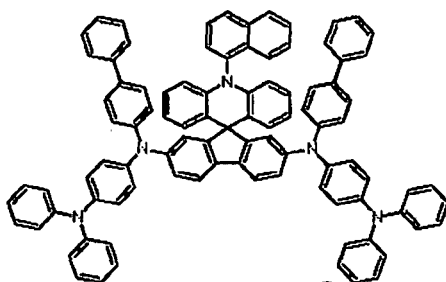
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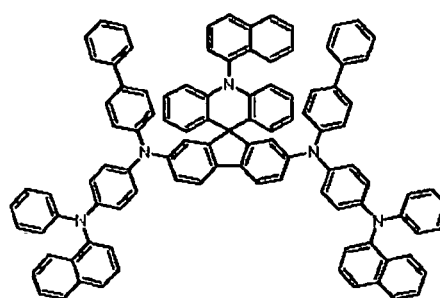
* Not according to the invention

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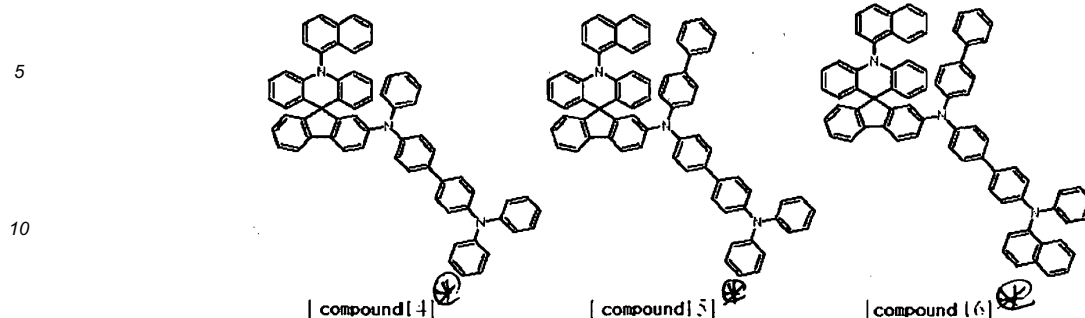
[compound 12] *



[compound 13] *

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15 * Not according to the invention

Brief Description of the Drawings

20 [0022]

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

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.

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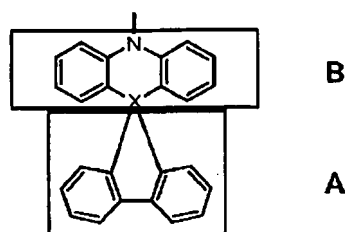
Best Mode for Carrying Out the Invention

[0023] Hereinafter, a detailed description will be given of the present invention.

30 [0024] Various substituent groups are introduced into a core structure shown in Formula 1, in detail, the core structure in which a fluorene group is bonded to a combination of an acridine group and a carbazolyl group up to form a spiro structure, thereby the compound of Formula 1 has characteristics suitable for application to an organic material layer used in an organic light emitting device. This will be described in detail, below.

[0025] The steric core structure of the compound of Formula 1, for convenience of explanation, can be divided into two portions, A and B, as shown in the following Formula.

35



45 [0026] The compound of Formula 1 has the steric core structure in which a plane A meets with a plane B at right angles around X, 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.

[0027] 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 of Formula 1, the core structure has a large energy band gap.

50 [0028] As described above, in the present invention, various substituent groups are introduced to R1 to R17 positions of the core structure having the large energy band gap so as to produce compounds having various energy band gaps. Generally, it is easy to control an energy band gap by introducing substituent groups into a core structure having a large energy band gap, but it is difficult to significantly control the energy band gap by introducing substituent groups into a core structure having a small energy band gap. Furthermore, in the present invention, it is possible to control HOMO and LUMO energy levels of the compound by introducing various substituent groups into R1 to R17 of the core structure.

55 [0029] Additionally, by introducing various substituent groups into the core structure, compounds having intrinsic characteristics of the substituent groups can be obtained. For example, substituent groups, which are frequently applied

to hole injection layer material, hole transport layer material, light emitting layer material, and electron transport layer materials during the production of the organic light emitting device, are introduced into the core structure so as to produce substances capable of satisfying the requirements of each organic material layer. Particularly, since the core structure of the compound of Formula 1 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 group among the compounds represented by Formula 1 to be used in the organic light emitting device, thereby it is possible to realize a device having a low actuating voltage and a high light efficiency.

[0030] Furthermore, various substituent groups are symmetrically or asymmetrically introduced into the core structure so as to precisely control the energy band gap, and to improve interfacial characteristics with organic materials, thereby apply the compound to various fields.

[0031] Additionally, various substituent groups are introduced into the steric structure of the compound of Formula 1 using spiro bonding to control the three-dimensional structure of the organic material so as to minimize π - π interaction in the organic material, thereby formation of excimers is prevented.

[0032] Meanwhile, since the compound of Formula 1 has a high glass transition temperature (T_g), it has excellent thermal stability. For example, the glass transition temperature of the compound of Formula 3-2 is 150°C, which is still higher than that of conventionally used NPB (T_g : 96°C). Such increase in thermal stability is an important factor providing actuating stability to the device.

[0033] Furthermore, the compound of Formula 1 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.

[0034] For example, the compound of Formula 2 has excellent solubility to 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. Additionally, a light emitting wavelength of a thin film or a solid formed using the solution coating process is typically shifted to a longer wavelength due to interaction between molecules, in comparison with a light emitting wavelength in a solution state. Little shift in the wavelength occurs in the compound having the structure shown in Formula 1.

[0035] 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 of Formula 1. The production will be described in detail in the preparation examples later.

[0036] In the organic light emitting device of the present invention, a compound, in which a thermosetting or photo-crosslinkable functional group is introduced into the compound of Formula 1, for example the compound of Formula 12, may be used instead of the compound of Formula 1. The former compound has the basic physical properties of the compound of Formula 1, and may be used to form a thin film using a solution coating process and then be cured so as to form an organic material layer during the production of the device.

[0037] The method of forming the organic material layer, which comprises introducing the curable functional group into the organic material during the production of the organic light emitting device, forming the organic thin film using the solution coating process, and curing the resulting film, is disclosed in US Pat. No. 2003-0044518 and EP Pat. No. 1146574 A2.

[0038] The above documents state that, if the organic material layer is formed through the above-mentioned method using a material having a thermosetting or photo-crosslinkable vinyl or acryl group so as to produce an organic light emitting device, it is possible to produce an organic light emitting device having a low voltage and high brightness as well as an organic light emitting device having a multilayered structure using the solution coating process. This operation mechanism may be applied to the compound of the present invention.

[0039] In the present invention, the thermosetting or photo-crosslinkable functional group may be a vinyl or an acryl group.

[0040] The organic light emitting device of the present invention can be produced using known materials through a known process, modified only in that at least one layer of organic material layer(s) include the compound of the present invention, that is, the compound of Formula 1.

[0041] The organic material layer(s) of the organic light emitting device according to the present invention may have a single layer structure, or alternatively, a multilayered structure in which at least two organic material layers are layered. For example, 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 layer(s). However, the structure of the organic light emitting device is not limited to this, but may comprise a smaller number of organic material layers.

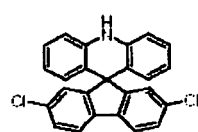
[0042] Furthermore, the organic light emitting device of the present invention may be produced, for example, by sequentially layering a first electrode, organic material layer(s), and a second electrode on a substrate. In connection with this, a physical vapor deposition (PVD) method, such as a sputtering method or an e-beam evaporation method, may be used, but the method is not limited to these.

[0043] A method of producing the compound of Formula 1 and the production 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.

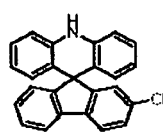
Mode for the Invention

[0044] A better understanding of a method of producing an organic compound represented by Formula 1 and the production 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.

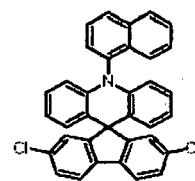
[0045] In order to produce the compound represented by Formula 1, any one of the compounds of the following Formulae, a to g, may be used as a starting material.



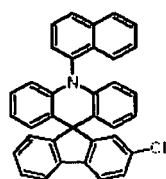
[formula a]



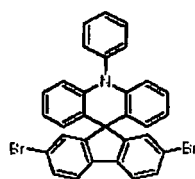
[formula b]



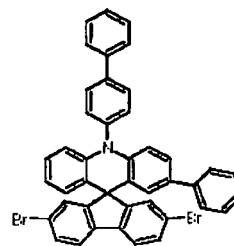
[formula c]



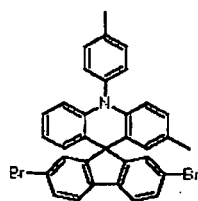
[formula d]



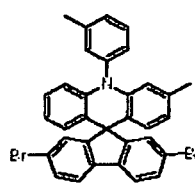
[formula e]



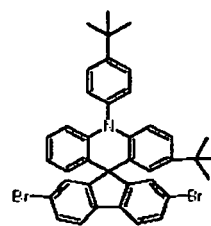
[formula f]



[formula g]



[formula h]



[formula i]

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

[0046]

1) After 10 g of diphenylamine (59 mmol) and 8.04 ml of bromomethyl methyl ether (88.6 mmol) were dissolved in 100 ml of tetrahydrofuran, 12.4 ml of triethylamine (88.6 mmol) were added thereto. Stirring was conducted in a nitrogen atmosphere 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 %).

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 dropped 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

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thereto. After stirring 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 added 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: $[M+H]^+ = 401$.

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

[0047]

1) After 10 g of diphenylamine (59 mmol) and 8.04 ml of bromomethyl methyl ether (88.6 mmol) were dissolved in 100 ml of tetrahydrofuran, 12.4 ml of triethylamine (88.6 mmol) were added thereto. Stirring was conducted in a nitrogen atmosphere 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 %).

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 dropped thereon. Stirring was conducted at the same temperature for 30 min, and a 2-chloro-9-fluorenone compound (12.1 g, 56.3 mmol) was added thereto. After stirring 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 added thereto and reflux was conducted for 4 hours. The resulting solid was filtered, washed with ethanol, and vacuum dried to produce 20 g of amine (97 % yield). MS: $[M+H]^+ = 366$.

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

[0048] 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 was conducted, 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: $[M+H]^+ = 527$.

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

[0049] A compound of Formula a (8.23 g, 22.5 mmol), iodobenzene (9.18 g, 45 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 was conducted, 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 (5.2 g, 47 % yield). MS: $[M+H]^+ = 493$.

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

[0050]

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

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% yield). MS: $[M+H]^+ = 323$.

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 the 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.

3) The unpurified compound produced in 2) was dissolved in 5 ml of acetic acid, 5 drops of concentrated sulfuric acid were loaded therein, and stirring was conducted at $60^\circ C$ for one day. After cooling, filtration was conducted, washing was conducted 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 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: $[M+H]^+ = 566$.

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

[0051]

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), 0.11 g of tris(dibenzylideneacetone)dipalladium(0) (0.12 mmol), and 0.036 g of tri-t-butylphosphine (0.18 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 3 hours. Distilled water was loaded 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: $[M+H]^+ = 322$.

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 : $[M+H]^+ = 477$.

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 $-78^\circ C$ 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 was conducted 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 $60^\circ C$ 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: $[M+H]^+ = 718$.

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

[0052]

1) 1.97 g of 4,4-ditolyamine (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: $[M]^+ = 353$.

3) 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

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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.

4) The compound produced in 3) 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, filtration was conducted, washing 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.

PREPARATION EXAMPLE 8: Preparation of a starting material represented by Formula h

[0053]

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).

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 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 1 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.

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, filtration was conducted, washing was conducted using water and a 5 % NaHCO₃ 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).

PREPARATION EXAMPLE 9: Preparation of a starting material represented by Formula i

[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.

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-fluorenone (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

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separation process was conducted using a solvent of n-hexane and ethyl acetate at a ratio of 9:1 to produce a compound.

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.

EXAMPLE 1: Preparation of the compound represented by Formula 1

[0055] After 5.02 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 1 (5.2 g, yield 78.9 %). MS: [M+H]⁺= 742.

EXAMPLE 2: Preparation of the compound represented by Formula 2 not according to the invention

[0056] After 4.68 g of 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 (5.1 g, yield 72.5 %). MS: [M+H]⁺= 793.

EXAMPLE 3: Preparation of the compound represented by Formula 3 not according to the invention

[0057] After 4.68 g of the compound of Formula c (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 (5.6 g, yield 71 %). MS: [M+H]⁺= 893.

EXAMPLE 4: Preparation of the compound represented by Formula 4

[0058] After 5.28 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 4 (4.9 g, yield 71.7 %). MS: [M+H]⁺= 771.

EXAMPLE 5: Preparation of the compound represented by Formula 5

[0059] After 5.27 g of the compound of Formula h (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 5 (5.0 g, yield 73 %). MS: [M+H]⁺= 771.

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EXAMPLE 6: Preparation of the compound represented by Formula 6

5 [0060] After 6.02 g of the compound of Formula i (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 6 (5.6 g, yield 73.8 %). MS: $[M+H]^+ = 855$.

10 EXAMPLE 7: Preparation of the compound represented by Formula 7 Not according to the invention

15 [0061] After 6.37 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 7 (6.2 g, yield 77 %). MS: $[M+H]^+ = 895$.

20 EXAMPLE 8: Preparation of the compound represented by Formula 8

25 [0062] After 4.37 g of the compound of Formula d (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 8 (5.2 g, yield 86.8 %). MS: $[M+H]^+ = 675$.

30 EXAMPLE 9: Preparation of the compound represented by Formula 9 Not according to the invention

[0063]

35 1) Synthesis of arylamine (1,4-naphthylbiphenylamine) to produce the compound represented by Formula 9: 1-aminonaphthalene (7.4 g, 51.48 mmol) and 4-bromobiphenyl (12 g, 51.48 mmol) were dissolved in 200 ml of toluene, and bis(dibenzylidene acetone)palladium(0) ($Pd(dba)_2$, 0.89 g, 1.54 mmol), 50 wt% tri-tert-butylphosphine (0.60 ml, 1.54 mmol), and sodium-tert-butoxide (9.90 g, 103.0 mmol) were added thereto. Reflux was conducted in a nitrogen atmosphere for 2 hours, and distilled water was added to the reaction solution to complete the reaction. The organic layer was extracted, a column separation process was conducted using a developing solvent of n-hexane and tetrahydrofuran (n-hexane/THF = 15/1), stirring was conducted using petroleum ether, and vacuum drying was conducted to produce arylamine (6.3 g, yield 42 %). MS: $[M+H]^+ = 295$.

40 2) After 4.37 g of the compound of Formula d (8.88 mmol) and 3.16 g of naphthylbiphenylamine (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 9 (4.5 g, yield 67.5 %). MS: $[M+H]^+ = 751$.

50 EXAMPLE 10: Preparation of the compound represented by Formula 10 not according to the invention

[0064]

55 1) Synthesis of arylamine (4-(N-phenyl-N-phenylamino)phenyl-1-phenylamine) to produce the compound represented by Formula 10: 13.5 g of 4-bromophenyl-N-phenyl-N-phenylamine (41.6 mmol) and 3.98 ml of aniline (43.7 mmol) were dissolved in 120 ml of toluene, 10.00 g of sodium-tert-butoxide (104.1 mmol), 0.48 g of bis(dibenzylidene acetone)palladium(0) (0.83 mmol), and 0.58 ml of 50 wt% tri-tert-butylphosphine toluene solution (1.25 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 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (9.6 g, yield 69 %). MS: [M+H]⁺= 336.

2) 4.68 g of compound of Formula c (8.88 mmol) and 6.86 g of 4-(N-phenyl-N-phenylamino)phenyl-1-phenylamine (20.4 mmol) were dissolved in 120 ml of toluene, 5.89 g of sodium-tert-butoxide (61.3 mmol), 0.24 g of tris(dibenzylidene acetone)dipalladium(0) (0.41 mmol), and 0.25 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.61 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 4:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 10 (5.2 g, yield 52 %). MS: [M+H]⁺= 1127.

EXAMPLE 11: Preparation of the compound represented by Formula 11 not according to the invention

[0065]

1) Synthesis of arylamine (4-(N-phenyl-N-phenylamino)phenyl-1-naphthylamine) to produce the compound represented by Formula 11: 15.0 g of 4-bromophenyl-N-phenyl-N-phenylamine (46.3 mmol) and 7.29 g of 1-naphthylamine (50.9 mmol) were dissolved in 200 ml of toluene, 13.34 g of sodium-tert-butoxide (138.8 mmol), 0.53 g of bis(dibenzylidene acetone)palladium(0) (0.93 mmol), and 0.56 ml of 50 wt% tri-tert-butylphosphine toluene solution (1.39 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 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (13 g, yield 73 %). MS: [M+H]⁺= 386.

2) 4.68 g of compound of Formula c (8.88 mmol) and 7.88 g of 4-(N-phenyl-N-phenylamino)phenyl-1-naphthylamine (20.4 mmol) were dissolved in 120 ml of toluene, 5.89 g of sodium-tert-butoxide (61.3 mmol), 0.24 g of tris(dibenzylidene acetone)dipalladium(0) (0.41 mmol), and 0.25 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.61 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 4:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 11 (5.4 g, yield 50 %). MS: [M+H]⁺= 1227.

EXAMPLE 12: Preparation of the compound represented by Formula 12 not according to the invention

[0066]

1) Synthesis of arylamine (4-(N-phenyl-N-phenylamino)phenyl-1-biphenylamine) to produce the compound represented by Formula 12: 17.4 g of 4-bromophenyl-N-phenyl-N-phenylamine (53.7 mmol) and 9.99 g of 4-aminobiphenyl (59.0 mmol) were dissolved in 250 ml of toluene, 17.02 g of sodium-tert-butoxide (177.1 mmol), 0.68 g of bis(dibenzylidene acetone)palladium(0) (1.2 mmol), and 0.72 ml of 50 wt% tri-tert-butylphosphine toluene solution (1.8 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 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (16 g, yield 73 %). MS: [M+H]⁺= 412.

2) 4.68 g of compound of Formula c (8.88 mmol) and 8.42 g of 4-(N,N-diphenylamino)phenyl-4-biphenylamine (20.4 mmol) were dissolved in 120 ml of toluene, 5.89 g of sodium-tert-butoxide (61.3 mmol), 0.24 g of tris(dibenzylidene acetone)dipalladium(0) (0.41 mmol), and 0.25 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.61 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 4:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 12 (5.2 g, yield 45.8 %). MS: [M+H]⁺= 1279.

EXAMPLE 13: Preparation of the compound represented by Formula 13 not according to the invention

[0067]

- 5 1) Synthesis of arylamine (4-(N-phenyl-N-naphthylamino)phenyl-1-biphenylamine) to produce the compound represented by Formula 13: 14.0 g of 4-bromophenyl-N-phenyl-N-naphthylamine (37.4 mmol) and 6.96 g of 4-amino-biphenyl (41.2 mmol) were dissolved in 200 ml of toluene, and 0.47 g of bis(dibenzylidene acetone)palladium(0) (0.82 mmol), 0.50 ml of 50 wt% tri-tert-butylphosphine toluene solution (1.2 mmol), and 11.86 g of sodium-tert-butoxide (123.4 mmol) were added thereto. After reflux was conducted in a nitrogen atmosphere for 2 hours, distilled water was added to the reaction solution to complete the reaction. The organic layer was extracted, a column separation process was conducted using a developing solvent of n-hexane and tetrahydrofuran at a ratio of 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (7.5 g, yield 43%). MS: $[M+H]^+ = 462$.
- 10 2) 4.68 g of compound of Formula c (8.88 mmol) and 9.44 g of 4-(N-phenyl-1-naphthylamino)phenyl-4-biphenylamine (20.4 mmol) were dissolved in 120 ml of toluene, 5.89 g of sodium-tert-butoxide (61.3 mmol), 0.24 g of tris(dibenzylidene acetone)dipalladium(0) (0.41 mmol), and 0.25 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.61 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 4:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce the compound of Formula 13 (5.5 g, yield 45 %). MS: $[M+H]^+ = 1379$.
- 15 20

EXAMPLE 14: Preparation of the compound represented by Formula 14 not according to the invention

[0068]

- 25 1) Synthesis of arylamine (4-(N,N-diphenylamino)-biphenyl-N-phenylamine) to produce the compound represented by Formula 14: 4.00 g of 4-chlorobiphenyl-N,N-diphenylamine (11.2 mmol) and 1.13 ml of aniline (12.4 mmol) were dissolved in 100 ml of toluene, 2.70 g of sodium-tert-butoxide (28.1 mmol), 0.13 g of bis(dibenzylidene acetone)palladium(0) (0.23 mmol), and 0.17 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.34 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 5 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 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (3.8 g, yield 81 %). MS: $[M+H]^+ = 413$.
- 30 35 2) 4.369 g of compound of Formula d (8.88 mmol) and 4.414 g of 4-(N,N-diphenylamino)-biphenyl-N-phenylamine (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 14 (5.2 g, yield 65 %). MS: $[M+H]^+ = 869$.
- 40

EXAMPLE 15: Preparation of the compound represented by Formula 15 not according to the invention

[0069]

- 45 1) Synthesis of arylamine (4-(N,N-diphenylamino)-biphenyl-N-biphenylamine) to produce the compound represented by Formula 15: 8.80 g of 4-chlorobiphenyl-N,N-diphenylamine (24.7 mmol) and 6.28 g of 4-aminobiphenyl (37.1 mmol) were dissolved in 200 ml of toluene, 5.94 g of sodium-tert-butoxide (61.8 mmol), 0.43 g of bis(dibenzylidene acetone)palladium(0) (0.74 mmol), and 0.61 ml of 50 wt% tri-tert-butylphosphine toluene solution (1.24 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 5 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 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (7.0 g, yield 58 %). MS: $[M+H]^+ = 489$.
- 50 55 2) 4.37 g of compound of Formula d (8.88 mmol) and 5.23 g of 4-(N,N-diphenylamino)-biphenyl-N-biphenylamine (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(diben-

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zylidene 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 15 (5.4 g, yield 64 %). MS: [M+H]⁺= 943.

[0070]

EXAMPLE 16: Preparation of the compound represented by Formula 16 not according to the invention

[0071]

1) Synthesis of arylamine 10 (4-(N-phenyl-N-naphthylamino)-biphenyl-N-biphenylamine) to produce the compound represented by Formula 16: 4.08 g of 4-chlorobiphenyl-N-phenyl-N-naphthylamine (10.1 mmol) and 2.55 g of 4-aminobiphenyl (15.1 mmol) were dissolved in 100 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.17 g of bis(dibenzylidene acetone)palladium(0) (0.30 mmol), and 0.26 ml of 50 wt% tri-tert-butylphosphine toluene solution (0.53 mmol) were added thereto, and reflux was conducted in a nitrogen atmosphere for 7 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 10:1, stirring was conducted using petroleum ether, and vacuum drying was conducted to produce an arylamine connection group (3.8 g, yield 70 %). MS: [M+H]⁺= 539.

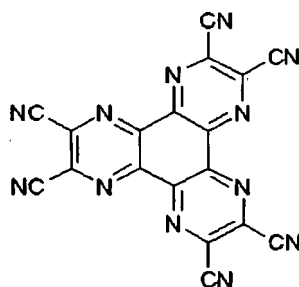
2) 4.369 g of compound of Formula d (8.88 mmol) and 5.76 g of 4-(N-phenyl-N-naphthylamino)-biphenyl-N-biphenylamine (10.7 mmol) were dissolved in 120 ml of toluene, 2.90 g of sodium-tert-butoxide (30.2 mmol), 0.12 g of bis(dibenzylidene acetone)palladium(0) (0.21 mmol), and 0.16 ml of 50 wt% tri-tert-butylphosphine 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 16 (4.9 g, yield 56 %). MS: [M+H]⁺= 995.

EXAMPLE 17: 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.

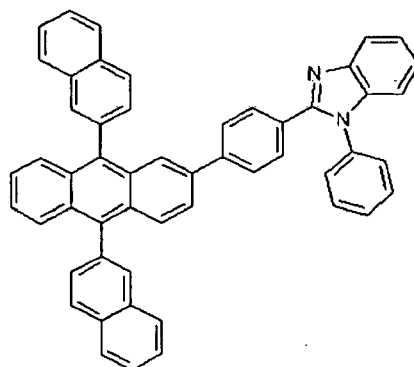
[0073] Hexanitride hexaazatriphenylene (hereinafter, referred to as "HAT") of the following Formula was vacuum deposited to a thickness of 500 Å by heating on a transparent ITO electrode, which was prepared through the above procedure, so as to form an anode including an ITO conductive layer and an N-type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

[HAT]



[0074] The compound of Formula 6 (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.

Electron transport layer material



[0075] 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.

[0076] 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 $1. - 3 \times 10^{-7}$.

[0077] The resulting device had an electric field of 5.47 V at a forward current density of 100 mA/cm², and emitted green light at a light efficiency of 2.91 lm/W. The operation and light emission of the device at the above-mentioned actuating voltage mean that the compound of Formula 6, which formed the layer between the hole injection layer and the light emitting layer, functions to transport holes.

EXAMPLE 18: Production of an organic light emitting device

[0078] 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 17 and a compound of Formula 6 was deposited thereon to a thickness of 1100 Å to form a layer for both injecting and transporting holes.

[0079] The resulting device had an electric field of 6.75 V at a forward current density of 100 mA/cm², 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 both 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.

Industrial Applicability

[0080] 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.

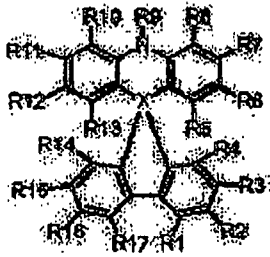
Claims

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 the following Formula 1:

5



10

Formula 1

15

wherein:

X is C;

R1, R2, R4 to R8, R10 to R14, R16 and R17 are each independently hydrogen or an unsubstituted alkyl group;
 R3 and R15 are each an unsubstituted arylamine group; and

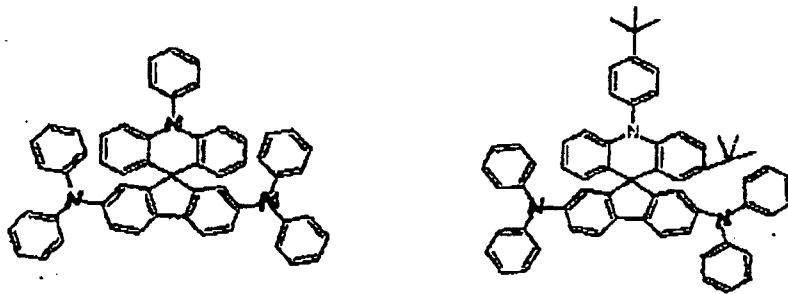
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R9 is a phenyl group which is unsubstituted or substituted with one or more of a halogen atom, an alkyl group, an alkenyl group, an alkoxy group, an unsubstituted arylamine group, an unsubstituted aryl group, an unsubstituted heterocyclic group and a nitrile group.

25

- An organic light-emitting device according to Claim 1, wherein the compound represented by Formula 1 is one of the following compounds:

30



35

- An organic light-emitting device according to Claim 1 or Claim 2, wherein the organic material layers comprise a layer which both injects and transports electrons and which includes a compound of Formula 1.
- An organic light-emitting device according to Claim 1 or Claim 2, wherein the light-emitting layer includes a compound of Formula 1.

45

Patentansprüche

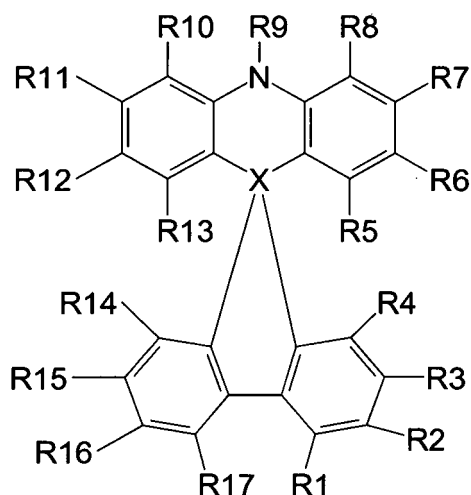
- Organische lichtemittierende Vorrichtung, umfassend:

50

eine erste Elektrode;
 organische Materialschichten, umfassend eine lichtemittierende Schicht und eine von einer Lochinjektions-
 schicht, Lochtransportschicht und einer Schicht mit sowohl Injektions- als auch Transportlöchern; und
 eine zweite Elektrode;

55

worin die erste Elektrode, die organischen Materialschichten und die zweite Elektrode eine Schichtstruktur
 bilden, und
 die Lochinjektionsschicht, Lochtransportschicht oder die Schicht mit sowohl Injektions- als auch Transportlö-
 chern eine Verbindung mit der folgenden Formel 1 enthält:



Formel 1

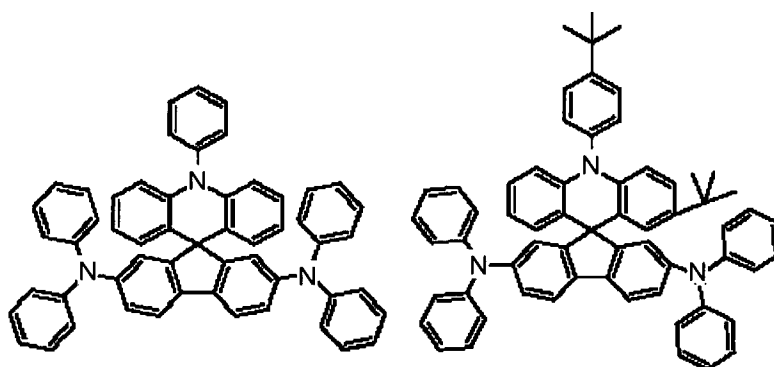
X C ist;

R1, R2, R4 bis R8, R10 bis R14, R16 und R17 jeweils unabhängig Wasserstoff oder eine unsubstituierte Alkylgruppe sind;

R3 und R15 jeweils eine unsubstituierte Arylamingruppe sind; und

R9 eine Phenylgruppe ist, die unsubstituiert oder mit einem oder mehreren von einem Halogenatom, Alkylgruppe, Alkenylgruppe, Alkoxygruppe, unsubstituierten Arylamingruppe, unsubstituierten Arylgruppe, unsubstituierten heterocyclischen Gruppe und Nitrilgruppe substituiert ist.

2. Organische lichtemittierende Vorrichtung nach Anspruch 1, worin die Verbindung mit der Formel 1 eine der folgenden Verbindungen ist:



3. Organische lichtemittierende Vorrichtung nach Anspruch 1 oder 2, worin die organischen Materialschichten eine Schicht mit sowohl Injektions- als auch Transportelextronen enthalten und die eine Verbindung der Formel 1 enthalten.

4. Organische lichtemittierende Vorrichtung nach Anspruch 1 oder 2, worin die lichtemittierende Schicht eine Verbindung der Formel 1 enthält.

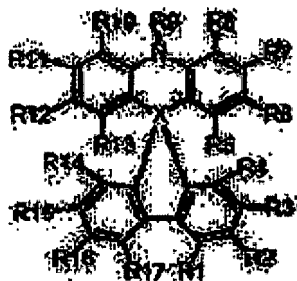
Revendications

1. Dispositif électroluminescent organique, comprenant :

une première électrode ;

des couches de matière organique comprenant une couche électroluminescente et une couche choisie parmi une couche d'injection de trous, une couche de transport de trous et 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 la formule 1 suivante :



Formule 1

dans laquelle :

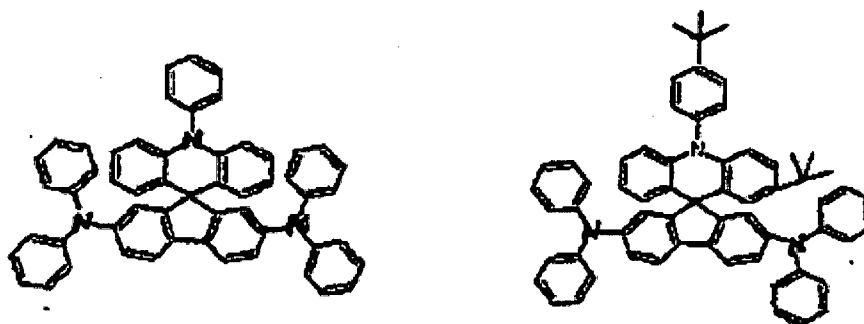
X est C ;

R1, R2, R4 à R8, R10 à R14, R16 et R17 sont chacun indépendamment un atome d'hydrogène ou un groupe alkyle non substitué ;

R3 et R15 sont chacun un groupe arylamine non substitué ; et

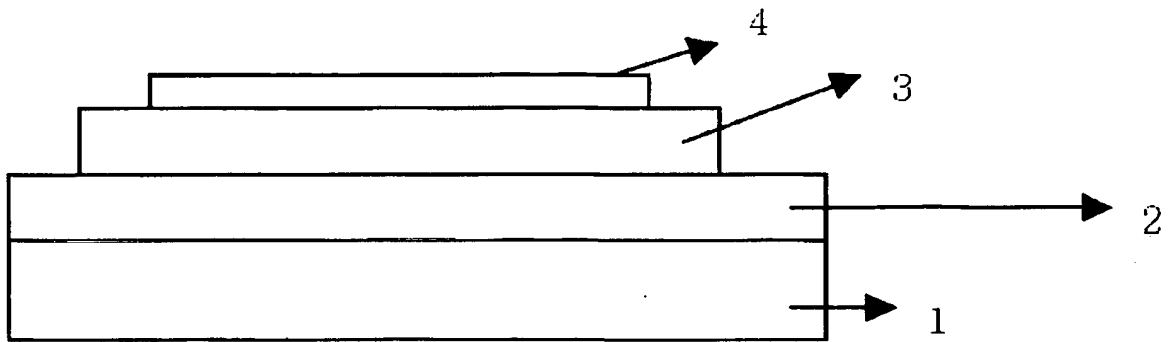
R9 est un groupe phényle qui n'est pas substitué ou est substitué avec un ou plusieurs substituants choisis parmi un atome d'halogène, un groupe alkyle, un groupe alcényle, un groupe alcoxy, un groupe arylamine non substitué, un groupe aryle non substitué, un groupe hétérocyclique non substitué et un groupe nitrile.

2. Dispositif électroluminescent organique selon la revendication 1, dans lequel le composé représenté par la formule 1 est un des composés suivants :

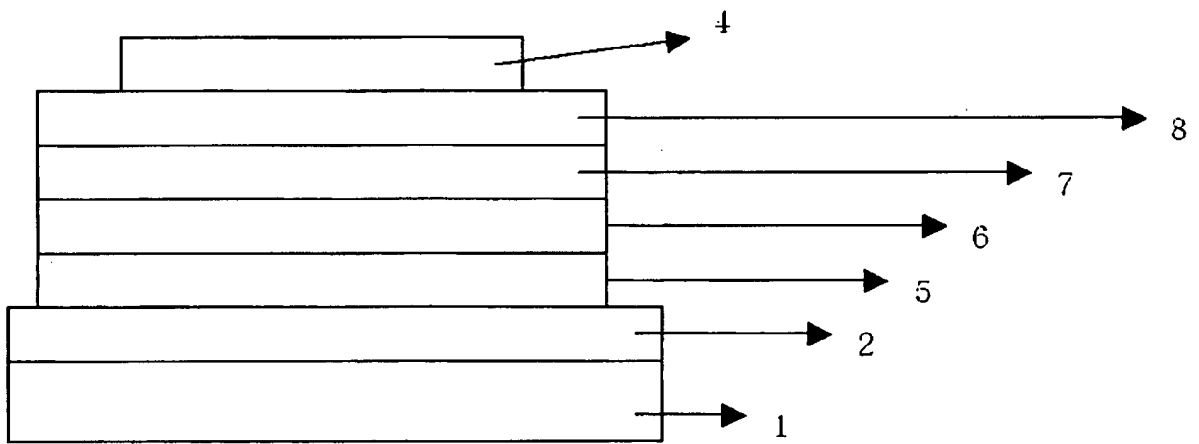


3. Dispositif électroluminescent organique selon la revendication 1 ou 2, dans lequel les couches de matière organique comprennent une couche qui à la fois injecte et transporte des électrons et qui comprend un composé de formule 1.
4. Dispositif électroluminescent organique selon la revendication 1 ou 2, dans lequel la couche électroluminescente comprend un composé de formule 1.

[Fig. 1]



[Fig. 2]



REFERENCES CITED IN THE DESCRIPTION

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- WO 2004020371 A [0008]
- US 20030044518 A [0037]
- EP 1146574 A2 [0037]

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申请(专利权)人(译)	LG化学有限公司.		
当前申请(专利权)人(译)	LG化学有限公司.		
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其他公开文献	EP1791926A1 EP1791926A4		
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。

