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**Hwang et al.**

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(54) **PHENYL CARBAZOLE-BASED COMPOUND  
AND ORGANIC ELECTROLUMINESCENT  
DEVICE EMPLOYING THE SAME**

(75) Inventors: **Seok-Hwan Hwang**, Suwon-si (KR);  
**Young-Kook Kim**, Suwon-si (KR);  
**Chang-Ho Lee**, Suwon-si (KR);  
**Seok-Jong Lee**, Suwon-si (KR);  
**Seung-Gak Yang**, Suwon-si (KR);  
**Hee-Yeon Kim**, Suwon-si (KR)

(73) Assignee: **Samsung Mobile Display Co., Ltd.**,  
Giheung-Gu, Yongin, Gyeonggi-Do (KR)

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patent is extended or adjusted under 35  
U.S.C. 154(b) by 758 days.

This patent is subject to a terminal dis-  
claimer.

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**Related U.S. Application Data**

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filed on Nov. 25, 2005.

(30) **Foreign Application Priority Data**

Nov. 29, 2004 (KR) ..... 10-2004-0098747

(51) **Int. Cl.**  
**H01L 51/54** (2006.01)  
**C07D 207/444** (2006.01)

(52) **U.S. Cl.** ..... **428/690**; 428/917; 548/442; 313/504;  
313/506

(58) **Field of Classification Search** ..... None  
See application file for complete search history.

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*Primary Examiner* — Dawn L. Garrett

(74) *Attorney, Agent, or Firm* — Robert E. Bushnell, Esq.

(57) **ABSTRACT**

A phenylcarbazole-based compound is represented by For-  
mula 1, and has superior electric properties and charge trans-  
port abilities, and thus is useful as a hole injection material, a  
hole transport material, and/or an emitting material which is  
suitable for fluorescent and phosphorescent devices of all  
colors, including red, green, blue, and white colors. The phe-  
nylcarbazole-based compound is synthesized by reacting car-  
bazole with diamine. The organic electroluminescent device  
manufactured using the phenylcarbazole-based compound  
has high efficiency, low voltage, high luminance, and a long  
lifespan.

**15 Claims, 16 Drawing Sheets**

CATHODE
EIL
ETL
EML
HTL
HIL
ANODE
SUBSTRATE

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FIG. 1

CATHODE
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ANODE
SUBSTRATE

FIG. 2

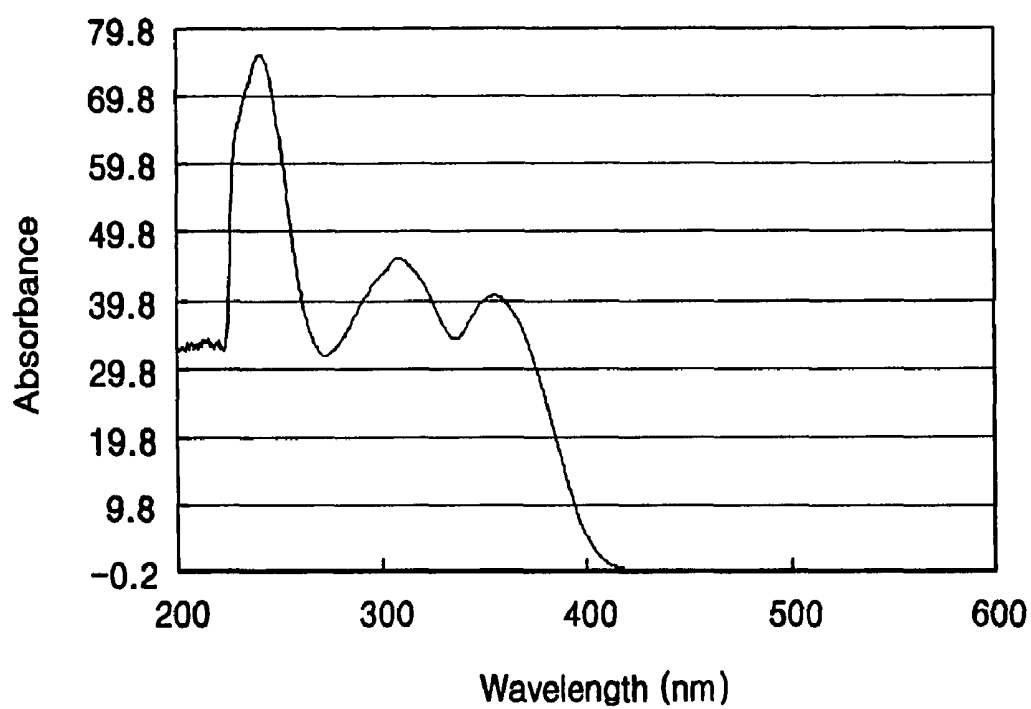


FIG. 3

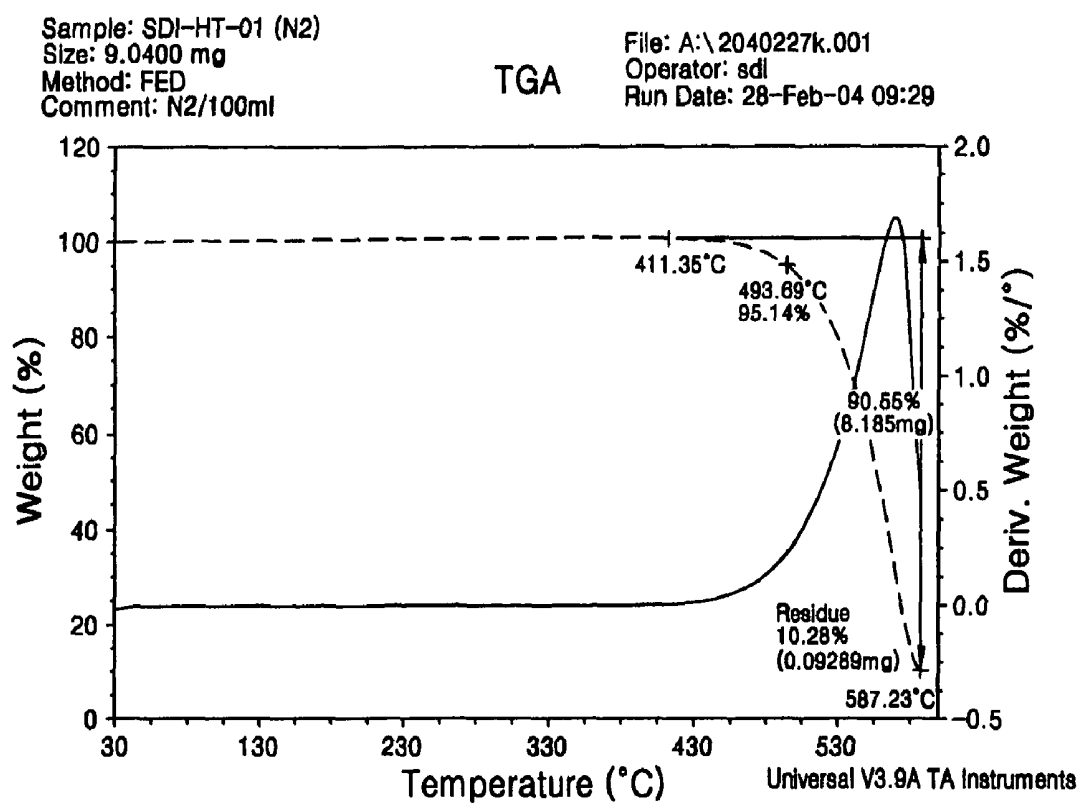


FIG. 4

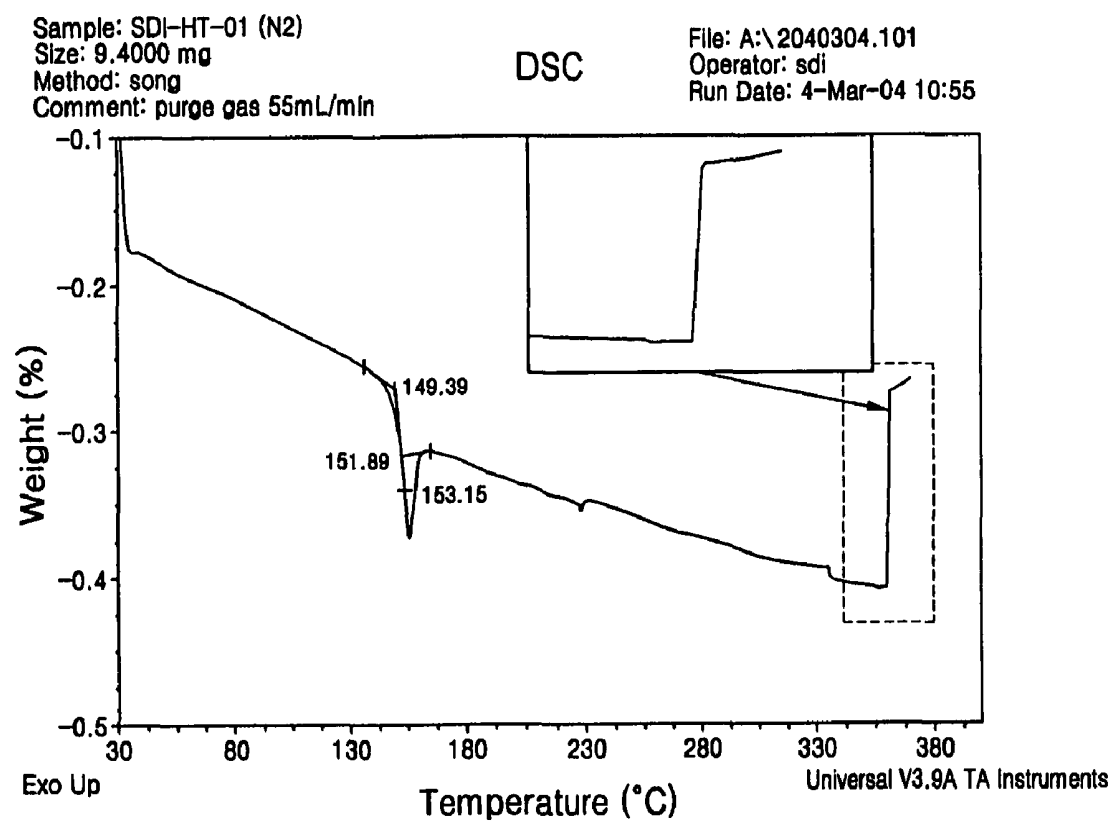


FIG. 5

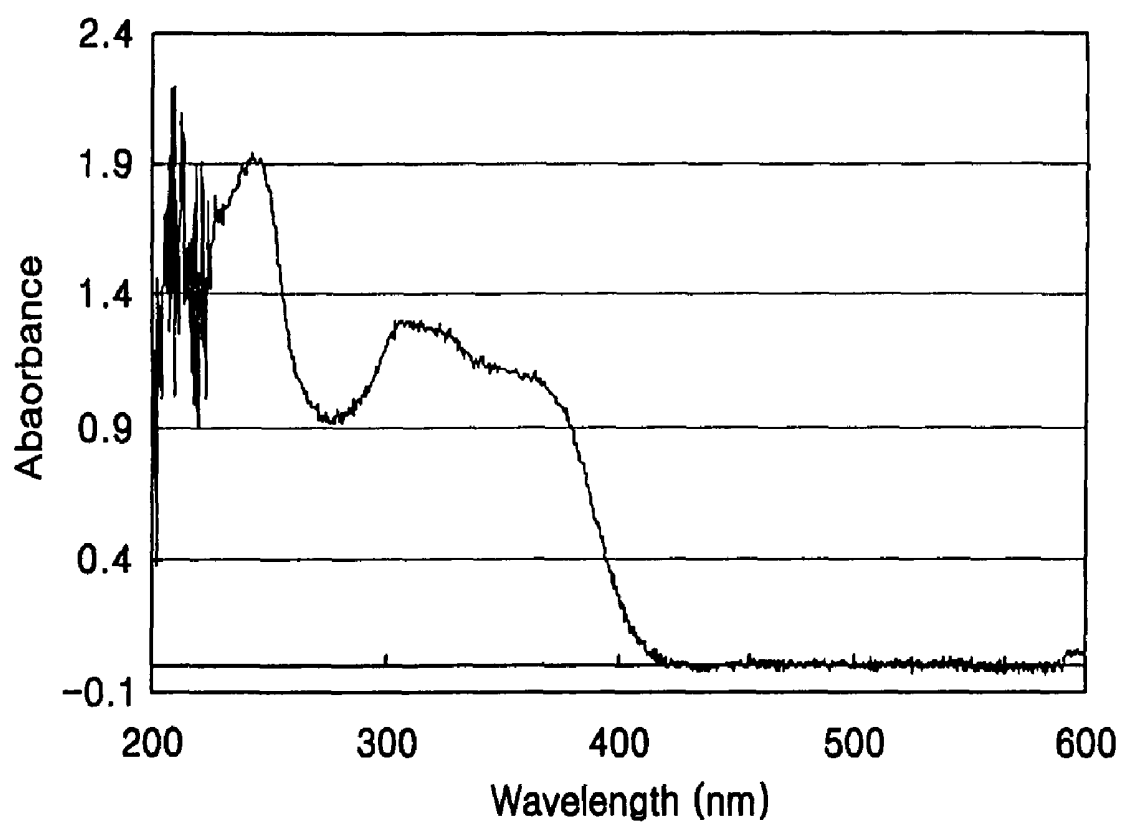


FIG. 6

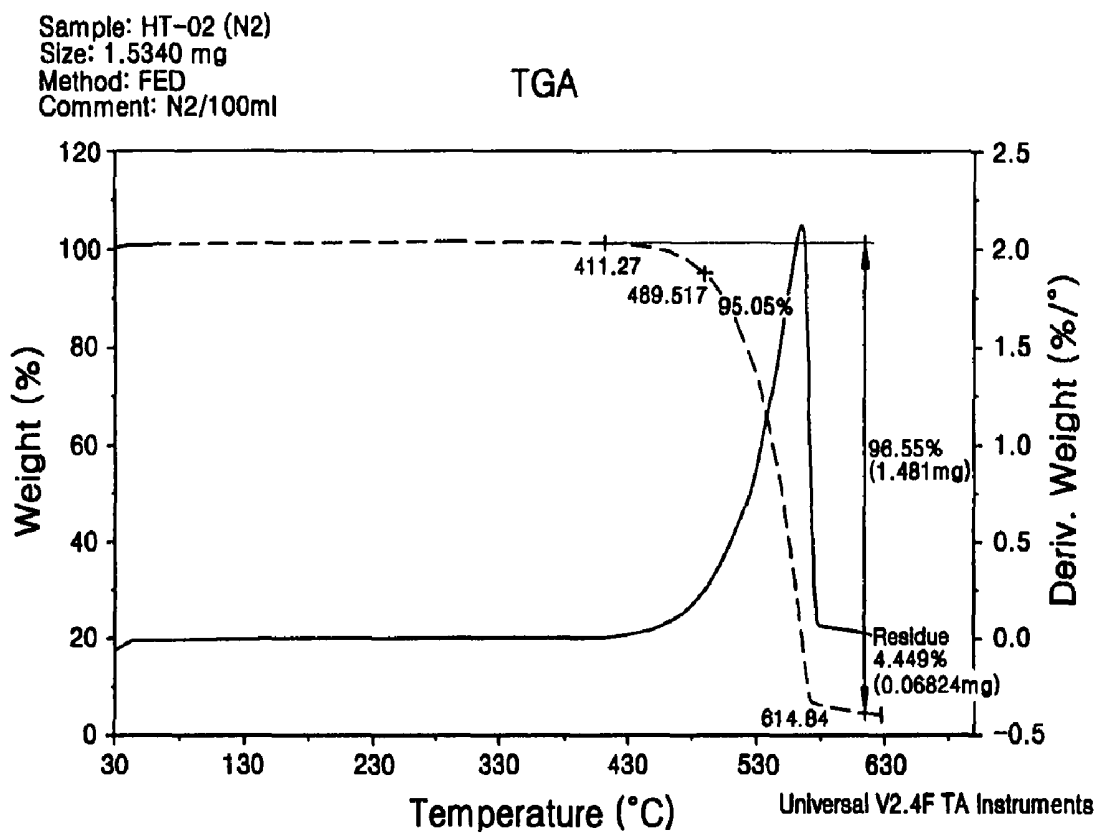


FIG. 7

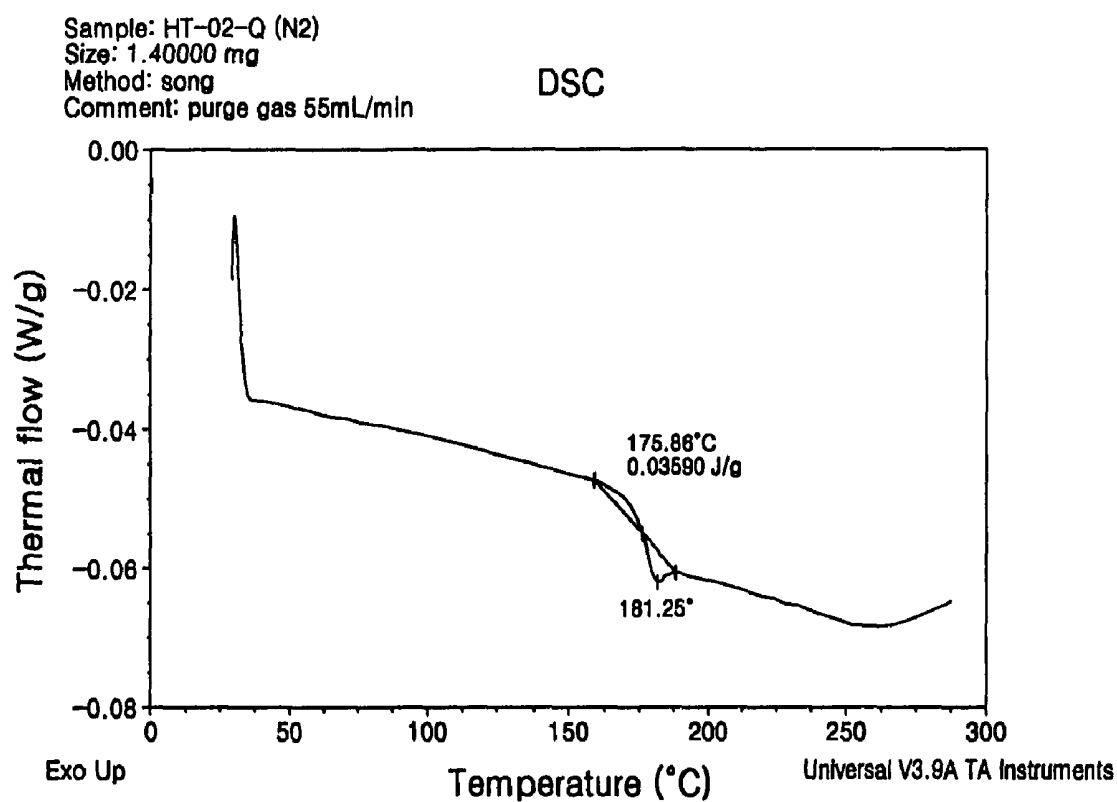




FIG. 8

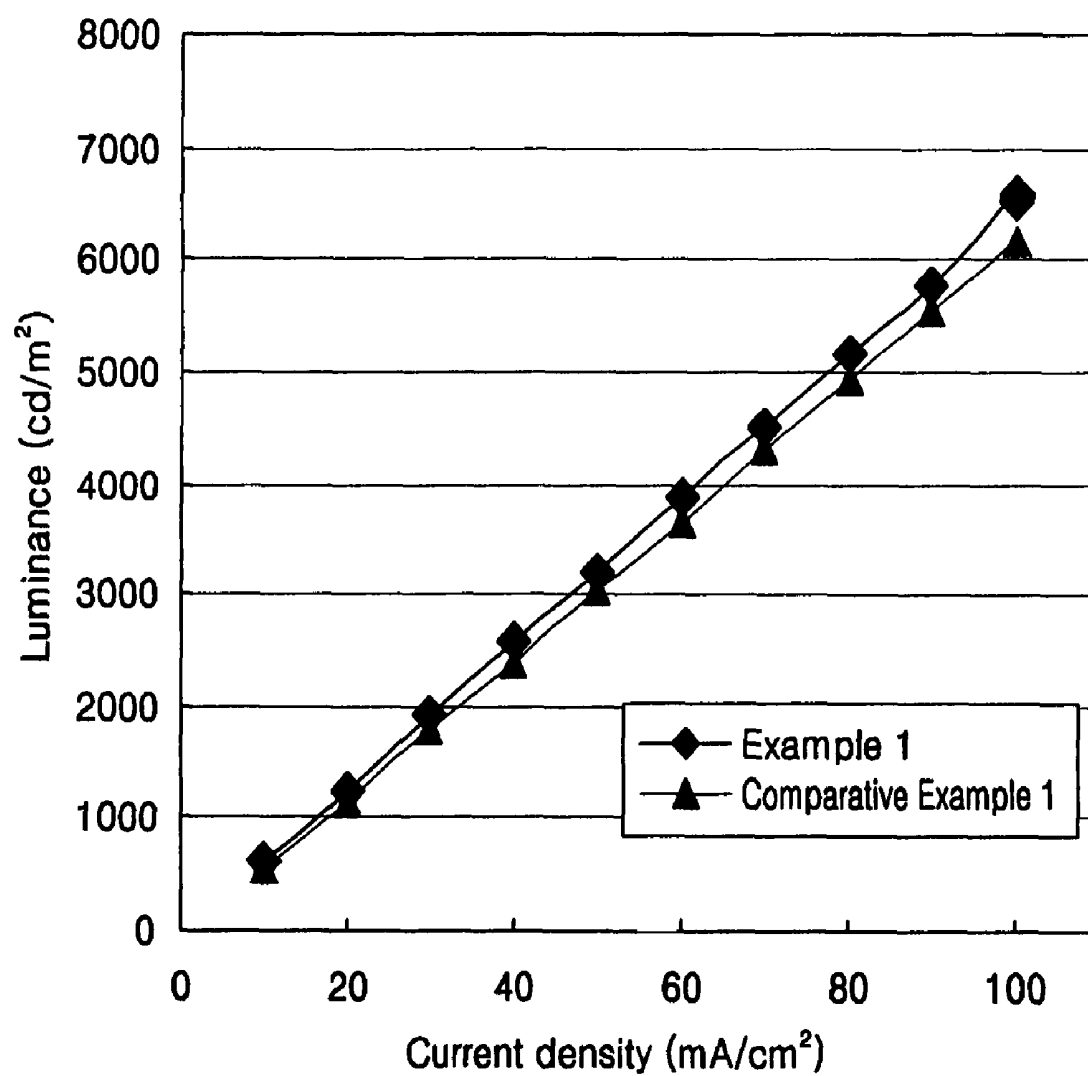


FIG. 9

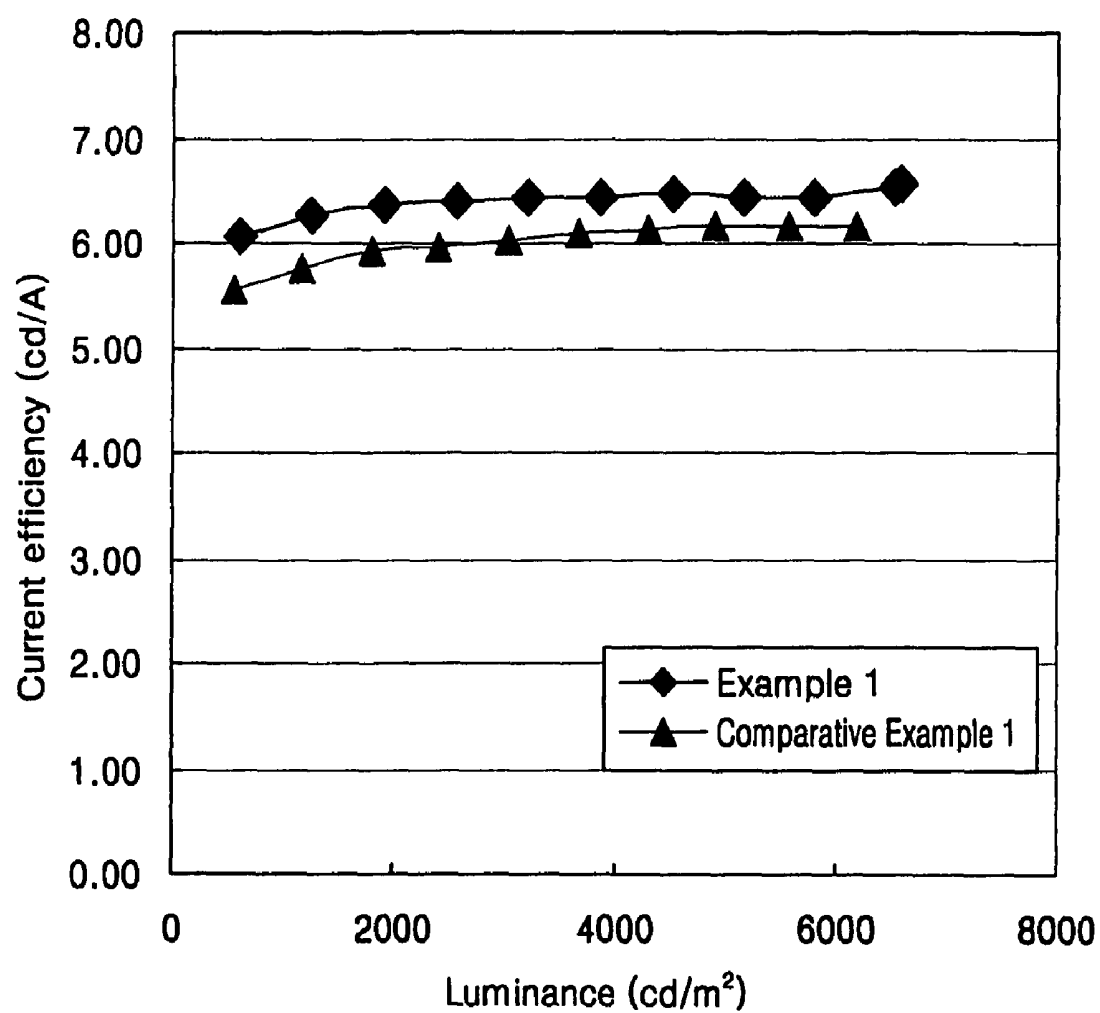


FIG. 10

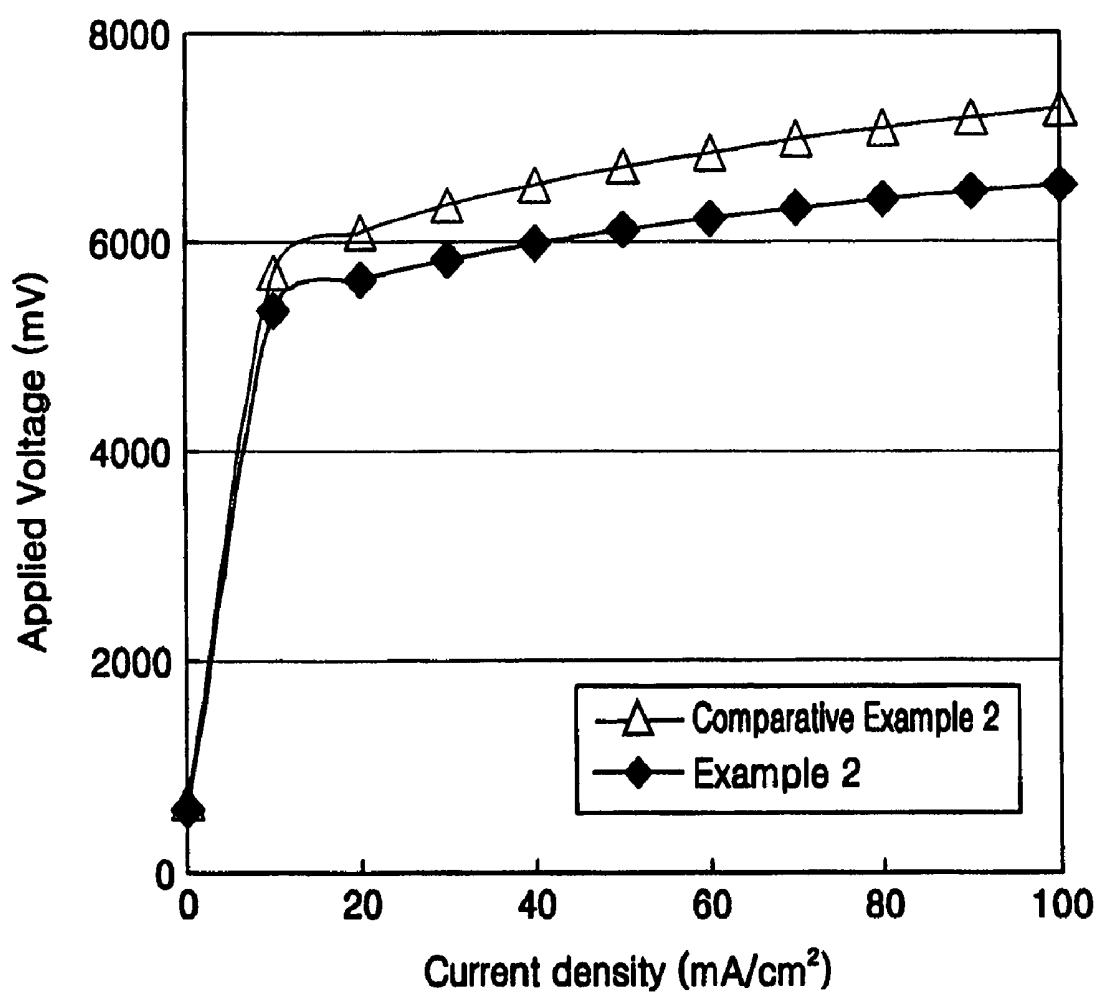


FIG. 11

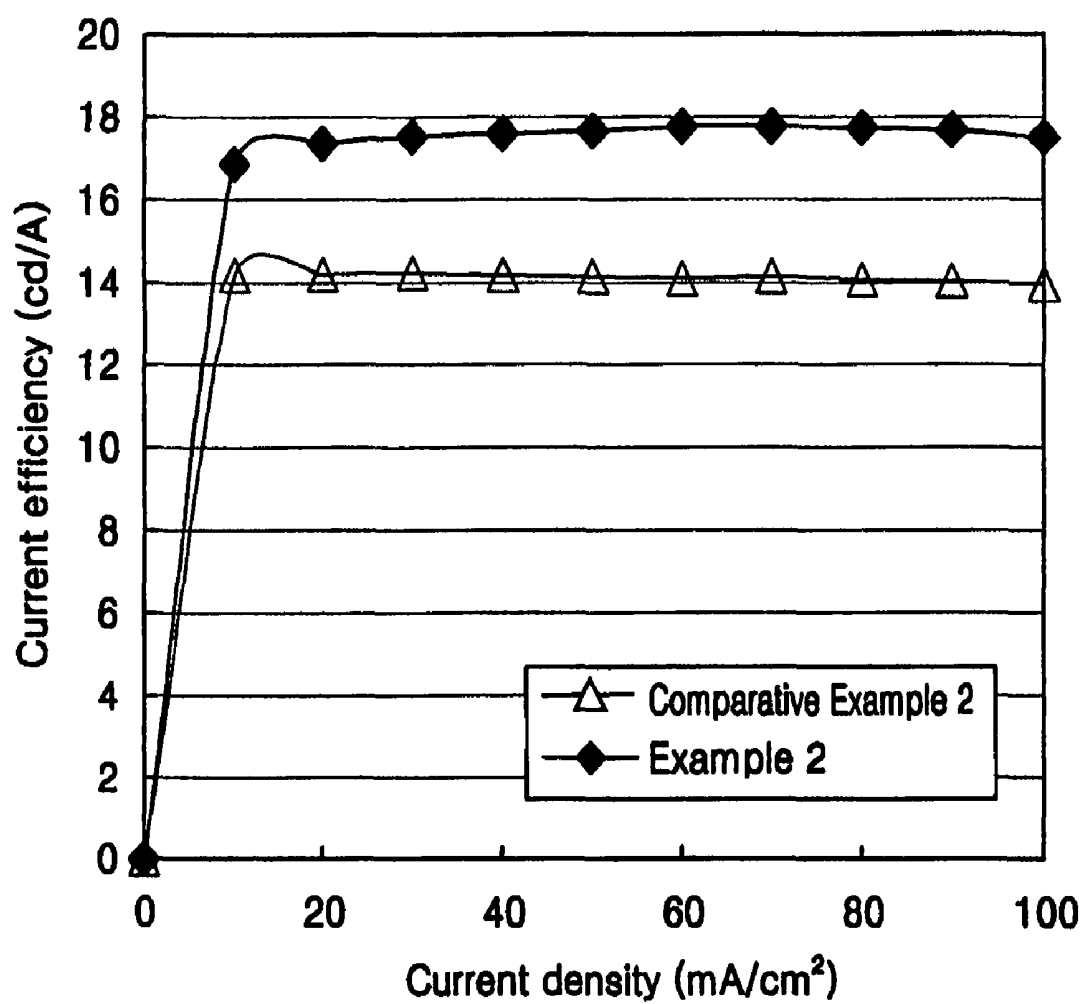


FIG. 12

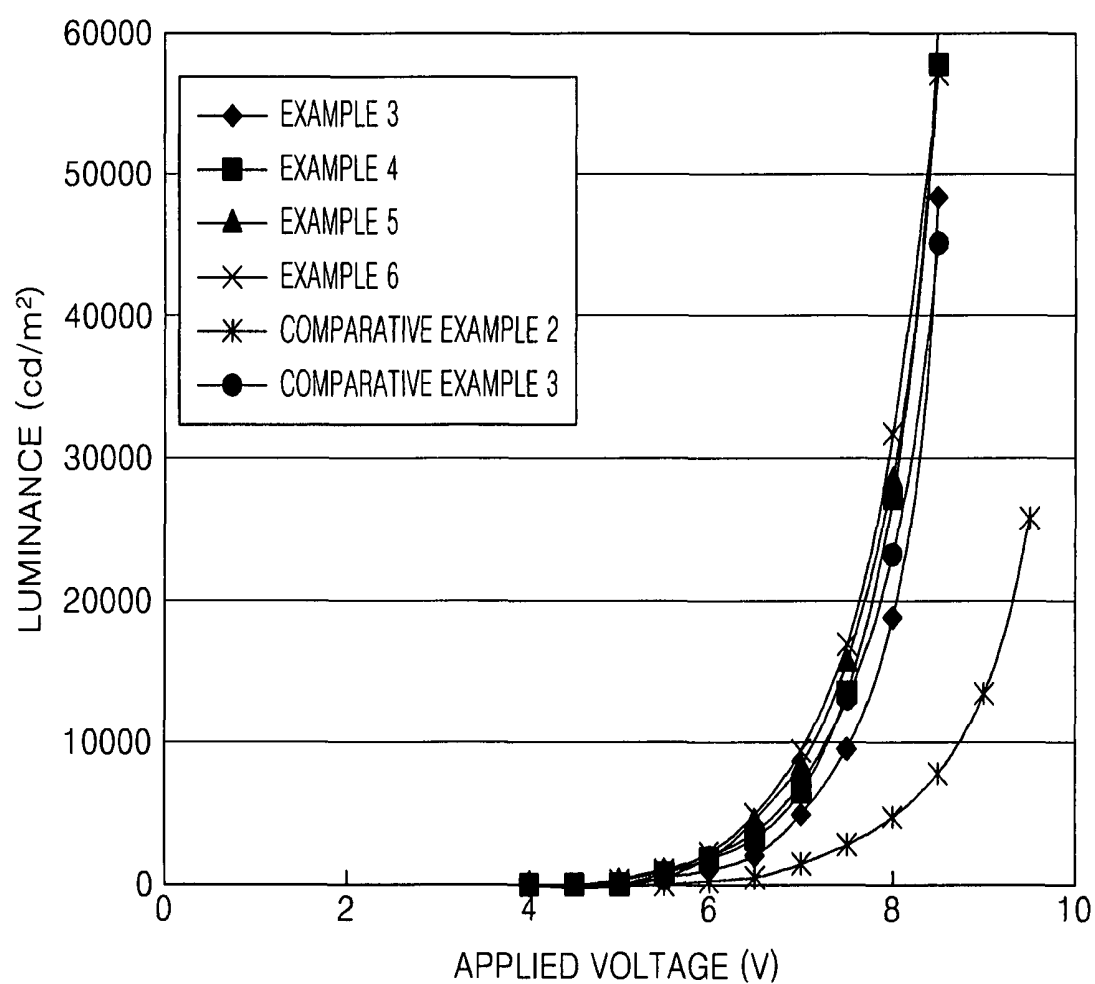
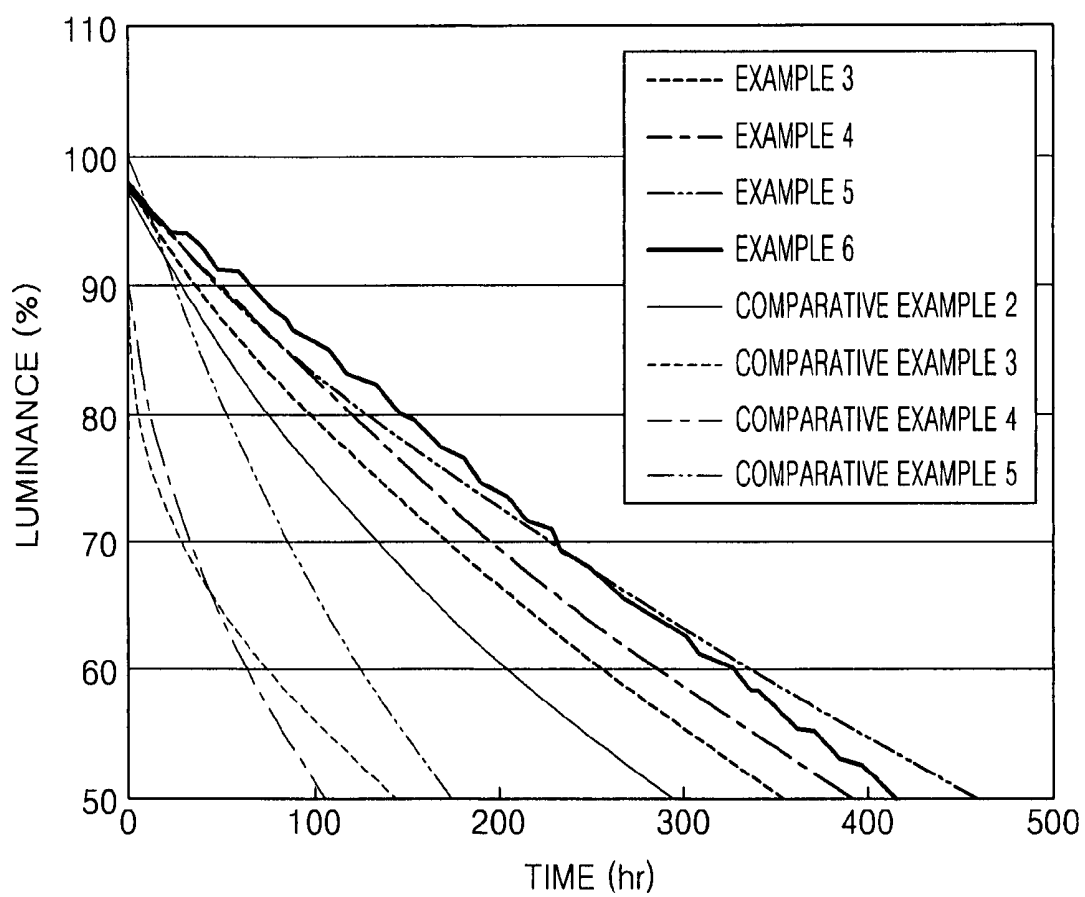
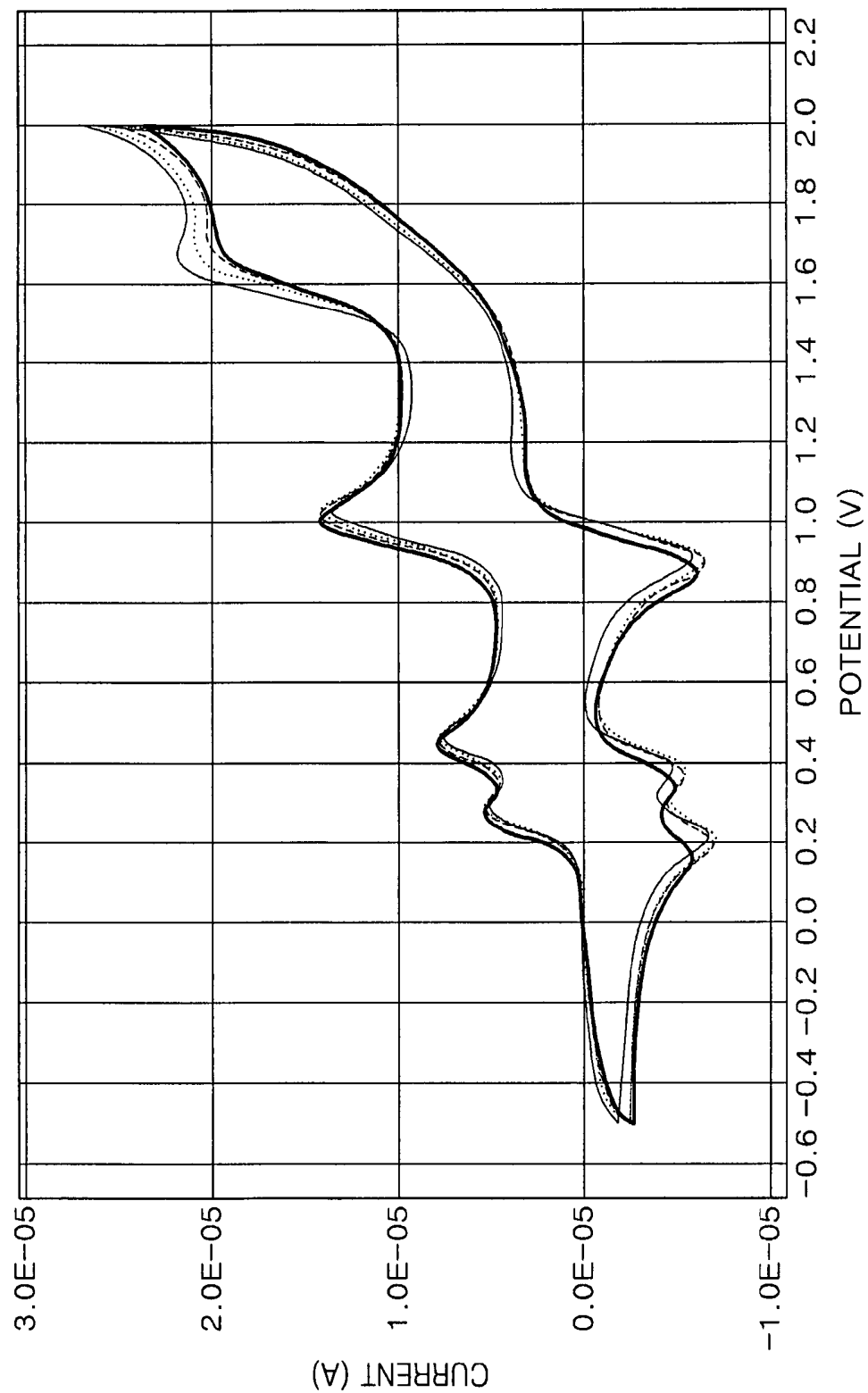


FIG. 13



**FIG. 14**

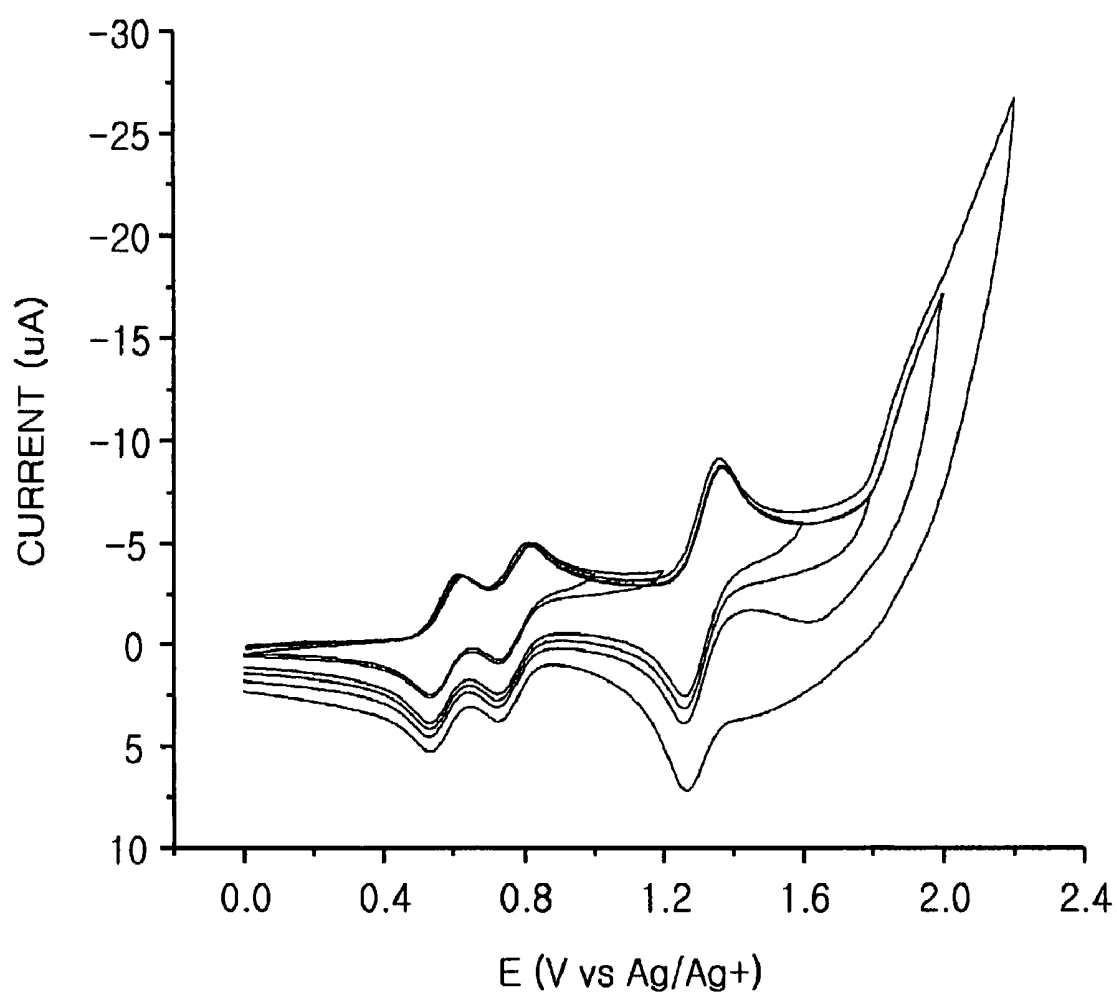
**FIG. 15**



FIG. 16

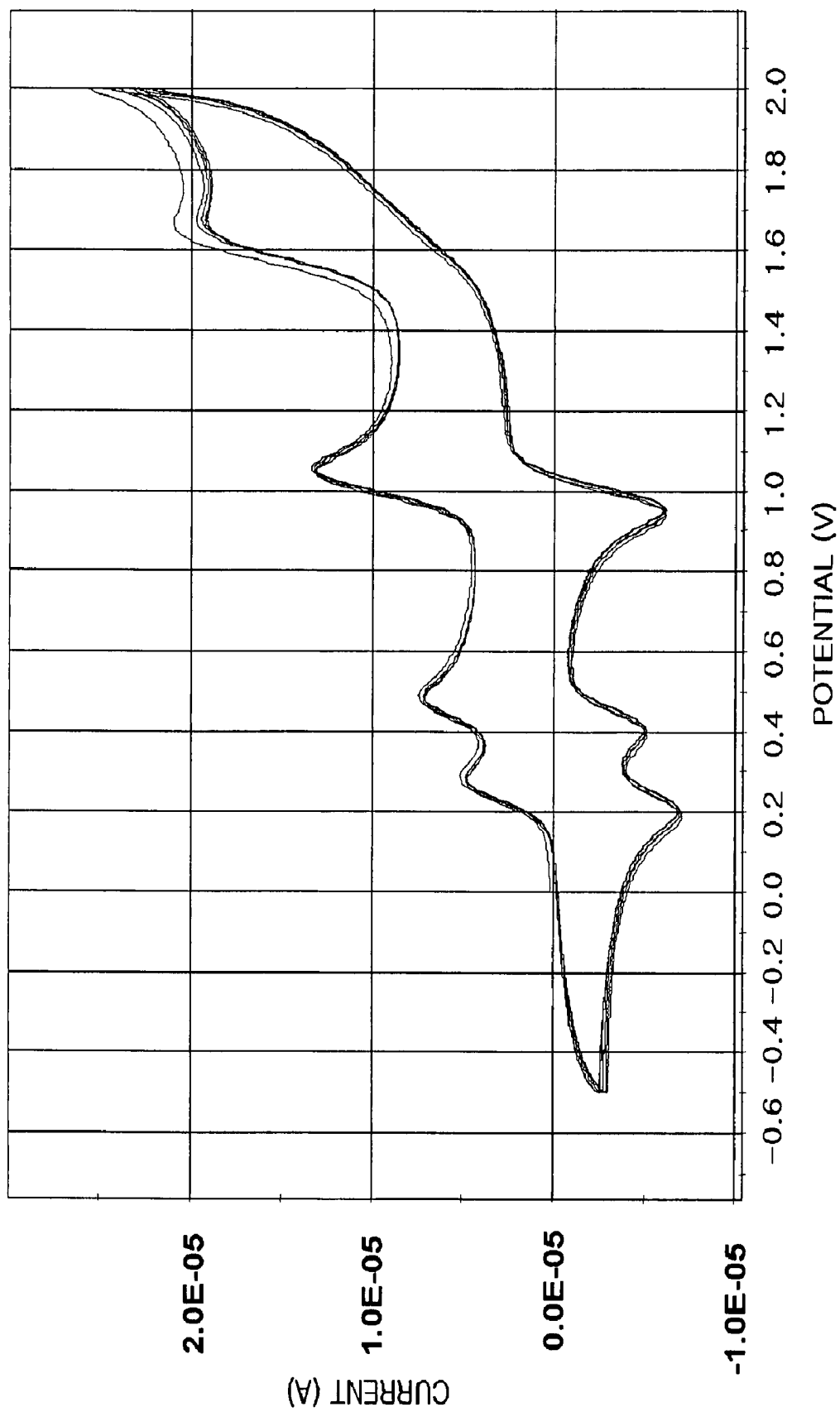
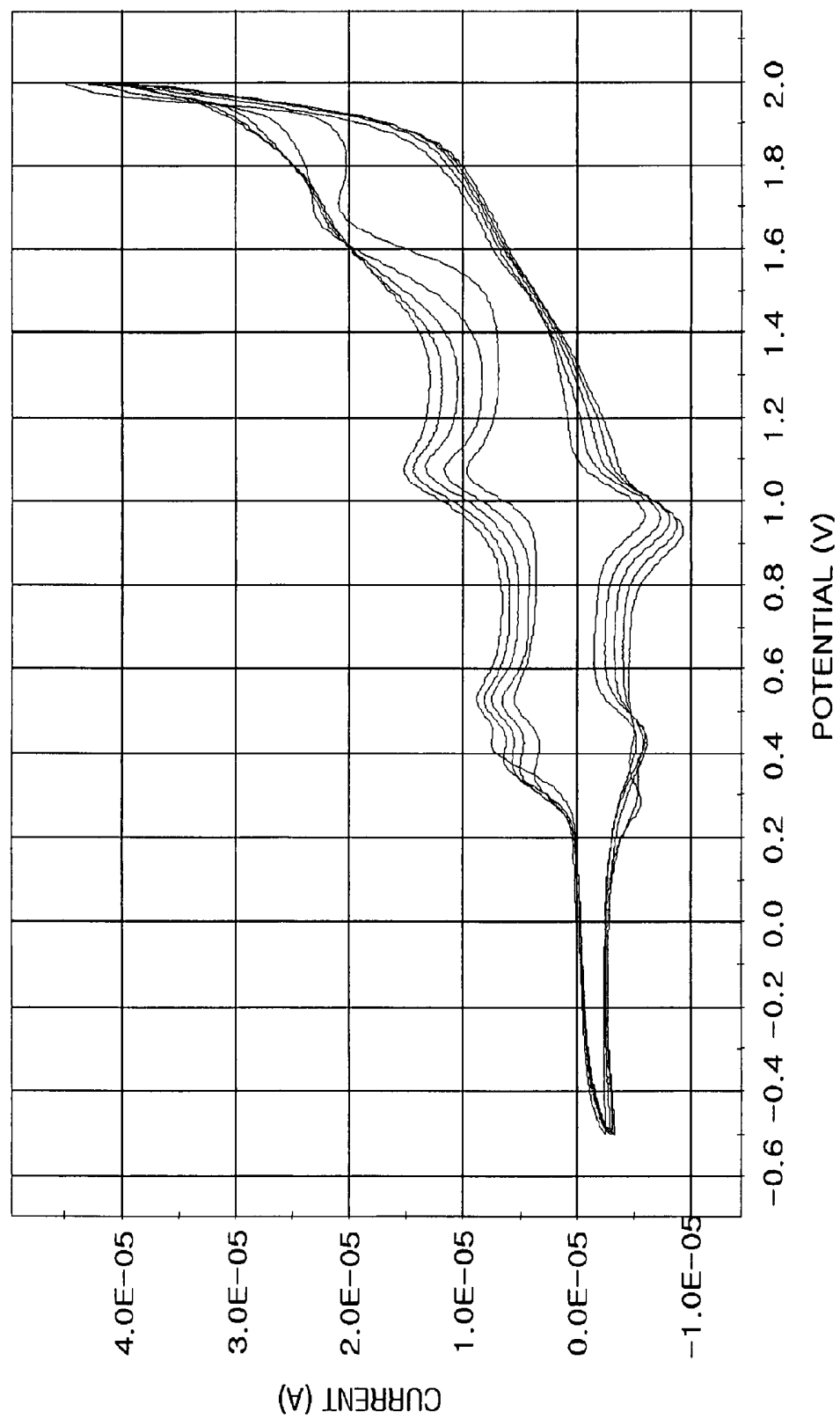


FIG. 17



# PHENYLCARBAZOLE-BASED COMPOUND AND ORGANIC ELECTROLUMINESCENT DEVICE EMPLOYING THE SAME

CROSS-REFERENCE TO RELATED PATENT  
APPLICATION AND CLAIM OF PRIORITY

This application claims the benefit of Korean Patent Application No. 10-2004-0098747, filed on Nov. 29, 2004, in the Korean Intellectual Property Office, the disclosure of which is incorporated herein in its entirety by reference. This application is a Continuation-in-Part of application Ser. No. 11/286,421 filed on the 25 Nov. 2005, and assigned to the assignee of the present invention. All benefits accruing under 35 U.S.C. §120 from the present application are also hereby claimed.

## BACKGROUND OF THE INVENTION

### 1. Field of the Invention

The present invention relates to a phenylcarbazole-based compound and an organic electroluminescent device employing the same, and more particularly, to a phenylcarbazole-based compound which has electric stability, superior charge transport ability and high glass transition temperature and can prevent crystallization, and an organic electroluminescent device using an organic layer including the same.

### 2. Description of the Related Art

An electroluminescent (EL) device is a self-emission type display device and has received significant attention owing to its merits of a wide viewing angle, superior contrast, and rapid response. EL devices are divided into inorganic EL devices in which an emitting layer is composed of an inorganic compound, and organic EL devices in which an emitting layer is composed of an organic compound. An organic EL device has superior luminance, driving voltage, and response rate to an inorganic EL device and can display multicolors, and thus much research into organic EL devices has been conducted.

The organic EL device generally has a layered structure of anode/organic emitting layer/cathode. When a hole transport layer and/or an electron injection layer is further interposed between the anode and the emitting layer or between the emitting layer and the cathode, an anode/hole transport layer/organic emitting layer/cathode structure or an anode/hole transport layer/organic emitting layer/electron injection layer/cathode structure is formed.

The hole transport layer is known to be composed of polyphenyl hydrocarbon or an anthracene derivative (see, for example, U.S. Pat. Nos. 6,596,415 and 6,465,115).

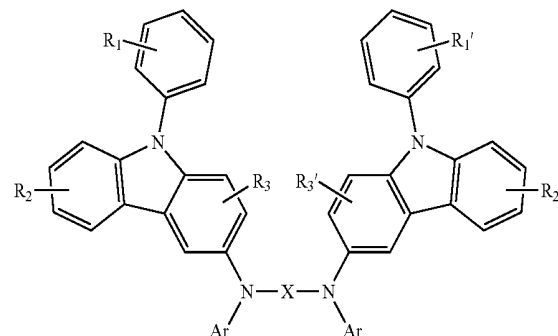
Organic EL devices including hole transport layers composed of conventional materials are not satisfactory in terms of lifespan, efficiency and power consumption, and thus a material for a hole transport layer with a significant improvement in such characteristics is required.

## SUMMARY OF THE INVENTION

The present invention provides an organic layer material which has electric stability, superior charge transport ability, and high glass transition temperature, can prevent crystallization, and is suitable for fluorescent and phosphorescent devices having all colors, including red, green, blue, and white colors, etc., and a method of preparing the same.

The present invention also provides an organic EL device using an organic layer composed of the above-described material and having high efficiency, low voltage, high luminance and long lifespan.

According to an aspect of the present invention, there is provided a phenylcarbazole-based compound represented by Formula (1):



where X is a substituted or unsubstituted C1-C30 alkylene group, a substituted or unsubstituted C2-C30 alkenylene group, a substituted or unsubstituted C6-C30 arylene group, a substituted or unsubstituted C2-C30 heteroarylene group, or a substituted or unsubstituted C2-C30 heterocyclic group; each of R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>1</sub>', R<sub>2</sub>' and R<sub>3</sub>' is independently a hydrogen atom, a substituted or unsubstituted C1-C30 alkyl group, a substituted or unsubstituted C1-C30 alkoxy group, a substituted or unsubstituted C6-C30 aryl group, a substituted or unsubstituted C6-C30 aryloxy group, a substituted or unsubstituted C2-C30 heterocyclic group, a substituted or unsubstituted C6-C30 condensed polycyclic group, a hydroxy group, a cyano group, or a substituted or unsubstituted amino group, and, alternatively, two or more adjacent groups among R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>1</sub>', R<sub>2</sub>' and R<sub>3</sub>' can be connected to each other to form a saturated or unsaturated carbocycle; and each Ar is independently a substituted or unsubstituted C6-C30 aryl group or a substituted or unsubstituted C2-C30 heteroaryl group.

According to another aspect of the present invention, there is provided an organic EL device including a first electrode, a second electrode, and an organic layer interposed therebetween, in which the organic layer contains the phenylcarbazole-based compound.

The organic layer may be a hole injection layer, a hole transport layer, or a single layer serving as both the hole injection layer and the hole transport layer.

In an embodiment of the present invention, the organic layer is a hole injection layer or a hole transport layer, and the organic EL device has a first electrode/hole injection layer/emitting layer/second electrode structure, a first electrode/hole injection layer/emitting layer/hole transport layer/second electrode structure, or a first electrode/emitting layer/hole transport layer/second electrode structure.

The organic layer is an emitting layer, in which the emitting layer is composed of a phosphorescent or fluorescent material.

In the emitting layer, the phenylcarbazole-based compound represented by Formula (1) is used as a fluorescent or phosphorescent host.

## BRIEF DESCRIPTION OF THE DRAWINGS

A more complete appreciation of the present invention, and many of the above and other features and advantages of the present invention, will be readily apparent as the same becomes better understood by reference to the following

detailed description when considered in conjunction with the accompanying drawings in which like reference symbols indicate the same or similar components, wherein:

FIG. 1 is a cross-sectional view of an organic electroluminescent device according to an embodiment of the present invention;

FIG. 2 illustrates a UV spectrum of Compound represented by Formula 3 obtained according to an embodiment of the present invention;

FIGS. 3 and 4 illustrate differential scanning calorimetry (DSC) and thermo gravimetric analysis (TGA) results of Compound represented by Formula 3 obtained according to an embodiment of the present invention;

FIG. 5 illustrates a UV spectrum of Compound represented by Formula 4 obtained according to another embodiment of the present invention;

FIGS. 6 and 7 illustrate DSC and TGA analysis results of Compound represented by Formula 4 obtained according to an embodiment of the present invention;

FIG. 8 is a graph illustrating the variation in luminance with respect to the current density in organic EL devices obtained in Example 1 of the present invention and Comparative Example 1;

FIG. 9 is a graph illustrating the variation in current efficiency with respect to the luminance in organic EL devices obtained in Example 1 of the present invention and Comparative Example 1;

FIG. 10 is a graph illustrating the driving voltage at the same current density for the organic EL devices according to the Example 2 and Comparative Example 2;

FIG. 11 is a graph illustrating the current efficiency at the same current density for the organic EL devices according to the Example 2 and Comparative Example 2;

FIG. 12 shows luminance according to driving voltages in the organic EL devices of Examples 3-6 according to the embodiments of the present invention and Comparative Examples 3 and 4;

FIG. 13 shows lifetime characteristics of the organic EL devices of Examples 3, 4, 5 and 6 according to embodiments of the present invention and the organic EL devices of Comparative Examples 3 through 6;

FIG. 14 shows a cyclic voltammogram of Compound 3 according to an embodiment of the present invention;

FIG. 15 shows a cyclic voltammogram of TLB1;

FIG. 16 shows a cyclic voltammogram of Compound 5 according to an embodiment of the present invention; and

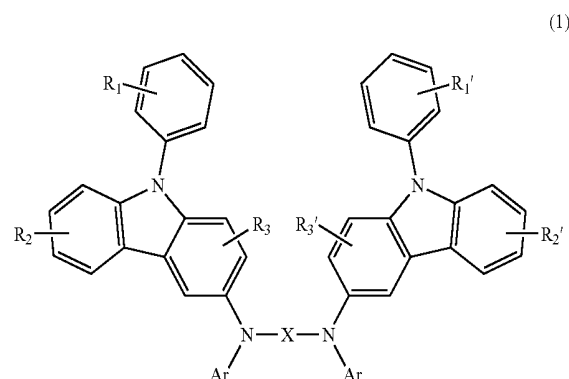
FIG. 17 shows a cyclic voltammogram of Compound 28.

### DETAILED DESCRIPTION OF THE INVENTION

Hereinafter, the present invention will be described in more detail.

The present invention provides a phenylcarbazole-based compound having at least two phenylcarbazole derivatives as side chains in a molecule, a method of preparing the same, and an organic EL device using the compound as a material for an organic layer such as a hole injection layer, a hole transport layer, or an emitting layer.

The phenylcarbazole-based compound may be preferably represented by Formula 1:



where X is a substituted or unsubstituted C1-C30 alkylene group, a substituted or unsubstituted C2-C30 alkenylene group, a substituted or unsubstituted C6-C30 arylene group, a substituted or unsubstituted C2-C30 heteroarylene group, or a substituted or unsubstituted C2-C30 heterocyclic group; each of  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_1'$ ,  $R_2'$  and  $R_3'$  is independently a hydrogen atom, a substituted or unsubstituted C1-C30 alkyl group, a substituted or unsubstituted C1-C30 alkoxy group, a substituted or unsubstituted C6-C30 aryl group, a substituted or unsubstituted C6-C30 aryloxy group, a substituted or unsubstituted C2-C30 heterocyclic group, a substituted or unsubstituted C6-C30 condensed polycyclic group, a hydroxy group, a cyano group, or a substituted or unsubstituted amino group, and, alternatively, two or more adjacent groups among  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_1'$ ,  $R_2'$  and  $R_3'$  can be connected to each other to form a saturated or unsaturated carbocycle; and each Ar is independently a substituted or unsubstituted C6-C30 aryl group or a substituted or unsubstituted C2-C30 heteroaryl group.

For example, each Ar may be a phenyl group, an ethylphenyl group, an ethylbiphenyl group, an o-, m-, or p-fluorophenyl group, a dichlorophenyl group, a dicyanophenyl group, a trifluoromethoxyphenyl group, an o-, m-, or p-tolyl group, an o-, m-, and p-cumenyl group, a mesityl group, a phenoxyphenyl group, a ( $\alpha,\alpha$ -dimethylbenzene)phenyl group, a (N,N'-dimethyl)aminophenyl group, a (N,N'-diphenyl)aminophenyl group, a pentalenyl group, an indenyl group, a naphthyl group, a methylnaphthyl group, an anthracenyl group, an azulenyl group, a heptalenyl group, an acenaphthylenyl group, a phenanthrenyl group, a fluorenyl group, an anthraquinolyl group, a triphenylene group, a pyrenyl group, a perylenyl group, a chloroperlylenyl group, a pentaphenyl group, a pentacenyl group, a tetraphenylenyl group, a hexaphenyl group, a coronenyl group, a trinaphthylenyl group, a heptaphenyl group, or a carbazolyl group.

Preferably, Ar may be a phenyl group, a lower alkylphenyl group, a lower alkoxyphenyl group, a cyanophenyl group, a phenoxyphenyl group, a halophenyl group, a naphthyl group, a lower alkyl naphthyl group, a lower alkoxy naphthyl group, a cyanonaphthyl group, a halonaphthyl group, a fluorenyl group, a carbazolyl group, a lower alkyl carbazolyl group, a biphenyl group, a lower alkyl biphenyl group, a lower alkoxy biphenyl group, a thiophenyl group, an indolyl group, or a pyridyl group. The lower alkyl and lower alkoxy may have 1-5 carbon atoms. More preferably, Ar is a monocyclic, bicy-

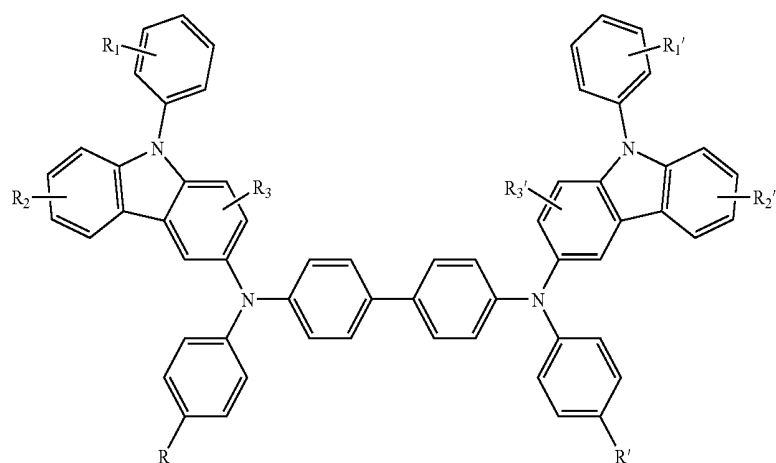
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clic, or tricyclic aryl group selected from a fluorenyl group, a carbazolyl group, a phenyl group, a naphthyl group, and a phenanthrenyl group, or an aryl group substituted with one to three, preferably, one C1-C3 lower alkyl, C1-C3 lower alkoxy, cyano, phenoxy, phenyl, or halogen on an aromatic ring thereof.

In Formula (1), one or more atoms of Ar may be substituted with a C1-C10 alkyl group, a C1-C10 alkoxy group, a nitro group, a halogen atom, an amino group, a C6-C10 aryl group, a C2-C10 heteroaryl group, a cyano group, a hydroxy group, etc.

The phenylcarbazole-based compound represented by Formula (1) has a high glass transition point or melting point due to a rigid carbazole group in its structure. Thus, the organic layer composed of the phenylcarbazole-based compound represented by Formula (1) has an increased resistance to joule heat generated in organic layers, between such organic layers, or between such an organic layer and a metal electrode when electroluminescence occurs, and an increased resistance to high temperature conditions, compared with the conventional organic layer. Therefore, when the compound represented by Formula (1) is used as a host material of a hole injection layer, a hole transport layer, or an emitting layer, it exhibits high luminance and can emit light for a long time. In particular, since this compound has at least two rigid carbazole groups in its molecule, the effects are further increased.

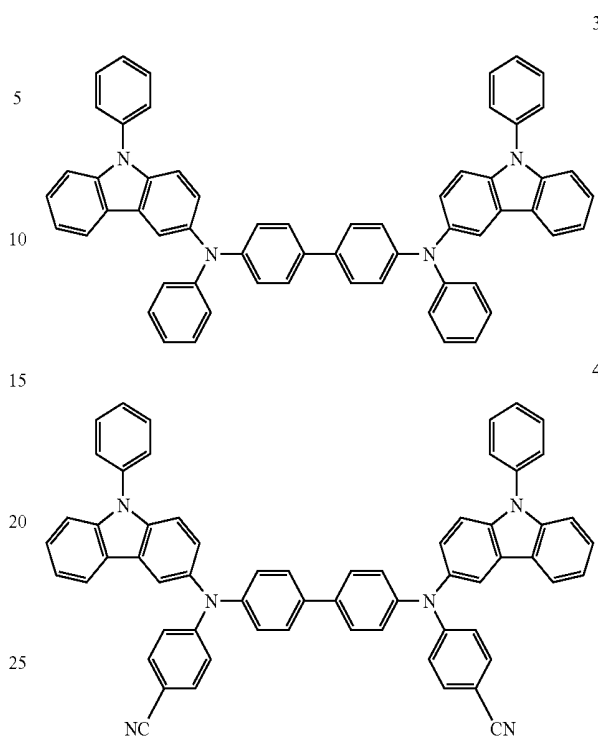
The compound represented by Formula (1) may be preferably a compound represented by Formula (2):



where each of  $R_1$  to  $R_3$  and  $R_1'$  to  $R_3'$  is independently a hydrogen atom, a fluorine atom, a cyano group, a substituted or unsubstituted C1-C30 alkyl group, a substituted or unsubstituted C6-C30 aryl group, a substituted or unsubstituted C2-C30 heterocyclic group, or a substituted or unsubstituted amino group; and each of  $R$  and  $R'$  is a hydrogen atom, a cyano group, a fluorine atom, a substituted or unsubstituted C1-C30 alkyl group, a substituted or unsubstituted C6-C30 aryl group, a substituted or unsubstituted C2-C30 heterocyclic group, or a substituted or unsubstituted amino group.

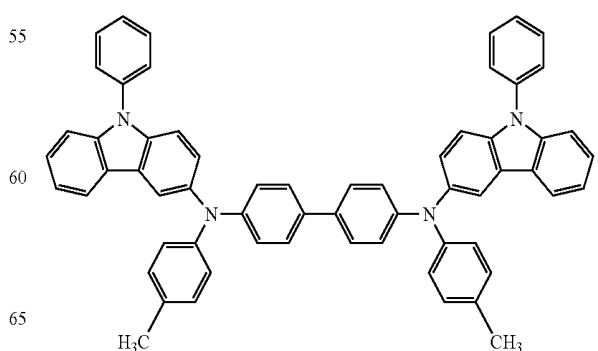
An organic EL device of the present invention has high durability when it is stored and operated. This is because the phenylcarbazole derivative has a high glass transition temperature  $T_g$ . The compound represented by Formula (1) serves as a hole injection material, a hole transport material, and/or an emitting material. Representative structures of embodiments of the present invention are represented by Formulae 3 through 27, but are not limited thereto:

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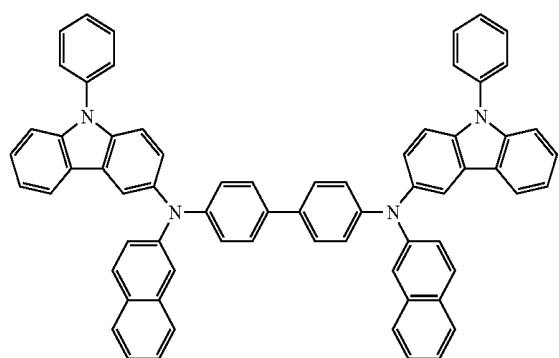
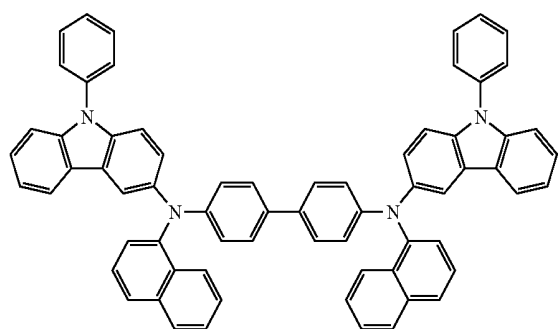
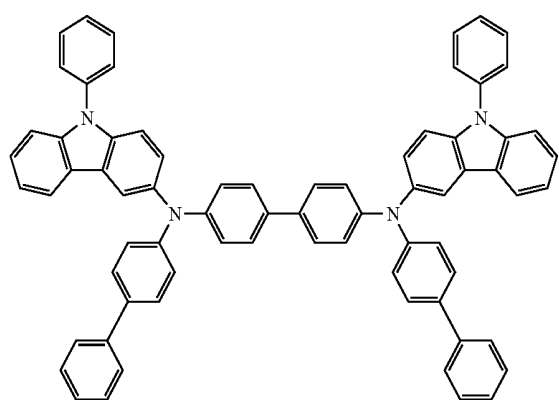
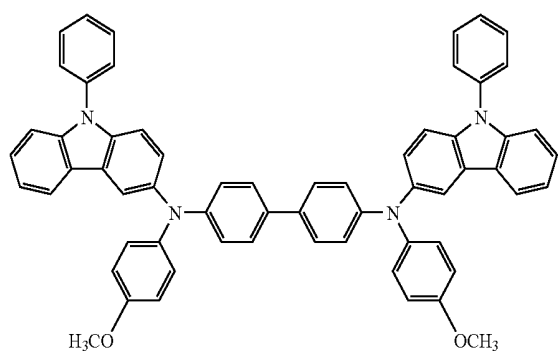
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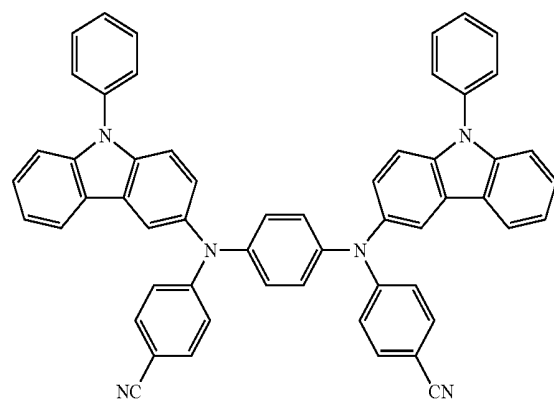
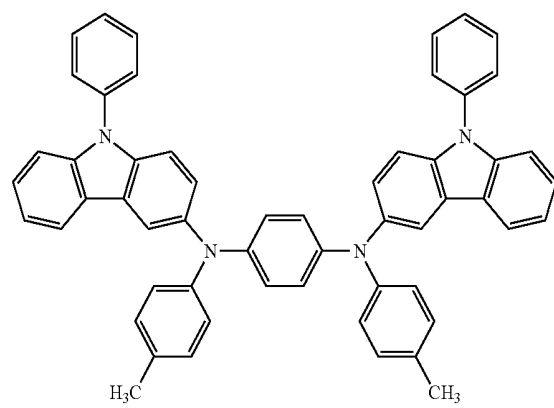
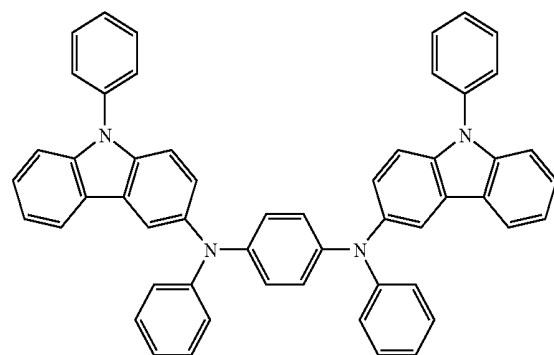
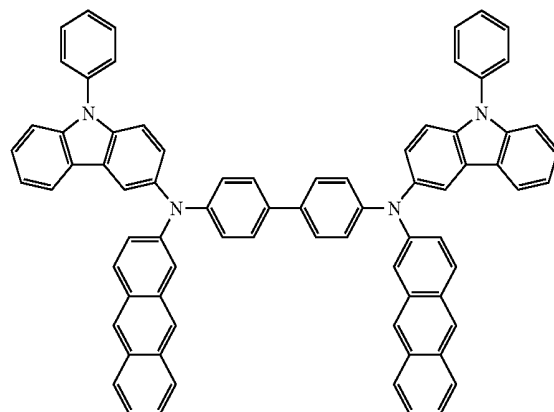
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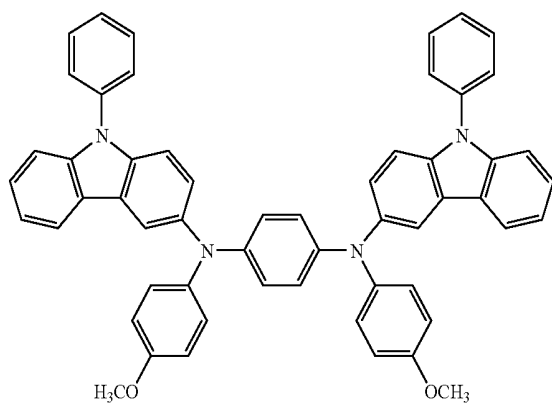
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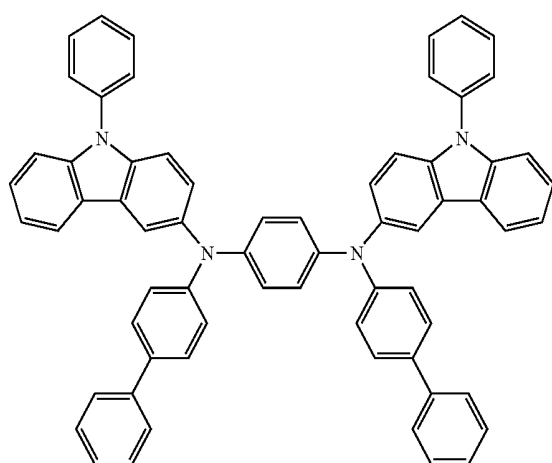
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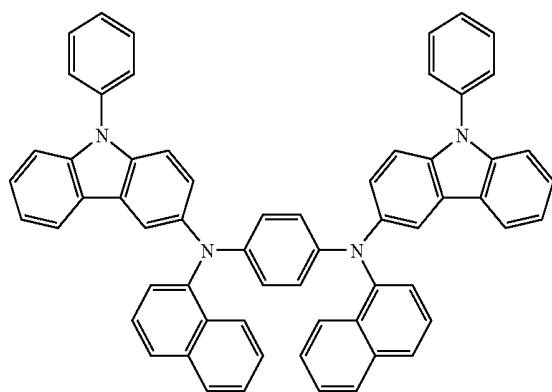
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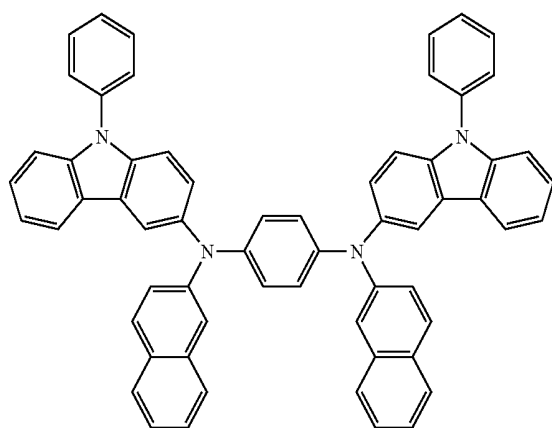


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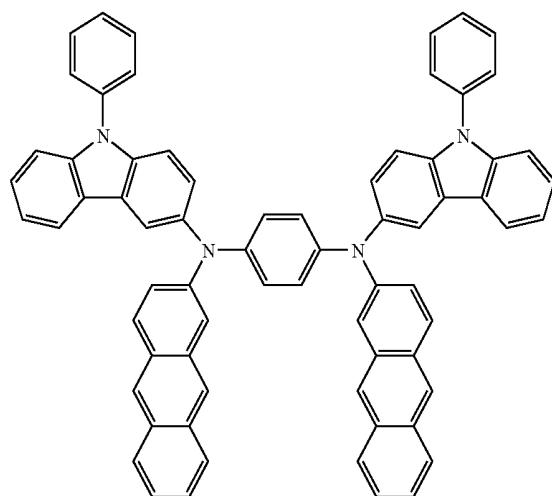
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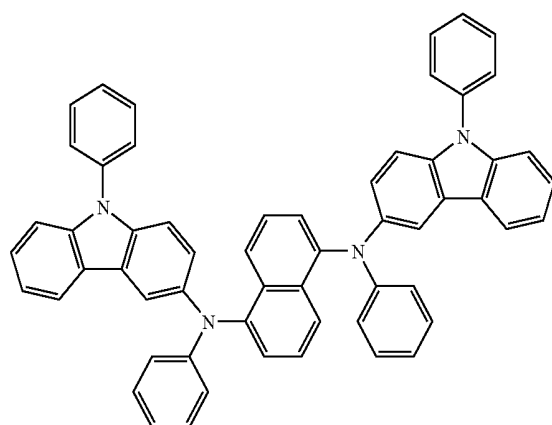
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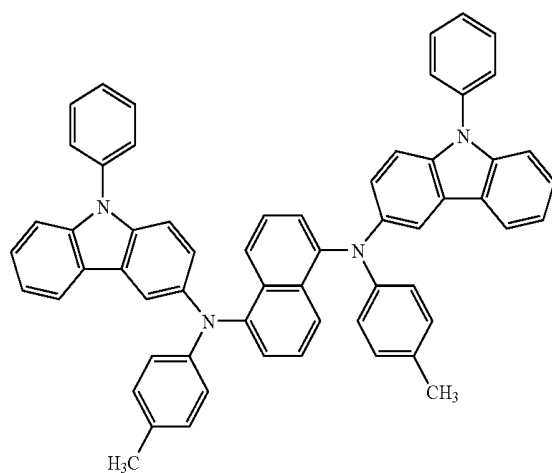
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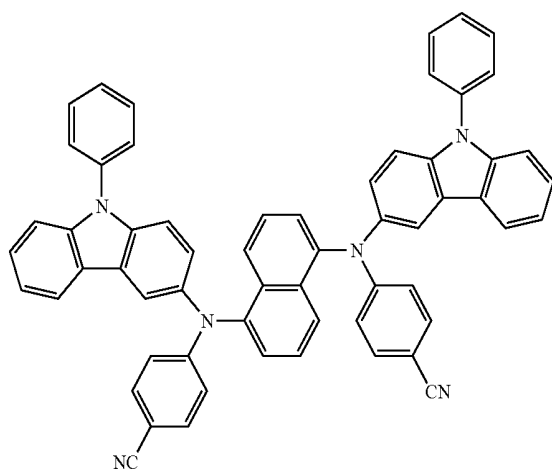
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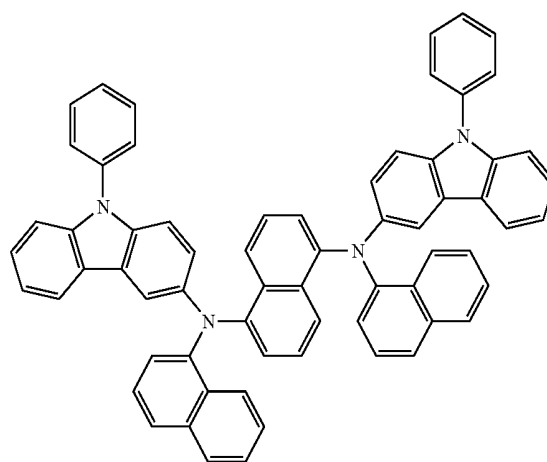
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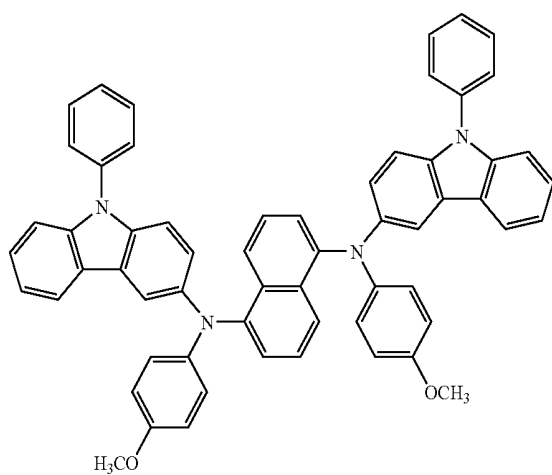
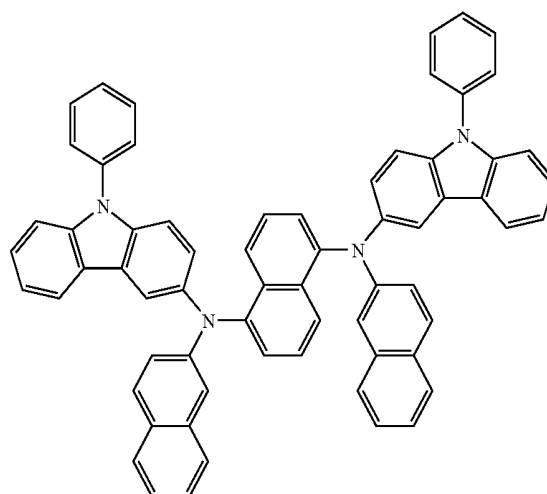
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H<sub>3</sub>CO

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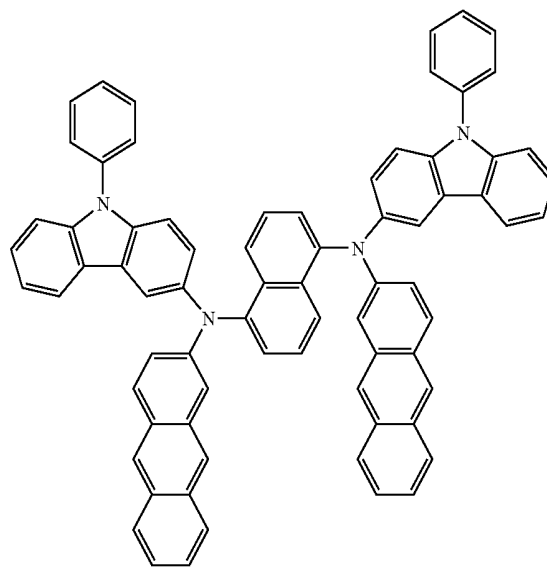
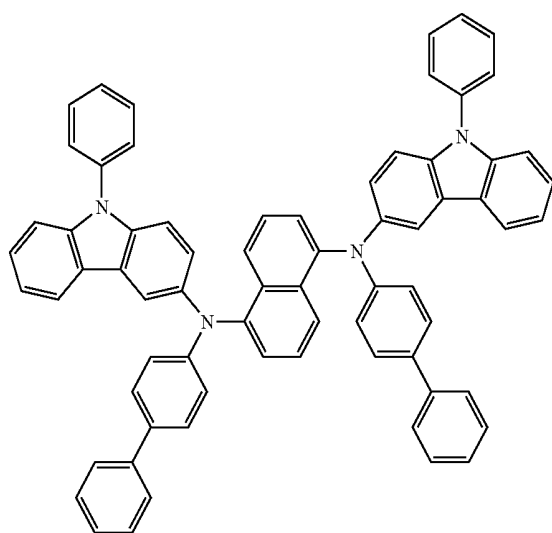
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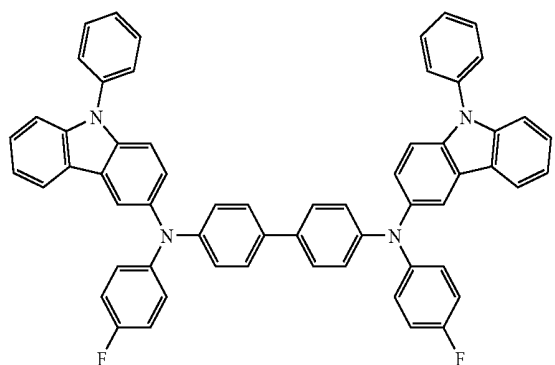


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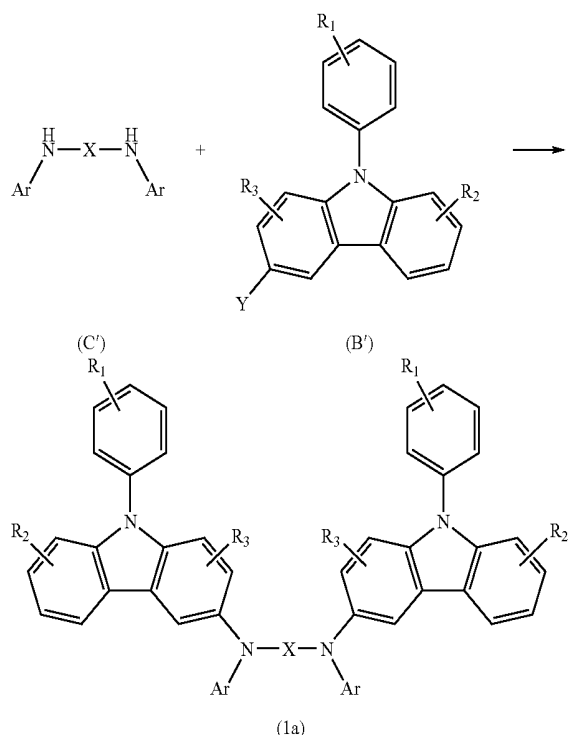


In the specification, the compounds represented by Formulae 3 through 27 are also referred to as the compounds 3 through 27, respectively.

An exemplary method of preparing one exemplary phenylcarbazole-based compound represented by Formula (1) will now be described.

As set forth in the reaction scheme 1, the phenylcarbazole-based compound represented by Formula (1a) is obtained by reacting carbazole (B')

Reaction Scheme 1



In above Reaction Scheme 1, X is a substituted or unsubstituted C1-C30 alkylene group, a substituted or unsubstituted C2-C30 alkenylene group, a substituted or unsubstituted C6-C30 arylene group, a substituted or unsubstituted C2-C30 heteroarylene group, or a substituted or unsubstituted C2-C30 heterocyclic group; each of R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> is independently a hydrogen atom, a substituted or unsubstituted C1-C30 alkyl group, a substituted or unsubstituted C1-C30 alkoxy group, a substituted or unsubstituted C6-C30 aryl group, a substituted or unsubstituted C6-C30 aryloxy

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group, a substituted or unsubstituted C2-C30 heterocyclic group, a substituted or unsubstituted C6-C30 condensed polycyclic group, a hydroxy group, a cyano group, or a substituted or unsubstituted amino group, and, alternatively, two or more adjacent groups among R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> on each side can be connected to each other to form a saturated or unsaturated carbocycle; each Ar is independently a substituted or unsubstituted C6-C30 aryl group or a substituted or unsubstituted C2-C30 heteroaryl group; and Y is a halogen atom.

The reaction is carried out in the presence of Pd<sub>2</sub>(dba)<sub>3</sub> (dba=dibenzylideneacetone), sodium tert-butoxide, and tri (tert-butyl)phosphine at a temperature of 50 to 150° C.

In the organic EL device of the present invention, an organic layer containing the phenylcarbazole-based compound represented by Formula (1) may be a hole injection layer or a hole transport layer, or a single layer serving as both a hole injection layer and a hole transport layer. An organic layer containing the phenylcarbazole-based compound represented by Formula (1) may be an emitting layer.

When the organic layer containing the phenylcarbazole-based compound represented by Formula (1) is a hole injection layer or a hole transport layer, the device has a first electrode/hole injection layer/emitting layer/second electrode structure, a first electrode/hole injection layer/emitting layer/hole transport layer/second electrode structure, or a first electrode/emitting layer/hole transport layer/second electrode structure, but not limited thereto.

The emitting layer is composed of a phosphorescent or fluorescent material.

When the organic layer containing the phenylcarbazole-based compound represented by Formula (1) is the emitting layer, the phenylcarbazole-based compound is used as a fluorescent or phosphorescent host.

A method of manufacturing an organic electroluminescent device according to an embodiment of the present invention will now be described.

FIG. 1 is a cross-sectional view of an organic electroluminescent device according to an embodiment of the present invention.

First, a material for forming an anode, which has a high work function, is deposited or sputtered on a substrate to form an anode. Any substrate used in a conventional organic EL device can be used, and a glass substrate or a transparent plastic substrate having good mechanical strength, thermal stability, transparency, surface softness, manageability, and water-proofness may be preferably used. The anode may be composed of indium tin oxide (ITO), indium zinc oxide (IZO), tin oxide (SnO<sub>2</sub>), or zinc oxide (ZnO), which is transparent and have good conductivity.

Next, a hole injection layer (HIL) is formed on the anode using a vacuum evaporation, spin coating, casting, or Langmuir-Blodgett (LB) method. The HIL may be formed using the vacuum evaporation method, since it is easy to obtain uniform film quality and the generation of pinholes is suppressed.

When the HIL is formed using the vacuum evaporation method, the deposition conditions can be varied according to the type of compound used as a hole injection material, and the desired structure and thermal property of the HIL, but may include a deposition temperature of 50 to 500° C., a vacuum of 10<sup>-8</sup> to 10<sup>-3</sup> torr, a deposition rate of 0.01 to 100 Å/sec, and a film thickness of 10 Å to 5 μm.

A HIL material is not particularly restricted, but may be the compound represented by Formula (1), a phthalocyanine-based compound, such as CuPc, as disclosed in U.S. Pat. No. 4,356,429 which is incorporated herein by reference, or a Starburst type amine derivatives, for example, TCTA, m-MT-

DATA, or m-MTDAPB, as described in Advanced Material, 6, p. 677 (1994) which is incorporated herein by reference.

Then, a hole transport layer (HTL) is formed on the HIL using a vacuum evaporation, spin coating, casting, or Langmuir-Blodgett method. The HTL may be formed using the vacuum evaporation method, since it is easy to obtain uniform film quality and the generation of pinholes is suppressed. When the HTL is formed using the vacuum evaporation method, the deposition conditions vary according to the type of compound to be used, but may be almost identical to the deposition conditions for the HIL.

A HTL material is not particularly restricted, but may be the phenylcarbazole-based compound represented by Formula (1), or any compound known to be used in the HTL. For example, carbazole derivatives, such as N-phenylcarbazole and polyvinylcarbazole, general amine derivatives having an aromatic condensed ring, such as N,N'-bis(3-methylphenyl)-N,N'-diphenyl-[1,1-biphenyl]-4,4'-diamine (TPD), N,N'-di(naphthalen-1-yl)-N,N'-diphenyl benzidine ( $\alpha$ -NPD), etc. are used.

Then, an emitting layer (EML) is formed on the HTL using a vacuum evaporation, spin coating, casting, or Langmuir-Blodgett method. The EML may be formed using the vacuum evaporation method since it is easy to obtain uniform film quality and the generation of pinholes is suppressed. When the EML is formed using the vacuum evaporation method, the deposition conditions vary according to the type of compound to be used, but may be almost identical to the deposition conditions for the HIL.

An EML material is not particularly restricted, but may be the phenylcarbazole-based compound represented by Formula (1) as a fluorescent or phosphorescent host. Alq<sub>3</sub> (tris(8-quinolinolate) aluminum) may be used as a fluorescent host. IDE102 and IDE105 available from Idemitsu Kosan Co., Ltd., and C545T available from Hayashibara may be used as fluorescent dopants, and Ir(PPy)<sub>3</sub> (PPy=phenylpyridine) (green), F2Irpic (bis[2-(4,6-difluorophenyl)pyridinato-N,C2']iridium picolinate) (blue), and RD61 (red) may be vacuum evaporated (doped) in combination as phosphorescent dopants.

The concentration of the dopant is not particularly restricted, but is typically 0.01-15 parts by weight based of total 100 parts by weight of the host and the dopant.

When the phosphorescent dopant is used in the EML, a hole blocking material is vacuum evaporated or spin coated on the EML to form a hole blocking layer (HBL), in order to prevent a triplet exciton or a hole from diffusing into the ETL. The hole blocking material is not particularly restricted, but may be selected from materials which are used as a hole blocking material in the art. For example, oxadiazole derivatives or triazole derivatives, phenanthroline derivatives, or hole blocking materials described in JP 11-329734 (A1) which is incorporated herein by reference and the like may be used and, more particularly, bis(2-methyl-8-quinolinolate) (4-phenylphenolato)aluminum (BALq), 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP) are used.

Then, an electron transport layer (ETL) is formed using a vacuum evaporating, spin coating, or casting method. Preferably, the ETL is formed using the vacuum evaporating method. An ETL material may be any material which can stably transport electrons injected from an electron injection electrode (cathode) and, more particularly, tris(8-quinolinolate) aluminum (Alq<sub>3</sub>) may be used. In addition, an electron injection layer (EIL) for facilitating the injection of electrons from the cathode may be deposited on the ETL and a material for the injection layer is not particularly restricted.

LiF, NaCl, CsF, Li<sub>2</sub>O, BaO, etc. may be used as the EIL material. The deposition conditions of the HBL, the ETL, and the EIL vary according to the type of compound to be used, but may be almost identical to the deposition conditions for HIL.

Finally, a metal for a cathode is vacuum evaporated or sputtered on the EIL to form a cathode. The metal for a cathode may be a metal having a low work function, alloy, electric conducting compound, and a mixture thereof. Examples of such a material include Li, Mg, Al, Al—Li, Ca, Mg—In, or Mg—Ag. Also, a transmittance type cathode composed of ITO or IZO may form a front surface of the light emitting device.

The organic EL device according to an embodiment of the present invention may include one or two intermediate layers in addition to the anode, the HIL, the HTL, the EML, the ETL, the EIL, and the cathode as illustrated in FIG. 1.

The compound represented by Formula (1) is useful as a light emitting material having superior light emitting property and hole transport property, in particular, as a host, and may also be used as a hole injection material and/or a hole transport material of blue, green, red fluorescent and phosphorescent devices.

Representative groups used in Formula (1) will now be defined.

Specific examples of the unsubstituted C1-C30 alkyl group in Formula (1) include methyl, ethyl, propyl, isobutyl, sec-butyl, pentyl, iso-amyl, hexyl, and the like. The hydrogen atoms of the alkyl group may be unsubstituted, or at least one hydrogen atom on the alkyl group may be substituted with a halogen atom, a C1-C30 alkyl group, a C1-C30 alkoxy group, a lower alkylamino group, a hydroxy group, a nitro group, a cyano group, an amino group, an amidino group, hydrazine, hydrazone, a carboxyl group, a sulfonic acid group, a phosphoric acid group, etc.

Specific examples of the unsubstituted C2-C30 alkenyl group in Formula (1) include an ethylene group, a propylene group, an isobutylene group, a vinyl group, and an allyl group. The hydrogen atoms of the alkenyl group may be unsubstituted, or at least one hydrogen atom on the alkenyl group may be substituted with a halogen atom, a C1-C30 alkyl group, a C1-C30 alkoxy group, a lower alkylamino group, a hydroxy group, a nitro group, a cyano group, an amino group, an amidino group, hydrazine, hydrazone, a carboxyl group, a sulfonic acid group, a phosphoric acid group, etc.

Specific examples of the unsubstituted C1-C30 alkoxy group in Formula (1) include methoxy, ethoxy, propoxy, isobutyloxy, sec-butyloxy, pentyloxy, iso-amtyloxy, and hexyloxy. The hydrogen atoms of the alkoxy group may be unsubstituted, or at least one hydrogen atom on the alkoxy group may be substituted with the same substituent as in the above-described C1-C20 alkyl group.

The aryl group in Formula (1) is a carbocyclic aromatic system that includes one or more rings, in which the rings may be attached or fused by a pendent method. Specific examples of the aryl group include phenyl, naphthyl, tetrahydronaphthyl, and the like. The hydrogen atoms of the aryl group may be unsubstituted, or at least one hydrogen atom on the aryl group may be substituted with the same substituent as in the above-described C1-C20 alkyl group.

The heteroaryl group in Formula (1) is a monovalent monocyclic ring system, which contains one, two or three heteroatoms selected from N, O, P, and S and has carbon atoms as the remaining ring atoms, in which the rings may be attached or fused by a pendent method. Examples of such a heteroaryl group include pyridyl, thienyl, furyl, and the like.

The heterocyclic group in Formula (1) is a monocyclic system, which contains one, two or three heteroatoms

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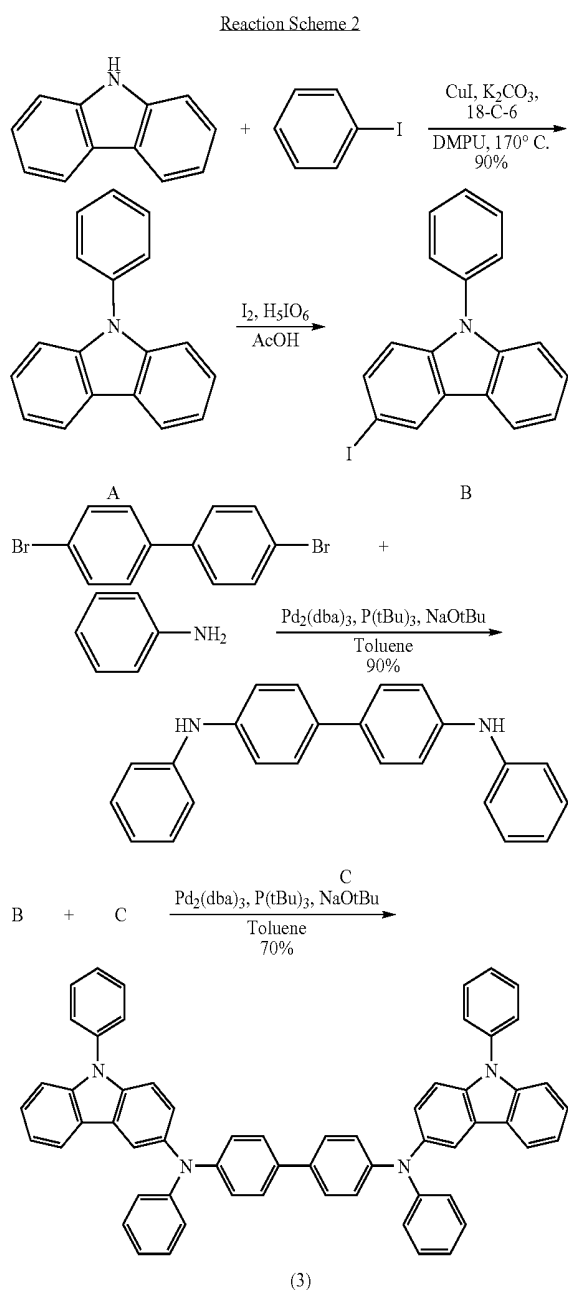
selected from N, O, P, and S and has carbon atoms as the remaining ring atoms, in which some hydrogen atoms on the cycloalkyl group are substituted with lower alkyl groups. At least one hydrogen atom on the cycloalkyl group may be substituted with the same substituent as in the above-described C1-C20 alkyl group.

The present invention will be described in greater detail with reference to the following examples. The following examples are for illustrative purposes and are not intended to limit the scope of the invention.

## SYNTHESIS EXAMPLE 1

## Preparation of Compound Represented by Formula 3

Compound represented by Formula 3 (i.e., Compound 3) was synthesized according to the reaction scheme 2.



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## Synthesis of Intermediate Compound A

Carbazole (16.7 g, 100 mmol), iodobenzene (26.5 g, 130 mmol), CuI (1.9 g, 10 mmol), K<sub>2</sub>CO<sub>3</sub> (138 g, 1 mol), and 18-crown-6 (530 mg, 2 mmol) were dissolved in 1,3-dimethyl-3,4,5,6-tetrahydro-(1H)-pyrimidinone (DMPU) (500 mL), and then heated at 170° C. for 8 hrs.

After the reaction was completed, the reaction mixture was cooled to room temperature and solid materials were filtered. A small amount of aqueous ammonia was added to the filtrate, and the resultant was three times washed with diethyl ether (300 mL). The washed diethyl ether layer was dried on MgSO<sub>4</sub> and dried under reduced pressure to obtain a crude product. The crude product was purified with a silica gel column chromatography to obtain 22 g of the intermediate compound A as a white solid (yield: 90%).

<sup>1</sup>H NMR (CDCl<sub>3</sub>, 400 MHz) δ (ppm) 8.12 (d, 2H), 7.58-7.53 (m, 4H), 7.46-7.42 (m, 1H), 7.38 (d, 4H), 7.30-7.26 (m, 2H); <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100 MHz) δ (ppm) 141.0, 137.9, 130.0, 127.5, 127.3, 126.0, 123.5, 120.4, 120.0, 109.9.

## Synthesis of Intermediate Compound B

2.433 g (10 mmol) of the intermediate compound A was added to 100 mL of 80% acetic acid, and then 1.357 g (5.35 mmol) of iodine (I<sub>2</sub>) and 0.333 g (1.46 mmol) of orthoperiodic acid (H<sub>5</sub>IO<sub>6</sub>) were added thereto. The resultant was stirred under nitrogen atmosphere at 80° C. for 2 hrs.

After the reaction was completed, the reaction mixture was three times extracted with ethyl ether (50 mL). The collected organic layer was dried on magnesium sulfate and the solvent was evaporated. The residue was purified with a silica gel column chromatography to obtain 3.23 g of the intermediate compound B as a white solid (yield: 87%).

<sup>1</sup>H NMR (CDCl<sub>3</sub>, 300 MHz) δ (ppm) 8.43 (d, 1H), 8.05 (d, 1H), 7.62 (dd, 1H), 7.61-7.75 (m, 2H), 7.51-7.43 (m, 3H), 7.41-7.35 (m, 2H), 7.27 (dd, 1H), 7.14 (d, 1H).

## Synthesis of Intermediate Compound C

3.12 g (10 mmol) of 4,4'-dibromodiphenyl, 2.3 mL (25 mmol) of aniline, 2.9 g (30 mmol) of t-BuONa, 183 mg (0.2 mmol) of Pd<sub>2</sub>(dba)<sub>3</sub>, and 20 mg (0.1 mmol) of P(t-Bu)<sub>3</sub> were dissolved in 30 mL of toluene, and then stirred at 90° C. for 3 hrs.

The reaction mixture was cooled to room temperature and three times extracted with distilled water and diethyl ether. Precipitates in an organic layer were filtered, washed with acetone and diethyl ether, and then dried in vacuum to obtain 0.3 g of the intermediate compound C (yield: 90%). The structure of the intermediate compound C was identified with <sup>1</sup>H NMR.

<sup>1</sup>H NMR (DMSO-d<sub>6</sub>, 400 MHz) δ (ppm) 8.22 (s, 2H), 7.48 (d, 4H), 7.23 (t, 4H), 7.10 (dd, 8H), 6.82 (t, 2H); <sup>13</sup>C NMR (DMSO-d<sub>6</sub>, 100 MHz) δ (ppm) 145.7, 144.3, 133.7, 131.4, 128.7, 121.2, 119.2, 118.9.

## Synthesis of Compound Represented by Formula 3

912 mg (2.47 mmol) of the intermediate compound B, 336.4 mg (1 mmol) of the intermediate compound C, 300 mg (3 mmol) of t-BuONa, 40 mg (0.02 mmol) of Pd<sub>2</sub>(dba)<sub>3</sub>, and 3 mg (0.01 mmol) of P(t-Bu)<sub>3</sub> were dissolved in 5 mL of toluene, and then stirred at 90° C. for 3 hrs.

After the reaction was completed, the resultant was cooled to room temperature and three times extracted with distilled water and diethyl ether. The collected organic layer was dried on magnesium sulfate and the solvent was evaporated. The residue was purified with a silica gel column chromatography to obtain 570 mg of Compound represented by Formula 3 as a yellow solid (yield: 70%).

<sup>1</sup>H NMR (CDCl<sub>3</sub>, 300 MHz) δ (ppm) 7.99 (d, 2H), 7.95 (s, 2H), 7.61-7.57 (m, 8H), 7.48-7.32 (m, 12H), 7.27-7.19 (m, 8H), 7.18-7.10 (m, 8H), 6.96 (t, 2H); <sup>13</sup>C NMR (CDCl<sub>3</sub>, 100 MHz) δ (ppm) 148.4, 147.3, 141.3, 140.4, 138.0, 137.6, 133.9, 129.9, 129.1, 127.4, 127.1, 127.0, 126.1, 125.6, 124.3, 123.0, 122.9, 122.8, 121.7, 120.5, 119.9, 118.5, 110.7, 109.9.

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The obtained Compound represented by Formula 3 was diluted with  $\text{CHCl}_3$  to a concentration of 0.2 mM and the UV spectrum thereof was obtained. A maximum absorption wavelength of 351 nm was observed in the UV spectrum (FIG. 2).

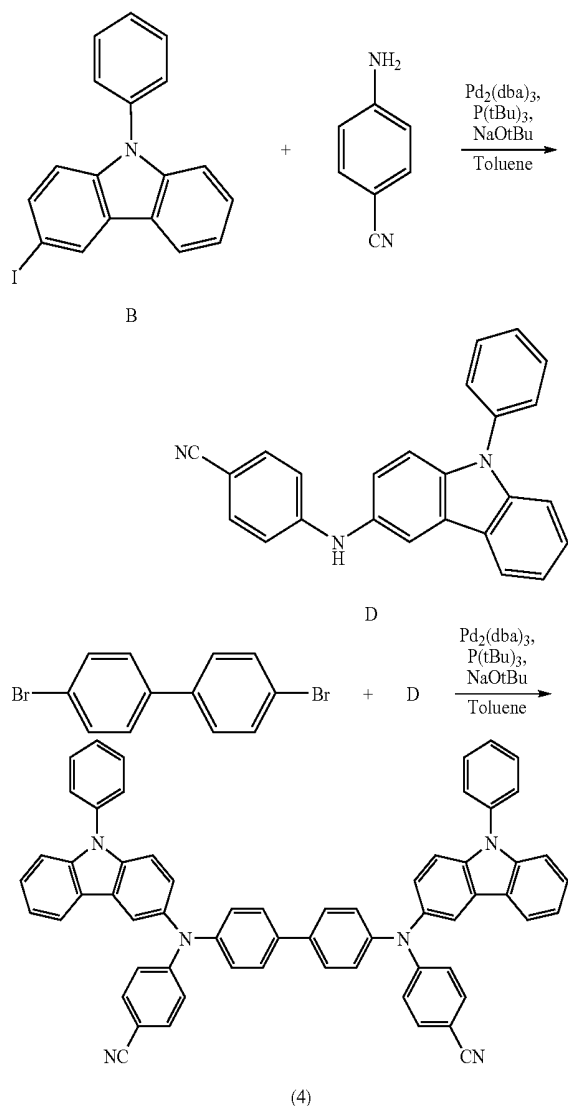
Further, Compound represented by Formula 3 was subjected to thermal analysis using TGA (Thermo Gravimetric Analysis) and DSC (Differential Scanning Calorimetry) ( $\text{N}_2$  atmosphere, temperature range: room temperature—600° C. (10° C./min)—TGA, room temperature—400° C.—DSC, Pan type: Pt pan in disposable Al pan (TGA), disposable Al pan (DSC)) to obtain Td 494° C. and Tg 153° C. (FIGS. 3 and 4).

A HOMO (Highest Occupied Molecular Orbital) energy level of 5.16 eV and a LUMO (Lowest Occupied Molecular Orbital) energy level of 2.16 eV were obtained using UV absorption spectrum and a potentiometer AC-2.

## SYNTHESIS EXAMPLE 2

## Preparation of Compound Represented by Formula 4

Compound represented by Formula 4 (i.e., Compound 4) was synthesized according to the reaction scheme 3.



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## Synthesis of Intermediate Compound D

3.69 g (10 mmol) of the intermediate compound B, 1.42 g (12 mmol) of 4-aminobenzonitrile, 1.44 g (15 mmol) of  $\text{t-BuONa}$ , 183 mg (0.2 mmol) of  $\text{Pd}_2(\text{dba})_3$ , and 40 mg (0.2 mmol) of  $\text{P}(\text{t-Bu})_3$  were dissolved in 50 mL of toluene, and then stirred at 90° C. for 3 hrs.

After the reaction was completed, the reaction mixture was cooled to room temperature and three times extracted with distilled water and diethyl ether. The collected organic layer was dried on magnesium sulfate and the solvent was evaporated. The residue was purified with a silica gel column chromatography to obtain 1.8 g of the intermediate compound D (yield: 50%).

## Synthesis of Compound represented by Formula 4

222 mg (0.61 mmol) of the intermediate compound D, 78 mg (0.25 mmol) of 4,4'-dibromodiphenyl, 80 mg (0.75 mmol) of  $\text{t-BuONa}$ , 310 mg (0.01 mmol) of  $\text{Pd}_2(\text{dba})_3$ , and 2 mg (0.01 mmol) of  $\text{P}(\text{t-Bu})_3$  were dissolved in 5 mL of toluene, and then stirred at 90° C. for 3 hrs.

After the reaction was completed, the reaction mixture was cooled to room temperature and three times extracted with distilled water and diethyl ether. The collected organic layer was dried on magnesium sulfate and the solvent was evaporated. The residue was purified with a silica gel column chromatography to obtain 186 mg of Compound represented by Formula 4 as a yellow solid (yield: 86%).

$^1\text{H}$  NMR ( $\text{CDCl}_3$ , 300 MHz)  $\delta$  (ppm) 8.02 (d, 2H), 7.97 (d, 2H), 7.64-7.48 (m, 14H), 7.43-7.39 (m, 10H), 7.29-7.22 (m, 8H), 7.03 (d, 4H);  $^{13}\text{C}$  NMR ( $\text{CDCl}_3$ , 100 MHz)  $\delta$  (ppm) 152.1, 145.6, 141.5, 138.9, 138.2, 137.3, 136.3, 133.2, 130.0, 127.9, 127.8, 127.0, 126.6, 125.8, 125.5, 124.6, 122.7, 120.5, 120.2, 119.9, 119.4, 118.9, 111.2, 110.1, 101.8.

The obtained Compound represented by Formula 4 was diluted with  $\text{CHCl}_3$  to a concentration of 0.2 mM and the UV spectrum thereof was obtained. A maximum absorption wavelength of 351 nm was observed in the UV spectrum (FIG. 5).

Further, Compound represented by Formula 4 was subjected to thermal analysis using TGA and DSC ( $\text{N}_2$  atmosphere, temperature range: room temperature—600° C. (10° C./min)—TGA, room temperature—400° C.—DSC, Pan type: Pt pan in disposable Al pan (TGA), disposable Al pan (DSC)) to obtain Td 490° C., Tg 178° C., and Tm of 263° C. (FIGS. 6 and 7).

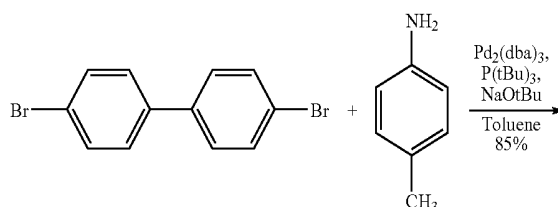
A HOMO energy level of 5.30 eV and a LUMO energy level of 2.37 eV were obtained using UV absorption spectrum and a potentiometer AC-2.

## SYNTHESIS EXAMPLE 3

## Preparation of Compound Represented by Formula 5

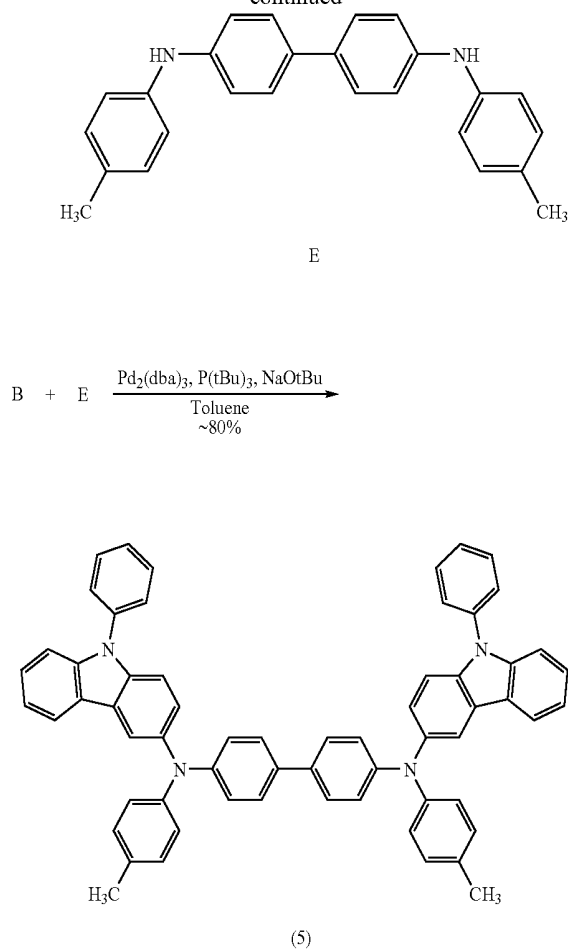
Compound represented by Formula 5 (i.e., Compound 5) was synthesized through Reaction Scheme 3 below.

Reaction Scheme 3



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-continued



Intermediate E was synthesized by reacting 4,4'-dibromodiphenyl and p-tolylamine in the same manner as in the synthesis of Intermediate C (yield: 85%). 678 mg of Compound 5 as a yellow solid was obtained by reacting Intermediates B and E in the same manner as in the synthesis of Compound 3 (yield: 80%).

$^1\text{H}$  NMR ( $\text{C}_6\text{D}_6$ , 400 MHz)  $\delta$  (ppm) 8.14 (d, 2H), 7.64 (d, 2H), 7.47 (d, 4H), 7.38-7.28 (m, 6H), 7.27-7.25 (m, 8H), 7.23-7.01 (m, 16H), 6.96 (d, 2H), 2.19 (s, 6H);  $^{13}\text{C}$  NMR ( $\text{C}_6\text{D}_6$ , 100 MHz)  $\delta$  (ppm) 149.0, 147.5, 142.6, 142.2, 139.1, 138.9, 135.1, 132.6, 130.1, 130.7, 128.1, 127.9, 127.2, 126.5, 125.9, 125.0, 124.5, 123.6, 121.8, 121.1, 119.2, 111.8, 110.8, 21.5.

The obtained Compound 5 was diluted with  $\text{CHCl}_3$  to a concentration of 0.2 mM and the UV spectrum therefor was obtained. Maximum absorption wavelengths of 358, 309, and 253 nm were observed in the UV spectrum.

Further, Compound 5 was subjected to thermal analysis using Thermo Gravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) ( $\text{N}_2$  atmosphere, temperature range: room temperature—600° C. (10° C./min)—TGA, room temperature—400° C.—DSC, Pan Type Pt Pan in disposal Al Pan (TGA), disposable Al pan (DSC)) to obtain Td 480° C. and Tg 155° C.

A Highest Occupied Molecular Orbital (HOMO) energy level of 5.0 eV and a Lowest Occupied Molecular Orbital (LUMO) energy level of 2.02 eV were obtained using a UV absorption spectrum and a potentiometer AC-2.

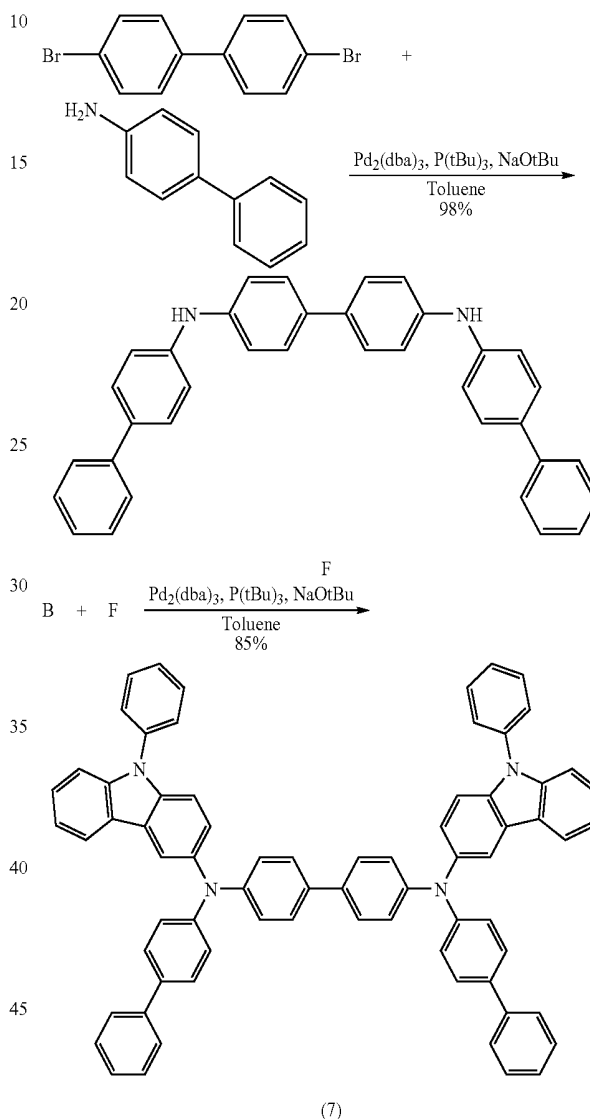
22

## SYNTHESIS EXAMPLE 4

## Preparation of Compound Represented by Formula 7

Compound represented by Formula 7 (i.e., Compound 7) was synthesized through Reaction Scheme 5 below.

Reaction Scheme 5



Intermediate F was synthesized by reacting 4,4'-dibromodiphenyl and 4-aminobiphenyl in the same manner as in the synthesis of Intermediate C (yield: 98%). 826 mg of Compound 7 as a yellow solid was obtained by reacting Intermediates B and F in the same manner as in the synthesis of Compound 3 (yield: 85%).

$^1\text{H}$  NMR ( $\text{CD}_2\text{Cl}_2$ , 400 MHz)  $\delta$  (ppm) 8.02-8.01 (m, 4H), 7.65-7.56 (m, 12H), 7.51-7.46 (m, 10H), 7.43-7.36 (m, 10H), 7.32-7.17 (m, 14H);  $^{13}\text{C}$  NMR ( $\text{CD}_2\text{Cl}_2$ , 100 MHz)  $\delta$  (ppm) 148.2, 147.6, 141.8, 141.0, 140.6, 138.6, 137.9, 134.5, 134.4, 130.3, 129.1, 127.9, 127.8, 127.4, 127.3, 127.0, 126.8, 126.6, 126.1, 124.7, 123.5, 123.4, 123.0, 120.8, 120.3, 119.0, 111.1, 110.3.

The obtained Compound 7 was diluted with  $\text{CHCl}_3$  to a concentration of 0.2 mM and the UV spectrum therefor was obtained. A maximum absorption wavelength of 329 nm was observed in the UV spectrum.

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Further, Compound 7 was subjected to thermal analysis using TGA and DSC (N<sub>2</sub> atmosphere, temperature range: room temperature—600° C. (10° C./min)—TGA, room temperature—400° C.—DSC, Pan Type: Pt Pan in disposal Al Pan (TGA), disposal Al pan (DSC)) to obtain Td 533° C. and Tg 174° C.

A HOMO energy level of 5.2 eV and a LUMO energy level of 2.27 eV were obtained using a UV absorption spectrum and a potentiometer AC-2.

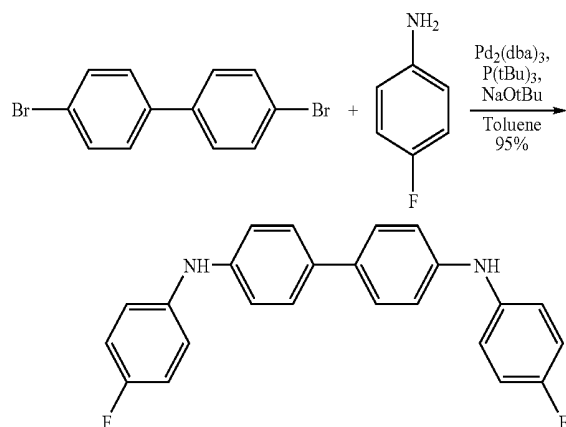
## SYNTHESIS EXAMPLE 5

Preparation of Compound Represented by Formula

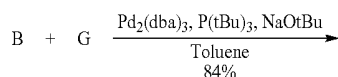
27

Compound represented by Formula 27 (i.e., Compound 27) was synthesized through Reaction Scheme 6 below.

Reaction Scheme 6

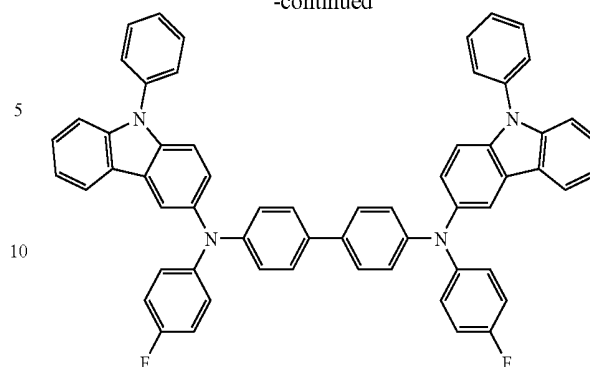


G



24

-continued



(27)

Intermediate G was synthesized by reacting 4,4'-dibromodiphenyl and 4-fluorophenylamine in the same manner as in the synthesis of Intermediate C (yield: 95%). 718 mg of Compound 27 as a yellow solid was obtained by reacting Intermediates B and G in the same manner as in the synthesis of Compound 3 (yield: 84%).

<sup>1</sup>H NMR (C<sub>6</sub>D<sub>6</sub>, 400 MHz) δ (ppm) 8.05 (s, 2H), 7.68 (d, 2H), 7.48 (d, 4H), 7.29-7.11 (m, 22H), 7.09-7.01 (m, 6H), 6.78 (t, 4H).

The obtained Compound 27 was diluted with CHCl<sub>3</sub> to a concentration of 0.2 mM and the UV spectrum thereof was obtained. Maximum absorption wavelengths of 351, 297, and 248 nm were observed in the UV spectrum.

Further, Compound 27 was subjected to TGA and DSC (N<sub>2</sub> atmosphere, temperature range: room temperature—600° C. (10° C./min)—TGA, room temperature—400° C.—DSC, Pan Type Pt Pan in disposal Al Pan (TGA), disposal Al pan (DSC)) to obtain Td 464° C., Tg 151° C., and Tm 299° C.

A HOMO energy level of 5.1 eV and a LUMO energy level of 2.28 eV were obtained using a UV absorption spectrum and a potentiometer AC-2.

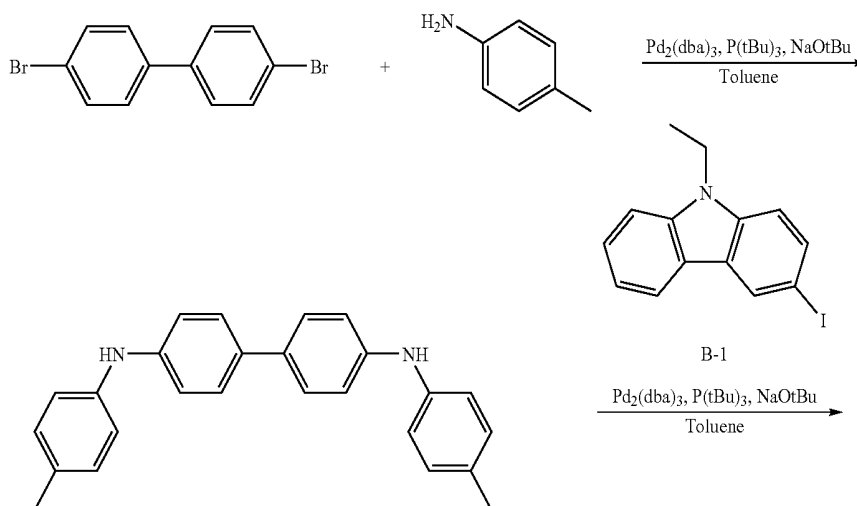
## SYNTHESIS REFERENCE EXAMPLE 1

Preparation of Compound Represented by Formula

28

Compound represented by Formula 28 (i.e., Compound 28) was synthesized through Reaction Scheme 7 below.

Reaction Scheme 7

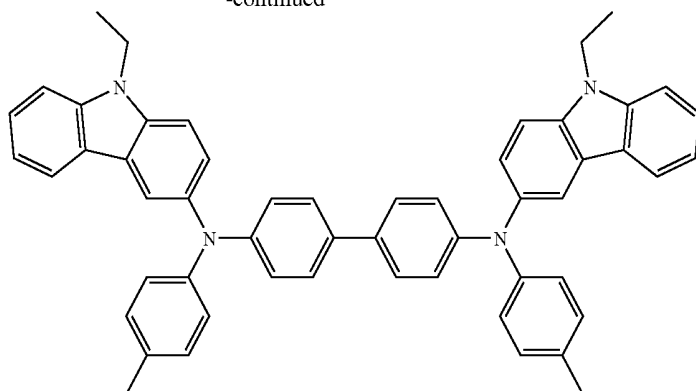


E

25

26

-continued



(28)

Intermediate B-1 was synthesized in the same manner as in the synthesis of Intermediate B (yield: 92%). 586 mg of

Compound 28 as a yellow solid was obtained by reacting Intermediates B-1 and E in the same manner as in the synthesis of Compound 3 (yield: 78%).

$^1\text{H}$  NMR ( $\text{CD}_2\text{Cl}_2$ , 400 MHz)  $\delta$  (ppm) 7.96-7.91 (m, 4H), 7.43-7.06 (m, 26H), 4.37 (q, 4H), 2.31 (s, 6H), 1.43 (t, 6H);  $^{13}\text{C}$  NMR ( $\text{CD}_2\text{Cl}_2$ , 100 MHz)  $\delta$  (ppm) 148.1, 146.3, 140.8, 139.9, 137.5, 133.4, 132.1, 130.0, 127.1, 126.1, 125.7, 124.1,

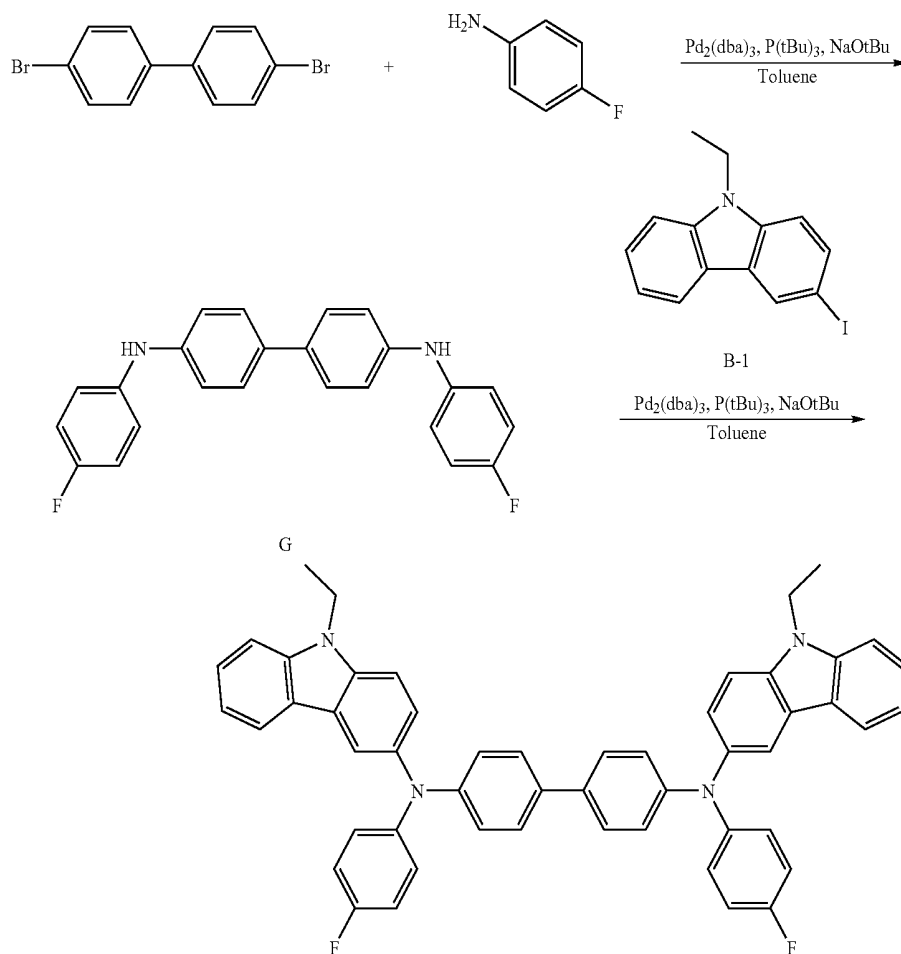
123.9, 122.9, 122.0, 120.7, 119.0, 118.9, 109.7, 109.0, 38.0, 20.8, 14.0.

## SYNTHESIS REFERENCE EXAMPLE 2

## Preparation of Compound Represented by Formula 29

Compound represented by Formula 29 (i.e., Compound 29) was synthesized through Reaction Scheme 8 below.

Reaction Scheme 8



(29)

27

Intermediate B-1 was synthesized in the same manner as in the synthesis of Intermediate B (yield: 92%). 630 mg of Compound 29 as a yellow solid was obtained by reacting Intermediates B-1 and G in the same manner as in the synthesis of Compound 3 (yield: 83%).

$^1\text{H}$  NMR ( $\text{CD}_2\text{Cl}_2$ , 400 MHz)  $\delta$  (ppm) 7.96 (d, 2H), 7.88 (d, 2H), 7.46-6.92 (m, 26H), 4.35 (q, 4H), 1.44 (t, 3H);  $^{13}\text{C}$  NMR ( $\text{CD}_2\text{Cl}_2$ , 100 MHz)  $\delta$  (ppm) 158.2 (d), 147.6, 144.6, 140.0, 139.3, 137.1, 133.4, 127.0, 125.8, 125.2, 125.1, 125.0, 123.2 (d), 121.6, 120.6, 118.8, 118.5, 115.8 (d), 109.3, 108.5, 37.6, 13.9.

#### Example 1

A coming 15  $\Omega/\text{cm}^2$  (1200 Å) ITO glass substrate as an anode was cut to a size of 50 mm $\times$ 50 mm $\times$ 0.7 mm and ultrasonically washed with isopropyl alcohol and pure water, for 5 min each wash. Then, the washed glass substrate was irradiated with a UV radiation for 30 min and washed by exposing to ozone, and then, installed in a vacuum evaporator.

Compound represented by Formula 3 was vacuum evaporated on the substrate to form a 600 Å thick HIL. Then, 4,4-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB) was vacuum evaporated on the HIL to form a 300 Å thick HTL.

IDE140 (available from Idemitsu Kosan Co., Ltd.), which was known as a blue fluorescent host, and IDE105 (available from Idemitsu Kosan Co., Ltd.), which was known as a blue fluorescent dopant, were co-deposited (weight ratio 98:2) on the HTL to form a 200 Å thick EML.

Alq<sub>3</sub> was deposited on the EML to form a 300 Å thick ETL, and then LiF was deposited on the ETL to form a 10 Å thick EIL and Al was deposited thereon to form a 3000 Å thick anode, thereby completing an organic electroluminescent device.

This device has a driving voltage of 7.1 V, a luminance of 3,214 cd/m<sup>2</sup>, a color coordination (0.14, 0.15), and luminous efficiency of 6.43 cd/A at a current density of 50 mA/cm<sup>2</sup>.

#### COMPARATIVE EXAMPLE 1

An organic EL device was manufactured in the same manner as in Example 1, except that IDE 406 (available from Idemitsu Kosan Co., Ltd.) instead of Compound represented by Formula 3 was used to form a HIL.

This device has a driving voltage of 8.0 V, a luminance of 3,024 cd/m<sup>2</sup>, a color coordination (0.14, 0.15), and luminous efficiency of 6.05 cd/A at a current density of 50 mA/cm<sup>2</sup>.

The driving voltage of the organic EL device which used Compound represented by Formula 3 according to an embodiment of the present invention as a HIL material was approximately 1 V lower than the driving voltage of the organic EL device of Comparative Example 1 at the same current density due to an improved charge injection ability. Further, the organic EL device of Example 1 had higher current efficiency and luminance than the organic EL device of Comparative Example 1.

The luminance and current efficiency at the same current density for the organic EL devices are illustrated in FIGS. 8 and 9.

#### Example 2

A coming 15  $\Omega/\text{cm}^2$  (1200 Å) ITO glass substrate as an anode was cut to a size of 50 mm $\times$ 50 mm $\times$ 0.7 mm and ultrasonically washed with isopropyl alcohol and pure water, for 5 min each wash. Then, the washed glass substrate was

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irradiated with a UV radiation for 30 min and washed by exposing to ozone, and then, installed in a vacuum evaporator.

Compound represented by Formula 3 was vacuum evaporated on the substrate to form a 600 Å thick HIL. Then, 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB) was vacuum evaporated on the HIL to form a 300 Å thick HTL. Alq<sub>3</sub>, which was a green fluorescent host and C545T, which was a green fluorescent dopant, were co-deposited (weight ratio 98:2) on the HTL to form a 250 Å thick EML.

Alq<sub>3</sub> was deposited on the EML to form a 300 Å thick ETL, and then LiF was deposited on the ETL to form a 10 Å thick EIL and Al was deposited thereon to form a 3000 Å thick anode, thereby completing an organic EL device.

This device has a driving voltage of 6.12 V, a luminance of 8,834 cd/m<sup>2</sup>, a color coordination (0.31, 0.63), and luminous efficiency of 17.67 cd/A at a current density of 50 mA/cm<sup>2</sup>.

#### COMPARATIVE EXAMPLE 2

An organic EL device was prepared in the same manner as in Example 2, except for using IDE406 (available from Idemitsu Kosan Co., Ltd.) instead of Compound represented by Formula 3 in forming a hole injection layer.

This device has a driving voltage of 6.73 V, a luminance of 7,083 cd/m<sup>2</sup>, a color coordination (0.31, 0.63), and luminous efficiency of 14.17 cd/A at a current density of 50 mA/cm<sup>2</sup>.

The driving voltage and current efficiency at the same current density for the organic EL devices according to the Example 2 and Comparative Example 2 are illustrated in FIGS. 10 and 11.

#### Example 3

An organic EL device was manufactured in the same manner as in Example 1, except that IDE215 (available from Idemitsu Kosan Co., Ltd.) and IDE118 (available from Idemitsu Kosan Co., Ltd.), which are known as blue fluorescent dopants instead of IDE 140 (available from Idemitsu Kosan Co., Ltd.) and IDE105 (available from Idemitsu Kosan Co., Ltd.), which are the blue fluorescent dopants used in Example 1, were co-deposited on a hole transport layer (weight ratio 98:2) to form an emitting layer (EML) having a thickness of 300 Å.

The device had a driving voltage of 7.18 V, a luminance of 7,253 cd/m<sup>2</sup>, a color coordination (0.144, 0.244), and luminous efficiency of 7.25 cd/A at a current density of 100 mA/cm<sup>2</sup>.

#### Example 4

An organic EL device was manufactured in the same manner as in Example 3, except that Compound 5 instead of Compound 3 was used to form a HIL.

The device has a driving voltage of 7.07 V, a luminance of 7,715 cd/m<sup>2</sup>, a color coordination (0.144, 0.235), and luminous efficiency of 7.72 cd/A at a current density of 100 mA/cm<sup>2</sup>.

#### Example 5

An organic EL device was manufactured in the same manner as in Example 3, except that Compound 7 instead of Compound 3 was used to form a HIL.

This device had a driving voltage of 6.72 V, a luminance of 7,104 cd/m<sup>2</sup>, a color coordination (0.143, 0.232), and luminous efficiency of 7.10 cd/A at a current density of 100 mA/cm<sup>2</sup>.



An organic EL device was manufactured in the same manner as in Example 3, except that Compound 27 instead of Compound 3 was used to form a HIL.

This device had a driving voltage of 6.87 V, a luminance of 7,931 cd/m<sup>2</sup>, a color coordination (0.145, 0.246), and luminous efficiency of 7.93 cd/A at a current density of 100 mA/cm<sup>2</sup>.

## COMPARATIVE EXAMPLE 3

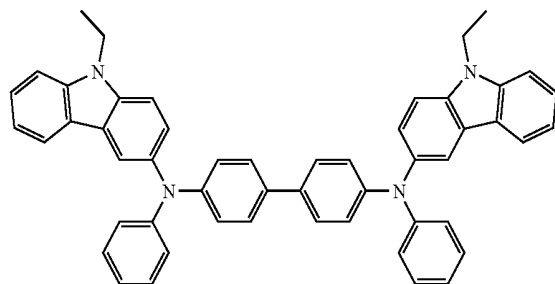
An organic EL device was manufactured in the same manner as Example 3, except that IDE406 (available from Idemitsu) instead of Compound 3 was used to form a HIL.

This device had a driving voltage of 8.38 V, a luminance of 6,528 cd/m<sup>2</sup>, a color coordination (0.143, 0.243), and luminous efficiency of 6.53 cd/A at a current density of 100 mA/cm<sup>2</sup>.

## COMPARATIVE EXAMPLE 4

An organic EL device was manufactured in the same manner as Example 3, except that N,N'-bis(9-ethylcarbazol-3-yl)-

N,N'-diphenylbenzidine (TLB1) disclosed in International Publication No. WO03/008515 instead of Compound 3 was used to form a HIL.



TLB1

This device had a driving voltage of 7.25 V, a luminance of 7,104 cd/m<sup>2</sup>, a color coordination (0.143, 0.235), and luminous efficiency of 7.10 cd/A at a current density of 100 mA/cm<sup>2</sup>.

An organic EL device was manufactured in the same manner as in Example 3, except that Compound 28 of Synthesis Reference Example 1, which is a derivative of TLB1, instead of Compound 3 was used to form a HIL.

This device had a driving voltage of 7.25 V, a luminance of 7,342 cd/m<sup>2</sup>, a color coordination (0.145, 0.239), and luminous efficiency of 7.34 cd/A at a current density of 100 mA/cm<sup>2</sup>.

## COMPARATIVE EXAMPLE 6

An organic EL device was manufactured in the same manner as in Example 3, except that Compound 29 of Synthesis Reference Example 2, which is a derivative of TLB1, instead of Compound 3 was used to form a HIL.

This device had a driving voltage of 7.18 V, a luminance of 7,681 cd/m<sup>2</sup>, a color coordination (0.144, 0.245), and luminous efficiency of 7.68 cd/A at a current density of 100 mA/cm<sup>2</sup>.

Characteristics of driving voltage, luminance, color coordination, and luminous efficiency of organic EL devices of Examples 3-6 according to the embodiments of the present invention and Comparative Examples 3-6 are shown in Table 1.

TABLE 1

	Hole injection material	Driving voltage (V) (@100 A/cm <sup>2</sup> )	Luminance (cd/m <sup>2</sup> )	Color coordination (x, y)	Luminous efficiency (cd/A)
Example 3	Compound 3	7.18	7,253	(0.144, 0.244)	7.25
Example 4	Compound 5	7.07	7,715	(0.144, 0.235)	7.72
Example 5	Compound 7	6.72	7,104	(0.143, 0.232)	7.10
Example 6	Compound 27	6.87	7,931	(0.145, 0.246)	7.93
Comparative Example 3	IDE 406	8.38	6,528	(0.143, 0.243)	6.53
Comparative Example 4	TLB1	7.25	7,104	(0.143, 0.235)	7.10
Comparative Example 5	Compound 28	7.25	7,342	(0.145, 0.239)	7.34
Comparative Example 6	Compound 29	7.18	7,681	(0.144, 0.245)	7.68

For reference, in Table 1, characteristics of driving voltage, luminance, color coordination and luminous efficiency of Compounds 3, 5, 7 and 27 according to the embodiments of the present invention are respectively and directly compared to those of TLB1 and Compounds 28 and 29.

According to Table 1, when Compounds 3, 5, 7 and 27 were used as a material for a hole injection layer, charge injection capability was improved, and thus driving voltages were respectively reduced to 1.13 V, 1.31 V, 1.66 V, and 1.51 V at the same current level, current efficiency was improved, and luminance was improved compared to when using IDE406.

FIG. 12 shows luminance according to driving voltages in the organic EL devices of Examples 3-6 according to the embodiments of the present invention and Comparative Examples 3 and 4.

Referring to FIG. 12, the organic EL devices of Examples 3-6 had improved luminance compared to the organic EL devices of Comparative Example 3, and had similar luminance to that of Comparative Example 4.

FIG. 13 shows lifetime characteristics of the organic EL devices of Examples 3, 4, 5 and 6 according to the embodiments of the present invention and the organic EL devices of Comparative Examples 3 through 6.

Referring to FIG. 13, lifetimes of the organic EL devices of Examples 3-6 and Comparative Examples 3-6 were measured

at a current density of 100 mA/cm<sup>2</sup> which is a condition for an accelerated lifetime test. The lifetime was evaluated by measuring time taken for the display/pixel to fall to half its initial stated luminance. The organic EL devices of Examples 3, 4, 5 and 6 had extended lifetime compared to those of Comparative Example 3 and 4. The lifetime characteristics of the organic EL devices of Examples 3 through 6 according to the embodiments of the present invention were respectively improved by 20%, 33%, 42% and 56% compared to those of the organic EL devices of Comparative Example 3 which uses a known hole injection material of IDE406 (available from Idemitsu Kosan Co., Ltd.). It is well known to those of ordinary skill in the art that the accelerated lifetime is directly in proportion to the real lifetime of the organic EL devices.

Particularly, initial characteristics of the organic EL devices in Comparative Examples 4 and 5 which use Compounds 28 and 29 were similar to initial characteristics of the organic EL devices according to the embodiments of the present invention, but lifetime thereof was considerably different from lifetime of the organic EL devices according to the embodiments of the present invention.

Lifetimes of the organic EL devices of Examples 3-6 according to the present invention were longer than that of Comparative Example 4 using TLB1 by 244%, 269%, 287%, and 315%, respectively. Also, as a result of respectively comparing Compound 5 with Compound 28 and Compound 27 with Compound 29 in which substituents at the N site in carbazole were varied, the lifetimes of the organic EL devices of Compound 5 (393 hours) and Compound 27 (418 hours) according to the embodiments of the present invention were longer than those of Compound 28 (106 hours) and Compound 29 (175 hours) by 371% and 239%, respectively.

In addition, Comparative Examples 4 through 6 clearly show a decrease in lifetime characteristics compared to a known hole injection material of IDE406 (available from Idemitsu Kosan Co., Ltd.) in FIG. 13. When the lifetime of a device is less than 300 hours, the device is hardly commercialized. In particular, an image sticking phenomenon may occur due to considerable reduction in the initial luminance in Comparative Examples 4 through 6 compared to Examples 3 through 6. "Image sticking" is a phenomenon in which the same image is continuously displayed and causes pixels or sub-pixels to continuously output undesired signals. In particular, image sticking is closely associated with lifetime in self-emission systems.

These results are caused by the substituents connected to the N site of carbazole. TLB1 has a very unstable structure in which a radical generated at the N site during transporting holes is easily disappeared since an ethyl group which is unstable in oxidation is substituted at the N site of carbazole. In contrast, the compounds according to the present invention in which a substituted or unsubstituted phenyl group is substituted at the N site of carbazole has a fully conjugated structure of the entire phenylcarbazole, and thus the compounds are protected against oxidation through external attacks and have a stable structure of radicals generated at the N site during the transportation of holes.

These were measured using cyclic voltammetry (CV) which is an electrochemical method for obtaining oxidation and reduction properties and stability of a compound. Using CV, electrochemical movement of oxidation and reduction species can be rapidly observed in a wide range of electric potential. CV is a widely used electrochemical method of determining the mechanism of oxidation and reduction reactions. CV of Compound 3, TLB 1, Compounds 5 and 28 was measured using a PAR 273A Potentiostat (AMETEK EG&G).

A sample having a concentration of  $1 \times 10^{-3}$  M was measured at a scan speed of 100 mV/sec using Ag<sup>+</sup>/Ag as a reference electrode, Pt (BioAnalytical System, USA) as a working electrode and auxiliary electrode, and dichloromethane (HPLC grade, Mallinkrodt) solution including 0.1 M tetrabutylammonium hexafluorophosphate (99%, Fluka) as an electrolyte solution.

As shown in FIG. 14, Compound 3 according to the embodiment of the present invention showed a stable state even after several oxidation-reduction cycles. In contrast, TLB1 showed a rapid collapse of oxidized state (FIG. 15). Further, as shown in FIG. 16, Compound 3 according to the embodiment of the present invention also showed a stable oxidation-reduction curve. In contrast, Compound 28 illustrated in FIG. 17 showed a very unstable state with a collapse of peaks due to repeated oxidation-reduction cycles.

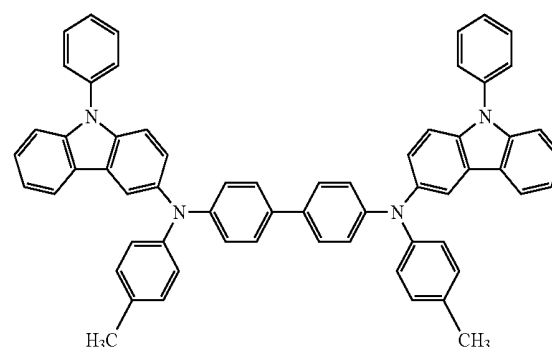
One of the distinguishable properties of hole transport materials of the embodiments of the present invention is stability against external thermal stimulus. While a glass transition temperature (T<sub>g</sub>) of TLB1 was 136° C., T<sub>g</sub> of the phenylcarbazole-based compounds according to the embodiments of the present invention was higher than 150° C. Considerable improvement in lifetime characteristics can be obtained because of differences in such structural and thermal stability.

As described above, the compounds according to the present invention are excellent hole injection materials having low driving voltage, high efficiency, and long lifetime compared to the compounds according to the cited reference. As described above, the phenylcarbazole-based compounds according to an embodiment of the present invention have superior electric properties and charge transport abilities, and thus are useful as a hole injection material, a hole transport material, and/or an emitting material which is suitable for fluorescent and phosphorescent devices of all colors, including red, green, blue, and white colors. The organic EL device manufactured using the phenylcarbazole-based compound has high efficiency, low voltage, high luminance, and a long lifespan.

While the present invention has been particularly shown and described with reference to exemplary embodiments thereof, it will be understood by those of ordinary skill in the art that various changes in form and details may be made therein without departing from the spirit and scope of the present invention as defined by the following claims.

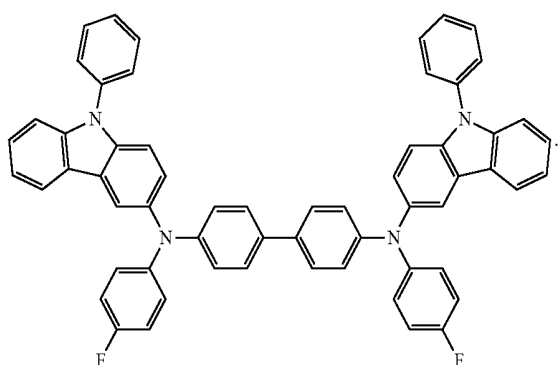
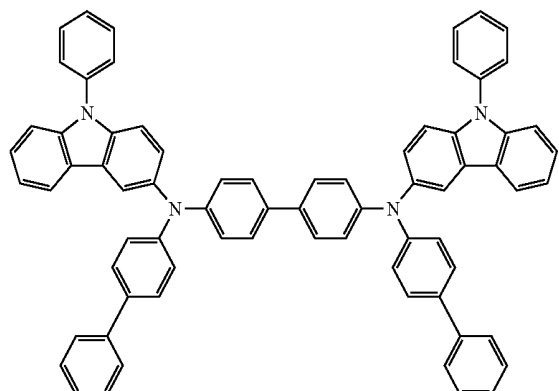
What is claimed is:

1. A compound represented by one of formulae (5), (7) and (27):



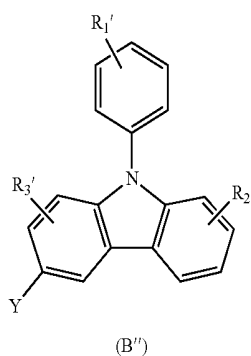
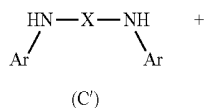
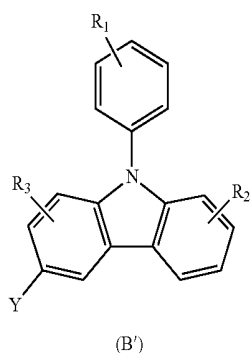
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2. A method of preparing a compound represented by Formula (1), comprising:

reacting carbazole (B' and B'') with a diamine compound (C'):



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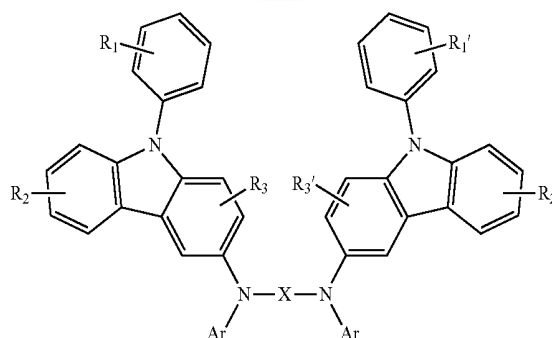
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(1)

where X is a biphenyl group;

each of  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_1'$ ,  $R_2'$  and  $R_3'$  is independently selected from the group consisting of a hydrogen atom, a substituted or unsubstituted C1-C30 alkyl group, a substituted or unsubstituted C1-C30 alkoxy group, a substituted or unsubstituted C6-C30 aryl group, a substituted or unsubstituted C6-C30 aryloxy group, a substituted or unsubstituted C2-C30 heterocyclic group, a substituted or unsubstituted C6-C30 condensed polycyclic group, a hydroxy group, a cyano group, and a substituted or unsubstituted amino group, and, alternatively, two or more adjacent groups among  $R_1$ ,  $R_2$ ,  $R_3$ ,  $R_1'$ ,  $R_2'$  and  $R_3'$  can be connected to each other to form a saturated or unsaturated carbocycle;

each Ar is independently a substituted or unsubstituted C6-C30 aryl group or a substituted or unsubstituted C2-C30 heteroaryl group; and

Y is a halogen atom.

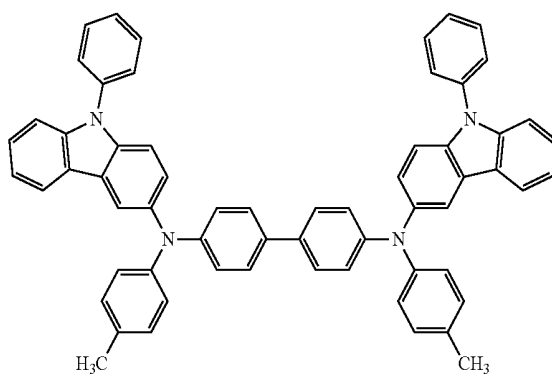
3. The method of claim 2, wherein the reaction is carried out in the presence of  $\text{Pd}_2(\text{dba})_3$  where dba is dibenzylideneacetone, sodium tert-butoxide, and tri(tert-butyl)phosphine at a temperature of 50 to 150° C.

4. An organic electroluminescent display device, comprising:

a first electrode;

a second electrode; and

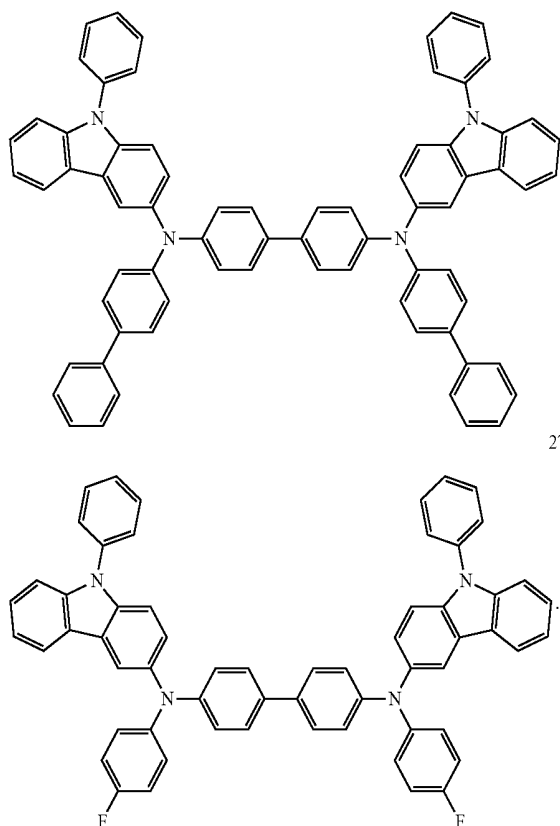
an organic layer interposed between the first electrode and the second electrode, the organic layer comprising a compound represented by Formulae 5, 7 and 27:



(5)

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5. The organic electroluminescent device of claim 4, wherein the organic layer is a hole injection layer or a hole transport layer.

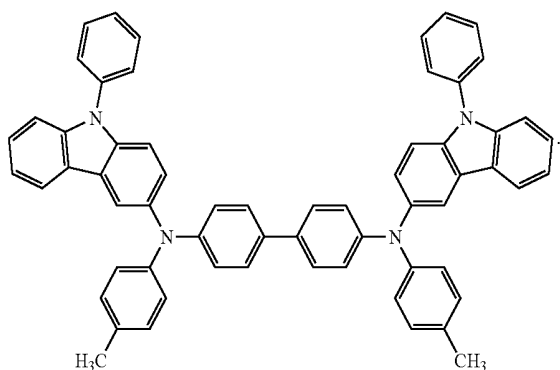
6. The organic electroluminescent device of claim 4, wherein the organic layer is a single layer serving as both a hole injection layer and a hole transport layer.

7. The organic electroluminescent device of claim 4, wherein the organic layer is an emitting layer.

8. The organic electroluminescent device of claim 7, wherein the emitting layer is composed of a phosphorescent or fluorescent material.

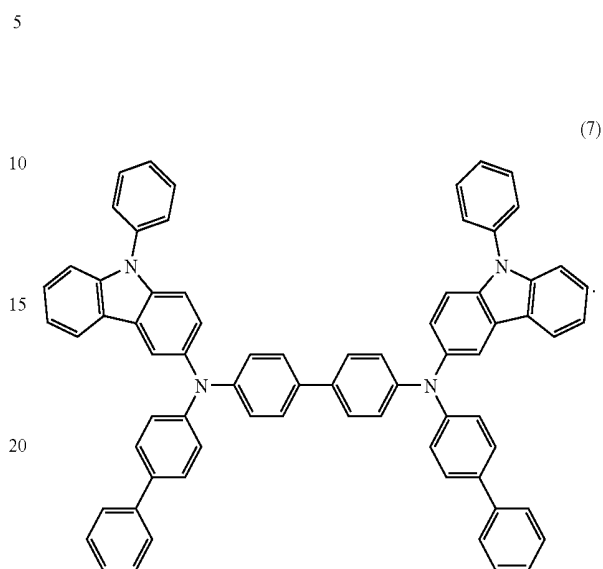
9. The organic electroluminescent device of claim 7, wherein the compound is used as a fluorescent or phosphorescent host in the emitting layer.

10. The organic electroluminescent device of claim 4, wherein the compound is a compound represented by Formula (5):

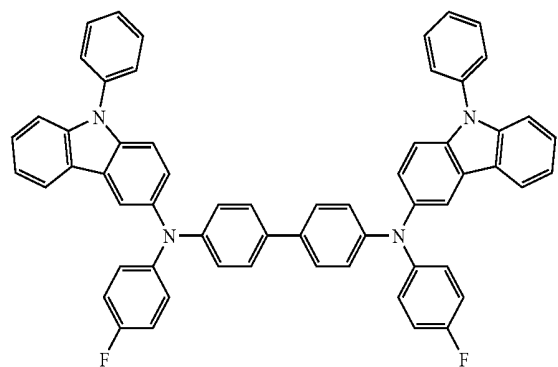


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11. The organic electroluminescent device of claim 4, wherein the compound is a compound represented by Formula (7):



12. The organic electroluminescent device of claim 4, wherein the compound is a compound represented by Formula (27):



13. The compound of claim 1, wherein the compound is represented by formulae (5).

14. The compound of claim 1, wherein the compound is represented by formulae (7).

15. The compound of claim 1, wherein the compound is represented by formulae (27).

专利名称(译)	基于苯基咪唑的化合物和使用其的有机电致发光器件		
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[标]申请(专利权)人(译)	HWANG SEOK HWAN 金荣KOOK 李昌镐 LEE JONG SEOK 杨SEUNG珏 金喜善YEON		
申请(专利权)人(译)	HWANG SEOK-HWAN 金荣KOOK 李昌镐 李硕JONG 杨升GAK 金喜善妍		
当前申请(专利权)人(译)	三星移动显示器有限公司.		
[标]发明人	HWANG SEOK HWAN KIM YOUNG KOOK LEE CHANG HO LEE SEOK JONG YANG SEUNG GAK KIM HEE YEON		
发明人	HWANG, SEOK-HWAN KIM, YOUNG-KOOK LEE, CHANG-HO LEE, SEOK-JONG YANG, SEUNG-GAK KIM, HEE-YEON		
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其他公开文献	US20080107919A1		
外部链接	<a href="#">Espacenet</a> <a href="#">USPTO</a>		

#### 摘要(译)

苯基咪唑类化合物由式1表示,并且具有优异的电性能和电荷传输能力,因此可用作空穴注入材料,空穴传输材料和/或适用于荧光和磷光装置的发光材料。所有颜色,包括红色,绿色,蓝色和白色。苯基咪唑类化合物通过咪唑与二胺反应合成。使用苯基咪唑类化合物制造的有机电致发光器件具有高效率,低电压,高亮度和长寿命。

CATHODE
EIL
ETL
EML
HTL
HIL
ANODE
SUBSTRATE