

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
5 June 2008 (05.06.2008)

PCT

(10) International Publication Number  
**WO 2008/066196 A1**

(51) International Patent Classification:  
*C09K 11/06* (2006.01) *H01L 51/50* (2006.01)

(74) Agents: TAKAMATSU, Takeshi et al.; Koh-Ei Patent Firm, Kawabe Bldg., 7-9, Shimbashi 3-chome, Minato-ku, Tokyo 105-0004 (JP).

(21) International Application Number:  
PCT/JP2007/073279

(81) Designated States (*unless otherwise indicated, for every kind of national protection available*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BH, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IS, KE, KG, KM, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LT, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RS, RU, SC, SD, SE, SG, SK, SL, SM, SV, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(22) International Filing Date:  
27 November 2007 (27.11.2007)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:  
2006-318772 27 November 2006 (27.11.2006) JP  
2007-221472 28 August 2007 (28.08.2007) JP

(84) Designated States (*unless otherwise indicated, for every kind of regional protection available*): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IS, IT, LT, LU, LV, MC, MT, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

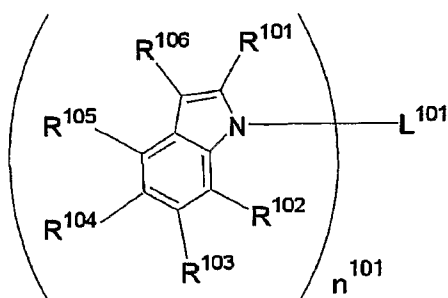
(71) Applicant (*for all designated States except US*): FUJIFILM Corporation [JP/JP]; 26-30, Nishiazabu 2-chome, Minato-ku, Tokyo, 1060031 (JP).

(72) Inventors; and

(75) Inventors/Applicants (*for US only*): IGARASHI, Tatsuya. YAGI, Kazunari.

Published:  
— with international search report

(54) Title: ORGANIC ELECTROLUMINESCENT DEVICE AND INDOLE DERIVATIVE



(1)

(57) Abstract: An organic electroluminescent device is provided and includes: a pair of electrodes; and at least one organic layer between the pair of electrodes, the at least one organic layer including a light-emitting layer containing a light-emitting material. The at least one organic layer includes at least one layer containing an indole derivative represented by formula (1). Formula (1):  $R^{101}$ ,  $R^{102}$ ,  $R^{103}$ ,  $R^{104}$  and  $R^{105}$  each independently represents a hydrogen atom or a substituent;  $R^{106}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{101}$  and  $R^{106}$  may be bonded to each other to form a ring;  $L^{101}$  represents a

linking group; and  $n^{101}$  represents an integer of 2 or higher.

## DESCRIPTION

## ORGANIC ELECTROLUMINESCENT DEVICE AND INDOLE DERIVATIVE

## 5 Technical Field

The present invention relates to a light-emitting device capable of emitting light by converting electric energy into light, in particular, the invention relates to an organic electroluminescent device (a light-emitting device, or an EL device). The invention further relates to an indole derivative useful to a light-emitting device.

10

## Background Art

Organic electroluminescent (EL) devices are attracting public attention as promising display devices for capable of emitting light of high luminance with low voltage. An important characteristic of organic electroluminescent devices is consumed electric power. Consumed electric power is represented by: “Consumed electric power = Voltage × electric current”, so that the lower the value of voltage that is necessary to obtain desired brightness and the smaller the value of electric current, the lower is the consumed electric power of the device.

As one trial to lower the value of electric current flowing in a device, a light-emitting device utilizing luminescence from ortho-metalated iridium complex (Ir(ppy)<sub>3</sub>: Tris-Ortho-Metalated Complex of Iridium(III) with 2-Phenylpyridine) is reported (e.g., refer to JP-A-2001-247859). The phosphorescent devices described therein are greatly improved in external quantum efficiency as compared with existing singlet luminescent devices, and have succeeded in making the value of electric current smaller. However, they cannot be said to have sufficient performances with respect to durability and efficiency, and color tone worsens with the deterioration of the device, so that further improvement is required.

For the purpose of improving the efficiency and durability of a phosphorescent device, a device containing an indole derivative (JP-A-2002-305084) is reported. However,

in view of durability and efficiency, a further improvement is required.

#### Disclosure of the Invention

An object of the invention is to provide a light-emitting device showing good  
 5 durability and efficiency, and little in variation of chromaticity by aging. A further object is to provide a novel indole derivative.

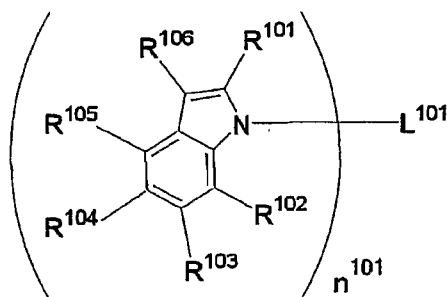
The above objects can be achieved by the following means.

(1) An organic electroluminescent device comprising:

a pair of electrodes; and

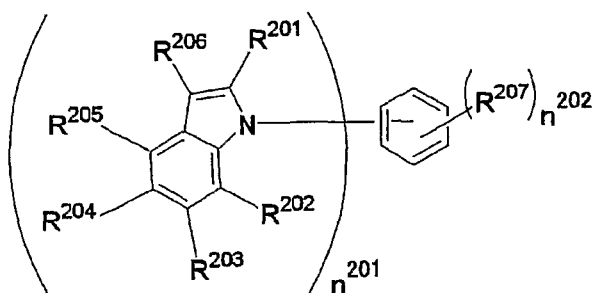
10 at least one organic layer between the pair of electrodes, the at least one organic layer including a light-emitting layer containing a light-emitting material,

wherein the at least one organic layer includes at least one layer containing an indole derivative represented by formula (1):



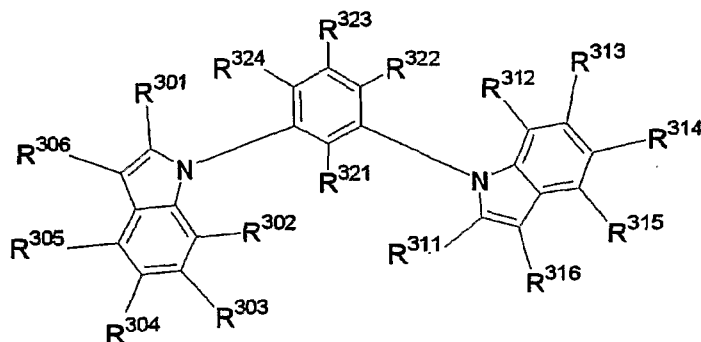
15 wherein R<sup>101</sup>, R<sup>102</sup>, R<sup>103</sup>, R<sup>104</sup> and R<sup>105</sup> each independently represents a hydrogen atom or a substituent; R<sup>106</sup> represents an alkyl group having a tertiary or quaternary carbon atom; R<sup>101</sup> and R<sup>106</sup> may be bonded to each other to form a ring; L<sup>101</sup> represents a linking group; and n<sup>101</sup> represents an integer of 2 or higher.

(2) The organic electroluminescent device described in (1), wherein the indole  
 20 derivative is a compound represented by formula (2):



wherein  $R^{201}$ ,  $R^{202}$ ,  $R^{203}$ ,  $R^{204}$  and  $R^{205}$  each independently represents a hydrogen atom or a substituent;  $R^{206}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{201}$  and  $R^{206}$  may be bonded to each other to form a ring;  $R^{207}$  represents a substituent;  $n^{201}$  represents an integer of from 2 to 6; and  $n^{202}$  represents an integer of from 0 to 4, provided that  $n^{201} + n^{202} \leq 6$ .

(3) The organic electroluminescent device described in (1), wherein the indole derivative is a compound represented by the following formula (3):



wherein  $R^{301}$ ,  $R^{302}$ ,  $R^{303}$ ,  $R^{304}$  and  $R^{305}$  each independently represents a hydrogen atom or a substituent;  $R^{306}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{301}$  and  $R^{306}$  may be bonded to each other to form a ring;  $R^{311}$ ,  $R^{312}$ ,  $R^{313}$ ,  $R^{314}$  and  $R^{315}$  each independently represents a hydrogen atom or a substituent;  $R^{316}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{311}$  and  $R^{316}$  may be bonded to each other to form a ring; and  $R^{321}$ ,  $R^{322}$ ,  $R^{323}$  and  $R^{324}$  each independently represents a hydrogen atom or a substituent.

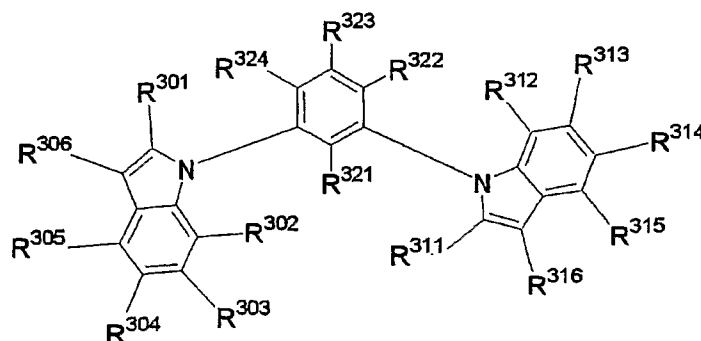
(4) The organic electroluminescent described in any one of (1) to (3), wherein the indole derivative is contained in the light-emitting layer.

(5) The organic electroluminescent device described in any one of (1) to (4), wherein the indole derivative is contained in a layer contiguous to the light-emitting layer.

(6) The organic electroluminescent device described in any one of (1) to (5), wherein the light-emitting material is a phosphorescent material.

5 (7) The organic electroluminescent device described in any one of (1) to (6), wherein the light-emitting material is a platinum complex.

(8) A compound represented by formula (3):



10 wherein  $R^{301}$ ,  $R^{302}$ ,  $R^{303}$ ,  $R^{304}$  and  $R^{305}$  each independently represents a hydrogen atom or a substituent;  $R^{306}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{301}$  and  $R^{306}$  may be bonded to each other to form a ring;  $R^{311}$ ,  $R^{312}$ ,  $R^{313}$ ,  $R^{314}$  and  $R^{315}$  each independently represents a hydrogen atom or a substituent;  $R^{316}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{311}$  and  $R^{316}$  may be bonded to each other to form a ring; and  $R^{321}$ ,  $R^{322}$ ,  $R^{323}$  and  $R^{324}$  each independently represents a hydrogen atom or a substituent.

15

### Best Mode for Carrying Out the Invention

A light-emitting device according to an aspect of the invention is capable of light emission of high efficiency, excellent in durability, and little in hue variation by aging.

20 Further, a novel indole derivative according to an aspect of the invention is useful as a material of the light-emitting device.

An aspect of the invention relates to an organic electroluminescent device including:

a pair of electrodes; and at least one organic layer including a light-emitting layer, between the pair of electrodes. The at least one organic layer includes at least one layer containing an indole derivative represented by formula (1).

The compound represented by formula (1) will be explained below.

5         $R^{101}$  to  $R^{105}$  each independently represents a hydrogen atom or a substituent.

The examples of the substituents include an alkyl group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 10 carbon atoms, e.g., methyl, ethyl, isopropyl, n-octyl, n-decyl, n-hexadecyl, cyclopropyl, cyclopentyl, cyclohexyl, etc., are exemplified), an alkenyl group (preferably  
10        having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, and especially preferably from 2 to 10 carbon atoms, e.g., vinyl, allyl, 2-butenyl, 3-pentenyl, etc., are exemplified), an alkynyl group (preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, and especially preferably from 2 to 10 carbon atoms, e.g., propargyl, 3-pentynyl, etc., are exemplified), an aryl group (preferably having from 6 to  
15        30 carbon atoms, more preferably from 6 to 20 carbon atoms, and especially preferably from 6 to 12 carbon atoms, e.g., phenyl, p-methylphenyl, naphthyl, anthranyl, etc., are exemplified), an amino group (preferably having from 0 to 30 carbon atoms, more preferably from 0 to 20 carbon atoms, and especially preferably from 0 to 10 carbon atoms, e.g., amino, methylamino, dimethylamino, diethylamino, dibenzylamino, diphenylamino, ditolylamino,  
20        etc., are exemplified), an alkoxyl group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 10 carbon atoms, e.g., methoxy, ethoxy, butoxy, 2-ethylhexyloxy, etc., are exemplified), an aryloxy group (preferably having from 6 to 30 carbon atoms, more preferably from 6 to 20 carbon atoms, and especially preferably from 6 to 12 carbon atoms, e.g., phenyloxy, 1-naphthyloxy,  
25        2-naphthyloxy, etc., are exemplified), a heterocyclic oxy group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., pyridyloxy, pyrazyloxy, pyrimidyloxy, quinolyloxy, etc., are exemplified), an acyl group (preferably having from 1 to 30 carbon atoms, more preferably

from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., acetyl, benzoyl, formyl, pivaloyl, etc., are exemplified), an alkoxycarbonyl group (preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, and especially preferably from 2 to 12 carbon atoms, e.g., methoxycarbonyl, ethoxycarbonyl, etc., are exemplified), an aryloxy carbonyl group (preferably having from 7 to 30 carbon atoms, more preferably from 7 to 20 carbon atoms, and especially preferably from 7 to 12 carbon atoms, e.g., phenyloxycarbonyl, etc., are exemplified), an acyloxy group (preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, and especially preferably from 2 to 10 carbon atoms, e.g., acetoxyl, benzoyloxy, etc., are exemplified), an acylamino group (preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, and especially preferably from 2 to 10 carbon atoms, e.g., acetylamino, benzoylamino, etc., are exemplified), an alkoxycarbonylamino group (preferably having from 2 to 30 carbon atoms, more preferably from 2 to 20 carbon atoms, and especially preferably from 2 to 12 carbon atoms, e.g., methoxycarbonylamino, etc., are exemplified), an aryloxy carbonylamino group (preferably having from 7 to 30 carbon atoms, more preferably from 7 to 20 carbon atoms, and especially preferably from 7 to 12 carbon atoms, e.g., phenyloxycarbonylamino, etc., are exemplified), a sulfonylamino group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., methanesulfonylamino, benzenesulfonylamino, etc., are exemplified), a sulfamoyl group (preferably having from 0 to 30 carbon atoms, more preferably from 0 to 20 carbon atoms, and especially preferably from 0 to 12 carbon atoms, e.g., sulfamoyl, methylsulfamoyl, dimethylsulfamoyl, phenylsulfamoyl, etc., are exemplified), a carbamoyl group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., carbamoyl, methylcarbamoyl, diethylcarbamoyl, phenylcarbamoyl, etc., are exemplified), an alkylthio group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., methylthio, ethylthio, etc., are exemplified), an arylthio group (preferably having from 6 to 30 carbon atoms, more

preferably from 6 to 20 carbon atoms, and especially preferably from 6 to 12 carbon atoms, e.g., phenylthio, etc., are exemplified), a heterocyclic thio group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., pyridylthio, 2-benzimidazolylthio, 2-benzoxazolylthio, 2-benzothiazolylthio, etc., are exemplified), a sulfonyl group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., mesyl, tosyl, etc., are exemplified), a sulfinyl group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., methanesulfinyl, benzenesulfinyl, etc., are exemplified), a ureido group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., ureido, methylureido, phenylureido, etc., are exemplified), a phosphoric amido group (preferably having from 1 to 30 carbon atoms, more preferably from 1 to 20 carbon atoms, and especially preferably from 1 to 12 carbon atoms, e.g., diethylphosphoric amido, phenylphosphoric amido, etc., are exemplified), a hydroxyl group, a mercapto group, a halogen atom (e.g., a fluorine atom, a chlorine atom, a bromine atom, an iodine atom), a cyano group, a sulfo group, a carboxyl group, a nitro group, a hydroxamic acid group, a sulfino group, a hydrazino group, an imino group, a heterocyclic group (preferably having from 1 to 30 carbon atoms, and more preferably from 1 to 20 carbon atoms, and as the hetero atoms, e.g., a nitrogen atom, an oxygen atom, a sulfur atom are exemplified, specifically, e.g., imidazolyl, pyridyl, quinolyl, furyl, thienyl, piperidyl, morpholino, benzoxazolyl, benzimidazolyl, benzothiazolyl, carbazolyl, azepinyl, etc., are exemplified), a silyl group (preferably having from 3 to 40 carbon atoms, more preferably from 3 to 30 carbon atoms, and especially preferably from 3 to 24 carbon atoms, e.g., trimethylsilyl, triphenylsilyl, etc., are exemplified), and a silyloxy group (preferably having from 3 to 40 carbon atoms, more preferably from 3 to 30 carbon atoms, and especially preferably from 3 to 24 carbon atoms, e.g., trimethylsilyloxy, triphenylsilyloxy, etc., are exemplified). These substituents may further be substituted.

$R^{101}$  to  $R^{105}$  each preferably represents a hydrogen atom, an alkyl group, an aryl

group, or a hetero aryl group, more preferably a hydrogen atom, an alkyl group, or an aryl group, still more preferably a hydrogen atom or an alkyl group, and especially preferably a hydrogen atom.

$R^{106}$  represents an alkyl group having a tertiary or quaternary carbon atom. The  
5 tertiary or quaternary carbon atom in  $R^{106}$  is preferably directly bonded to the indole ring  
(e.g., a t-butyl group, an isopropyl group, an isobutyl group, an isopentyl group, a  
cyclopropyl group, a cyclopentyl group, a cyclohexyl group, a 2-phenyl-2-propyl group, a  
2-(2-pyridyl)propyl group, etc.).  $R^{106}$  preferably represents an alkyl group having a  
quaternary carbon atom, more preferably an alkyl group in which a quaternary carbon atom is  
10 bonded to the indole ring, still more preferably an alkyl group having from 4 to 10 carbon  
atoms in which a quaternary carbon atom is bonded to the indole ring, and especially  
preferably an alkyl group having from 4 to 6 carbon atoms in which a quaternary carbon  
atom is bonded to the indole ring (preferably a t-butyl group). Further,  $R^{101}$  and  $R^{106}$  may  
be bonded to each other to form a ring. As the ring formed by  $R^{101}$  and  $R^{106}$  to be bonded to  
15 each other, e.g., a cyclopentene ring, a cyclohexene ring, a 1,4-cyclohexadiene ring, a  
cycloheptene ring, a cyclooctene ring, etc., are exemplified.

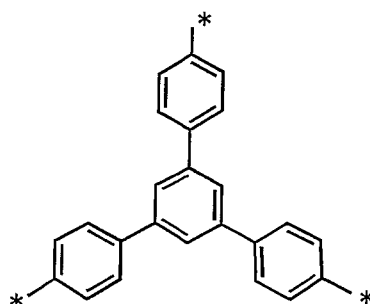
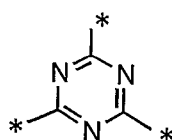
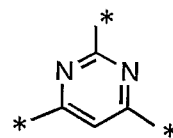
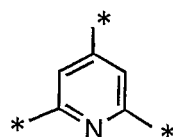
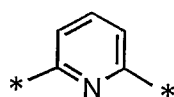
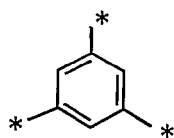
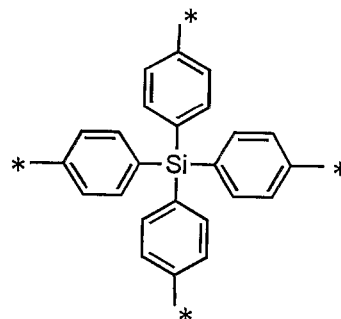
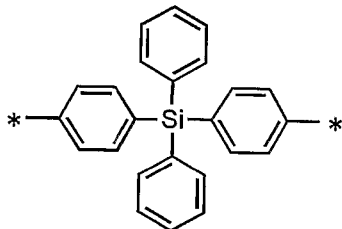
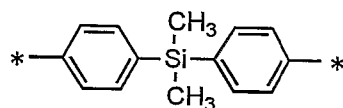
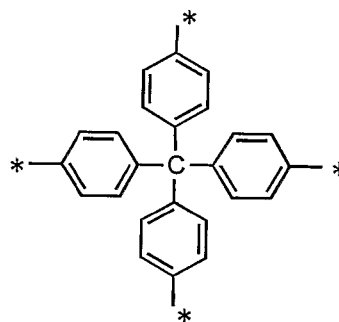
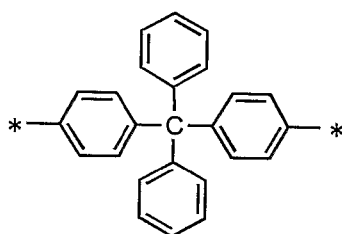
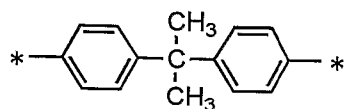
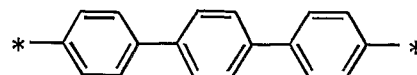
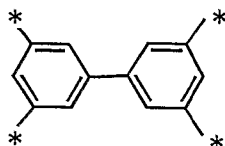
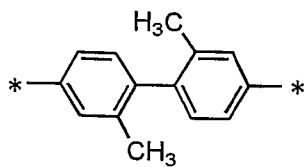
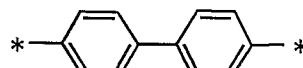
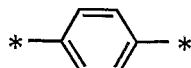
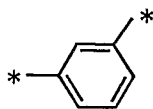
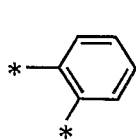
$R^{106}$  may have a substituent. As the substituents, those exemplified as the  
substituents represented by  $R^{101}$  to  $R^{105}$  are applicable.

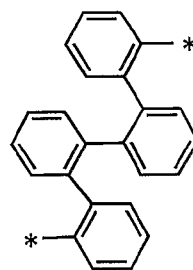
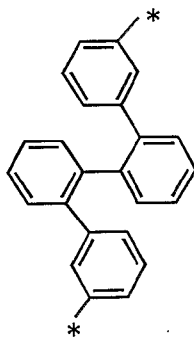
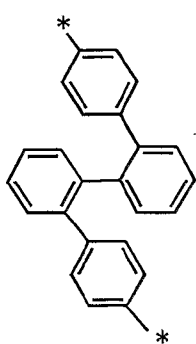
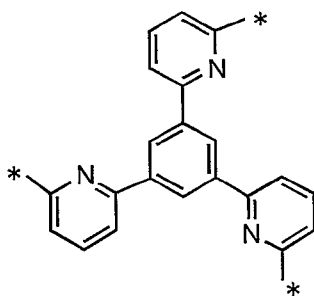
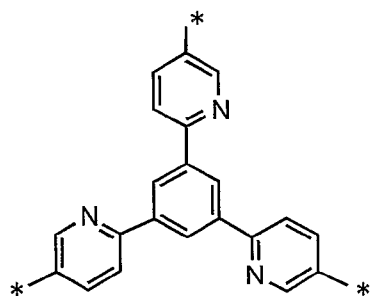
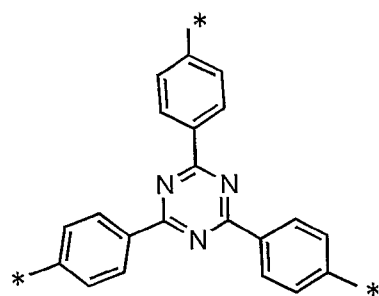
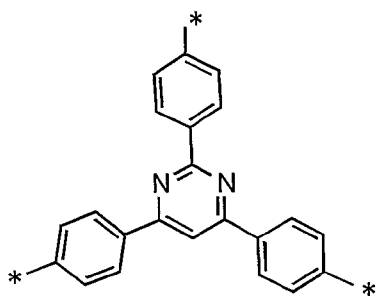
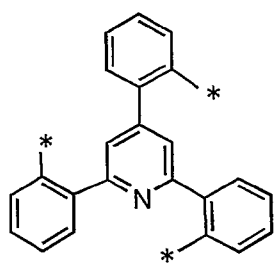
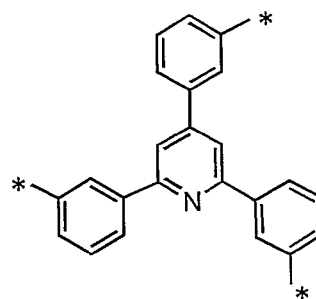
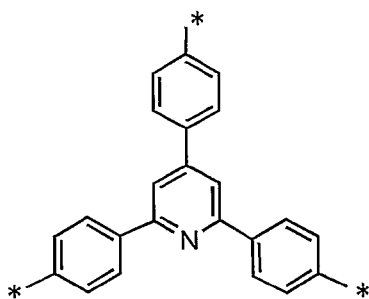
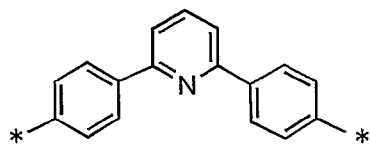
$L^{101}$  represents a linking group. As the linking groups, linking groups containing  
20 an atom selected from a carbon atom, a nitrogen atom, an oxygen atom, a sulfur atom, a  
silicon atom, and a phosphorus atom are preferred, an alkyl linking group (a linking group  
mainly including an alkyl group), an aryl linking group (a linking group mainly including  
aryl group), and a hetero aryl linking group (a linking group mainly including an hetero aryl  
group) are more preferred, an aryl linking group and a hetero aryl linking group are still more  
25 preferred, and an aryl linking group is especially preferred.

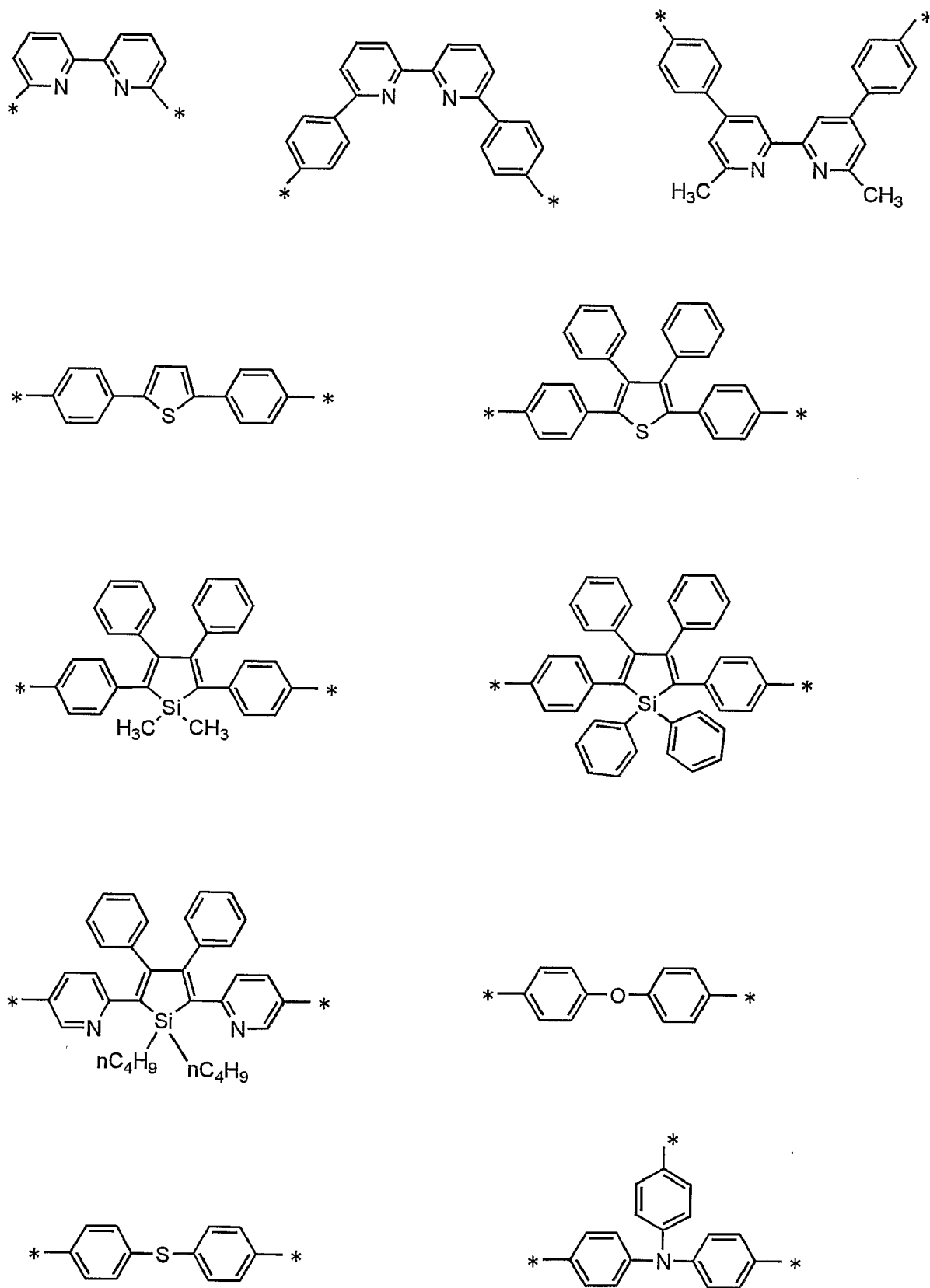
The linking group represented by  $L^{101}$  may have a substituent. As the substituents,  
those exemplified as the substituents represented by  $R^{101}$  to  $R^{105}$  are applicable.

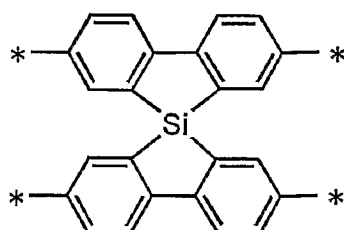
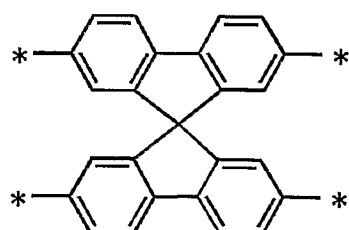
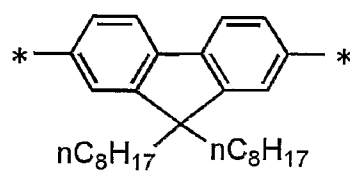
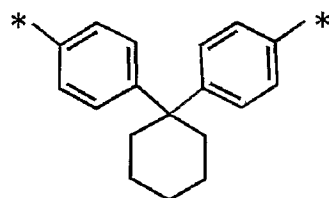
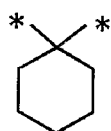
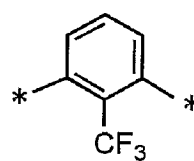
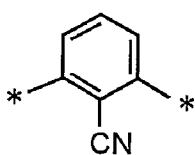
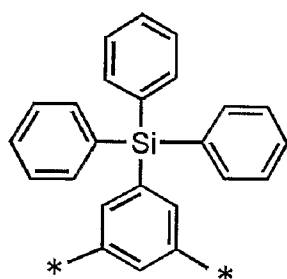
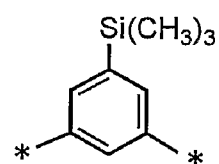
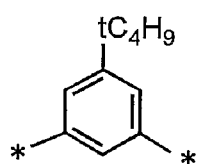
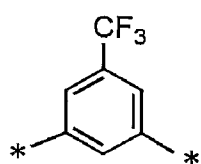
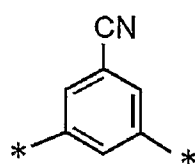
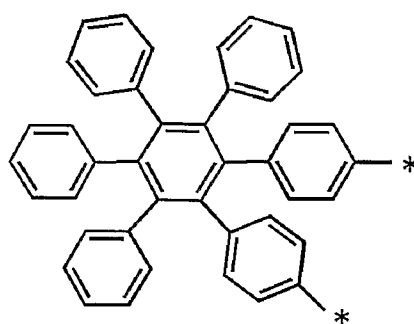
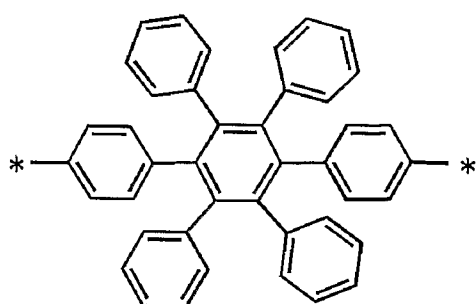
As the specific examples of the linking groups represented by  $L^{101}$ , for example, the

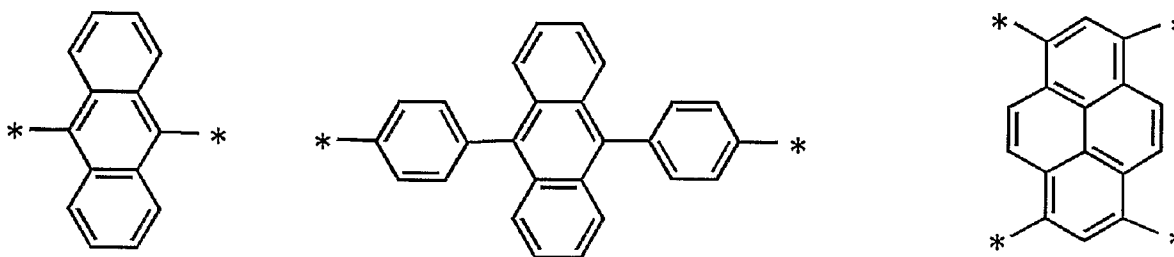
following compounds are exemplified. In the examples below, \* indicates a portion to which the indole ring binds.











$n^{101}$  represents an integer of 2 or higher, preferably an integer of from 2 to 4, more preferably 2 or 3, and still more preferably 2.

The structures of a plurality of indole skeletal parts in formula (1) may be the same or different from each other.

The compound represented by formula (1) is preferably a compound represented by formula (2), and more preferably a compound represented by formula (3).

The compound represented by formula (2) will be described below.

$R^{201}$  to  $R^{206}$  each has the same meaning as the meaning of  $R^{101}$  to  $R^{106}$ , and the preferred range is also the same.  $R^{201}$  and  $R^{206}$  may be bonded to each other to form a ring. As the ring formed by  $R^{201}$  and  $R^{206}$  to be bonded to each other, e.g., a cyclopentene ring, a cyclohexene ring, a 1,4-cyclohexadiene ring, a cycloheptene ring, a cyclooctene ring, etc., are exemplified.

$R^{206}$  may have a substituent. As the substituents, those exemplified as the substituents represented by  $R^{101}$  to  $R^{105}$  are applicable.

$R^{207}$  represents a substituent. As the substituents, the substituents described in  $R^{101}$  are exemplified. An alkyl group, an aryl group, and a silyl group are preferred as the substituents, an alkyl group and an aryl-substituted silyl group are more preferred, and an alkyl group and a triphenylsilyl group are still more preferred.

$n^{201}$  represents an integer of from 2 to 6, and  $n^{202}$  represents an integer of from 0 to 4, provided that  $n^{201} + n^{202} \leq 6$ .  $n^{201}$  is preferably an integer of from 2 to 4, more preferably 2 or 3, and still more preferably 2.  $n^{202}$  is preferably an integer of from 0 to 3, more preferably from 0 to 2, still more preferably 0 or 1, and especially preferably 0. When  $n^{201}$  is an integer of from 2 to 4, a plurality of  $R^{207}$ 's may be the same or different from each other.

The structures of a plurality of indole skeletal parts in formula (2) may be the same or different from each other.

The compound represented by formula (3) will be described below.  $R^{301}$  to  $R^{306}$  each has the same meaning as the meaning of  $R^{101}$  to  $R^{106}$ , and the preferred range is also the same.  $R^{301}$  and  $R^{306}$  may be bonded to each other to form a ring. As the ring formed by  $R^{301}$  and  $R^{306}$  to be bonded to each other, e.g., a cyclopentene ring, a cyclohexene ring, a 1,4-cyclohexadiene ring, a cycloheptene ring, a cyclooctene ring, etc., are exemplified.

$R^{306}$  may have a substituent. As the substituents, those exemplified as the substituents represented by  $R^{101}$  to  $R^{105}$  are applicable.

$R^{311}$  to  $R^{316}$  each has the same meaning as the meaning of  $R^{101}$  to  $R^{106}$ , and the preferred range is also the same.  $R^{311}$  and  $R^{316}$  may be bonded to each other to form a ring. As the ring formed by  $R^{311}$  and  $R^{316}$  to be bonded to each other, e.g., a cyclopentene ring, a cyclohexene ring, a 1,4-cyclohexadiene ring, a cycloheptene ring, a cyclooctene ring, etc., are exemplified.

$R^{316}$  may have a substituent. As the substituents, those exemplified as the substituents represented by  $R^{101}$  to  $R^{105}$  are applicable.

$R^{321}$  to  $R^{324}$  each represents a hydrogen atom or a substituent. As the examples of the substituents, the same groups as described in  $R^{101}$  above are exemplified.  $R^{321}$ ,  $R^{322}$  and  $R^{324}$  each preferably represents a hydrogen atom, an alkyl group or an aryl group, more preferably a hydrogen atom or an alkyl group, and still more preferably a hydrogen atom.  $R^{323}$  preferably represents a hydrogen atom, an alkyl group, or an aryl group, more preferably a hydrogen atom, an alkyl group, or an aryl-substituted silyl group, and still more preferably a hydrogen atom or a triphenylsilyl group.

The indole derivatives for use in the invention may be low molecular weight compounds, may be high molecular weight compounds in which the residue is directly bonded to the polymer main chain (preferably having a mass average molecular weight of from 1,000 to 5,000,000, more preferably from 5,000 to 2,000,000, and still more preferably from 10,000 to 1,000,000), or may be high molecular weight compounds having the indole

derivative of the invention on the main chain (preferably having a mass average molecular weight of from 1,000 to 5,000,000, more preferably from 5,000 to 2,000,000, and still more preferably from 10,000 to 1,000,000). In the case where the indole derivatives are high molecular weight compounds, they may be homopolymers, or may be copolymers with other polymers. When the indole derivatives are copolymers, they may be random copolymers or block copolymers. Further, when they are copolymers, at least one of a compound having a luminescent function and a compound having a charge transporting function may be contained in the polymers.

The indole derivative represented by any of formulae (1) to (3) is preferably contained in the light-emitting layer or a layer contiguous to the light-emitting layer, and more preferably contained in the light-emitting layer or a layer contiguous to the light-emitting layer on the anode side. It is also preferred to introduce the indole derivative represented by any of formulae (1) to (3) into both of the light-emitting layer and a layer contiguous to the light-emitting layer on the anode side.

When the indole derivative represented by any of formulae (1) to (3) is contained in the light-emitting layer, the content is preferably from 50 to 99 mass% (weight%), more preferably from 60 to 95 mass%, and still more preferably from 70 to 90 mass%. When the indole derivative is contained in layers other than the light-emitting layer, the content is preferably from 20 to 100 mass%, more preferably from 60 to 100 mass%, and still more preferably from 90 to 100 mass%.

By the introduction of the indole derivative represented by any of formulae (1) to (3) into the light-emitting device, it becomes possible to obtain good efficiency and durability and prevent variation of chromaticity by aging.

It is preferred for the light-emitting device in the invention to contain a phosphorescent material, e.g., a platinum phosphorescent material, and it is more preferred to contain a platinum complex having a tetradentate ligand.

The maximum emission wavelength of the platinum complex phosphorescent material having a tetradentate ligand is preferably 500 nm or less, more preferably 480 nm or

less, still more preferably 470 nm or less, and especially preferably 460 nm or less.

The external quantum efficiency of the light-emitting device is preferably 5% or more, more preferably 10% or more, and still more preferably 13% or more. As the numerical value of external quantum efficiency, the maximum value of the external quantum efficiency at the time of driving a device at 20 °C, or the value of the external quantum efficiency near 100 to 300 cd/m<sup>2</sup> at the time of driving a device at 20 °C can be used.

The inner quantum efficiency of the light-emitting device is preferably 30% or more, more preferably 50% or more, and still more preferably 70% or more. The inner quantum efficiency of a device is computed by the expression: inner quantum efficiency = external quantum efficiency/coupling out efficiency of light. In ordinary organic EL device, coupling out efficiency of light is about 20%, but it is possible to make coupling out efficiency of light 20% or more by various contrivances such as the shape of a substrate, the shape of electrodes, the thickness of an organic layer, the thickness of an inorganic layer, the refractive index of an organic layer, and the refractive index of an inorganic layer.

The light-emitting device is preferably a device having at least three layers of a hole transporting layer, a light-emitting layer and an electron transporting layer. It is more preferred to additionally provide, between the hole transporting layer and the light-emitting layer, a layer to accelerate hole injection to the light-emitting layer, a layer to block electrons, and a layer to block excitons.

The degree of charge transfer of a host material contained in the light-emitting layer is preferably  $1 \times 10^{-6}$  cm<sup>2</sup>/Vs or more and  $1 \times 10^{-1}$  cm<sup>2</sup>/Vs or less, more preferably  $5 \times 10^{-6}$  cm<sup>2</sup>/Vs or more and  $1 \times 10^{-2}$  cm<sup>2</sup>/Vs or less, still more preferably  $1 \times 10^{-5}$  cm<sup>2</sup>/Vs or more and  $1 \times 10^{-2}$  cm<sup>2</sup>/Vs or less, and especially preferably  $5 \times 10^{-5}$  cm<sup>2</sup>/Vs or more and  $1 \times 10^{-2}$  cm<sup>2</sup>/Vs or less.

The glass transition points of host materials, electron transporting materials and hole transporting materials contained in the organic electroluminescent device are preferably 90 °C or more and 400 °C or less, more preferably 100 °C or more and 380 °C or less, still more preferably 120 °C or more and 370 °C or less, and especially preferably 140 °C or more and

360 °C or less.

The organic electroluminescent device of the invention may be a white luminescent device.

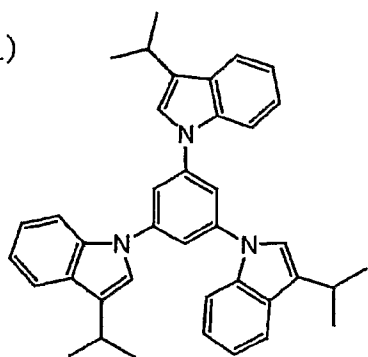
The  $T_1$  level (the energy level in the state of minimum triplet excitation) of the host material contained in the light-emitting device of the invention is preferably 60 kcal/mol or more (251.4 kJ/mol or more) and 90 kcal/mol or less (377.1 kJ/mol or less), more preferably 62 kcal/mol or more (259.78 kJ/mol or more) and 85 kcal/mol or less (356.15 kJ/mol or less), and still more preferably 65 kcal/mol or more (272.35 kJ/mol or more) and 80 kcal/mol or less (335.2 kJ/mol or less).

The  $T_1$  level (the energy level in the state of minimum triplet excitation) of the layer contiguous to the light-emitting layer is preferably 60 kcal/mol or more (251.4 kJ/mol or more) and 90 kcal/mol or less (377.1 kJ/mol or less), more preferably 62 kcal/mol or more (259.78 kJ/mol or more) and 85 kcal/mol or less (356.15 kJ/mol or less), and still more preferably 65 kcal/mol or more (272.35 kJ/mol or more) and 80 kcal/mol or less (335.2 kJ/mol or less).

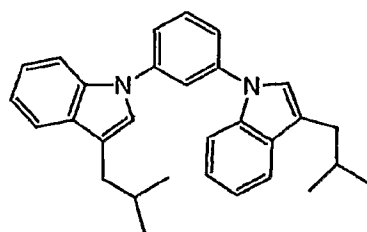
$T_1$  energy can be found by measuring the spectrum of emission of phosphorescence of a thin film of the material, and from the end of the short wavelength. For example, a film is formed with the material on a cleaned quartz glass substrate in a thickness of about 50 nm by vacuum deposition. The spectrum of emission of phosphorescence of the thin film is measured with a fluorescence spectrophotometer Model F-7000 (manufactured by Hitachi High Technologies) under a liquid nitrogen temperature. The  $T_1$  energy can be found by converting the rising wavelength on the short wave side of the obtained emission spectrum into an energy unit.

The compounds according to the invention are shown below, but the invention is not restricted to these compounds.

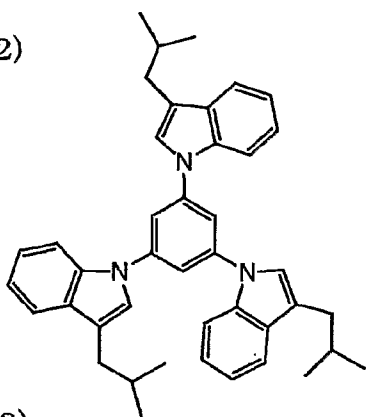
(1-1)



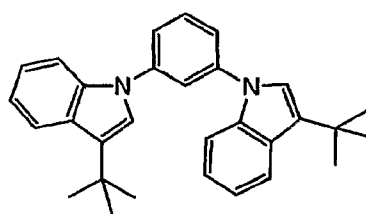
(1-5)



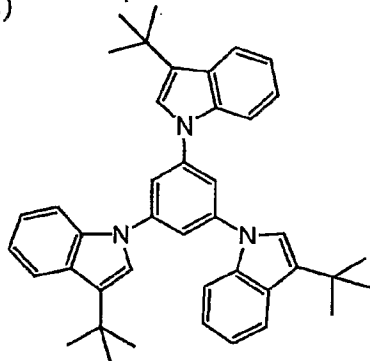
(1-2)



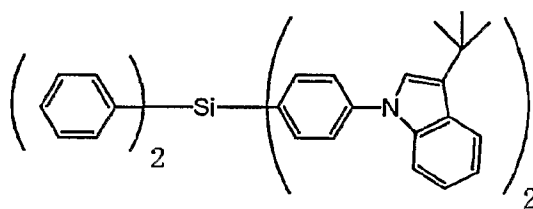
(1-6)



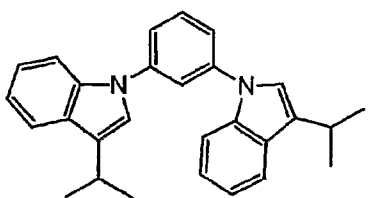
(1-3)



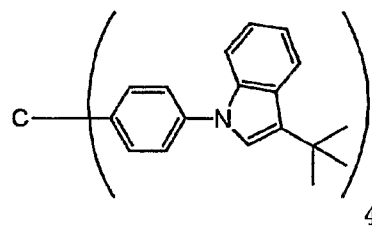
(1-7)



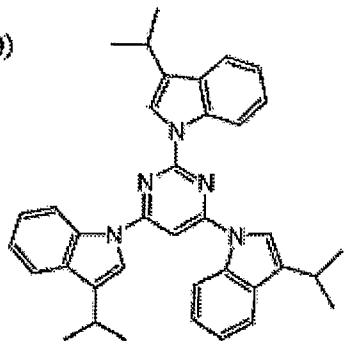
(1-4)



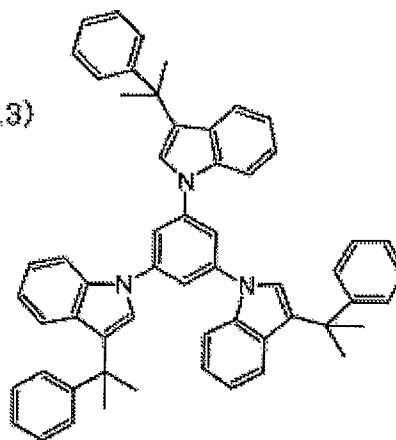
(1-8)



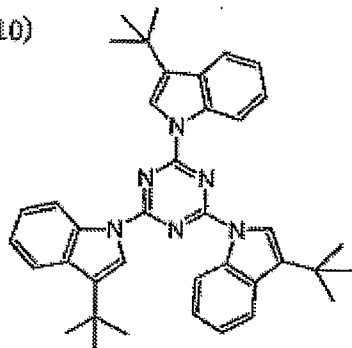
(1-9)



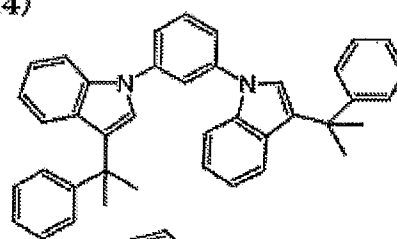
(1-13)



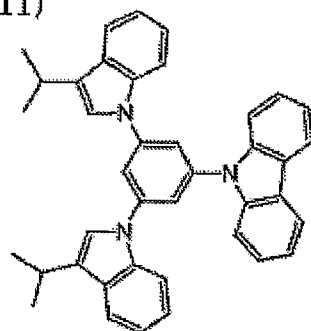
(1-10)



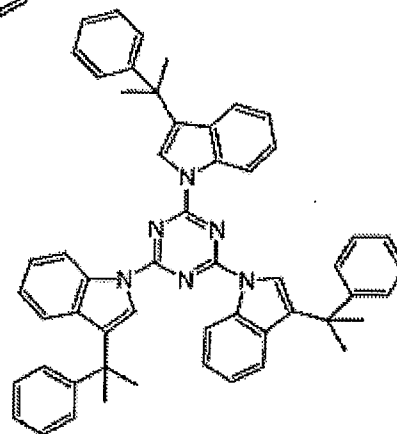
(1-14)



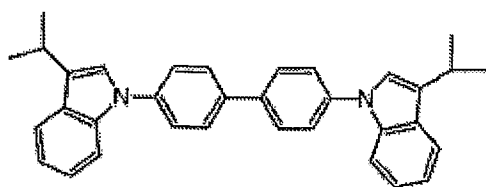
(1-11)



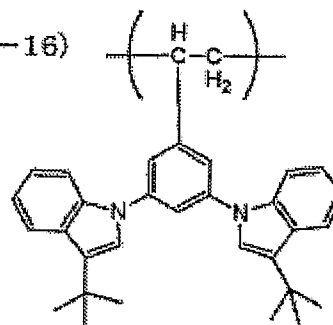
(1-15)



(1-12)

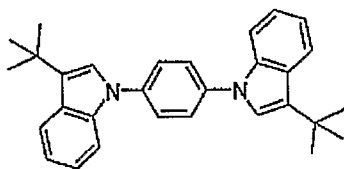


(1-16)

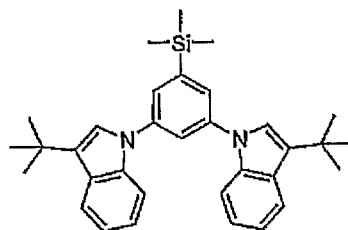


Mass-average molecular weight: 18,000

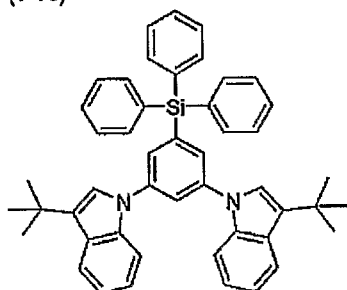
(1-17)



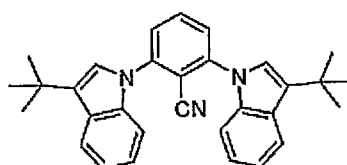
(1-21)



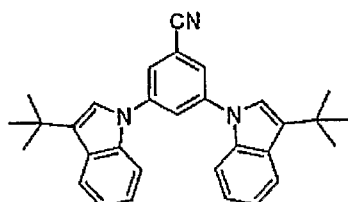
(1-18)



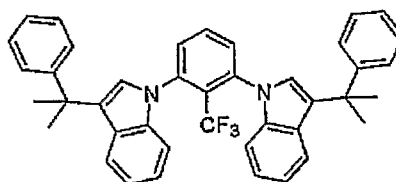
(1-22)



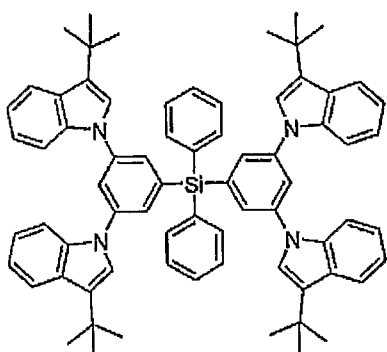
(1-19)



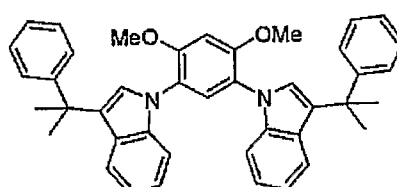
(1-23)



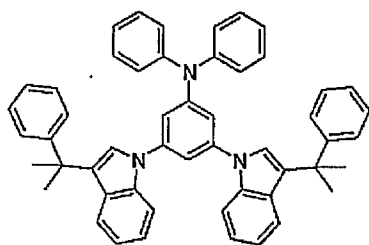
(1-20)



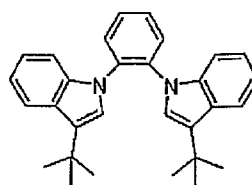
(1-24)



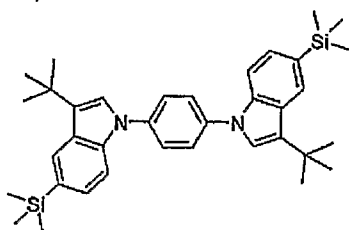
(1-25)



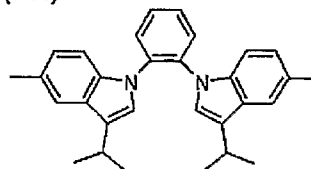
(1-30)



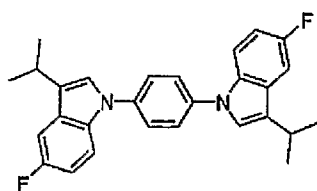
(1-26)



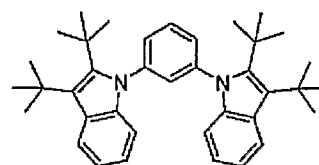
(1-31)



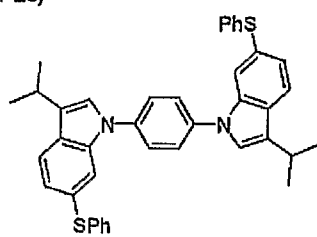
(1-27)



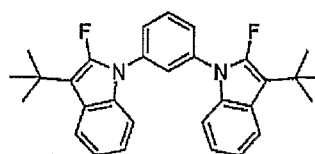
(1-32)



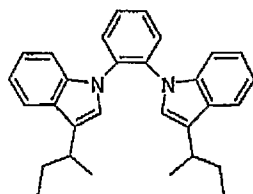
(1-28)



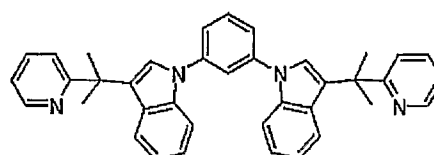
(1-33)



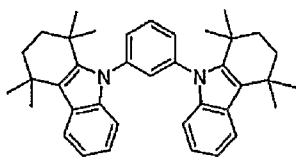
(1-29)



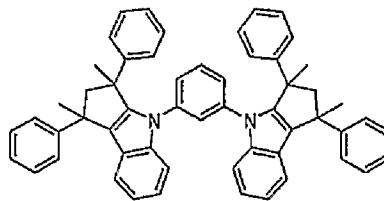
(1-34)



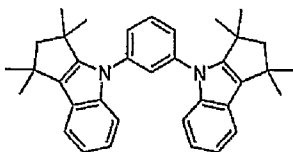
(1-35)



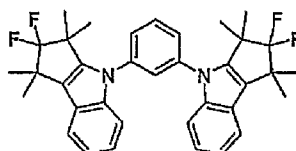
(1-36)



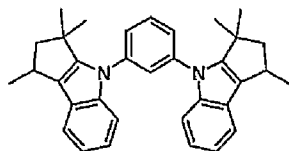
(1-37)



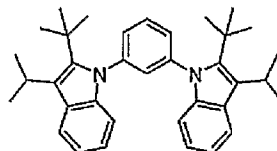
(1-38)



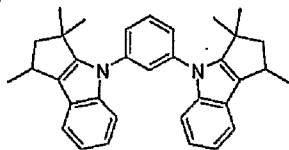
(1-39)



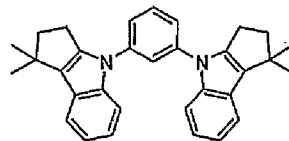
(1-40)



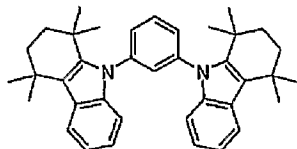
(1-41)



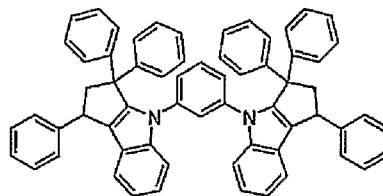
(1-42)



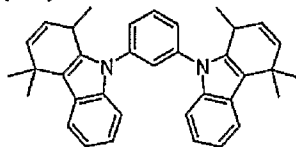
(1-43)



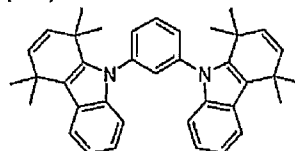
(1-44)



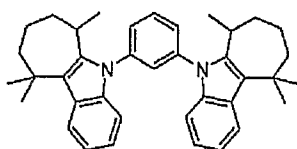
(1-45)



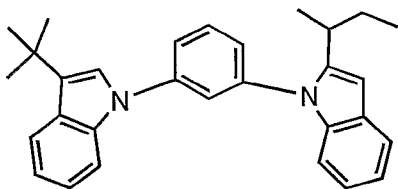
(1-46)



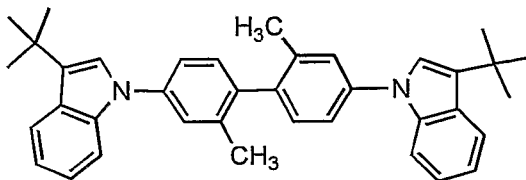
(1-47)



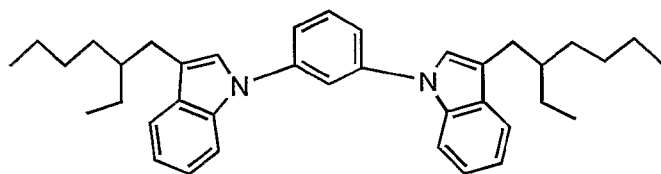
(1-48)



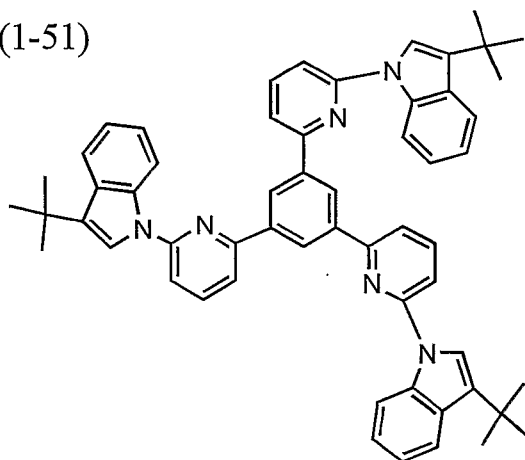
(1-49)



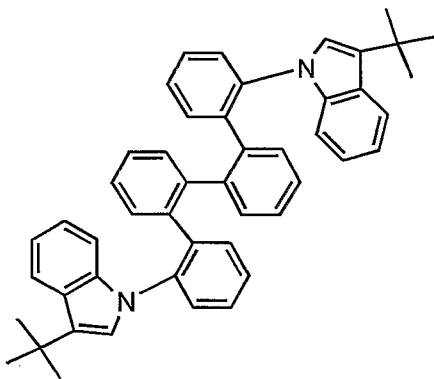
(1-50)



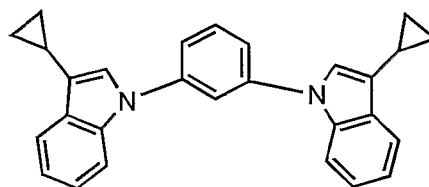
(1-51)



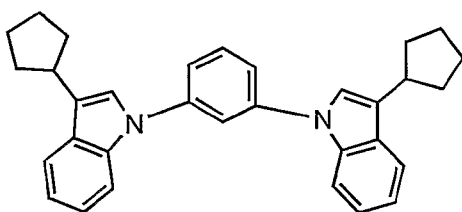
(1-52)



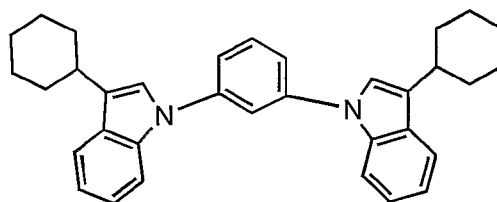
(1-53)



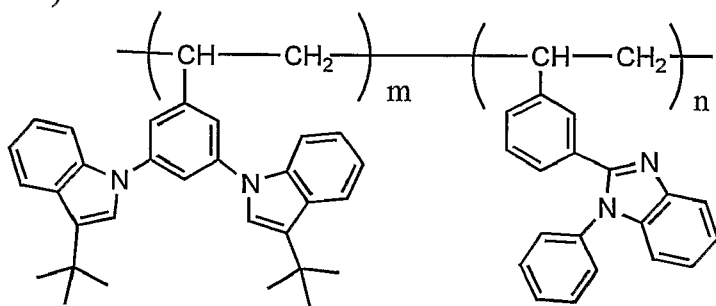
(1-54)



(1-55)

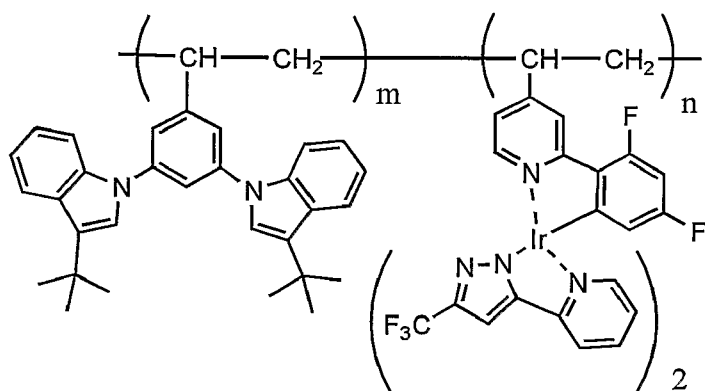


(1-56)



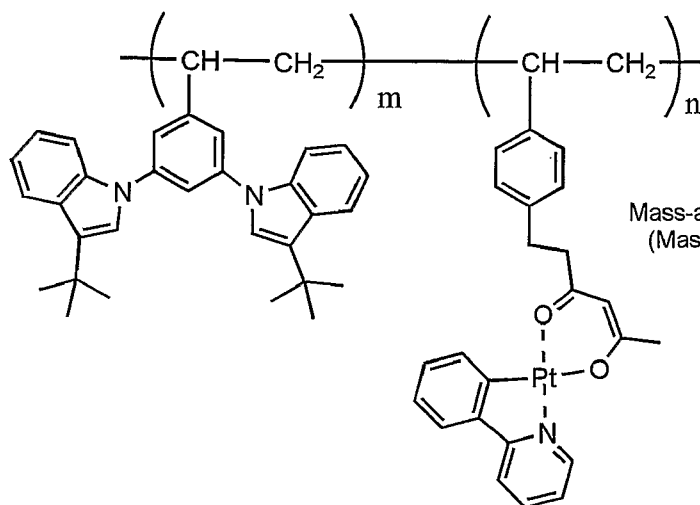
Mass-average molecular weight = 18,000  
(Mass% ratio: m/n = 60/40)

(1-57)



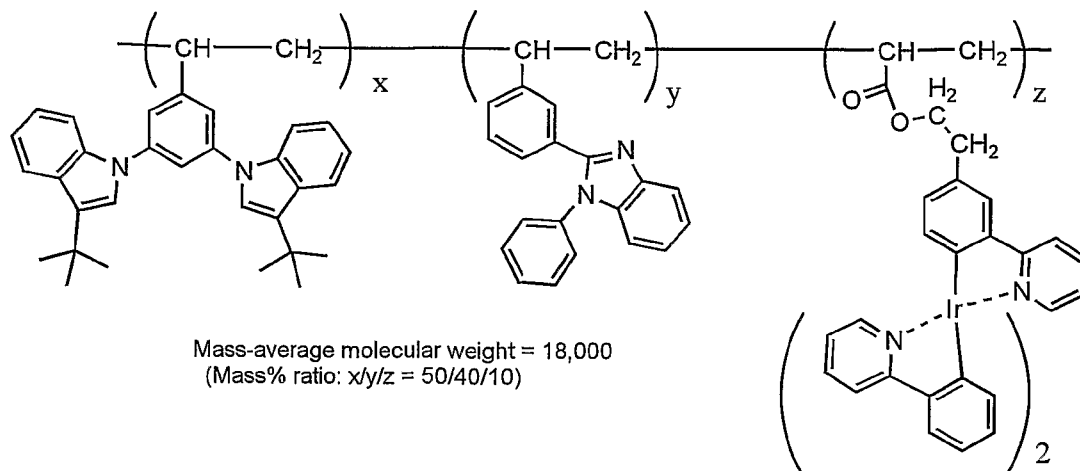
Mass-average molecular weight = 12,000  
(Mass% ratio: m/n = 85/15)

(1-58)

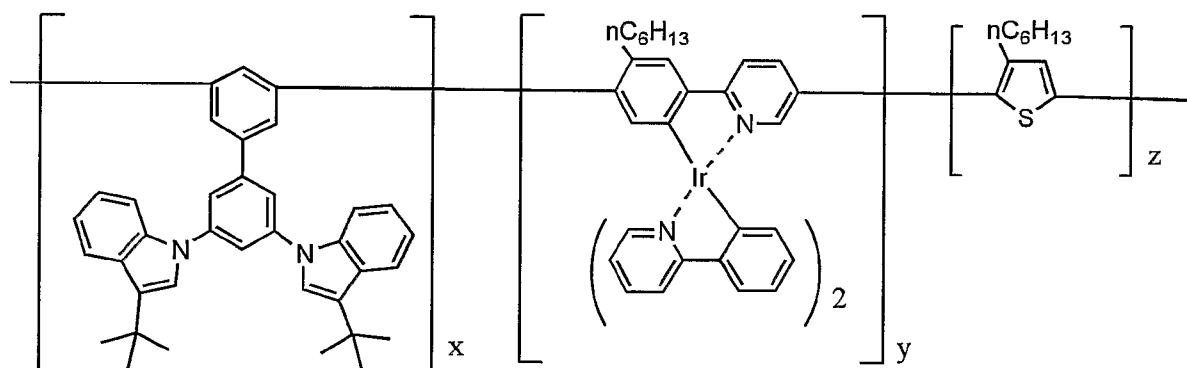


Mass-average molecular weight = 21,000  
(Mass% ratio: m/n = 85/15)

(1-59)



(1-60)



The ordinary synthesizing examples of the compounds according to the invention are shown below. The synthesizing methods are by no means restricted thereto.

Indole substituted with an alkyl group on the 3-position can be synthesized in accordance with various methods and, for example, a method of introduction of the alkyl group by electrophilic substitution reaction, and a method of synthesis by condensation reaction are exemplified. In connection with the method of introduction of the alkyl group by electrophilic substitution reaction, the indole can be synthesized by mixing an appropriate Lewis acid (e.g., aluminum chloride, zinc trifluoromethanesulfonate, zinc chloride, boron bromide, etc.) and a proper alkyl cation precursor (e.g., alkyl halide, etc.) in a solvent (refer

to J. Org. Chem., 2002, 67, 2705-2708). As the method of synthesis by condensation reaction, for example, Fischer's indole synthesis is exemplified. Indole substituted with an alkyl group on the 3-position can be obtained by heating phenylhydrazine and corresponding aldehyde in the presence of an acid (refer to Katsuyuki Ogura, Kagakusha no Tameno Kiso  
5 Koza 9 – Yuki Jinmei Hannou (Elementary Course for Chemists 9 - Organic Reactions Having the Names of Chemists), p. 182, Asakura Shoten (1997).

The compound of the invention can be obtained by a cross coupling reaction of the obtained indole substituted with an alkyl group on the 3-position and a proper aryl halide with a transition metal catalyst (a palladium catalyst, a copper catalyst, etc.) (refer to J. Org.  
10 Chem., 2004, 69, 5578).

Organic electroluminescent device:

The organic electroluminescent devices in the invention are described in detail below.

The light-emitting device includes a substrate having thereon a cathode and an  
15 anode, and organic layers (the organic layers may be organic layers including an organic compound alone, or may be organic layers containing an inorganic compound) including an organic light-emitting layer (sometimes referred to as merely "a light-emitting layer") between the electrodes. From the properties of the light-emitting device, it is preferred that at least one electrode of the cathode and anode is transparent.

As the embodiment of stacking of the organic layers in the invention, stacking is  
20 preferably in order of a hole transporting layer, a light-emitting layer, and an electron transporting layer from the anode side. Further, a charge blocking layer may be provided between the hole transporting layer and the light-emitting layer, or between the light-emitting layer and the electron transporting layer. A hole injecting layer may be provided between  
25 the anode and the hole transporting layer, and an electron injecting layer may be provided between the cathode and the electron transporting layer. Each layer may be divided into a plurality of secondary layers.

Constituents of the light-emitting device in the invention are described in detail

below.

Substrate:

The substrate for use in the invention is preferably a substrate that does not scatter or attenuate the light emitted from the organic layer. The specific examples of the materials of the substrate include inorganic materials, e.g., yttria stabilized zirconia (YSZ), glass, etc., and organic materials, such as polyester, e.g., polyethylene terephthalate, polybutylene phthalate, polyethylene naphthalate, etc., polystyrene, polycarbonate, polyether sulfone, polyallylate, polyimide, polycycloolefin, norbornene resin, poly(chloro- trifluoroethylene), etc.

When glass is used as a substrate, non-alkali glass is preferably used as the material for reducing elution of ions from the glass. Further, when soda lime glass is used, it is preferred to provide a barrier coat such as silica. In the case of organic materials, materials excellent in heat resistance, dimensional stability, solvent resistance, electrical insulating properties and processability are preferably used.

The form, structure and size of a substrate are not especially restricted, and these can be arbitrarily selected in accordance with the intended use and purpose of the light-emitting device. In general, a substrate is preferably in a plate-like form. The structure of a substrate may be a single layer structure or may be a stacking structure, and may consist of a single member or may be formed of two or more members.

A substrate may be colorless and transparent, or may be colored and transparent, but from the point of not scattering or attenuating the light emitted from an organic light-emitting layer, a colorless and transparent substrate is preferably used.

A substrate can be provided with a moisture permeation preventing layer (a gas barrier layer) on the front surface or rear surface.

As the materials of the moisture permeation preventing layer (the gas barrier layer), inorganic materials such as silicon nitride and silicon oxide are preferably used. The moisture permeation preventing layer (the gas barrier layer) can be formed, for example, by a high frequency sputtering method.

When a thermoplastic substrate is used, if necessary, a hard coat layer and an

undercoat layer may further be provided.

Anode:

An anode is generally sufficient to have the function of the electrode to supply positive holes to an organic layer. The form, structure and size of an anode are not especially restricted, and these can be arbitrarily selected from known materials of electrode in accordance with the intended use and purpose of the light-emitting device. As described above, an anode is generally provided as a transparent anode.

As the materials of anode, for example, metals, alloys, metal oxides, electrically conductive compounds, and mixtures of these materials are preferably exemplified. The specific examples of the materials of anode include electrically conductive metal oxides, e.g., tin oxide doped with antimony or fluorine (ATO, FTO), tin oxide, zinc oxide, indium oxide, indium tin oxide (ITO), indium zinc oxide (IZO), etc., metals, e.g., gold, silver, chromium, nickel, etc., mixtures or layered products of these metals with electrically conductive metal oxides, inorganic electrically conductive substances, e.g., copper iodide, copper sulfide, etc., organic electrically conductive materials, e.g., polyaniline, polythiophene, polypyrrole, etc., laminates of these materials with ITO, etc. Of these materials, electrically conductive metal oxides are preferred, and ITO is especially preferred in view of productivity, high conductivity, transparency and the like.

An anode can be formed on the substrate in accordance with various methods arbitrarily selected from, for example, wet methods, e.g., a printing method, a coating method, etc., physical methods, e.g., a vacuum deposition method, a sputtering method, an ion plating method, etc., and chemical methods, e.g., a CVD method, a plasma CVD method, etc., taking the suitability with the material to be used in the anode into consideration. For example, in the case of selecting ITO as the material of an anode, the anode can be formed according to a direct current or high frequency sputtering method, a vacuum deposition method, an ion plating method, etc.

In the organic electroluminescent device, the position of the anode to be formed is not especially restricted and can be formed anywhere. The position can be arbitrarily

selected in accordance with the intended use and purpose of the light-emitting device, but preferably provided on the substrate. In this case, the anode may be formed on the entire surface of one side of the substrate, or may be formed at a part.

As patterning in forming an anode, patterning may be performed by chemical etching such as by photo-lithography, may be carried out by physical etching such as by laser, may be performed by vacuum deposition or sputtering on a superposed mask, or a lift-off method and a printing method may be used.

The thickness of an anode can be optionally selected in accordance with the materials of the anode, so that cannot be regulated unconditionally, but the thickness is generally from 10 nm to 50  $\mu\text{m}$  or so, and is preferably from 50 nm to 20  $\mu\text{m}$ .

The value of resistance of an anode is preferably  $10^3 \Omega/\square$  or less, and more preferably  $10^2 \Omega/\square$  or less. In the case where an anode is transparent, the anode may be colorless and transparent, or colored and transparent. For the coupling out of luminescence from the transparent anode side, transmittance is preferably 60% or more, and more preferably 70% or more.

In connection with transparent anodes, description is found in Yutaka Sawada supervised, Tomei Denkyoku-Maku no Shintenkai (New Development in Transparent Electrode Films), CMC Publishing Co., Ltd. (1999), and the description therein can be referred to. In the case of using a plastic substrate low in heat resistance, a transparent anode film-formed with ITO or IZO at a low temperature of 150  $^{\circ}\text{C}$  or less is preferred.

#### Cathode:

A cathode is generally sufficient to have the function of the electrode to supply electrons to an organic layer. The form, structure and size of a cathode are not especially restricted, and these can be arbitrarily selected from known materials of electrode in accordance with the intended use and purpose of the light-emitting device.

As the materials of cathode, for example, metals, alloys, metal oxides, electrically conductive compounds, and mixtures of these materials are exemplified. The specific examples of the materials of cathode include alkali metals (e.g., Li, Na, K, Cs, etc.), alkaline

earth metals (e.g., Mg, Ca, etc.), gold, silver, lead, aluminum, sodium-potassium alloy, lithium- aluminum alloy, magnesium-silver alloy, indium, rare earth metals, e.g., ytterbium, etc. These materials may be used by one kind alone, but from the viewpoint of the compatibility of stability and an electron injecting property, two or more kinds of materials  
5 are preferably used in combination.

As the materials constituting a cathode, alkali metals and alkaline earth metals are preferred of these materials in the point of electron injection, and materials mainly comprising aluminum are preferred for their excellent preservation stability.

The materials mainly comprising aluminum mean aluminum alone, alloys of  
10 aluminum with 0.01 to 10 mass% of alkali metal or alkaline earth metal, or mixtures of these (e.g., lithium-aluminum alloy, magnesium-aluminum alloy, etc.).

The materials of cathode are disclosed in JP-A-2-15595 and JP-A-5-121172, and the materials described in these patents can also be used in the invention.

A cathode can be formed by known methods with no particular restriction. For  
15 example, a cathode can be formed according to wet methods, e.g., a printing method, a coating method, etc., physical methods, e.g., a vacuum deposition method, a sputtering method, an ion plating method, etc., and chemical methods, e.g., a CVD method, a plasma CVD method, etc., taking the suitability with the material constituting the cathode into consideration. For example, in the case of selecting metals as the material of a cathode, the  
20 cathode can be formed with one or two or more kinds of materials at the same time or in order by sputtering, etc.

As patterning in forming a cathode, patterning may be performed by chemical etching such as by photo-lithography, may be carried out by physical etching such as by laser, may be performed by vacuum deposition or sputtering on a superposed mask, or a lift-off  
25 method and a printing method may be used.

The position of the cathode to be formed is not especially restricted and can be formed anywhere in the invention. The cathode may be formed on the entire surface of the organic layer, or may be formed at a part.

A dielectric layer comprising fluoride or oxide of alkali metal or alkaline earth metal may be inserted between the cathode and the organic layer in a thickness of from 0.1 to 5 nm. The dielectric layer can be regarded as one kind of an electron injecting layer. The dielectric layer can be formed, for example, according to a vacuum deposition method, a sputtering method, an ion plating method, etc.

The thickness of a cathode can be optionally selected in accordance with the materials of the cathode, so that cannot be regulated unconditionally, but the thickness is generally from 10 nm to 5  $\mu$ m or so, and is preferably from 50 nm to 1  $\mu$ m.

A cathode may be transparent or opaque. A transparent cathode can be formed by forming a thin film of the materials of the cathode in a thickness of from 1 to 10 nm, and further laminating transparent conductive materials such as ITO and IZO.

#### Organic layer:

Organic layers in the invention will be described below.

The organic electroluminescent device of the invention has at least one organic layer including a light-emitting layer. As organic layers other than the light-emitting layer, as described above, a hole transporting layer, an electron transporting layer, a charge blocking layer, a hole injecting layer and an electron injecting layer are exemplified.

#### Formation of organic layers:

In the organic electroluminescent device of the invention, each layer constituting the organic layers can be preferably formed by any of dry film-forming methods such as a vacuum deposition method, a sputtering method, etc., a transfer method, and a printing method.

#### Organic light-emitting layer:

The organic light-emitting layer is a layer having functions to receive, at the time of electric field application, positive holes from the anode, hole injecting layer or hole transporting layer, and electrons from the cathode, electron injecting layer or electron transporting layer, and offer the field of recombination of positive holes and electrons to emit light.

The light-emitting layer in the invention may consist of light-emitting materials alone, or may comprise a mixed layer of a host material and a light-emitting material. The light-emitting material may be a fluorescent material or may be a phosphorescent material. Dopant may be one or two or more kinds. The host material is preferably a charge  
5 transporting material, and one or two or more host materials may be used. For example, the constitution of the mixture of an electron transporting host material and a hole transporting host material is exemplified. Further, a material not having an electron transporting property and not emitting light may be contained in the light-emitting layer.

The light-emitting layer may comprise one layer, or may be two or more layers, and  
10 each layer may emit light in different luminescent color.

The examples of fluorescent materials that can be used in the invention include various metal complexes represented by metal complexes of benzoxazole derivatives, benzimidazole derivatives, benzothiazole derivatives, styrylbenzene derivatives, polyphenyl derivatives, diphenylbutadiene derivatives, tetraphenylbutadiene derivatives, naphthalimide  
15 derivatives, coumarin derivatives, condensed aromatic compounds, perinone derivatives, oxadiazole derivatives, oxazine derivatives, aldazine derivatives, pyridine derivatives, cyclopentadiene derivatives, bisstyryl- anthracene derivatives, quinacridone derivatives, pyrrolo- pyridine derivatives, thiadiazolopyridine derivatives, cyclopentadiene derivatives, styrylamine derivatives, diketopyrrolopyrrole derivatives, aromatic dimethyldyne  
20 compounds, 8-quinolinol derivatives, and pyrromethene derivatives, polymer compounds such as polythiophene, polyphenylene, polyphenylenevinylene, and compounds such as organic silane derivatives.

The examples of phosphorescent materials that can be used in the invention include complexes containing a transition metal atom or a lanthanoid atom.

25 The transition metal atoms are not especially restricted, but ruthenium, rhodium, palladium, tungsten, rhenium, osmium, iridium and platinum are preferably exemplified, and rhenium, iridium and platinum are more preferred.

It is preferred to contain a platinum phosphorescent material, and more preferably a

platinum complex having a tetradentate ligand.

As lanthanoid atoms, lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, and lutecium are exemplified. Of these lanthanoid atoms, neodymium, europium and  
5 gadolinium are preferred.

As the examples of ligands of complexes, the ligands described, for example, in G. Wilkinson et al., Comprehensive Coordination Chemistry, Pergamon Press (1987), H. Yersin, Photochemistry and Photophysics of Coordination Compounds, Springer-Verlag (1987), and Akio Yamamoto, Yuki Kinzoku Kagaku -Kiso to Oyo- (Organic Metal Chemistry -Elements  
10 and Applications), Shokabo Publishing Co. (1982) are exemplified.

As the specific examples of ligands, halogen ligands (preferably a chlorine ligand), nitrogen-containing heterocyclic ligands (e.g., phenylpyridine, benzoquinoline, quinolinol, bipyridyl, phenanthroline, etc.), diketone ligands (e.g., acetylacetone, etc.), carboxylic acid ligands (e.g., acetic acid ligand, etc.), carbon monoxide ligands, isonitrile ligands, and cyano  
15 ligands are preferably exemplified, and more preferably nitrogen-containing heterocyclic ligands. These complexes may have one transition metal atom in a compound, or may be the so-called polynuclear complexes having two or more transition metal atoms. They may contain metal atoms of different kinds at the same time.

It is preferred for a phosphorescent material to be contained in the light-emitting  
20 layer in an amount of from 0.1 to 40 mass%, and more preferably from 0.5 to 20 mass%.

As the platinum complex phosphorescent materials having a tetradentate ligand, the compounds disclosed in U.S. Patent 6,653,654, WO 04/108857, WO 04/081017, WO 05/042444, JP-A-2006-232784, WO 05/042550, JP-A-2005-310733, JP-A-2005-317516, JP-A-2006-261623 and WO 06/098505 are specifically exemplified.

25 As host materials to be contained in the light-emitting layer in the invention, e.g., materials having a carbazole skeleton, having a diarylamine skeleton, having a pyridine skeleton, having a pyrazine skeleton, having a triazine skeleton, having an arylsilane skeleton, and those described later in the items of a hole injecting layer, a hole transporting layer, an

electron injecting layer, and an electron transporting layer are exemplified. As the host material, a compound having an indole skeleton is preferred, a compound represented by formula (1) is more preferred, a compound represented by formula (2) is still more preferred, and a compound represented by formula (3) is especially preferred.

5           The thickness of the light-emitting layer is not especially limited, but is generally preferably from 1 to 500 nm, more preferably from 5 to 200 nm, and still more preferably from 10 to 100 nm.

Hole injecting layer and hole transporting layer:

10           The hole injecting layer and the hole transporting layer are layers having a function to receive positive holes from the anode or anode side and transport the positive holes to the cathode side. The hole injecting layer and the hole transporting layer are specifically preferably the layers containing carbazole derivatives, azacarbazole derivatives, indole derivatives, azaindole derivatives, imidazole derivatives, polyaryllalkane derivatives, pyrazoline derivatives, pyrazolone derivatives, phenylenediamine derivatives, arylamine derivatives, amino-substituted chalcone derivatives, styrylanthracene derivatives, fluorenone derivatives, hydrazone derivatives, stilbene derivatives, silazane derivatives, aromatic tertiary amine compounds, styrylamine compounds, aromatic dimethyldiyne compounds, porphyrin compounds, organic silane derivatives, carbon, various kinds of metal complexes represented by Ir complex having phenylazole, or phenylazine as the ligand.

20           An electron accepting dopant can be contained in the positive hole injecting layer or positive hole transporting layer of the organic EL device of the invention. As the electron accepting dopants to be introduced to the hole injecting layer or hole transporting layer, inorganic compounds and organic compounds can be used so long as they are electron accepting and have a property of capable of oxidizing an organic compound.

25           Specifically, as the inorganic compounds, halogenated metals, e.g., ferric chloride, aluminum chloride, gallium chloride, indium chloride, antimony pentachloride, etc., and metal oxides, e.g., vanadium pentoxide, molybdenum trioxide, etc., are exemplified.

When dopants are organic compounds, the compounds having as a substituent a

nitro group, halogen, a cyano group, or a trifluoromethyl group, quinone compounds, acid anhydride compounds, and fullerene are preferably used.

Besides the above compounds, the compounds disclosed in JP-A-6-212153, JP-A-11-111463, JP-A-11-251067, JP-A-2000-196140, JP-A-2000-286054, JP-A-2000-315580, JP-A-2001-102175, JP-A-2001-160493, JP-A-2002-252085, JP-A-2002-56985, JP-A-2003-157981, JP-A-2003-217862, JP-A-2003-229278, JP-A-2004-342614, JP-A-2005-72012, JP-A-2005-166637 and JP-A-2005-209643 can be preferably used.

Of these compounds, hexacyanobutadiene, hexacyanobenzene, tetracyanoethylene, tetracyanoquinodimethane, tetrafluorotetracyanoquinodimethane, p-fluoranyl, p-chloranyl, p-bromanyl, p-benzoquinone, 2,6-dichlorobenzoquinone, 2,5-dichlorobenzoquinone, 1,2,4,5-tetracyanobenzene, 1,4-dicyanotetrafluorobenzene, 2,3-dichloro-5,6-dicyanobenzoquinone, p-dinitrobenzene, m-dinitrobenzene, o-dinitrobenzene, 1,4-naphthoquinone, 2,3-dichloronaphthoquinone, 1,3-dinitronaphthoquinone, 1,5-dinitronaphthalene, 9,10-anthraquinone, 1,3,6,8-tetranitrocarbazole, 2,4,7-trinitro-9-fluorenone, 2,3,5,6-tetracyanopyridine, and fullerene C<sub>60</sub> are preferred, hexacyanobutadiene, hexacyanobenzene, tetracyanoethylene, tetracyanoquinodimethane, tetrafluorotetracyanoquinodimethane, p-fluoranyl, p-chloranyl, p-bromanyl, 2,6-dichlorobenzoquinone, 2,5-dichlorobenzoquinone, 2,3-dichloronaphthoquinone, 1,2,4,5-tetracyanobenzene, 2,3-dichloro-5,6-dicyanobenzoquinone, and 2,3,5,6-tetracyanopyridine are more preferred, and tetrafluorotetracyanoquinodimethane is especially preferred.

These electron accepting dopants may be used by one kind alone, or two or more kinds may be used in combination. The amount of the electron accepting dopant to be used differs according to the kind of the material, but the amount is preferably from 0.01 to 50 mass% to the material of the positive hole transporting layer, more preferably from 0.05 to 20 mass%, and still more preferably from 0.1 to 10 mass%.

The thickness of the hole injecting layer and hole transporting layer is preferably

500 nm or less from the viewpoint of lowering driving voltage.

The thickness of the hole transporting layer is preferably from 1 to 500 nm, more preferably from 5 to 200 nm, and still more preferably from 10 to 100 nm. The thickness of the hole injecting layer is preferably from 0.1 to 200 nm, more preferably from 0.5 to 100 nm, and still more preferably from 1 to 100 nm.

The hole injecting layer and the hole transporting layer may be a single layer structure comprising one or two or more of the above materials, or may be a multilayer structure comprising a plurality of layers of the same or different compositions.

Electron injecting layer and electron transporting layer:

The electron injecting layer and the electron transporting layer are layers having a function to receive electrons from the cathode or cathode side and transport the electrons to the anode side. The electron injecting layer and the electron transporting layer are specifically preferably layers containing triazole derivatives, oxazole derivatives, oxadiazole derivatives, imidazole derivatives, fluorenone derivatives, anthraquinodimethane derivatives, anthrone derivatives, diphenylquinone derivatives, thiopyran dioxide derivatives, carbodiimide derivatives, fluorenylidene methane derivatives, distyrylpyrazine derivatives, tetracarboxylic anhydride of aromatic rings such as naphthalene, perylene, etc., a phthalocyanine derivative, various metal complexes represented by metal complexes of 8-quinolinol derivatives, metalphthalocyanine, metal complexes having benzoxazole, benzothiazole as the ligand, organic silane derivative, etc.

An electron donating dopant can be contained in the electron injecting layer or electron transporting layer of the organic EL elemental device of the invention. Any compound can be used as the electron donating dopant to be introduced to the electron injecting layer or electron transporting layer, so long as the compound is electron accepting and has a property of capable of reducing an organic compound, and alkali metal salts, e.g., Li, alkaline earth metals, e.g., Mg, transition metals containing a rare earth metal, and reducing organic compounds are preferably used as the electron donating dopant. As the metals, metals having a work function of 4.2 eV or less can be preferably used, and

specifically Li, Na, K, Be, Mg, Ca, Sr, Ba, Y, Cs, La, Sm, Gd and Yb are exemplified. As the reducing organic compounds, e.g., nitrogen-containing compounds, sulfur-containing compounds, and phosphorus-containing compounds are exemplified.

Besides the above, the materials disclosed in JP-A-6-212153, JP-A-2000-196140, JP-A-2003-68468, JP-A-2003-229278, and JP-A-2004-342614 can be used.

These electron donating dopants may be used alone, or two or more kinds may be used in combination. The use amount of the electron donating dopants differs according to the kind of the material, but the amount is preferably from 0.1 to 99 mass% to the material of the electron transporting layer, more preferably from 1.0 to 80 mass%, and especially preferably from 2.0 to 70 mass%.

The thickness of each of the electron injecting layer and electron transporting layer is preferably 500 nm or less from the viewpoint of lowering driving voltage.

The thickness of the electron transporting layer is preferably from 1 to 500 nm, more preferably from 5 to 200 nm, and still more preferably from 10 to 100 nm. The thickness of the electron injecting layer is preferably from 0.1 to 200 nm, more preferably from 0.2 to 100 nm, and still more preferably from 0.5 to 50 nm.

The electron injecting layer and the electron transporting layer may be a single layer structure comprising one or two or more of the above materials, or may be a multilayer structure comprising a plurality of layers of the same or different compositions.

#### Hole blocking layer:

A hole blocking layer is a layer having a function of preventing the positive holes transported from the anode side to the light-emitting layer from passing through to the cathode side. In the invention, a hole blocking layer can be provided as the organic layer contiguous to the light-emitting layer on the cathode side.

As the examples of the organic compounds constituting the hole blocking layer, aluminum complexes, e.g., aluminum (III) bis(2-methyl-8-quinolino)-4-phenylphenolate (abbreviated to BAlq), etc., triazole derivatives, phenanthroline derivatives, e.g., 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (abbreviated to BCP), etc., are exemplified.

The thickness of the hole blocking layer is preferably from 1 to 500 nm, more preferably from 5 to 200 nm, and still more preferably from 10 to 100 nm.

The hole blocking layer may be a single layer structure comprising one or two or more of the above materials, or may be a multilayer structure comprising a plurality of layers  
5 of the same or different compositions.

Protective layer:

In the invention the organic EL device may be completely protected with a protective layer.

It is sufficient for the materials to be contained in the protective layer to have a  
10 function capable of restraining the substances accelerating deterioration of device, e.g., water, oxygen, etc., from entering the device.

The specific examples of such materials include metals, e.g., In, Sn, Pb, Au, Cu, Ag, Al, Ti, Ni, etc., metal oxides, e.g., MgO, SiO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, GeO, NiO, CaO, BaO, Fe<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, etc., metal nitrides, e.g., SiN<sub>x</sub>, SiN<sub>x</sub>O<sub>y</sub>, etc., metal fluorides, e.g., MgF<sub>2</sub>, LiF,  
15 AlF<sub>3</sub>, CaF<sub>2</sub>, etc., polyethylene, polypropylene, polymethyl methacrylate, polyimide, polyurea, polytetrafluoroethylene, polychlorotrifluoroethylene, polydichlorodifluoroethylene, copolymers of chlorotrifluoro- ethylene with dichlorodifluoroethylene, copolymers obtained by copolymerization of a monomer mixture containing tetrafluoroethylene and at least one comonomer, fluorine- containing copolymers having a cyclic structure on the main chain of  
20 the copolymer, water absorptive substances having a water absorption rate of not lower than 1%, moisture proofing substances having a water absorption rate of not higher than 0.1%.

The forming method of the protective layer is not especially restricted and, for example, a vacuum deposition method, a sputtering method, a reactive sputtering method, an MBE (molecular beam epitaxy) method, a cluster ion beam method, an ion plating method, a  
25 plasma polymerization method (a high frequency excitation ion plating method), a plasma CVD method, a laser CVD method, a heat CVD method, a gas source CVD method, a coating method, a printing method, a transfer method, etc., can be applied to the invention.

Sealing:

The organic electroluminescent device of the invention may be completely sealed in a sealing container.

Further, a water absorber or an inert liquid may be filled in the space between the sealing container and the light-emitting device. The water absorber is not especially restricted and, for example, barium oxide, sodium oxide, potassium oxide, calcium oxide, sodium sulfate, calcium sulfate, magnesium sulfate, phosphorus pentoxide, calcium chloride, magnesium chloride, copper chloride, cesium fluoride, niobium fluoride, calcium bromide, vanadium bromide, molecular sieve, zeolite, magnesium oxide, etc., can be exemplified. The inert liquid is not particularly limited and, for example, paraffins, liquid paraffins, fluorine solvents, such as perfluoroalkane, perfluoroamine, perfluoroether, etc., chlorine solvents, and silicone oils are exemplified.

Luminescence can be obtained by the application of DC (if necessary, an alternating current factor may be contained) voltage (generally from 2 to 15 V) or DC electric current between the anode and cathode of the organic electroluminescent device of the invention.

In connection with the driving methods of the organic electroluminescent device of the invention, the driving methods disclosed in JP-A-2-148687, JP-A-6-301355, JP-A-5-29080, JP-A-7-134558, JP-A-8-234685, JP-A-8-241047, Japanese Patent 2784615, and U.S. Patents 5,828,429 and 6,023,308 can be used.

## Examples

The invention will be described more specifically with reference to examples, but the invention should not be construed as being restricted thereto.

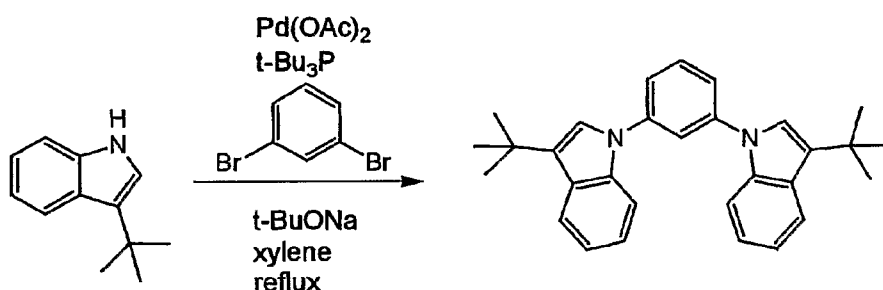
### Synthesis of Exemplified Compound (1-6):

3-tert-Butylindole is synthesized according to the method described in J. Org. Chem., 2002, 67, 2705-2708.

Under the nitrogen atmosphere, 0.06 ml (0.24 mmol) of tri-tert-butylphosphine is added to a mixed solution comprising 0.975 g (5.63 mmol) of 3-tert-butylindole, 0.604 g (2.56 mmol) of 1,3-dibromobenzene, 14 mg (0.06 mmol) of palladium acetate, 0.74 g (7.7

mmol) of sodium tert-butoxylate, and 25 ml of xylene, and the reaction mixture is stirred by heating while refluxing for 6 hours. Water is added to the obtained reaction solution, an organic layer extracted with ethyl acetate is dried with magnesium sulfate and concentrated under reduced pressure. The resulting residue is refined with silica gel column chromatography to obtain 1.05 g (2.51 mmol, yield: 98%) of exemplified compound (1-6).

<sup>1</sup>H-NMR of exemplified compound (1-6): 1.52 (s, 18H), 7.12 (s, 2H), 7.14-7.27 (m, 4H), 7.47 (d, 2H), 7.62 (dd, 4H), 7.89 (d, 2H) 400 MHz



#### Synthesis of Exemplified Compound (1-3):

Under the nitrogen atmosphere, 0.20 ml (0.80 mmol) of tri-tert-butylphosphine is added to a mixed solution comprising 4.30 g (24.8 mmol) of 3-tert-butylindole, 2.36 g (7.5 mmol) of 1,3,5-tribromobenzene, 45 mg (0.2 mmol) of palladium acetate, 3.27 g (34 mmol) of sodium tert-butoxylate, and 120 ml of xylene, and the reaction mixture is stirred by heating while refluxing for 6 hours. Water is added to the obtained reaction solution, an organic layer extracted with ethyl acetate is dried with magnesium sulfate and concentrated under reduced pressure. The resulting residue is refined with silica gel column chromatography to obtain 1.92 g (3.24 mmol) of exemplified compound (1-3).

<sup>1</sup>H-NMR of exemplified compound (1-3): 1.52 (s, 27H), 7.14-7.28 (m, 9H), 7.61 (s, 3H), 7.68 (d, 3H), 7.90 (s, 3H) 300 MHz

#### Synthesis of Exemplified Compound (1-10):

Under the nitrogen atmosphere, 0.96 g of sodium hydroxide (an oil additive, purity: 50-70%) is added to 20 ml of an N-methylpyrrolidone solution containing 3.47 g (20 mmol) of 3-tert-butylindole and stirred. To the resulting mixture is added 1.22 g (6.6 mmol) of

isocyanurate chloride. The reaction solution is heated at 200 °C and stirred for 5 hours. After being allowed to stand, the reaction solution is added to water, and the resulting solid is filtered. The solid is washed with isopropyl alcohol with heating to obtain 1.06 g of exemplified compound (1-10).

5 <sup>1</sup>H-NMR of exemplified compound (1-10): 1.58 (s, 27H), 7.33 (dd, 3H), 7.42 (dd, 3H), 7.87 (d, 2H), 8.10 (s, 1H), 8.92 (d, 2H) 300 MHz

Synthesis of Exemplified Compound (1-17):

Exemplified compound (1-17) is synthesized in the same manner as in the synthesis of compound (1-6) except for using 1,4-dibromobenzene in place of 1,3-dibromobenzene.

10 <sup>1</sup>H-NMR of exemplified compound (1-17): 1.52 (s, 18H), 7.13-7.28 (m, 6H), 7.58-7.67 (m, 6H), 7.90 (d, 2H) 400 MHz

Synthesis of Exemplified Compound (1-18):

Exemplified compound (1-18) is synthesized by referring to the method described in Eur. J. Org. Chem., 1999, 9, 2079 except for using 1,3-dibromo-5-triphenylsilylbenzene in  
15 place of 1,3-dibromobenzene.

Exemplified compound (1-18) is synthesized in the same manner as in the synthesis of compound (1-6) except for using 1,3-dibromo-5-triphenylsilylbenzene in place of 1,3-dibromobenzene.

20 <sup>1</sup>H-NMR of exemplified compound (1-18): 1.47 (s, 18H), 7.05 (s, 2H), 7.12-7.16 (m, 4H), 7.40-7.50 (m, 11H), 7.61 (dd, 1H), 7.64-7.68 (m, 8H), 7.82-7.85 (m 2H) 400 MHz

Synthesis of Exemplified Compound (1-22):

Under the nitrogen atmosphere, 4.32 g (21.7 mmol) of potassium hexamethyldisilazane is added to 40 ml of an N-methylpyrrolidone solution containing 3.43 g (19.8 mmol) of 3-tert-butylindole, and then 1.25 g (9.0 mmol) of 2,6-difluorobenzonitrile is  
25 added. The mixture is stirred at 200 °C for 30 minutes and then allowed to stand to be cooled to room temperature. The reaction solution is added to water and the resulting solid is filtered. The solid is refined with silica gel column chromatography and further recrystallized with a mixed solvent of isopropyl alcohol and hexane to obtain 0.90 g of

exemplified compound (1-22) as a white solid.

<sup>1</sup>H-NMR of exemplified compound (1-22): 1.51 (s, 18H), 7.08 (s, 2H), 7.18-7.32 (m, 4H), 7.60 (d, 2H), 7.72 (d, 2H), 7.87-7.92 (m, 3H) 300 MHz

Synthesis of Exemplified Compound (1-21):

5 Exemplified compound (1-21) is synthesized by referring to the method described in Eur. J. Org. Chem., 1999, 9, 2079 except for using 1,3-dibromo-5-trimethylsilylbenzene in place of 1,3-dibromobenzene.

Exemplified compound (1-21) is synthesized in the same manner as in the synthesis of compound (1-6) except for using 1,3-dibromo-5-trimethylsilylbenzene in place of  
10 1,3-dibromo- benzene.

<sup>1</sup>H-NMR of exemplified compound (1-21): 0.36 (s, 9H), 1.51 (s, 18H), 7.23-7.27 (m, 5H), 7.55-7.62 (m, 5H), 7.89 (d, 2H) 300 MHz

Synthesis of Exemplified Compound (1-19):

Exemplified compound (1-19) is synthesized in the same manner as in the synthesis  
15 of Compound (1-22) except for using 3,5-difluorobenzonitrile in place of 2,6-difluorobenzonitrile.

<sup>1</sup>H-NMR of exemplified compound (1-19): 1.52 (s, 18H), 7.18-7.30 (m, 6H), 7.44 (d, 2H), 7.59 (d, 2H), 7.78 (dd, 1H), 7.89 (d, 2H) 400 MHz

Other indole derivatives can also be synthesized similarly. For example,  
20 exemplified Compound (1-7) can be synthesized in the same manner with bis(4-bromophenyl)diphenylsilane as the starting material.

COMPARATIVE EXAMPLE 1

A cleaned ITO substrate is placed in a vacuum evaporator, copper phthalocyanine is  
25 deposited on the substrate in a thickness of 10 nm, and NPD (N,N'-di- $\alpha$ -naphthyl-N,N'-diphenyl)benzidine is deposited thereon in a thickness of 40 nm. Compound B-1 and compound A in the ratio of 12/88 (by mass) are deposited on the above deposited film in a thickness of 30 nm (a light-emitting layer), then Aluminum (III)

bis(2-methyl-8-quinolino)-4-phenylphenolate (referred to as BAlq) is deposited thereon in a thickness of 6 nm, and then Alq (tris(8-hydroxyquinoline) aluminum complex) is deposited on the above film in a thickness of 20 nm. Lithium fluoride is deposited thereon in a thickness of 3 nm, and then Al having a thickness of 60 nm is provided as a cathode by  
5 patterning with the shadow mask.

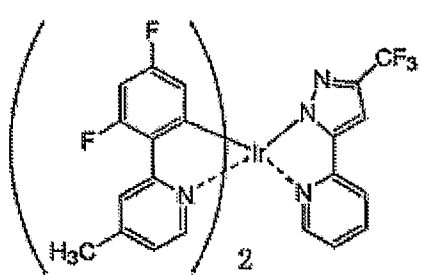
Each layer is provided according to resistance heating vacuum deposition.

The manufactured layered product is put in a glove box substituted with nitrogen gas, and sealed in a stainless steel sealing can with a UV-curable type adhesive (XNR5516HV, manufactured by Nagase Ciba).

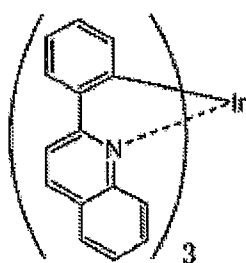
10 The obtained EL device is subjected to application of DC constant voltage with a source measure unit Model 2400 (manufactured by Toyo Technica Co., Ltd.) to emit light. It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained.

## 15 COMPARATIVE EXAMPLE 2

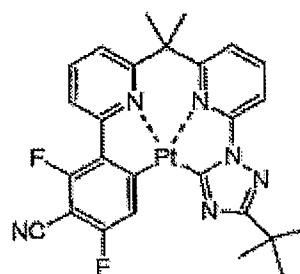
A device is prepared and evaluated in the same manner as in comparative Example 1 except for using compound B in place of compound A. It is confirmed that the emission of blue phosphorescence originating in compound B-1 is obtained.



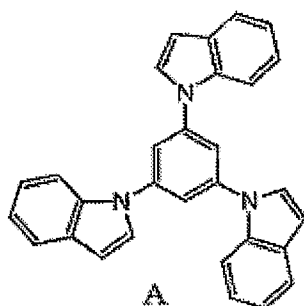
B-1



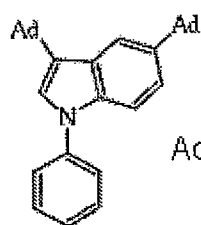
B-2



B-3

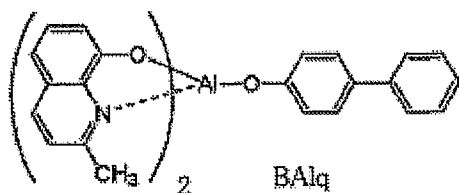


A

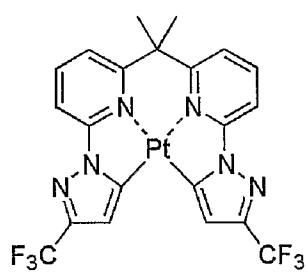


B

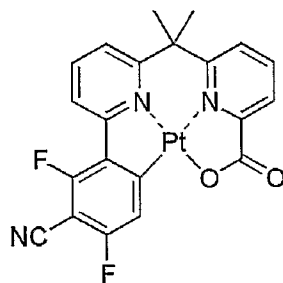
Ad: 1-Adamantyl group



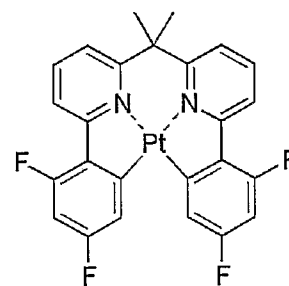
BAlq



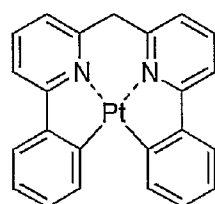
B-4



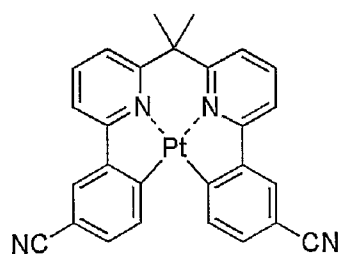
B-5



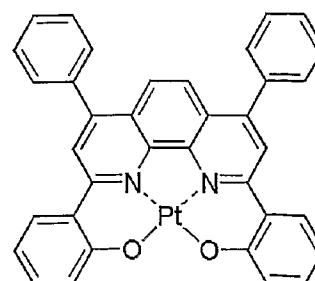
B-6



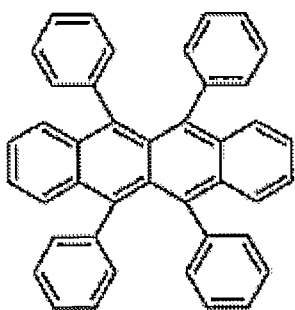
B-7



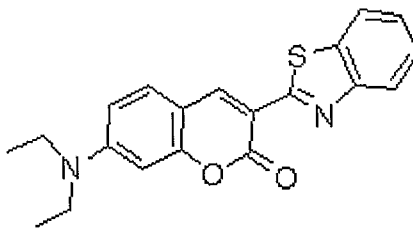
B-8



B-9



12-1



12-2

### EXAMPLE 1

Evaluation of a device is performed in the same manner as in Comparative Example 1 except for using exemplified compound (1-1) of the invention in place of compound A. It is confirmed that the emission of blue phosphorescence originating in compound B-1 is obtained.

### EXAMPLE 2

Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-3) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained.

### EXAMPLE 3

Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-6) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained.

### EXAMPLE 4

Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-7) in place of Compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained.

## EXAMPLE 5

Evaluation of a device is performed in the same manner as in Example 3 except for using compound B-2 in place of compound B-1. It is confirmed that the emission of red phosphorescence originating in compound B-2 is obtained.

## EXAMPLE 6

Evaluation of a device is performed in the same manner as in Example 3 except for using compound B-3 in place of compound B-1. It is confirmed that the emission of blue phosphorescence originating in compound B-3 is obtained.

## EXAMPLE 7

Evaluation of a device is performed in the same manner as in Example 3 except for inserting a layer containing exemplified compound (1-6) having a thickness of 3 nm between the NPD layer and the light-emitting layer. It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained.

Evaluation of light-emitting device:

Each of the obtained light-emitting devices is driven at 20 °C by the application of constant electric current. Luminance is measured with a luminance meter BM-8 (trade name, manufactured by Topcon Co.). Emission spectrum is measured with an emission spectrum measuring system (ELS1500, manufactured by Shimadzu Corporation). The half life time of luminance is found by measuring the time required to reach the half life of luminance from the initial luminance of 360 cd/m<sup>2</sup>. CIE Y value is found from the emission spectrum measured at 20 °C with an emission spectrum measuring system (ELS1500, manufactured by Shimadzu Corporation), and the variation in chromaticity is computed from the CIE Y value. The light-emitting device is driven at 20 °C and luminance of 360 cd/m<sup>2</sup> by the application of constant current, and the external quantum efficiency is computed from

the obtained emission spectrum and front luminance by a luminance conversion method.

The results obtained are shown in Table 1 below.

**TABLE 1**

5 Results of evaluation of devices prepared in Comparative Examples 1 and 2 and Examples 1 to 7

Example No.	External Quantum Efficiency (relative value)	Half Life Time of Luminance (relative value)	Variation in Chromaticity at Half Life Time of Luminance (rate of change of CIE y value in relative value)
Comparative Example 1	1.0	1.0	1.0
Comparative Example 2	1.4	0.9	0.8
Example 1	1.6	1.5	0.2
Example 2	2.0	2.1	0.2
Example 3	2.0	2.5	0.2
Example 4	1.8	1.7	0.2
Example 5	2.0	3.0	0.2
Example 6	2.0	4.1	0.1
Example 7	2.4	2.8	0.2

\* The evaluation value in each example is a relative value to the evaluation value of Comparative Example 1 as the reference value.

10 It can be seen from the results shown in Table 1 that the phosphorescent elemental devices in the invention using indole derivatives of the invention in the light-emitting layers are excellent in efficiency and durability and little in variation of chromaticity. Further, in the case where indole derivative of the invention is used in the light-emitting layer and at the same time in the layer contiguous to the light-emitting layer as in Example 7, the above  
15 effects can be obtained.

#### EXAMPLE 8

Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-10) in place of compound (1-1). It is confirmed that the  
20 emission of phosphorescence originating in compound B-1 is obtained. The results

obtained are shown in Table 2 below.

#### EXAMPLE 9

5 Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-17) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained. The results obtained are shown in Table 2.

#### EXAMPLE 10

10 Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-18) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained. The results obtained are shown in Table 2.

#### 15 EXAMPLE 11

Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-19) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained. The results obtained are shown in Table 2.

20

#### EXAMPLE 12

Evaluation of a device is performed in the same manner as in Example 1 except for using exemplified compound (1-21) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained. The results  
25 obtained are shown in Table 2.

#### EXAMPLE 13

Evaluation of a device is performed in the same manner as in Example 1 except for

using exemplified compound (1-22) in place of compound (1-1). It is confirmed that the emission of phosphorescence originating in compound B-1 is obtained. The results obtained are shown in Table 2.

## 5 EXAMPLE 14

Evaluation of a device is performed in the same manner as in Example 3 except for using compound B-4 in place of compound B-1. It is confirmed that the emission of blue phosphorescence originating in compound B-4 is obtained. The results obtained are shown in Table 2.

10

TABLE 2

Results of evaluation of devices prepared in Examples 8 to 14

Example No.	External Quantum Efficiency (relative value)	Half Life Time of Luminance (relative value)	Variation in Chromaticity at Half Life Time of Luminance (rate of change of CIE y value in relative value)
Example 8	2.0	2.9	0.3
Example 9	1.7	3.5	0.2
Example 10	2.0	2.3	0.2
Example 11	1.8	3.2	0.1
Example 12	2.0	1.5	0.2
Example 13	1.7	2.4	0.2
Example 14	1.5	3.2	0.2

\* The evaluation value in each example is a relative value to the evaluation value of Comparative Example 1 as the reference value.

15

From the results in Table 2, it can be seen that the light-emitting devices in the invention are excellent in efficiency and durability, and little in chromaticity variation.

## COMPARATIVE EXAMPLE 3

20

A cleaned ITO substrate is placed in a vacuum evaporator, copper phthalocyanine is deposited on the substrate in a thickness of 10 nm, and NPD (N,N'-di- $\alpha$ -naphthyl-N,N'-diphenyl)benzidine is deposited thereon in a thickness of 40 nm. Compound 12-1 and

compound A in the ratio of 5/95 (by mass) are deposited on the above deposited film in a thickness of 30 nm (a light-emitting layer), then BAlq is deposited thereon in a thickness of 6 nm, and then Alq (tris(8-hydroxyquinoline) aluminum complex) is deposited on the above film in a thickness of 20 nm. Lithium fluoride is deposited thereon in a thickness of 3 nm, followed by deposition of aluminum in a thickness of 60 nm to prepare an EL device. The obtained EL device is subjected to application of DC constant voltage with a source measure unit Model 2400 (manufactured by Toyo Technica Co., Ltd.) to emit light. It is confirmed that the emission of fluorescence originating in compound 12-1 is obtained.

The results obtained are shown in Table 3 below.

#### COMPARATIVE EXAMPLE 4

Preparation and evaluation of a device is performed in the same manner as in Comparative Example 1 except for using compound 12-2 in place of compound 12-1, and using compound B in place of compound A respectively in Comparative Example 3. It is confirmed that the emission of fluorescence originating in compound 12-2 is obtained.

The results of evaluation are shown in Table 3.

#### EXAMPLE 15

Evaluation of a device is performed in the same manner as in Comparative Example 1 except for using exemplified compound (1-1) of the invention in place of compound A in Comparative Example 3. It is confirmed that the emission of fluorescence originating in compound 12-1 is obtained.

The results of evaluation are shown in Table 3.

#### EXAMPLE 16

Evaluation of a device is performed in the same manner as in Comparative Example 1 except for using exemplified compound (1-6) of the invention in place of compound A in Comparative Example 3. It is confirmed that the emission of fluorescence originating in

compound 12-1 is obtained.

The results of evaluation are shown in Table 3.

#### EXAMPLE 17

5 Evaluation of a device is performed in the same manner as in Comparative Example 1 except for using exemplified compound (1-7) of the invention in place of compound A in Comparative Example 3. It is confirmed that the emission of fluorescence originating in compound 12-1 is obtained.

The results of evaluation are shown in Table 3.

10

#### EXAMPLE 18

Evaluation of a device is performed in the same manner as in Comparative Example 1 except for using exemplified compound (1-1) of the invention in place of compound B in Comparative Example 4. It is confirmed that the emission of fluorescence originating in  
15 compound 12-2 is obtained.

The results of evaluation are shown in Table 3.

#### EXAMPLE 19

Evaluation of a device is performed in the same manner as in Comparative Example  
20 1 except for using exemplified compound (1-6) of the invention in place of compound B in Comparative Example 4. It is confirmed that the emission of fluorescence originating in compound 12-2 is obtained.

The results of evaluation are shown in Table 3.

#### 25 EXAMPLE 20

Evaluation of a device is performed in the same manner as in Comparative Example 1 except for using exemplified compound (1-7) of the invention in place of compound B in Comparative Example 4. It is confirmed that the emission of fluorescence originating in

compound 12-2 is obtained.

The results of evaluation are shown in Table 3.

In Table 3, the device is driven at 20 °C by the application of constant current. The half life time of luminance is found by measuring the time required to reach the half life of  
 5 luminance from the initial luminance. The variation in chromaticity is measured at 20 °C.

TABLE 3

Results of evaluation of devices prepared in Comparative Examples 3 and 4 and Examples 15  
 to 20

Example No.	External Quantum Efficiency (relative value)	Half Life Time of Luminance (relative value)	Variation in Chromaticity at Half Life Time of Luminance (rate of change of CIE y value in relative value)
Comparative Example 3	1.0	1.0	1.0
Comparative Example 4	0.8	1.2	0.7
Example 15	1.2	1.9	0.4
Example 16	1.3	1.4	0.2
Example 17	1.2	1.7	0.5
Example 18	1.2	1.6	0.2
Example 19	1.1	1.8	0.4
Example 20	1.4	1.5	0.3

10 \* The evaluation value in each example is a relative value to the evaluation value of Comparative Example 1 as the reference value.

From the results in Table 3, it can be seen that the light-emitting devices in the invention are excellent in efficiency and durability, and little in chromaticity variation.

15 The same effects can be obtained with light-emitting devices using other compounds according to the invention.

#### EXAMPLE 21

Evaluation of a device is performed in the same manner as in Example 14 except for  
 20 using exemplified compound (1-3) in place of compound (1-6) in Example 14 and using compound B-5 in place of compound B-4 in Example 14. It is confirmed that the emission

of blue-green phosphorescence originating in compound B-5 is obtained. The results obtained are shown in Table 4 below.

#### EXAMPLE 22

5 Evaluation of a device is performed in the same manner as in Example 21 except for using exemplified compound (1-22) in place of compound (1-3) in Example 21 and using compound B-6 in place of compound B-5 in Example 21. It is confirmed that the emission of green phosphorescence originating in compound B-6 is obtained. The results obtained are shown in Table 4 below.

10

#### EXAMPLE 23

Evaluation of a device is performed in the same manner as in Example 21 except for using exemplified compound (1-19) in place of compound (1-3) in Example 21 and using compound B-7 in place of compound B-5 in Example 21. It is confirmed that the emission of green phosphorescence originating in compound B-7 is obtained. The results obtained are shown in Table 4 below.

15

#### EXAMPLE 24

Evaluation of a device is performed in the same manner as in Example 21 except for using exemplified compound (1-17) in place of compound (1-3) in Example 21 and using compound B-8 in place of compound B-5 in Example 21. It is confirmed that the emission of green phosphorescence originating in compound B-8 is obtained. The results obtained are shown in Table 4 below.

20

#### 25 EXAMPLE 25

Evaluation of a device is performed in the same manner as in Example 21 except for using exemplified compound (1-18) in place of compound (1-3) in Example 21 and using compound B-9 in place of compound B-5 in Example 21. It is confirmed that the emission

of red phosphorescence originating in compound B-9 is obtained. The results obtained are shown in Table 4 below.

#### EXAMPLE 26

5 A device is manufactured and evaluated in the same manner as in Comparative Example 1, except for inserting a layer containing exemplified compound (1-20) having a thickness of 3 nm between the NPD layer and the light-emitting layer in Comparative Example 1. The results obtained are shown in Table 4 below.

#### 10 EXAMPLE 27

A device is manufactured and evaluated in the same manner as in Comparative Example 1, except for inserting a layer containing exemplified compound (1-51) having a thickness of 3 nm between the light-emitting layer and the BA1q layer in Comparative Example 1. The results obtained are shown in Table 4 below.

15

**TABLE 4**

Evaluation of the devices in Examples 21 to 27

Example No.	External Quantum Efficiency (relative value)	Half Life Time of Luminance (relative value)	Variation in Chromaticity at Half Life Time of Luminance (rate of change of CIE y value in relative value)
Example 21	1.5	2.6	0.3
Example 22	1.9	2.8	0.3
Example 23	1.6	3.7	0.1
Example 24	1.6	3.1	0.2
Example 25	1.4	2.4	0.3
Example 26	1.9	2.0	0.2
Example 27	1.8	2.2	0.2

\* The evaluation value in each example is a relative value to the evaluation value of Comparative Example 1 as the reference value.

20

It can be seen from the results shown in Table 4 that the devices of the invention

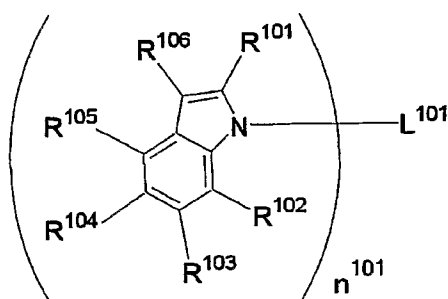
using the indole derivatives in the layers contiguous to the light-emitting layers are excellent in efficiency and durability and little in variation of chromaticity.

It will be apparent to those skilled in the art that various modifications and variations  
5 can be made to the described embodiments of the invention without departing from the spirit or scope of the invention. Thus, it is intended that the invention cover all modifications and variations of this invention consistent with the scope of the appended claims and their equivalents.

The present application claims foreign priority based on Japanese Patent Application  
10 Nos. JP2006-318772 and JP2007-221472, filed November 27, 2006 and August 28, 2007, respectively, the contents of which are incorporated herein by reference.

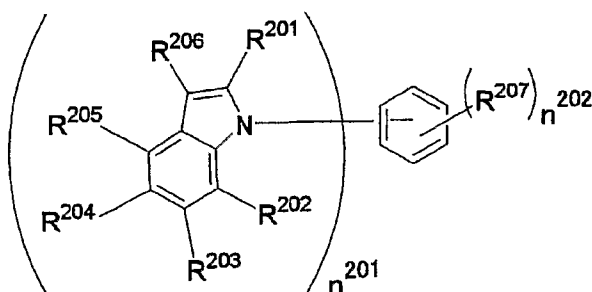
## CLAIMS

1. An organic electroluminescent device comprising:  
 a pair of electrodes; and  
 at least one organic layer between the pair of electrodes, the at least one organic layer including a light-emitting layer containing a light-emitting material,  
 wherein the at least one organic layer includes at least one layer containing an indole derivative represented by formula (1):



- 10 wherein  $R^{101}$ ,  $R^{102}$ ,  $R^{103}$ ,  $R^{104}$  and  $R^{105}$  each independently represents a hydrogen atom or a substituent;  $R^{106}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{101}$  and  $R^{106}$  may be bonded to each other to form a ring;  $L^{101}$  represents a linking group; and  $n^{101}$  represents an integer of 2 or higher.

- 15 2. The organic electroluminescent device according to claim 1, wherein the indole derivative is a compound represented by formula (2):

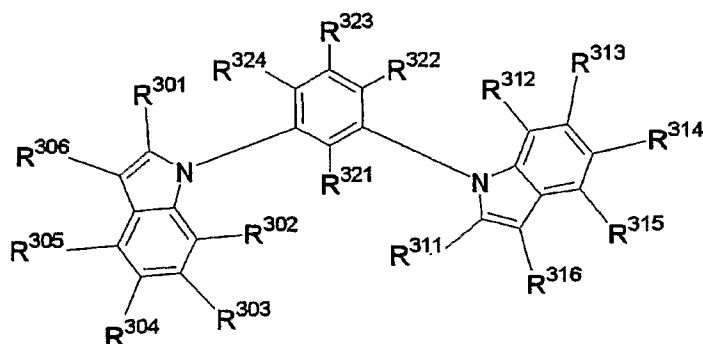


- wherein  $R^{201}$ ,  $R^{202}$ ,  $R^{203}$ ,  $R^{204}$  and  $R^{205}$  each independently represents a hydrogen atom or a

substituent;  $R^{206}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{201}$  and  $R^{206}$  may be bonded to each other to form a ring;  $R^{207}$  represents a substituent;  $n^{201}$  represents an integer of from 2 to 6; and  $n^{202}$  represents an integer of from 0 to 4, provided that  $n^{201} + n^{202} \leq 6$ .

5

3. The organic electroluminescent device according to claim 1, wherein the indole derivative is a compound represented by the following formula (3):



wherein  $R^{301}$ ,  $R^{302}$ ,  $R^{303}$ ,  $R^{304}$  and  $R^{305}$  each independently represents a hydrogen atom or a substituent;  $R^{306}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{301}$  and  $R^{306}$  may be bonded to each other to form a ring;  $R^{311}$ ,  $R^{312}$ ,  $R^{313}$ ,  $R^{314}$  and  $R^{315}$  each independently represents a hydrogen atom or a substituent;  $R^{316}$  represents an alkyl group having a tertiary or quaternary carbon atom;  $R^{311}$  and  $R^{316}$  may be bonded to each other to form a ring; and  $R^{321}$ ,  $R^{322}$ ,  $R^{323}$  and  $R^{324}$  each independently represents a hydrogen atom or a substituent.

15

4. The organic electroluminescent according to any one of claims 1 to 3, wherein the indole derivative is contained in the light-emitting layer.

5. The organic electroluminescent device according to any one of claims 1 to 4, wherein the indole derivative is contained in a layer contiguous to the light-emitting layer.

20

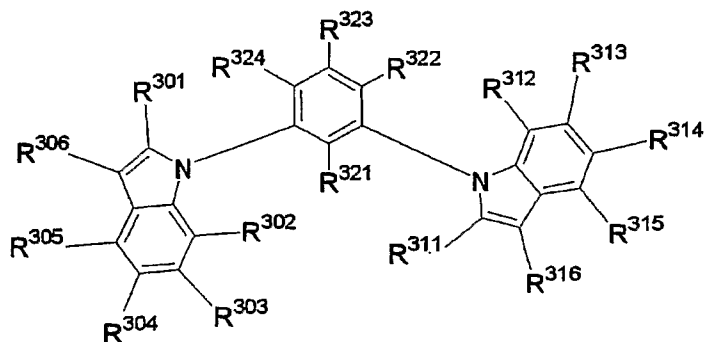
6. The organic electroluminescent device according to any one of claims 1 to 5,

wherein the light-emitting material is a phosphorescent material.

7. The organic electroluminescent device according to any one of claims 1 to 6, wherein the light-emitting material is a platinum complex.

5

8. A compound represented by formula (3):



wherein R<sup>301</sup>, R<sup>302</sup>, R<sup>303</sup>, R<sup>304</sup> and R<sup>305</sup> each independently represents a hydrogen atom or a substituent; R<sup>306</sup> represents an alkyl group having a tertiary or quaternary carbon atom; R<sup>301</sup> and R<sup>306</sup> may be bonded to each other to form a ring; R<sup>311</sup>, R<sup>312</sup>, R<sup>313</sup>, R<sup>314</sup> and R<sup>315</sup> each independently represents a hydrogen atom or a substituent; R<sup>316</sup> represents an alkyl group having a tertiary or quaternary carbon atom; R<sup>311</sup> and R<sup>316</sup> may be bonded to each other to form a ring; and R<sup>321</sup>, R<sup>322</sup>, R<sup>323</sup> and R<sup>324</sup> each independently represents a hydrogen atom or a substituent.

## INTERNATIONAL SEARCH REPORT

International application No  
PCT/JP2007/073279

A. CLASSIFICATION OF SUBJECT MATTER  
INV. C09K11/06 H01L51/50

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
C09K H01L

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

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

EPO-Internal, WPI Data

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2006/145145 A1 (NISHIO TETSUYA [JP]) 6 July 2006 (2006-07-06) claims 1,14	1,2,4-6
X	JP 2005 082701 A (TOYO INK MFG CO) 31 March 2005 (2005-03-31) compounds 4-6,10,16,18,21,23,25,26,28-31,33,34,41-43 ,45-47	1,4-7
X	JP 2005 002346 A (NISHIO TETSUYA) 6 January 2005 (2005-01-06) compounds 254,287,291	1,4-6
X	JP 2003 277744 A (IDEMITSU KOSAN CO) 2 October 2003 (2003-10-02) compounds A1-A6	1,4-6
	----- -/--	

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

☒ See patent family annex.

\* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
- \*L\* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

- \*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- \*X\* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- \*Y\* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- \*&\* document member of the same patent family

Date of the actual completion of the international search

11 February 2008

Date of mailing of the international search report

18/02/2008

Name and mailing address of the ISA/

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Saldamli, Saltuk

## INTERNATIONAL SEARCH REPORT

International application No

PCT/JP2007/073279

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 2007/063754 A (NIPPON STEEL CHEMICAL CO [JP]; KOMORI MASAKI [JP]; YAMAMOTO TOSHIHIRO) 7 June 2007 (2007-06-07) the whole document	1-8
X	US 2003/165712 A1 (LIN TUNG-SHEN [TW] ET AL) 4 September 2003 (2003-09-04) compounds 1-8	1-6, 8

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/JP2007/073279

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US 2006145145	A1	06-07-2006	NONE		
JP 2005082701	A	31-03-2005	NONE		
JP 2005002346	A	06-01-2005	NONE		
JP 2003277744	A	02-10-2003	JP	3985895 B2	03-10-2007
WO 2007063754	A	07-06-2007	NONE		
US 2003165712	A1	04-09-2003	NONE		