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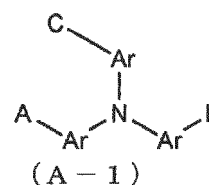
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(54) **ORGANIC EL ELEMENT, AMINE COMPOUND HAVING BENZAZOLE RING STRUCTURE, AND METHOD IN WHICH SAID AMINE COMPOUND IS USED IN CAPPING LAYER OF ORGANIC EL ELEMENT**

(57) An organic EL element having at least a positive electrode, a hole transport layer, a light-emitting layer, an electron transport layer, a negative electrode, and a capping layer in the stated order, wherein the capping layer contains an amine compound having a benzoazole ring structure represented by general formula (A-1).

[Chem. 44]



Description

Technical Field

5 **[0001]** The present invention relates to a compound and a device suitable for an organic electroluminescence device (hereinafter abbreviated as an organic EL device) which is a light emitting device suitable for various display devices, specifically relates to a compound having a benzazole ring structure, an organic EL device using the compound and a method of using the compound in an organic EL device.

10 Background Art

[0002] Since organic EL devices are self-luminescent devices, they are bright and excellent in visibility as compared with liquid-crystalline devices, and capable of giving clear display. Therefore, the organic EL devices have been actively studied.

15 **[0003]** In 1987, C. W. Tang et al. of Eastman Kodak Company put an organic EL device using organic materials into practical use by developing a device having a multilayered structure in which various roles are assigned to respective materials. They formed a lamination of a fluorescent material capable of transporting electrons and an organic material capable of transporting holes, so that both charges are injected into the layer of the fluorescent material to emit light, thereby achieving a high luminance of 1,000 cd/m² or more at a voltage of 10 V or lower (see, for example, Patent

20 Literatures 1 and 2).
[0004] To date, many improvements have been performed for practical utilization of the organic EL devices. And in an electroluminescent device in which an anode, a hole injection layer, a hole transport layer, a light emitting layer, an electron transport layer, an electron injection layer, and a cathode are sequentially provided on a substrate, to further subdivide various roles of the multilayered structure, high efficiency and high durability have been achieved by a light emitting device having a bottom emission structure that emits light from a bottom portion (see, for example, Non-Patent Literature 1).

25 **[0005]** In recent years, a light emitting device having a top emission structure that uses a metal having a high work function as an anode and emits light from an upper portion has been used. In the bottom emission structure in which light is extracted from a bottom portion having a pixel circuit, the area of the light emitting portion is limited. On the other hand, in the light emitting device having the top emission structure, since light is extracted from the upper portion, there is an advantage that the light emitting portion can be widened without blocked by the pixel circuit. In the light emitting device having a top emission structure, a translucent electrode such as LiF/Al/Ag (see, for example, Non-Patent Literature 2), Ca/Mg (see, for example, Non-Patent Literature 3), or LiF/MgAg is used as a cathode.

30 **[0006]** In such a light emitting device, in the case where light emitted on the light emitting layer is incident on another film, when incident at a certain angle or above, light is totally reflected at an interface between the light emitting layer and the other film. Therefore, only a part of the emitted light was available. In recent years, in order to improve light extraction efficiency, a light emitting device in which a "capping layer" having a high refractive index is provided outside the translucent electrode having a low refractive index has been proposed (see, for example, Non-Patent Literatures 2 and 3).

35 **[0007]** An effect of the capping layer in the light emitting device having a top emission structure is: in the light emitting device using Ir(ppy)₃ as a light emitting material, current efficiency was 38 cd/A in the case where there was no capping layer; whereas in a light emitting device using ZnSe having a film thickness of 60 nm as the capping layer, current efficiency was 64 cd/A and efficiency improvement of about 1.7 times was recognized. It has been also shown that a maximum point of transmittance of the translucent electrode and the capping layer does not necessarily coincide with a maximum point of efficiency, and it has been shown that a maximum point of light extraction efficiency is determined by an interference effect (see, for example, Non-Patent Literature 3).

40 **[0008]** Conventionally, it has been proposed to use a metal mask having high definition to form of the capping layer, but in use under high temperature conditions, there is a problem that alignment accuracy decreases since distortion due to heat occurs in the metal mask. Therefore, since ZnSe has a melting point as high as 1,100°C or higher (see, for example, Non-Patent Literature 3), it cannot be deposited at a correct position by a metal mask having high definition, and thus may affect the light emitting device itself. Further, since film formation by a sputtering method also affects the light emitting device, the capping layer containing an inorganic substance as a constituent material is not suitable for use.

45 **[0009]** In addition, in the case where tris(8-hydroxyquinoline) aluminum (hereinafter abbreviated as Alq₃) is used as the capping layer for adjusting a refractive index (see, for example, Non-Patent Literature 2), Alq₃ is known as an organic EL material commonly used as a green light emitting material or an electron transport material, but has weak absorption near 450 nm used as a blue light emitting material, so that there is a problem of reduction in color purity and reduction in light extraction efficiency in the case of a blue light emitting device.

50 **[0010]** In a device produced by using a conventional capping layer, since among sunlight, light having a wavelength

of 400 nm to 410 nm may pass therethrough and affect a material inside the device, there is also a problem of reduction in color purity and reduction in light extraction efficiency.

[0011] In order to improve device characteristics of the organic EL device, in particular, in order to absorb light having a wavelength of 400 nm to 410 nm among sunlight not to affect the material inside the device and considerably improve light extraction efficiency, a material having a high light absorption coefficient, a high refractive index, and excellent stability or durability of a thin film is demanded as a material of the capping layer.

Citation List

Patent Literature

[0012]

Patent Literature 1: JP-A-H08-048656

Patent Literature 2: Japanese Patent No. 3194657

Patent Literature 3: WO 2014/009310

Patent Literature 4: WO 2013/038627

Non-Patent Literature

[0013]

Non-Patent Literature 1: Japan Society of Applied Physics, 9th Class, Proceedings, Pages 55 to 61 (2001)

Non-Patent Literature 2: Appl. Phys. Lett., 78, 544 (2001)

Non-Patent Literature 3: Appl. Phys. Lett., 82, 466 (2003)

Non-Patent Literature 4: Aust. J. Chem., 45, 371 (1992)

Non-Patent Literature 5: J. Org. Chcm., 60, 7508 (1995)

Non-Patent Literature 6: Synth. Commun., 11, 513 (1981)

Non-Patent Literature 7: Appl. Phys. Lett., 98, 083302 (2011)

Summary of Invention

Technical Problem

[0014] In order to improve device characteristics of an organic EL device, in particular, in order to absorb light having a wavelength of 400 nm to 410 nm among sunlight not to affect the material inside the device and considerably improve light extraction efficiency, an object of the present invention is to provide an organic EL device including a capping layer constituted by a material having (1) a high light absorption coefficient, (2) a high refractive index, (3) good stability in a thin film, (4) excellent durability, (5) excellent light resistance, and (6) no absorption in each wavelength range of blue, green and red.

[0015] Physical characteristics of the material of the capping layer suitable for the present invention can include (1) a high light absorption coefficient, (2) a high refractive index, (3) capable of deposition, (4) a stable thin film state, and (5) a high glass transition temperature. Physical characteristics of the device suitable for the present invention can include (1) absorption of light from 400 nm to 410 nm, (2) high light extraction efficiency, (3) no reduction in color purity, (4) transmission of light without change over time, and (5) a long lifetime.

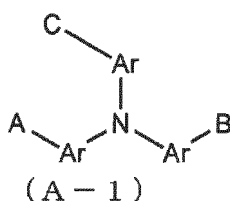
Solution to Problem

[0016] Therefore, in order to achieve the above object, the present inventors focused on the fact that aryl amine materials have excellent stability and durability of a thin film, and screened materials having a high absorbance at a wavelength of 400 nm to 410 nm in an absorption spectrum of a concentration of 10^{-5} mol/L regarding amine compounds having a specific benzazole ring structure with a high refractive index. They produced organic EL devices using the materials as a material constituting the capping layer, and characteristics of the device were evaluated earnestly. As a result, they have completed the present invention.

[0017] That is, according to the present invention, the following organic EL device is provided.

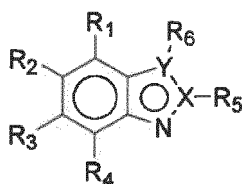
[0018] An organic EL device having at least an anodic electrode, a hole transport layer, a light emitting layer, an electron transport layer, a cathodic electrode, and a capping layer in this order, in which the capping layer contains an amine compound having a benzazole ring structure represented by the following general formula (A-1).

[Chem. 1]



[0019] (In the formula, A represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site; B and C may be the same as or different from each other, and each represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group; Ar's may be the same as or different from each other, and each represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group, or a single bond. Ar's adjacent to each other via a nitrogen atom may be bonded by a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring.)

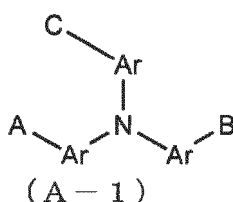
[Chem. 2]



[0020] (In the formula, R_1 to R_6 may be the same as or different from each other, and each represents a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkyloxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyloxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom. However, in the case where X and Y are nitrogen atoms, X or Y does not have R_5 or R_6 , and the case where X and Y are carbon atoms is excepted.)

[0021] According to the present invention, there is provided an amine compound having a benzazole ring structure represented by the following general formula (A-1).

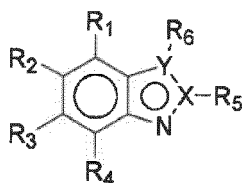
[Chem. 3]



[0022] (In the formula, A represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site; B and C may be the same as or different from each other, and each represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group,

or a substituted or unsubstituted condensed polycyclic aromatic group; Ar's may be the same as or different from each other, and each represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group, or a single bond. Ar's adjacent to each other via a nitrogen atom may be bonded by a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring.)

[Chem. 4]



(B - 1)

[0023] (In the formula, R_1 to R_6 may be the same as or different from each other, and each represents a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom. However, in the case where X and Y are nitrogen atoms, X or Y does not have R^5 or R^6 , and the case where X and Y are carbon atoms is excepted.)

Advantageous Effects of Invention

[0024] Since the organic EL device of the present invention has a capping layer which is provided outside the transparent or translucent electrode and has a refractive index higher than that of the translucent electrode, it is possible to obtain an organic EL device capable of significantly improving light extraction efficiency. In addition, since an amine compound having a benzazole ring compound represented by the general formula (A-1) is used in the capping layer, film formation can be performed at a temperature of not higher than 400°C, and thus the present invention can optimize light extraction efficiency of each color by using a high definition mask without damaging the light emitting device, can be suitably applied to a full color display, and can display a clear and bright image with high color purity.

[0025] Since the organic EL device of the present invention uses, as a material of the capping layer, an organic EL device material having a high light absorption coefficient, a high refractive index, excellent stability, durability and light resistance of a thin film, it is possible to maintain color purity and significantly improve light extraction efficiency without being affected by sunlight, as compared with a conventional organic EL device. Further, it is possible to realize an organic EL device having high efficiency and a long life.

Brief Description of Drawing

[0026] [FIG. 1] This is a diagram illustrating an organic EL device configuration of Examples 17 to 29 and Comparative Examples 1 and 2.

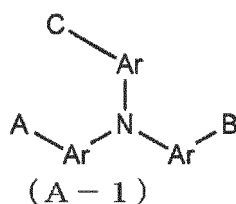
Description of Embodiments

[0027] An embodiment of the present invention will be described in detail below. First, aspects of the present embodiment will be listed and described. In the present application, a term "to" is a term representing a range. For example, description of "5 to 10" means "5 or more and 10 or less", and represents a range including numerical values themselves described before and after "to".

1) An organic EL device having at least an anodic electrode, a hole transport layer, a light emitting layer, an electron

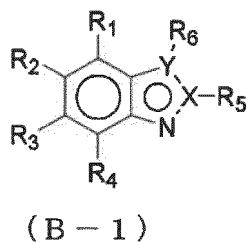
transport layer, a cathodic electrode, and a capping layer in this order, in which the capping layer contains an amine compound having a benzazole ring structure represented by the following general formula (A-1).

[Chem. 5]



(In the formula, A represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site; B and C may be the same as or different from each other, and each represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group; Ar's may be the same as or different from each other, and each represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group or a single bond. Ar's adjacent to each other via a nitrogen atom may be bonded by a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring.)

[Chem. 6]

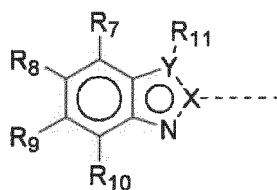


(In the formula, R_1 to R_6 may be the same as or different from each other, and each represents a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkyloxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyloxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom. However, in the case where X and Y are nitrogen atoms, X or Y does not have R^5 or R^6 , and the case where X and Y are carbon atoms is excepted.)

2) The organic EL device according to 1), in which the amine compound having a benzazole ring structure has an extinction coefficient of 0.20 or more at a wavelength of 400 nm to 410 nm and has an absorbance of 0.20 or more at a wavelength of 400 nm to 410 nm in an absorption spectrum of a toluene solution with a concentration of 1.0×10^{-5} mol/L.

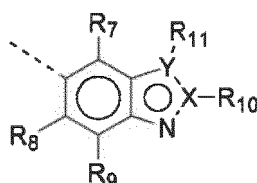
3) The organic EL device according to 1), in which the general formula (B-1) is represented by the following general formula (B-2) or (B-3).

[Chem. 7]



(B - 2)

[Chem. 8]

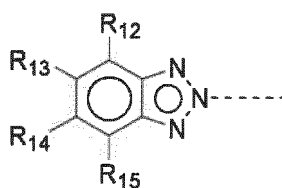


(B - 3)

(In the formulae, a broken line is a bonding site, R_7 to R_{11} may be the same as or different from each other, and each represents a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom. However, in the case where X and Y are nitrogen atoms, Y does not have R_{11} , and the case where X and Y are carbon atoms is excepted.)

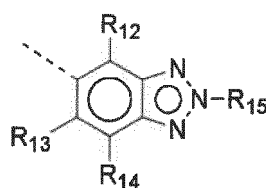
4) The organic EL device according to 1), in which the general formula (B-1) is represented by the following general formula (B-4) or (B-5).

[Chem. 9]



(B - 4)

[Chem. 10]



(B - 5)

(In the formulae, a broken line is a bonding site, R_{12} to R_{15} may be the same as or different from each other, and each represents a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group.)

5) The organic EL device according to any one of 1) to 4), in which A, B and C each have a benzazole ring structure in the general formula (A-1).

6) The organic EL device according to any one of 1) to 4), in which in the general formula (A-1), A and B each have a benzazole ring structure, and C represents a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group.

7) The organic EL device according to any one of 1) to 4), in which in the general formula (A-1), A has a benzazole ring structure, and B and C each represents a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group.

8) The organic EL device according to 1), in which A, B and C are the same as each other in the general formula (A-1).

9) The organic EL device according to 5) or 6), in which A and B are the same as each other in the general formula (A-1).

10) The organic EL device according to any one of 5) to 7), in which A, B and C are not the same as each other in the general formula (A-1).

11) The organic EL device according to any one of 5) to 7), in which A and B are not the same as each other in the general formula (A-1).

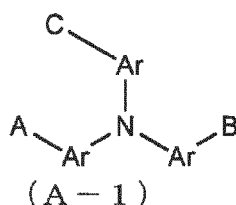
12) The organic EL device according to 1), in which the capping layer has a thickness in a range of 30 nm to 120 nm.

13) The organic EL device according to 1), in which the capping layer has a refractive index of 1.85 or more with respect to light having a wavelength in a range of 400 nm to 410 nm.

14) A method of using an amine compound having a benzotriazole ring structure where A or B in the general formula (A-1) as described in 1) is represented by the general formula (B-4) or (B-5) as described in 4), in a capping layer of an organic EL device.

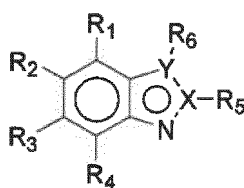
15) An amine compound having a benzazole ring structure represented by the following general formula (A-1).

[Chem. 11]



[0028] (In the formula, A represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site; B and C may be the same as or different from each other, and each represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group; Ar's may be the same as or different from each other, and each represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group, or a single bond. Ar's adjacent to each other via a nitrogen atom may be bonded by a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring.)

[Chem. 12]



(B - 1)

[0029] (In the formula, R_1 to R_6 may be the same as or different from each other, and each represents a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom. However, in the case where X and Y are nitrogen atoms, X or Y does not have R_5 or R_6 , and the case where X and Y are carbon atoms is excepted.)

[0030] The "aromatic hydrocarbon group", the "aromatic heterocyclic group" or the "condensed polycyclic aromatic group" in the "substituted or unsubstituted aromatic hydrocarbon group", the "substituted or unsubstituted aromatic heterocyclic group" or the "substituted or unsubstituted condensed polycyclic aromatic group", which is represented by B, C, Ar, and R_1 to R_{15} in the general formulae (A-1), (B-1), (B-2), (B-3), (B-4), and (B-5), is specifically selected from a phenyl group, a biphenyl group, a terphenyl group, a naphthyl group, an anthracenyl group, a phenanthrenyl group, a fluorenyl group, a spirobifluorenyl group, an indenyl group, a pyrenyl group, a perylenyl group, a fluoranthenyl group, a triphenylenyl group, a pyridyl group, a pyrimidinyl group, a thioridinyl group, a furyl group, a pyrrolyl group, a thienyl group, a quinolyl group, an isoquinolyl group, a benzofuranyl group, a benzothienyl group, an indolyl group, a carbazolyl group, a benzoxazolyl group, a benzothiazolyl group, a quinoxalyl group, a benzoimidazolyl group, a pyrazolyl group, a dibenzofuranyl group, a dibenzothienyl group, a naphthyridinyl group, a phenanthrolinyl group, an acridinyl group, a carborinyl group, and the like, and an aryl group having a carbon number of 6 to 30 or a heteroaryl group having a carbon number of 2 to 20.

[0031] As the "linear or branched alkyl group having a carbon number of 1 to 6", the "cycloalkyl group having a carbon number of 5 to 10", the "linear or branched alkenyl group having a carbon number of 2 to 6", the "linear or branched alkoxy group having a carbon number of 1 to 6", the "cycloalkoxy group having a carbon number of 5 to 10", or the "aryloxy group" in the "linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent", the "cycloalkyl group having a carbon number of 5 to 10 which may have a substituent", the "linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent", the "linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent", the "cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent", or the "substituted or unsubstituted aryloxy group", which is represented by R_1 to R_{15} in the general formulae (B-1), (B-2), (B-3), (B-4), and (B-5), specific examples thereof include a methyl group, an ethyl group, an n-propyl group, an isopropyl group, an n-butyl group, an isobutyl group, a tert-butyl group, an n-pentyl group, an isopentyl group, a neopentyl group, an n-hexyl group, and a cyclopentyl group, a cyclohexyl group, a 1-adamantyl group, a 2-adamantyl group, a vinyl group, an allyl group, an isopropenyl group, a 2-butenyl group, a methoxy group, an ethoxy group, an n-propoxy group, a cyclopentyloxy group, a cyclohexyloxy group, a 1-adamantyloxy group, a phenoxy group, a tolyloxy group, a biphenyloxy group, and the like.

[0032] As the "substituent" in the "substituted aromatic hydrocarbon group", the "substituted aromatic heterocyclic group", the "substituted condensed polycyclic aromatic group", the "linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent", the "cycloalkyl group having a carbon number of 5 to 10 which may have a substituent", the "linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent", the "linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent", the "cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent", the "substituted aryloxy group", or the "substituted methylene group", which is represented by B, C, Ar, and R_1 to R_{15} in the general formulae (A-1), (B-1), (B-2), (B-3), (B-4), and (B-5), specific examples thereof include a deuterium atom, a cyano group, a nitro group; halogen atoms such as a fluorine atom, a chlorine atom, a bromine atom, and an iodine atom; silyl groups such as a trimethyl silyl group and a triphenyl silyl group; a linear or branched alkyl group having a carbon number of 1 to 6 such as a methyl group, an ethyl group, and a propyl group; a linear or branched alkoxy group having a carbon number of 1 to 6 such as a methoxy

group, an ethyloxy group, and a propyloxy group; alkenyl groups such as a vinyl group and an allyl group; aryloxy groups such as a phenoxy group and a tolyloxy group; aryl alkyloxy groups such as a benzyloxy group and a phenethyloxy group; aromatic hydrocarbon groups or condensed polycyclic aromatic groups such as a phenyl group, a biphenyl group, a terphenyl group, a naphthyl group, an anthracenyl group, a phenanthrenyl group, a fluorenyl group, a spirobifluorenyl group, an indenyl group, a pyrenyl group, a perylenyl group, a fluoranthenyl group, and a triphenylenyl group; a pyridyl group, a thienyl group, a furyl group, a pyrrolyl group, a quinolyl group, an isoquinolyl group, a benzofuranyl group, a benzothienyl group, an indolyl group, a carbazolyl group, a benzoxazolyl group, and a benzothiazolyl group, a quinoxaliny group, a benzoimidazolyl group, a pyrazolyl group, a dibenzofuranyl group, a dibenzothienyl group, and a carborinyl group, and an aryl group having a carbon number of 6 to 30 or a heteroaryl group having a carbon number of 2 to 20, and these substituents may be further substituted with the exemplified substituents. In addition, the substituent and the substituted benzene ring or plurality of substituents substituted with the same benzene ring may be bonded to each other via a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring.

[0033] In the organic EL device of the present embodiment, Ar in the general formula (A-1) is preferably a substituted or unsubstituted aromatic hydrocarbon group, and more preferably a substituted or unsubstituted phenyl group.

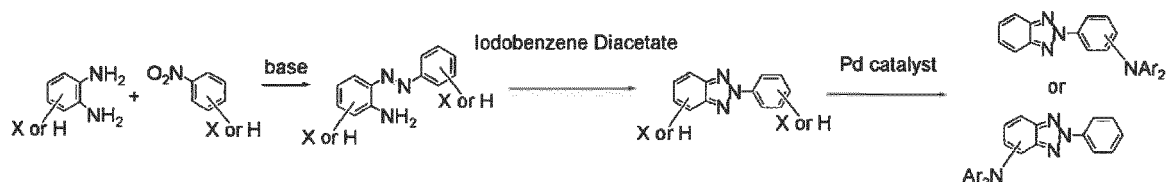
[0034] Further, in the organic EL device of the present embodiment, the thickness of the capping layer is preferably in a range of 30 nm to 120 nm and more preferably in a range of 40 nm to 80 nm.

[0035] In the organic EL device of the present embodiment, the refractive index of the capping layer with respect to light having a wavelength in a range of 400 nm to 410 nm is preferably 1.85 or more and more preferably 1.90 or more.

[0036] In the organic EL device of the present embodiment, the capping layer may be produced by laminating or mixing two or more kinds of different constituent materials.

[0037] The amine compound having the benzazole ring structure represented by the general formula (A-1) of the present embodiment is a new compound, and a benzazole derivative which is a main skeleton of these compounds can be synthesized by a per se known method, for example, as follows (for example, see Non-Patent Literature 4). Further, by performing a coupling reaction of the synthesized halogenated benzazole derivative and an aryl amine by using a copper catalyst, a palladium catalyst or the like, the amine compound having a benzazole ring structure represented by the general formula (A-1) of the present embodiment can be synthesized. In addition, also by converting the halogenated benzazole derivative to a boronic acid derivative or a boronic acid ester derivative, the amine compound having a benzazole ring structure represented by the general formula (A-1) of the present embodiment can be synthesized by a coupling reaction with a halogenated aryl amine (see, for example, Non-Patent Literatures 5 and 6).

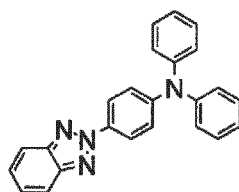
[Chem. 13]



[0038] Specific examples of a preferable compound among benzazole compounds represented by the general formula (A-1), which are suitably used in the organic EL device of the present embodiment, are shown below, but the present invention is not limited to these compounds.

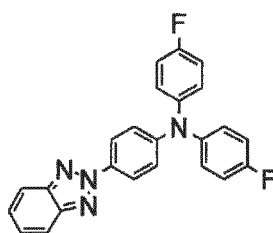
[Chem. 14]

5

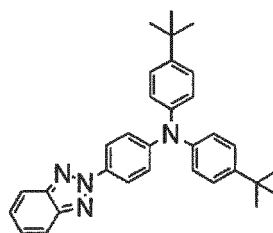


(Compound 1)

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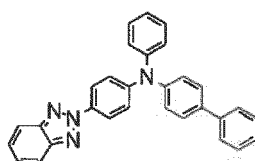


(Compound 2)



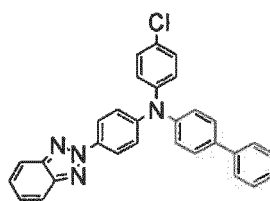
(Compound 3)

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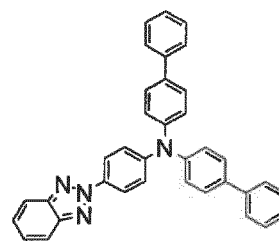


(Compound 4)

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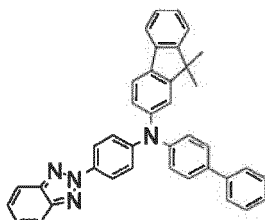


(Compound 5)



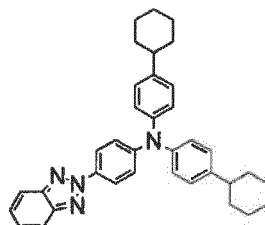
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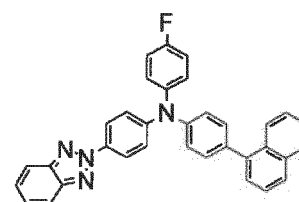


(Compound 7)

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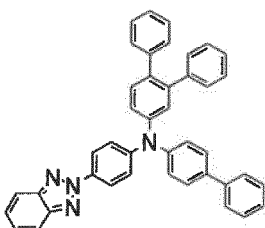


(Compound 8)



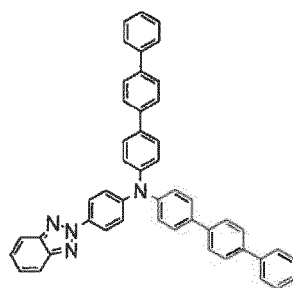
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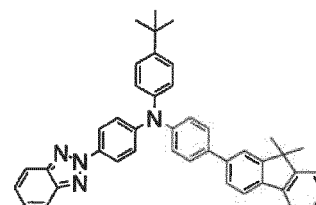


(Compound 10)

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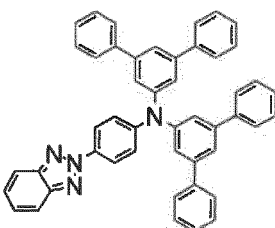


(Compound 11)



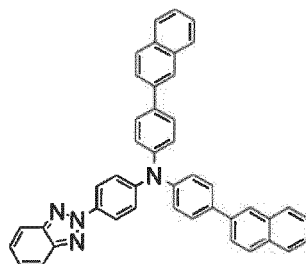
(Compound 12)

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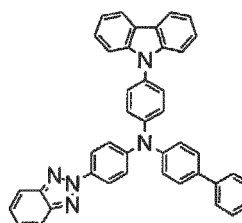


(Compound 13)

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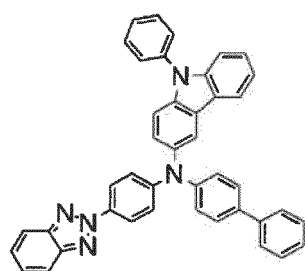
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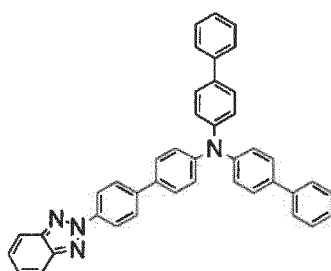
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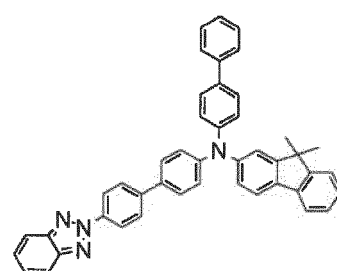
[Chem. 15]



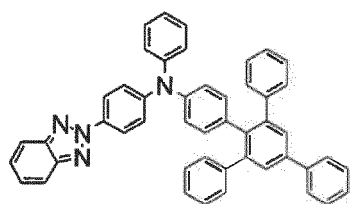
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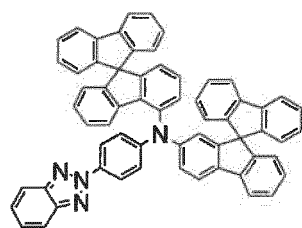
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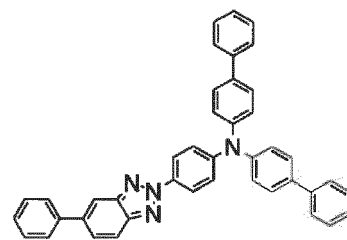
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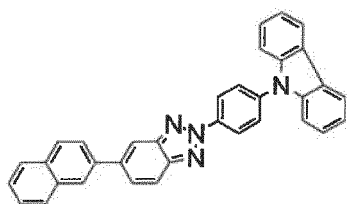
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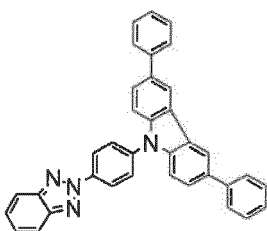
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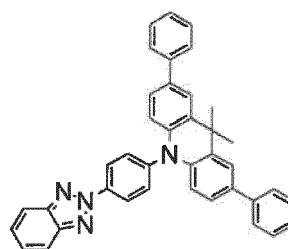
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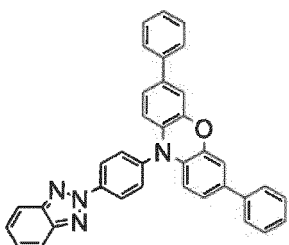
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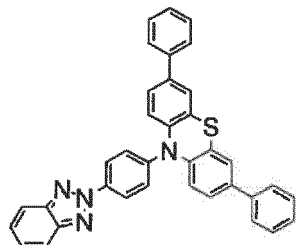
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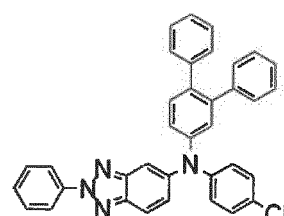
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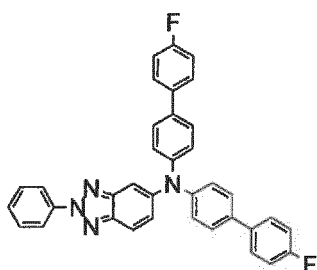
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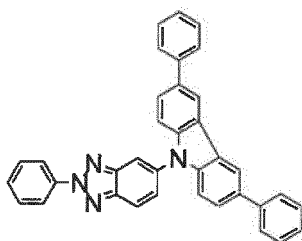
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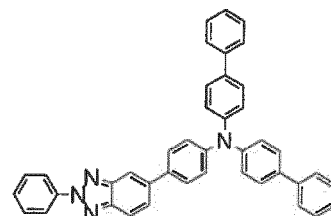
(Compound 27)



(Compound 28)

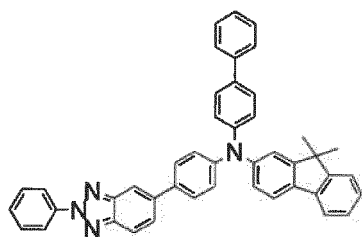


(Compound 29)

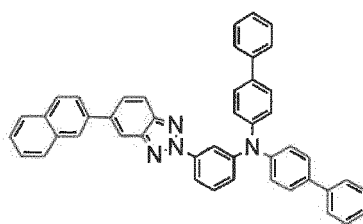


(Compound 30)

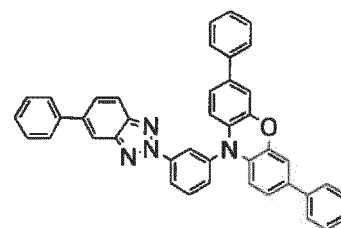
[Chem. 16]



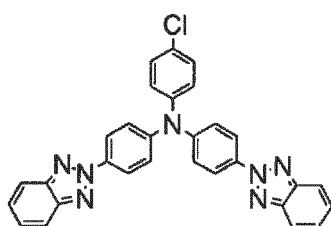
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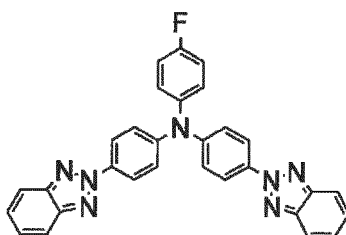
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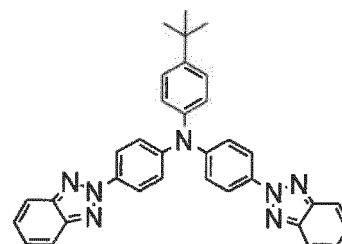
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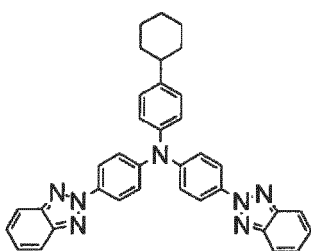
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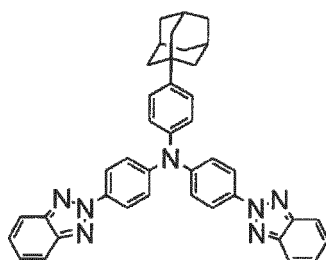
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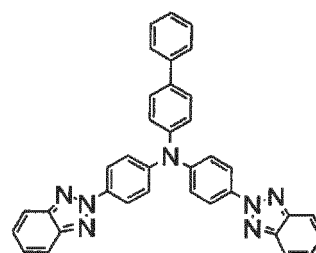
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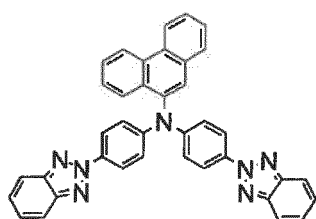
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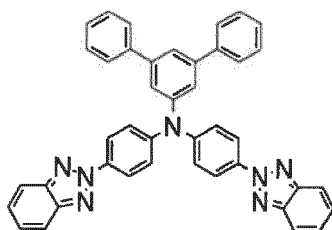
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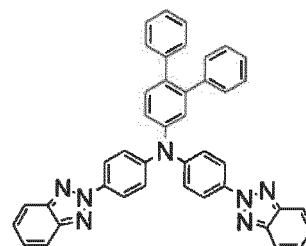
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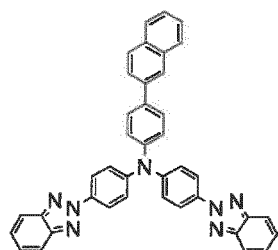
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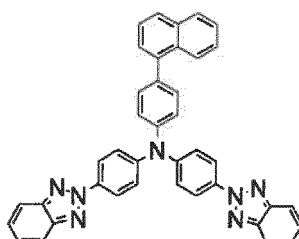
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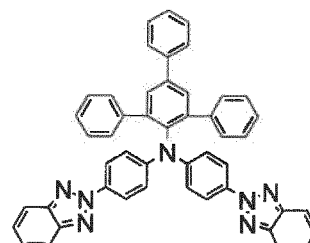
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(Compound 43)

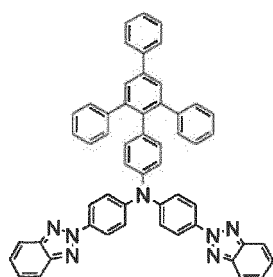


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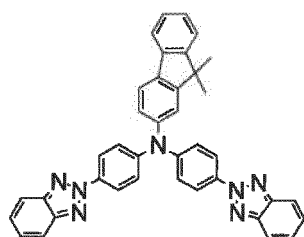


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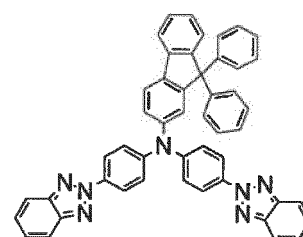
[Chem. 17]



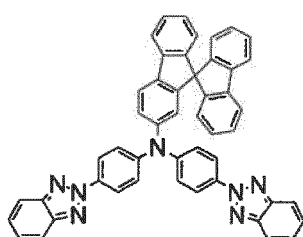
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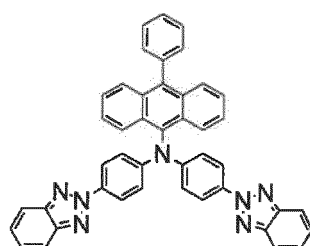
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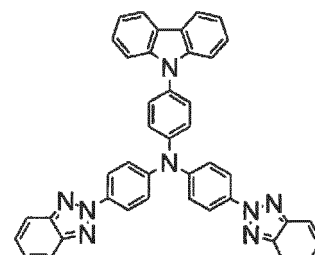
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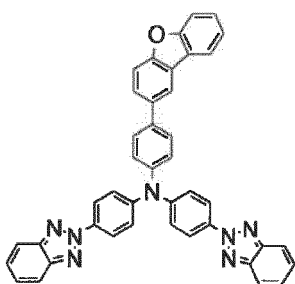
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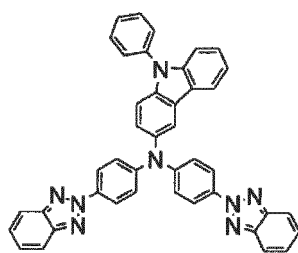
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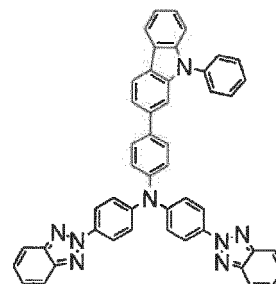
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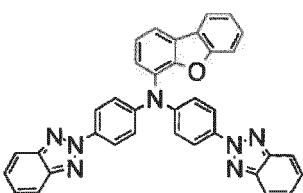
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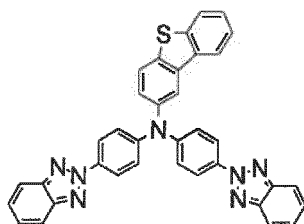
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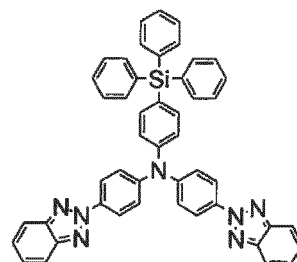
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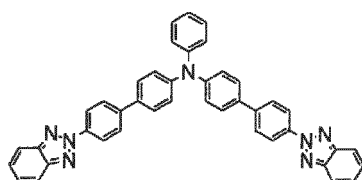
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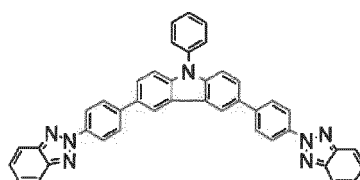
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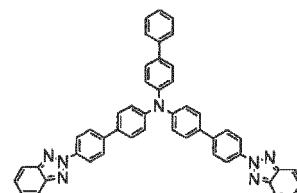
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(Compound 58)

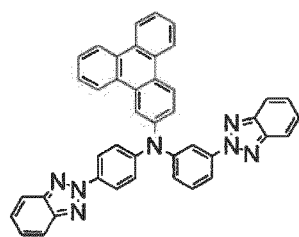


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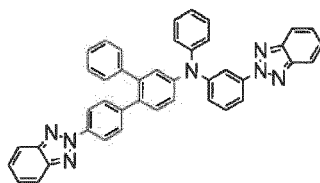


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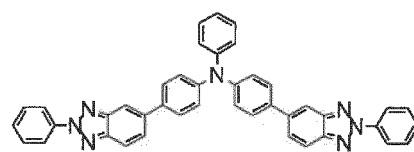
[Chem. 18]



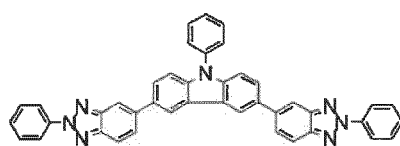
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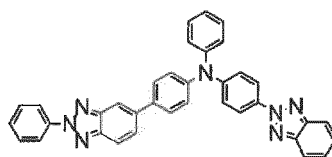
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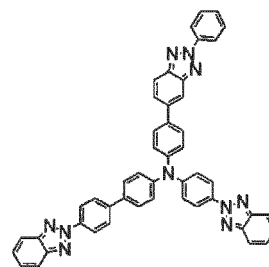
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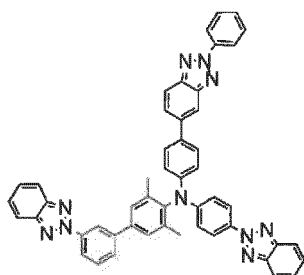
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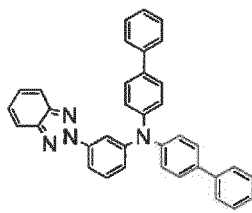
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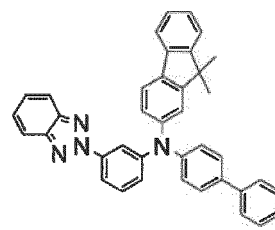
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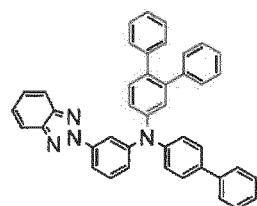
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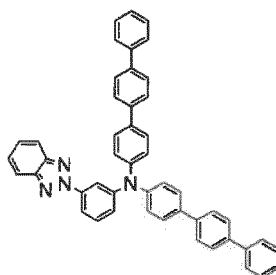
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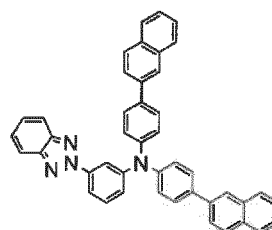
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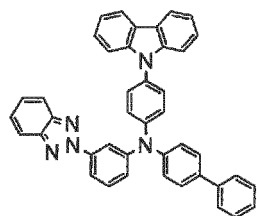
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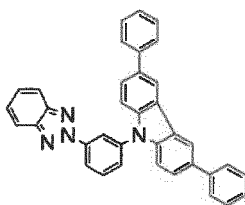
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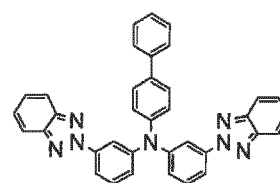
(Compound 72)



(Compound 73)

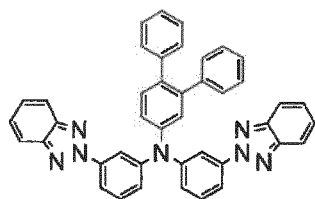


(Compound 74)

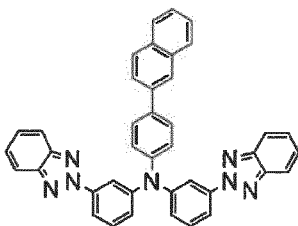


(Compound 75)

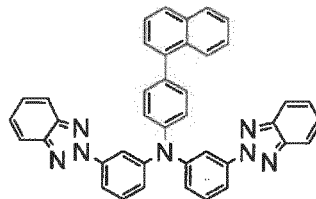
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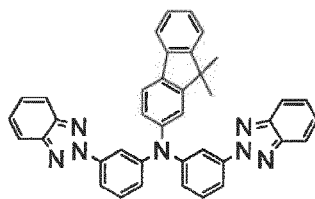
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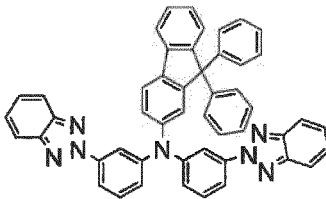
(Compound 77)



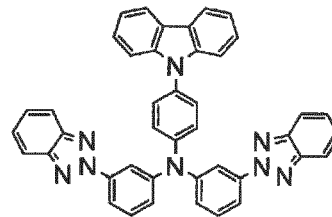
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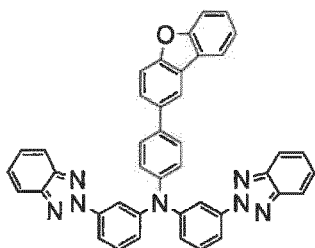
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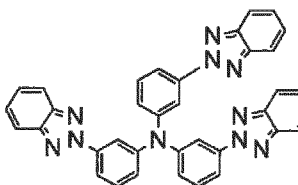
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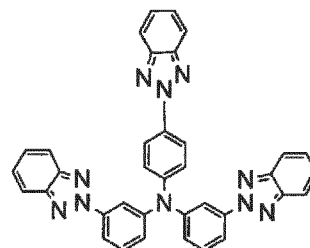
(Compound 81)



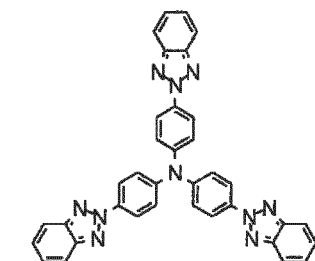
(Compound 82)



(Compound 83)



(Compound 84)



(Compound 85)

[0039] Purification of the compound represented by the general formula (A-1) is performed by purification by column chromatography, adsorption purification using silica gel, activated carbon, activated white clay or the like, recrystallization or crystallization using a solvent, sublimation purification, or the like. Identification of the compound is performed by NMR analysis. As physical property values, measurements of a melting point, a glass transition point (T_g) and a refractive index are performed in the following procedures. The melting point gives an index of deposition property, the glass transition point (T_g) gives an index of stability of a thin film, and the refractive index gives an index related to improvement in light extraction efficiency.

[0040] The melting point and the glass transition point (T_g) are measured with a high sensitivity differential scanning calorimeter (DSC 3100 SA manufactured by Bruker AXS GmbH) by using a powder of the compound represented by the general formula (A-1).

[0041] The refractive index and the extinction coefficient are measured by producing the thin film of the compound represented by the general formula (A-1) of 80 nm on a silicon substrate and using a spectroscopic measurement apparatus (F10-RT-UV manufactured by Filmetrics Japan, Inc.).

[0042] The absorbance is measured by using an ultraviolet-visible near-infrared spectrophotometer (V-650 manufac-

tured by JASCO Corporation) by adjusting a toluene solution containing the compound represented by the general formula (A-1) to a concentration of 1.0×10^{-5} mol/L. The absorption coefficient is calculated from a calibration curve obtained by adjusting a concentration of the toluene solution to four kinds of concentrations of 5.0×10^{-6} mol/L, 1.0×10^{-5} mol/L, 1.5×10^{-5} mol/L, and 2.0×10^{-5} mol/L, and measuring the absorbance at a peak wavelength with an ultraviolet-visible near-infrared spectrophotometer (V-650 manufactured by JASCO Corporation).

[0043] Examples of the structure of the organic EL device of the present embodiment include, in the case of a light emitting device having a top emission structure: a structure including an anode, a hole transport layer, a light emitting layer, an electron transport layer, a cathode, and a capping layer in this order on a glass substrate; a structure further including a hole injection layer between the anode and the hole transport layer; a structure further including an electron blocking layer between the hole transport layer and the light emitting layer; a structure further including a hole blocking layer between the light emitting layer and the electron transport layer; and a structure further including an electron injection layer between the electron transport layer and the cathode. In these multilayer structures, some of organic layers can be omitted or combined, and examples thereof can include: a configuration in which the hole injection layer and the hole transport layer are combined; a configuration in which the hole transport layer and the electron blocking layer are combined; a configuration in which the hole blocking layer and the electron transport layer are combined; a configuration in which the electron transport layer and the electron injection layer are combined, and the like. A configuration in which two or more organic layers having the same function are laminated can be also used, which can include a configuration in which two hole transport layers are laminated; a configuration in which two light emitting layers are laminated; a configuration in which two electron transport layers are laminated; a configuration in which two capping layers are laminated, or the like.

[0044] A total thickness of layers of the organic EL device is preferably about 200 nm to 750 nm and more preferably about 350 nm to 600 nm. A thickness of the capping layer is preferably 30 nm to 120 nm and more preferably 40 nm to 80 nm. In this case, good light extraction efficiency can be obtained. The thickness of the capping layer can be appropriately changed in accordance with the type of the light emitting material used for the light emitting device, the thickness of the organic EL device other than the capping layer, and the like.

[0045] As an anode of the organic EL device of the present embodiment, an electrode material having a large work function, such as ITO or gold is used.

[0046] As the hole injection layer of the organic EL device of the present embodiment, use can be made of an aryl amine compound having a structure in which three or more triphenyl amine structures are linked by a single bond or a divalent group containing no hetero atom in the molecule, such as star burst type triphenyl amine derivatives, a material such as various triphenyl amine tetramers, a porphyrin compound represented by copper phthalocyanine, and a heterocyclic compound having acceptor properties such as hexacyano azatriphenylene, a coating type polymer material, and the like. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used in a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed. These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0047] As the hole transport layer of the organic EL device of the present embodiment, use can be made preferably of benzidine derivatives such as N,N'-diphenyl-N,N'-di(m-tolyl) benzidine (hereinafter abbreviated as TPD), N,N'-diphenyl-N,N'-di(α -naphthyl) benzidine (hereinafter referred to as NPD) and N,N,N',N'-tetrabiphenyl benzidine, 1,1-bis[4-(di-4-tolyl amino) phenyl] cyclohexane (hereinafter abbreviated as TAPC), in particular, aryl amine compound having a structure in which two triphenyl amine structures are linked by a single bond or a divalent group containing no hetero atom in the molecule, such as N,N,N',N'-tetrabiphenyl benzidine, and the like. In addition, it is preferable to use an aryl amine compound having a structure in which three or more triphenyl amine structures are linked with a single bond or a divalent group containing no hetero atom in the molecule, such as various kinds of triphenyl amine trimer and tetramer, and the like. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used in a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed. As a hole injection and transport layer, a coating type polymer material such as poly(3,4-ethylene dioxythiophene) (hereinafter abbreviated as PEDOT)/poly(styrene sulfonate) (hereinafter abbreviated as PSS) can be used. These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0048] In addition, in the hole injection layer or the hole transport layer, a material that is generally used for these layers and is further P-doped with trisbromo phenyl amine hexachloro antimony, a radialene derivative (see, for example, Patent Literature 3), or the like, and a polymer compound having a structure of a benzidine derivative such as TPD in a partial structure thereof can be used.

[0049] As the electron blocking layer of the organic EL device of the present embodiment, use can be made of a compound having an electron blocking action, such as carbazole derivatives such as 4,4',4"-tri(N-carbazolyl) triphenyl amine (hereinafter referred to as TCTA), 9,9-bis[4-(carbazole-9-yl) phenyl] fluorene, 1,3-bis(carbazole-9-yl) benzene (hereinafter abbreviated as mCP), and 2,2-bis(4-carbazole-9-yl-phenyl) adamantane (hereinafter abbreviated as Ad-

Cz), and compounds having a triphenyl silyl group and a triaryl amine structure represented by 9-[4-(carbazole-9-yl)phenyl]-9-[4-(triphenyl silyl)phenyl]-9H-fluorene. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used in a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed. These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0050] As the light emitting layer of the organic EL device of the present embodiment, use can be made of, in addition to the metal complexes of quinolinol derivatives as typified by Alq_3 , various metal complexes, anthracene derivatives, bisstyryl benzene derivatives, pyrene derivatives, oxazole derivatives, poly para-phenylene vinylene derivatives, and the like. The light emitting layer may be constituted by a host material and a dopant material. As the host material, the anthracene derivatives are preferably used, and in addition to the above-described light emitting materials, heterocyclic compounds having an indole ring as a partial structure of a condensed ring, heterocyclic compounds having a carbazole ring as a partial structure of a condensed ring, carbazole derivatives, thiazole derivatives, benzimidazole derivatives, polydialkyl fluorene derivatives, and the like can be used. As the dopant material, quinacridone, coumarin, rubrene, perylene and derivatives thereof, benzopyran derivatives, rhodamine derivatives, amino styryl derivatives, and the like can be used. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used in a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed.

[0051] A phosphorescence emitter can also be used as the light emitting material. As the phosphorescence emitter, a phosphorescence emitter of a metal complex of iridium, platinum, or the like can be used. Green phosphorescence emitters such as Ir(ppy)_3 , blue phosphorescence emitters such as Flrpic and Flr6 , red phosphorescence emitters such as $\text{Btp}_2\text{Ir(acac)}$, and the like can be used, and as the host material at this time, carbazole derivatives such as 4,4'-di(N-carbazolyl) biphenyl (hereinafter abbreviated as CBP), TCTA and mCP can be used as the host material having hole injection and transport properties. As the electron transport host material, p-bis(triphenyl silyl) benzene (hereinafter abbreviated as UGH2), 2,2',2''-(1,3,5-phenylene)-tris(1-phenyl-1H-benzimidazole) (hereinafter abbreviated as TPBI) and the like can be used, and an organic EL device having a high performance can be produced.

[0052] In order to avoid concentration quenching, doping of the phosphorescent emitting material into the host material is preferably performed by co-deposition in a range of 1 to 30 weight percents with respect to the entire light emitting layer.

[0053] It is also possible to use materials that emit delayed fluorescence, such as PIC-TRZ, CC2TA, PXZ-TRZ, and CDCB derivatives such as 4CzIPN as the light emitting material. (See, for example, Non-Patent Literature 7.) These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0054] As the hole blocking layer of the organic EL device of the present embodiment, use can be made of compounds having a hole blocking effect, such as phenanthroline derivatives such as bathocuproine (hereinafter abbreviated as BCP), metal complexes of quinolinol derivatives such as aluminum (III) bis(2-methyl-8-quinolate)-4-phenyl phenolate (hereinafter abbreviated as BAlq), various rare earth complexes, triazole derivatives, triazine derivatives, pyrimidine derivatives, oxadiazole derivatives, benzazole derivatives, and the like. These materials may also double as materials of the electron transport layer. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used in a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed. These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0055] As the electron transport layer of the organic EL device of the present embodiment, use can be made of, in addition to metal complexes of quinolinol derivatives as typified by Alq_3 and BAlq, various metal complexes, triazole derivatives, triazine derivatives, pyrimidine derivatives, oxadiazole derivatives, pyridine derivatives, benzimidazole derivatives, benzazole derivatives, thiadiazole derivatives, anthracene derivatives, carbodiimide derivatives, quinoxaline derivatives, pyridindole derivatives, phenanthroline derivatives, silole derivatives, and the like. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used as a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed. These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0056] As the electron injection layer of the organic EL device of the present embodiment, use can be made of alkali metal salts such as lithium fluoride and cesium fluoride, alkaline earth metal salts such as magnesium fluoride, metal complexes of quinolinol derivatives such as lithium quinolinol, metal oxides such as aluminum oxide, metals such as ytterbium (Yb), samarium (Sm), calcium (Ca), strontium (Sr), and cesium (Cs), and the like, but this can be omitted in preferred selection of the electron transport layer and the cathode.

[0057] Further, in the electron injection layer or the electron transport layer, it is possible to use a material that is generally used for these layers and is further N-doped with a metal such as cesium.

[0058] As the cathode of the organic EL device of the present embodiment, an electrode material having a low work

function such as aluminum, an alloy having a lower work function such as a magnesium silver alloy, a magnesium calcium alloy, a magnesium indium alloy, or an aluminum magnesium alloy, ITO, IZO, and the like can be used as an electrode material.

[0059] As the capping layer of the organic EL device of the present embodiment, an amine compound having a benzazole ring structure represented by the general formula (A-1) or the like is preferably used. These may be used to form a film alone or may be used as a single layer formed by being mixed with another material, or may be used as a laminated structure of layers each formed by alone, layers each formed by being mixed, or a layer formed by alone and a layer formed by being mixed. These materials can form a thin film by a publicly known method such as a spin coating method or an ink jet method in addition to a deposition method.

[0060] Although the organic EL device having a top emission structure has been described above, the present invention is not limited thereto, and the organic EL device having a bottom emission structure and an organic EL device having a dual emission structure that emits light from both the top and bottom can be similarly applied. In these cases, an electrode in a direction in which light is extracted from the light emitting device to the outside is required to be transparent or translucent.

[0061] The refractive index of the material constituting the capping layer is preferably larger than a refractive index of the adjacent electrode. That is, although light extraction efficiency in the organic EL device is improved by the capping layer, the effect thereof is effective in the case where the reflectance at an interface between the capping layer and the material in contact with the capping layer is greater since an effect of optical interference is great. Therefore, the refractive index of the material constituting the capping layer is preferably larger than the refractive index of the adjacent electrode, and the refractive index may be 1.70 or more, more preferably 1.80 or more, and particularly preferably 1.85 or more.

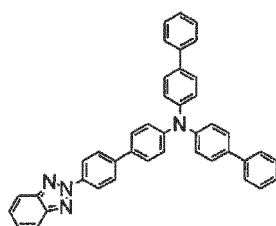
[0062] The embodiments of the present invention will be described more specifically by Examples below, but the present invention is by no means restricted to the following Examples so long as it does not exceed the gist thereof.

[Example 1]

<Synthesis of {4'-(benzotriazole-2-yl) biphenyl-4-yl}-bis-(biphenyl-4-yl)-amine (compound 17)>

[0063] Into a nitrogen substituted reaction vessel, 2-(4-bromophenyl)-2H-benzotriazole: 3.7 g, bis-(biphenyl-4-yl)-14-(4,4,5,5-tetramethyl-[1,3,2] dioxaborane-2-yl)-phenyl)-amine: 9.4 g, toluene: 80 mL, and ethanol: 20 mL were added, then an aqueous solution where potassium carbonate: 5.6 g was dissolved in H₂O: 20 mL in advance was added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tetrakis(triphenyl phosphine palladium: 0.8 g was added and stirred under heated reflux for 4 hours. After standing to cool, H₂O was added, an organic layer was extracted by a liquid separation operation and concentrated under reduced pressure to obtain a crude product. The obtained crude product was purified by column chromatography (carrier: silica gel, eluent: toluene/n-hexane) to obtain yellow powder of {4'-(benzotriazole-2-yl) biphenyl-4-yl}-bis-(biphenyl-4-yl)-amine (compound 17): 6.4 g (yield: 85%).

[Chem. 20]



(Compound 17)

[0064] A structure of the obtained yellow powder was identified with NMR.

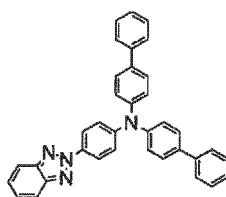
[0065] The following 30 hydrogen signals were detected with ¹H-NMR (THF-ds). δ (ppm) = 8.46 (2 H), 7.94 to 7.92 (2 H), 7.88 (2 H), 7.69 (2 H), 7.62 (4 H), 7.59 (4 H), 7.43 to 7.38 (6 H), 7.29 to 7.23 (8 H).

[Example 2]

<Synthesis of {4-(benzotriazole-2-yl) phenyl}-bis(biphenyl-4-yl)-amine (compound 6)>

[0066] Into a reaction vessel, 2-(4-bromophenyl)-2H-benzotriazole: 9.0 g, bis-(biphenyl-4-yl)-amine: 9.6 g, t-butoxy sodium: 4.3 g, and toluene: 150 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Palladium (II) acetate: 0.1 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.24 mL were added and stirred under heated reflux for 2 hours. After standing to cool, toluene was added to perform dispersion washing, insoluble matter was filtered off, and silica gel and active white clay were added to the filtrate to perform adsorption purification. Adsorbent was removed by filtration and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with a toluene/acetone mixed solvent to collect a precipitated solid, to obtain yellow powder of {4-(benzotriazole-2-yl) phenyl}-bis(biphenyl-4-yl)-amine (compound 6): 11.6 g (yield: 75.3%).

[Chem. 21]



(Compound 6)

[0067] A structure of the obtained yellow powder was identified with NMR.

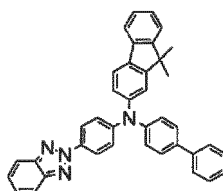
[0068] The following 26 hydrogen signals were detected with $^1\text{H-NMR}$ (CDCl_3). δ (ppm) = 8.27 to 8.31 (2 H), 7.95 to 7.99 (2 H), 7.58 to 7.66 (8 H), 7.28 to 7.51 (14 H).

[Example 3]

<Synthesis of {4-(benzotriazole-2-yl) phenyl}-(biphenyl-4-yl)-(9,9-dimethyl-9H-fluorene-2-yl)-amine (compound 7)>

[0069] Into a reaction vessel, 2-(4-bromophenyl)-2H-benzotriazole: 8.3 g, (biphenyl-4-yl)-(9,9-dimethyl-9H-fluorene-2-yl)-amine: 10.0 g, t-butoxy sodium: 4.0 g, and toluene: 150 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Palladium (II) acetate: 0.1 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.22 mL were added and stirred under heated reflux for 2 hours. After standing to cool, toluene was added to perform dispersion washing, insoluble matter was filtered off, and silica gel and active white clay were added to the filtrate to perform adsorption purification. Adsorbent was removed by filtration and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with acetone to collect a precipitated solid, to obtain yellow powder of {4-(benzotriazole-2-yl) phenyl}-(biphenyl-4-yl)-(9,9-dimethyl-9H-fluorene-2-yl)-amine (compound 7): 12.3 g (yield: 80.4%).

[Chem. 22]



(Compound 7)

[0070] A structure of the obtained yellow powder was identified with NMR.

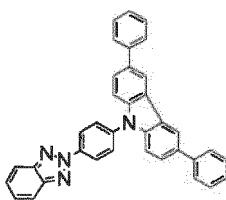
[0071] The following 30 hydrogen signals were detected with $^1\text{H-NMR}$ (CDCl_3). δ (ppm) = 8.24 to 8.27 (2 H), 7.94 to 7.97 (2 H), 7.57 to 7.72 (6 H), 7.28 to 7.50 (13 H), 7.16 to 7.20 (1 H), 1.48 (6 H).

[Example 4]

<Synthesis of 9-{4-(benzotriazole-2-yl) phenyl}-3,6-diphenyl-9H-carbazole (compound 23)>

[0072] Into a reaction vessel, 2-(4-bromophenyl)-2H-benzotriazole: 7.6 g, 3,6-biphenyl-9H-carbazole: 8.0 g, t-butoxy sodium: 3.6 g, and toluene: 130 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Palladium (II) acetate: 0.1 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.20 mL were added and stirred under heated reflux for 12 hours. After standing to cool, toluene was added to perform dispersion washing, insoluble matter was filtered off, and silica gel and active white clay were added to the filtrate to perform adsorption purification. Adsorbent was removed by filtration and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with acetone to collect a precipitated solid, to obtain yellow powder of 9-{4-(benzotriazole-2-yl) phenyl}-3,6-diphenyl-9H-carbazole (compound 23): 6.9 g (yield: 54%).

[Chem. 23]



(Compound 23)

[0073] A structure of the obtained yellow powder was identified with NMR.

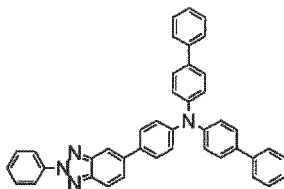
[0074] The following 30 hydrogen signals were detected with $^1\text{H-NMR}$ (CDCl_3). δ (ppm) = 8.67 to 8.68 (2 H), 8.45 (2 H), 8.00 to 8.03 (2 H), 7.73 to 7.87 (8 H), 7.39 to 7.57 (10 H).

[Example 5]

<Synthesis of {(2-phenyl benzotriazole-5-yl) phenyl-4-yl}-bis-(biphenyl-4-yl)-amine (compound 30)>

[0075] Into a nitrogen substituted reaction vessel, 5-bromo-2-phenyl-2H-benzotriazole: 2.3 g, bis-(biphenyl-4-yl)-{4-(4,4,5,5-tetramethyl-[1,3,2] dioxaborane-2-yl)-phenyl}-amine: 5.3 g, toluene: 52 mL, and ethanol: 13 mL were added, then an aqueous solution where potassium carbonate: 3.5 g was dissolved in H_2O : 13 mL in advance was added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tetrakis(triphenyl) phosphine palladium: 0.5 g was added and stirred under heated reflux for 6 hours. After standing to cool, H_2O was added, an organic layer was extracted by a liquid separation operation and concentrated under reduced pressure to obtain a crude product. The obtained crude product was purified by column chromatography (carrier: silica gel, eluent: toluene/n-hexane) to obtain yellow powder of {(2-phenyl benzotriazole-5-yl) phenyl-4-yl}-bis-(biphenyl-4-yl)-amine (compound 30): 3.5 g (yield: 71%).

[Chem. 24]



(Compound 30)

[0076] A structure of the obtained yellow powder was identified with NMR.

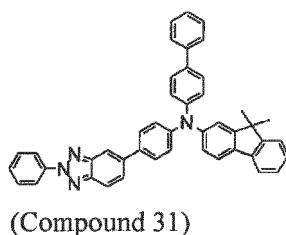
[0077] The following 30 hydrogen signals were detected with $^1\text{H-NMR}$ (THF-d_8). δ (ppm) = 8.41 (2 H), 8.15 (1 H), 7.98 (1 H), 7.80 (1 H), 7.72 (2 H), 7.56 to 7.65 (10 H), 7.47 (1 H), 7.40 (4 H), 7.24 to 7.29 (8 H).

[Example 6]

<Synthesis of {(2-phenyl benzotriazole-5-yl) phenyl-4-yl}-(biphenyl-4-yl)-9,9-dimethyl-9H-fluorene-2-yl) (compound 31)>

[0078] Into a nitrogen substituted reaction vessel, 5-bromo-2-phenyl-2H-benzotriazole: 2.2 g, bis-(biphenyl-4-yl)-{4-(4,4,5,5-tetramethyl-[1,3,2] dioxaborane-2-yl)-phenyl}-amine: 5.4 g, toluene: 48 mL, and ethanol: 12 mL were added, then an aqueous solution where potassium carbonate: 3.3 g was dissolved in H₂O: 12 mL in advance was added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tetrakis(triphenyl phosphine) palladium: 0.5 g was added and stirred under heated reflux for 5 hours. After standing to cool, H₂O was added, an organic layer was extracted by a liquid separation operation and concentrated under reduced pressure to obtain a crude product. The obtained crude product was purified by column chromatography (carrier: silica gel, eluent: toluene/n-hexane) to obtain yellow powder of {(2-phenyl benzotriazole-5-yl) phenyl-4-yl}-(biphenyl-4-yl)-9,9-dimethyl-9H-fluorene-2-yl) (compound 31): 3.9 g (yield: 77%).

[Chem. 25]



[0079] A structure of the obtained yellow powder was identified with NMR.

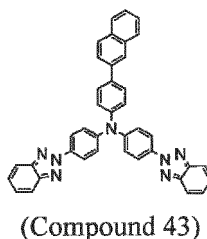
[0080] The following 34 hydrogen signals were detected with ¹H-NMR (THF-ds). δ (ppm) = 8.41 (2 H), 8.16 (1 H), 7.98 (1 H), 7.81 (1 H), 7.72 (2 H), 7.69 (2 H), 7.63 (2 H), 7.59 (2 H), 7.58 (2 H), 7.47 (1 H), 7.38 to 7.43 (4 H), 7.21 to 7.30 (7 H), 7.12 (1 H), 1.44 (6 H).

[Example 7]

<Synthesis of bis-{4-(benzotriazole-2-yl) phenyl}-{4-(naphthalene-2-yl) phenyl} amine (compound 43)>

[0081] Into a reaction vessel, 4-(naphthalen-2-yl) phenyl-amine: 5.0 g, 2-(4-bromophenyl)-2H-benzotriazole: 13.1 g, t-butoxy sodium: 6.6 g, and toluene: 130 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tris(dibenzylidene acetone) dipalladium (0): 0.21 g and 2,2'-bis(diphenyl phosphino)-1,1'-binaphthyl: 0.28 g were added and stirred under heated reflux for one day. After standing to cool to 80°C, insoluble matter was filtered off, and silica gel was added to the filtrate to perform adsorption purification. Adsorbent was removed by filtration and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with toluene to collect a precipitated solid, to obtain yellow powder of bis-{4-(benzotriazole-2-yl) phenyl}-{4-(naphthalene-2-yl) phenyl} amine (compound 43): 6.5 g (yield: 47%).

[Chem. 26]



[0082] A structure of the obtained yellow powder was identified with NMR.

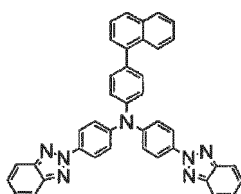
[0083] The following 27 hydrogen signals were detected with ¹H-NMR (CDCl₃). δ (ppm) = 8.33 to 8.35 (4 H), 8.31 (1 H), 7.73 to 7.98 (10 H), 7.34 to 7.55 (12 H).

[Example 8]

<Synthesis of bis-{4-(benzoxazole-2-yl) phenyl}-4-(naphthalene-1-yl) phenyl} amine (compound 44)>

[0084] Into a reaction vessel, 4-(naphthalen-1-yl) phenyl-amine: 5.0 g, 2-(4-bromophenyl)-2H-benzotriazole: 13.8 g, t-butoxy sodium: 6.6 g, and toluene: 50 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tris(dibenzylidene acetone) dipalladium (0): 0.6 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.6 mL were added and stirred under heated reflux for 8 hours. After standing to cool to 80°C, insoluble matter was filtered off, and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with acetone to collect a precipitated solid, to obtain yellow powder of bis-{4-(benzotriazole-2-yl) phenyl }-4-(naphthalene-1-yl) phenyl} amine (compound 44): 6.4 g (yield: 46%).

[Chem. 27]



(Compound 44)

[0085] A structure of the obtained yellow powder was identified with NMR.

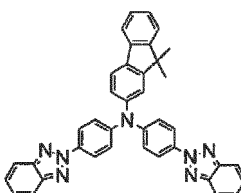
[0086] The following 27 hydrogen signals were detected with ¹H-NMR (DMSO-d₆). δ (ppm) = 8.33 to 8.36 (4 H), 7.96 to 8.06 (7 H), 7.37 to 7.63 (16 H).

[Example 9]

<Synthesis of bis-{4-(benzotriazole-2-yl) phenyl}-(9,9-dimethyl-9H-fluorene-2-yl)-amine (compound 47)>

[0087] Into a reaction vessel, 9,9-dimethyl-9H-fluorene-2-yl-amine: 3.8 g, 2-(4-bromophenyl)-2H-benzotriazole: 10.9 g, t-butoxy sodium: 5.2 g, and toluene: 40 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tris(dibenzylidene acetone) dipalladium (0): 0.5 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.44 mL were added and stirred under heated reflux for 7 hours. Toluene was added to perform dispersion washing, insoluble matter was thermal-filtered off, and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with a toluene/methanol mixed solvent to collect a precipitated solid, to obtain yellow powder of bis-{4-(benzotriazole-2-yl) phenyl}-(9,9-dimethyl-9H-fluorene-2-yl)-amine (compound 47): 6.8 g (yield: 63%).

[Chem. 28]



(Compound 47)

[0088] A structure of the obtained yellow powder was identified with NMR.

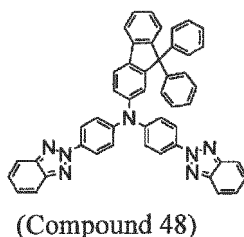
[0089] The following 28 hydrogen signals were detected with ¹H-NMR (DMSO-d₆). δ (ppm) = 8.29 to 8.31 (4 H), 8.01 to 8.04 (4 H), 7.81 to 7.90 (2 H), 7.19 to 7.56 (13 H), 1.43 (6H).

[Example 10]

<Synthesis of bis-{4-(benzoxazole-2-yl) phenyl}-(9,9-diphenyl-9H-fluorene-2-yl)-amine (compound 48)>

[0090] Into a reaction vessel, 9,9-diphenyl-9H-fluorene-2-yl-amine: 5.0 g, 2-(4-bromophenyl)-2H-benzotriazole: 9.0 g, t-butoxy sodium: 4.3 g, and toluene: 60 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tris(dibenzylidene acetone) dipalladium (0): 0.4 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.4 mL were added and stirred under heated reflux for 7 hours. After standing to cool, toluene was added to perform dispersion washing, insoluble matter was filtered off, and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with toluene to collect a precipitated solid, to obtain yellow powder of bis-{4-(benzoxazole-2-yl) phenyl}-(9,9-diphenyl-9H-fluorene-2-yl)-amine (compound 48): 8.1 g (yield: 75%).

[Chem. 29]



[0091] A structure of the obtained yellow powder was identified with NMR.

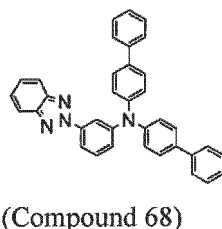
[0092] The following 32 hydrogen signals were detected with ¹H-NMR (DMSO-d₆). δ (ppm) = 8.24 to 8.27 (4 H), 7.91 to 8.04 (6 H), 7.40 to 7.54 (6 H), 7.23 to 7.34 (12 H), 7.09 to 7.12 (4 H).

[Example 11]

<Synthesis of {3-(benzotriazole-2-yl) phenyl}-bis(biphenyl-4-yl)-amine (compound 68)>

[0093] Into a reaction vessel, 2-(3-bromophenyl)-2H-benzotriazole: 9.6 g, bis-(biphenyl-4-yl)-amine: 9.0 g, t-butoxy sodium: 4.3 g, and toluene: 150 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Palladium (II) acetate: 0.1 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.24 mL were added and stirred under heated reflux for 2 hours. After standing to cool, toluene was added to perform dispersion washing, insoluble matter was filtered off, and silica gel and active white clay were added to the filtrate to perform adsorption purification. Adsorbent was removed by filtration and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with a toluene/acetone mixed solvent to collect a precipitated solid, to obtain yellow powder of {3-(benzotriazole-2-yl) phenyl}-bis(biphenyl-4-yl)-amine (compound 68): 8.2 g (yield: 53%).

[Chem. 30]



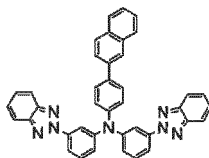
[Example 12]

<Synthesis of bis-{3-(benzotriazole-2-yl) phenyl}-{4-(naphthalene-2-yl) phenyl} amine (compound 77)>

[0094] Into a reaction vessel, 4-(naphthalen-2-yl) phenyl-amine: 6.1 g, 2-(3-bromophenyl)-2H-benzotriazole: 16.0 g,

t-butoxy sodium: 8.0 g, and toluene: 170 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tris(dibenzylidene acetone) dipalladium (0): 0.25 g and 2,2'-bis(diphenyl phosphino)-1,1'-binaphthyl: 0.35 g were added and stirred under heated reflux for one night. After adding toluene and standing to cool to 80°C, insoluble matter was filtered off, and silica gel was added to the filtrate to perform adsorption purification. Adsorbent was removed by filtration and the filtrate was concentrated to obtain a crude product. The crude product was purified by crystallization with toluene to collect a precipitated solid, to obtain yellow powder of bis-{3-(benzotriazole-2-yl) phenyl}-{4-(naphthalene-2-yl) phenyl} amine (compound 77): 6.2 g (yield: 37%).

[Chem. 31]



(Compound 77)

[0095] A structure of the obtained yellow powder was identified with NMR.

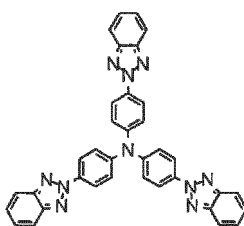
[0096] The following 27 hydrogen signals were detected with ¹H-NMR (CDCl₃). δ (ppm) = 8.24 to 8.25 (2 H), 8.08 to 8.11 (3 H), 7.89 to 7.96 (7 H), 7.72 to 7.81 (3 H), 7.30 to 7.55 (12 H).

[Example 13]

<Synthesis of tris-(4-benzotriazole-2-yl-phenyl)-amine (compound 85)>

[0097] Into a reaction vessel, 4-benzotriazole-2-yl-phenyl amine: 2.5 g, t-butoxy sodium: 3.4 g, and toluene: 60 mL were added, and nitrogen gas was aerated while the solution was irradiated with ultrasonic waves for 30 minutes. Tris(dibenzylidene acetone) dipalladium (0): 0.2 g and a 50% (w/v) toluene solution of tri-(t-butyl) phosphine: 0.5 mL were added and stirred under heated reflux for one night. Toluene was added to perform dispersion washing, insoluble matter was thermal-filtered off, and the filtrate was concentrated to obtain a crude product. The obtained crude product was purified by column chromatography (carrier: silica gel, eluent: toluene/n-hexane) to obtain yellow powder of tris-(4-benzotriazole-2-yl-phenyl)-amine (compound 85): 3.3 g (yield: 46.5%).

[Chem. 32]



(Compound 85)

[0098] A structure of the obtained yellow powder was identified with NMR.

[0099] The following 24 hydrogen signals were detected with ¹H-NMR (CDCl₃). δ (ppm) = 8.26 (6 H), 7.93 (6 H), 7.42 (6 H), 7.33 (6 H).

[Example 14]

[0100] A melting point and a glass transition point of each of the benzazole compounds represented by the general formula (A-1) were measured with a high sensitivity differential scanning calorimeter (DSC 3100 SA manufactured by Bruker AXS GmbH).

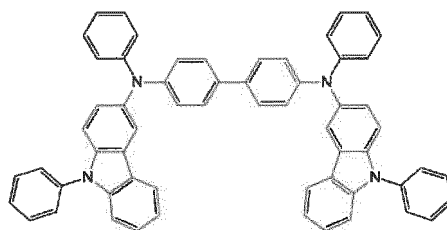
	Melting point	Glass transition point
Compound of Example 1	207°C	102°C
Compound of Example 2	187°C	100°C
Compound of Example 3	not observed	101°C
Compound of Example 4	not observed	106°C
Compound of Example 5	109°C	100°C
Compound of Example 6	125°C	106°C
Compound of Example 7	271°C	110°C
Compound of Example 8	224°C	113°C
Compound of Example 9	228°C	119°C
Compound of Example 10	267°C	146°C
Compound of Example 11	not observed	100°C
Compound of Example 12	not observed	105°C
Compound of Example 13	not observed	117°C

[0101] The compounds having a benzazole ring structure represented by the general formula (A-1) have a glass transition point of 100°C or higher, which indicates that the thin film state is stable.

[Example 15]

[0102] A deposited film having a film thickness of 80 nm was produced on a silicon substrate by using the amine compound having a benzazole ring structure represented by the general formula (A-1), and a refractive index n and an extinction coefficient k at wavelengths of 400 nm and 410 nm were measured with a spectroscopic measurement device (F10-RT-UV manufactured by Filmetrics Japan, Inc.). For comparison, a comparative compound (2-1) and Alq3 of the following structural formulae were also measured (see, for example, Patent Literature 4). Measurement results were summarized in Table 1.

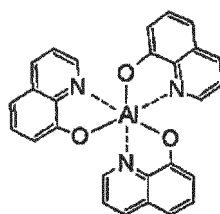
[Chem. 33]



Comparative Compound (2-1)

[0142]

[Chem. 34]



Alq₃

[Table 1]

	Refractive index n (λ : 400 nm)	Refractive index n (λ : 410 nm)	Extinction coefficient k (λ : 400 nm)	Extinction coefficient k (λ : 410 nm)
Compound 17	2.30	2.27	0.33	0.25
Compound 6	2.35	2.29	0.50	0.30
Compound 7	2.30	2.24	0.49	0.31
Compound 23	2.41	2.30	0.45	0.30
Compound 30	2.29	2.20	0.33	0.22
Compound 31	2.28	2.20	0.33	0.20
Compound 43	2.37	2.47	0.70	0.47
Compound 44	2.33	2.40	0.67	0.45
Compound 47	2.30	2.39	0.65	0.40
Compound 48	2.35	2.42	0.68	0.40
Compound 68	2.15	2.13	0.28	0.24
Compound 77	2.20	2.15	0.28	0.25
Compound 85	2.50	2.43	0.88	0.56
Comparative compound (2-1)	2.13	2.10	0.15	0.06
Alq ₃	1.86	1.89	0.16	0.14

[0103] Thus, the compounds of the present example have a refractive index of a value equal to or higher than that of the comparative compound (2-1) and Alq₃, and improvement in light extraction efficiency in the organic EL device can be expected. The extinction coefficient at a wavelength of 400 nm to 410 nm was larger than the extinction coefficient at a wavelength of 410 nm. While the extinction coefficient at a wavelength of 400 nm to 410 nm of the comparative compound (2-1) and Alq₃ is less than 0.20, the compounds of the present example have a large value, which indicates that light at a wavelengths of 400 nm to 410 nm among sunlight is well absorbed and does not affect a material inside the device.

[Example 16]

[0104] The compounds of the present example were adjusted as a toluene solution at a concentration of 1.0×10^{-5} mol/L to measure absorbance of the solution at wavelengths of 400 nm and 410 nm. An absorption coefficient was calculated from a calibration curve of absorbance at a peak wavelength by adjusting a concentration of the toluene solution to four kinds of concentration of 5.0×10^{-6} mol/L, 1.0×10^{-5} mol/L, 1.5×10^{-5} mol/L, and 2.0×10^{-5} mol/L and measuring the absorbance with an ultraviolet-visible near-infrared spectrophotometer (V-650 manufactured by JASCO Corporation). For comparison, the comparative compound (2-1) and Alq₃ of the above-mentioned structural formulae were also measured. Measurement results were summarized in Table 2.

[Table 2]

	Peak wavelength ($\lambda_{\max}$)	Absorbance (λ : 400 nm)	Absorbance (λ : 410 nm)	Absorption coefficient
Compound 17	380 nm	0.50	0.20	94400
Compound 6	374 nm	0.81	0.30	123605
Compound 7	370 nm	0.79	0.25	112990
Compound 23	355 nm	0.66	0.27	124458
Compound 30	349 nm	0.60	0.22	124960
Compound 31	345 nm	0.60	0.23	122199

(continued)

	Peak wavelength ($\lambda_{\max}$)	Absorbance (λ : 400 nm)	Absorbance (λ : 410 nm)	Absorption coefficient
Compound 43	377 nm	0.85	0.30	149105
Compound 44	370 nm	0.77	0.29	141650
Compound 47	370 nm	0.70	0.25	134470
Compound 48	373 nm	0.72	0.25	129256
Compound 68	305 nm	0.45	0.22	104014
Compound 77	310 nm	0.50	0.20	112015
Compound 85	385 nm	0.87	0.44	168087
Comparative compound (2-1)	358 nm	0.07	0.02	48856
Alq ₃	394 nm	0.07	0.06	7518

[0105] In an absorption spectrum, the absorbance at a wavelength of 400 nm to 410 nm was larger than the absorbance at a wavelength of 410 nm. In this way, while the absorbance at a wavelength of 400 nm to 410 nm of the comparative compound (2-1) and Alq₃ is 0.10 or less, the compounds of the present example have a large value of 0.20 or more, which indicates that light at a wavelength of 400 nm to 410 nm among sunlight is well absorbed. In addition, as compared with the comparative compound (2-1) and Alq₃, the compounds of the present example have a large value of absorption coefficient, and light is well absorbed under the same concentration condition. With respect to the thin film, it can be found that the more the film thickness is, the better light is absorbed, and the material is excellent in light resistance.

[Example 17]

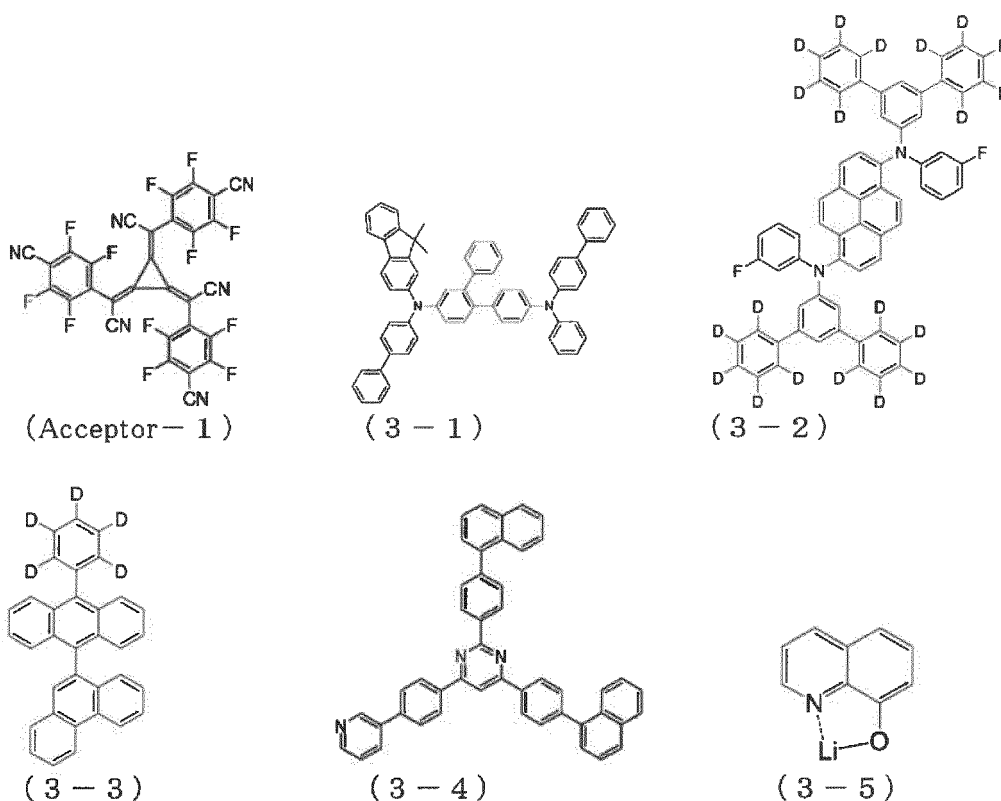
[0106] As illustrated in FIG. 1, the organic EL device was produced by depositing a hole injection layer 3, a hole transport layer 4, a light emitting layer 5, an electron transport layer 6, an electron injection layer 7, a cathode 8, and a capping layer 9 in this order on a preformed reflective ITO electrode as a metal anode 2 on a glass substrate 1.

[0107] Specifically, the glass substrate 1 on which ITO having a film thickness of 50 nm, a reflective film of a silver alloy having a film thickness of 100 nm and ITO having a film thickness of 5 nm were formed in order was subjected to ultrasonic washing in isopropyl alcohol for 20 minutes, and then dried for 10 minutes on a hot plate heated to 250°C. Then, after performing a UV ozone treatment for 2 minutes, the glass substrate with ITO was mounted in a vacuum deposition machine and depressurized to 0.001 Pa or less. Next, as the hole injection layer 3 to cover the transparent anode 2, an electron acceptor (Acceptor-1) of the following structural formula and a compound (3-1) of the following structural formula were subjected to binary deposition at a deposition rate where a ratio of deposition rates of Acceptor-1 : Compound (3-1) = 3 : 97 to form the hole injection layer 3 so as to have a film thickness of 10 nm. A layer of the compound (3-1) of the following structural formula was formed to have a film thickness of 140 nm as the hole transport layer 4 on the hole injection layer 3. A compound (3-2) of the following structural formula and a compound (3-3) of the following structural formula as the light emitting layer 5 were subjected to binary deposition at a deposition rate where a ratio of deposition rates of (3-2) : (3-3) = 5 : 95 on the hole transport layer 4 to form the light emitting layer 5 so as to have a film thickness of 20 nm. A compound (3-4) of the following structural formula and a compound (3-5) of the following structural formula as the electron transport layer 6 were subjected to binary deposition at a deposition rate where a ratio of deposition rates of (3-4) : (3-5) = 50 : 50 on the light emitting layer 5 to form the electron transport layer 6 so as to have a film thickness of 30 nm. A layer of lithium fluoride was formed to have a film thickness of 1 nm as the electron injection layer 7 on the electron transport layer 6.

[0108] A layer of a magnesium silver alloy was formed to have a film thickness of 12 nm as the cathode 8 on the electron injection layer 7. Finally, a layer of the compound 17 of Example 1 was formed to have a film thickness of 60 nm as the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature.

[0109] Table 3 summarized measurement results of light emitting characteristics in which a direct current voltage was applied to the produced organic EL device.

[Chem. 35]



[Example 18]

[0110] The organic EL device was produced under the same conditions as in Example 17 except that the compound 6 of Example 2 was used in place of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 19]

[0111] The organic EL device was produced under the same conditions as in Example 17 except that the compound 7 of Example 3 was used in place of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 20]

[0112] The organic EL device was produced under the same conditions as in Example 17 except that the compound 23 of Example 4 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 21]

[0113] The organic EL device was produced under the same conditions as in Example 17 except that the compound 30 of Example 5 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement

results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 22]

[0114] The organic EL device was produced under the same conditions as in Example 17 except that the compound 31 of Example 6 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 23]

[0115] The organic EL device was produced under the same conditions as in Example 17 except that the compound 43 of Example 7 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 24]

[0116] The organic EL device was produced under the same conditions as in Example 17 except that the compound 44 of Example 8 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 25]

[0117] The organic EL device was produced under the same conditions as in Example 17 except that the compound 47 of Example 9 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 26]

[0118] The organic EL device was produced under the same conditions as in Example 17 except that the compound 48 of Example 10 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 27]

[0119] The organic EL device was produced under the same conditions as in Example 17 except that the compound 68 of Example 11 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 28]

[0120] The organic EL device was produced under the same conditions as in Example 17 except that the compound 77 of Example 12 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Example 29]

[0121] The organic EL device was produced under the same conditions as in Example 17 except that the compound 85 of Example 13 was used instead of the compound 17 of Example 1 as the material of the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Measurement results of light emitting characteristics when a direct current voltage was applied to the produced organic EL device were summarized in Table 3.

[Comparative Example 1]

[0122] For comparison, an organic EL device was produced under the same conditions as in Example 17 except that a layer of the comparative compound (2-1) of the above-mentioned structural formula was formed to have a film thickness of 60 nm instead of the compound 17 of Example 1 as the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Table 3 summarized measurement results of light emitting characteristics in which a direct current voltage was applied to the produced organic EL device.

[Comparative Example 2]

[0123] For comparison, an organic EL device was produced under the same conditions as in Example 17 except that a layer of Alq_3 was formed to have a film thickness of 60 nm instead of the compound 17 of Example 1 as the capping layer 9. The produced organic EL device was subjected to characteristic measurement in the atmosphere at room temperature. Table 3 summarized measurement results of light emitting characteristics in which a direct current voltage was applied to the produced organic EL device.

[0124] Table 3 summarized results of measuring a device lifetime by using the organic EL devices produced in Examples 17 to 29 and Comparative Examples 1 and 2. The device lifetime was measured as time until decay to 95% decay when initial luminance was 100% when constant current driving was performed at 10 mA/cm^2 .

[Table 3]

	Capping layer	Luminance Voltage [V] [cd/m^2] (@10 mA/cm^2) (@10 mA/cm^2)		Power Efficiency Luminous efficiency [cd/A] [lm/W] (@10 mA/cm^2) (@10 mA/cm^2)		Device lifetime 95% decay
Example 17	Compound 17	3.66	685	6.90	5.92	163 hours
Example 18	Compound 6	3.66	700	7.00	6.01	165 hours
Example 19	Compound 7	3.65	697	6.97	6.01	161 hours
Example 20	Compound 23	3.66	681	6.81	5.85	158 hours
Example 21	Compound 30	3.66	675	6.75	5.79	155 hours
Example 22	Compound 31	3.64	675	6.75	5.83	154 hours
Example 23	Compound 43	3.65	715	7.15	6.15	159 hours
Example 24	Compound 44	3.65	711	7.11	6.12	163 hours
Example 25	Compound 47	3.64	706	7.06	6.09	160 hours
Example 26	Compound 48	3.64	708	7.08	6.11	160 hours
Example 27	Compound 68	3.64	674	6.74	5.82	156 hours
Example 28	Compound 77	3.64	684	6.84	5.90	151 hours
Example 29	Compound 85	3.64	722	7.22	6.23	164 hours
Comparative Example 1	Comparative compound (2-1)	3.69	668	6.68	5.68	121 hours
Comparative Example 2	Alq_3	3.67	647	6.47	5.54	106 hours

[0125] As shown in Table 3, a drive voltage at a current density of 10 mA/cm² is almost the same as the devices of Comparative Example 1 and Comparative Example 2 and the devices of Examples 17 to 29. On the other hand, luminance, luminous efficiency, power efficiency, and lifetime were improved in the devices of Examples 17 to 29 with respect to the devices of Comparative Example 1 and Comparative Example 2. This indicates that the light extraction efficiency can be greatly improved since the capping layer contains the material of the present example, which is suitably used for the organic EL device and has a high refractive index.

[0126] Although the present invention has been described in detail with reference to specific embodiments, it is apparent to those skilled in the art that it is possible to add various alterations and modifications without departing from the spirit and the scope of the present invention.

[0127] The present application is based on Japanese Patent Application (No. 2018-001564) filed on January 10, 2018, the entirety of which is incorporated by reference. In addition, all references cited here are entirely incorporated.

Industrial Applicability

[0128] As described above, the amine compound having a benzazole ring structure represented by the general formula (A-1) that is suitably used in the organic EL device of the present invention has a high light absorption coefficient and a high refractive index, can greatly improve light extraction efficiency, and is stable in a thin film state, and thus is excellent as a compound for the organic EL device. By producing the organic EL device using the compound, high efficiency can be obtained, and durability or light resistance can be improved so as to absorb light of sunlight and not to influence the material inside the device. Use of the compound that does not have absorption in respective wavelength ranges of blue, green and red is particularly suitable when it is desired to display a vivid and bright image with good color purity. For example, it is possible to develop a home electrical appliance or a lighting application.

Reference Sign List

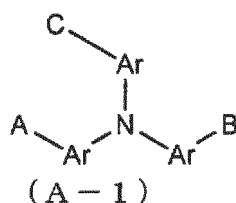
[0129]

- 1 Glass substrate
- 2 Transparent anode
- 3 Hole injection layer
- 4 Hole transport layer
- 5 Light emitting layer
- 6 Electron transport layer
- 7 Electron injection layer
- 8 Cathode
- 9 Capping layer

Claims

1. An organic EL device having at least an anodic electrode, a hole transport layer, a light emitting layer, an electron transport layer, a cathodic electrode, and a capping layer in this order, wherein the capping layer comprises an amine compound having a benzazole ring structure represented by the following general formula (A-1):

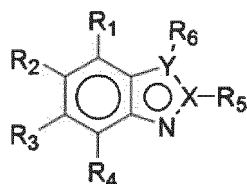
[Chem. 36]



(in the formula, A represents a monovalent group represented by the following general formula (B-1) where one position in R₁ to R₆ is a bonding site; B and C may be the same as or different from each other, and each represents a monovalent group represented by the following general formula (B-1) where one position in R₁ to R₆ is a bonding site, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic

group, or a substituted or unsubstituted condensed polycyclic aromatic group; Ar's may be the same as or different from each other, and each represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group, or a single bond; and Ar's adjacent to each other via a nitrogen atom may be bonded by a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring);

[Chem. 37]

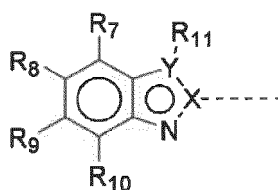


(B - 1)

(in the formula, R_1 to R_6 may be the same as or different from each other, and each represents a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkoxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkoxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom, provided that in a case where X and Y are nitrogen atoms, X or Y does not have R^5 or R^6 , and a case where X and Y are carbon atoms is excepted).

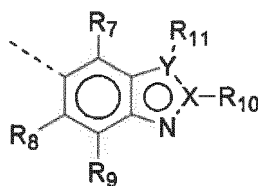
2. The organic EL device according to Claim 1, wherein the amine compound having a benzazole ring structure has an extinction coefficient of 0.20 or more at a wavelength of 400 nm to 410 nm and has an absorbance of 0.20 or more at a wavelength of 400 nm to 410 nm in an absorption spectrum of a toluene solution with a concentration of 1.0×10^{-5} mol/L.
3. The organic EL device according to Claim 1, wherein the general formula (B-1) is represented by the following general formula (B-2) or (B-3):

[Chem. 38]



(B - 2)

[Chem. 39]

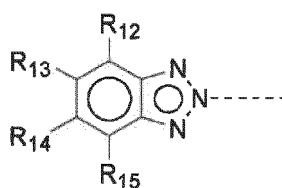


(B - 3)

(in the formulae, a broken line is a bonding site; R_7 to R_{11} may be the same as or different from each other, and each represents a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkyloxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyloxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom, provided that in a case where X and Y are nitrogen atoms, Y does not have R_{11} , and a case where X and Y are carbon atoms is excepted).

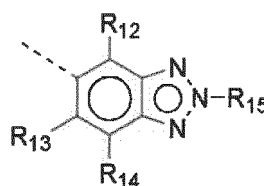
4. The organic EL device according to Claim 1, wherein the general formula (B-1) is represented by the following general formula (B-4) or (B-5):

[Chem. 40]



(B - 4)

[Chem. 41]



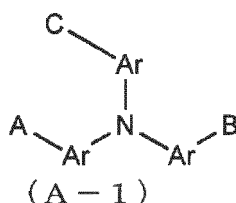
(B - 5)

(in the formulae, a broken line is a bonding site; R_{12} to R_{15} may be the same as or different from each other, and each represents a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon number of 2 to 6 which may have a substituent, a linear or branched alkyloxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyloxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group).

5. The organic EL device according to any one of Claims 1 to 4, wherein A, B and C each have a benzazole ring structure in the general formula (A-1).
6. The organic EL device according to any one of Claims 1 to 4, wherein in the general formula (A-1), A and B each have a benzazole ring structure, and C represents a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group.
7. The organic EL device according to any one of Claims 1 to 4, wherein in the general formula (A-1), A has a benzazole ring structure, and B and C each represents a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group.

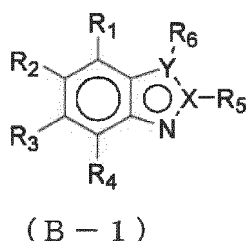
8. The organic EL device according to Claim 5, wherein A, B and C are the same as each other in the general formula (A-1).
9. The organic EL device according to Claim 5 or 6, wherein A and B are the same as each other in the general formula (A-1).
10. The organic EL device according to any one of Claims 5 to 7, wherein A, B and C are not the same as each other in the general formula (A-1).
11. The organic EL device according to any one of Claims 5 to 7, wherein A and B are not the same as each other in the general formula (A-1).
12. The organic EL device according to Claim 1, wherein the capping layer has a thickness in a range of 30 nm to 120 nm.
13. The organic EL device according to Claim 1, wherein the capping layer has a refractive index of 1.85 or more with respect to light having a wavelength in a range of 400 nm to 410 nm.
14. A method of using an amine compound having a benzotriazole ring structure where A or B in the general formula (A-1) as described in Claim 1 is represented by the general formula (B-4) or (B-5) as described in Claim 4, in a capping layer of an organic EL device.
15. An amine compound having a benzazole ring structure represented by the following general formula (A-1):

[Chem. 42]



(in the formula, A represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site; B and C may be the same as or different from each other, and each represents a monovalent group represented by the following general formula (B-1) where one position in R_1 to R_6 is a bonding site, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group; Ar's may be the same as or different from each other, and each represents a divalent group of a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted condensed polycyclic aromatic group, or a single bond; and Ar's adjacent to each other via a nitrogen atom may be bonded by a single bond, a substituted or unsubstituted methylene group, an oxygen atom, or a sulfur atom to form a ring);

[Chem. 43]



(in the formula, R_1 to R_6 may be the same as or different from each other, and each represents a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, a linear or branched alkyl group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyl group having a carbon number of 5 to 10 which may have a substituent, a linear or branched alkenyl group having a carbon

number of 2 to 6 which may have a substituent, a linear or branched alkyloxy group having a carbon number of 1 to 6 which may have a substituent, a cycloalkyloxy group having a carbon number of 5 to 10 which may have a substituent, a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, a substituted or unsubstituted condensed polycyclic aromatic group, or a substituted or unsubstituted aryloxy group; and X and Y represent a carbon atom or a nitrogen atom, provided that in a case where X and Y are nitrogen atoms, X or Y does not have R₅ or R₆, and a case where X and Y are carbon atoms is excepted).

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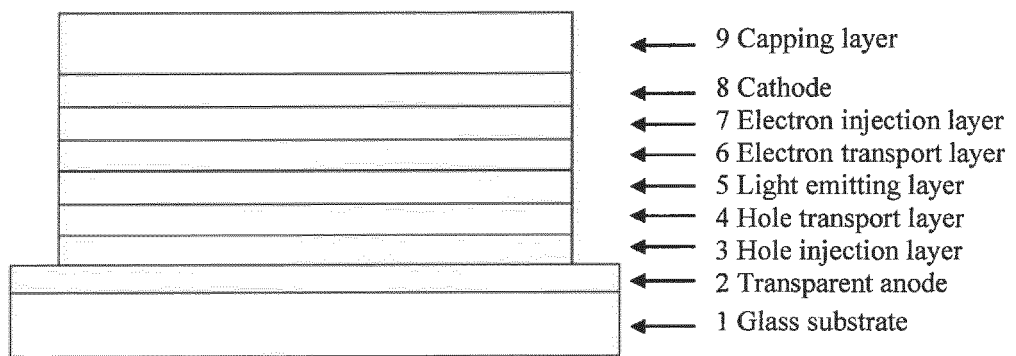
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[FIG. 1]



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2019/000452

A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl. H05B33/02 (2006.01) i, C07D249/20 (2006.01) i, C07D403/10 (2006.01) i, H01L51/50 (2006.01) i

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)

Int.Cl. H05B33/02, C07D249/20, C07D403/10, H01L51/50

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

Published examined utility model applications of Japan 1922-1996

Published unexamined utility model applications of Japan 1971-2019

Registered utility model specifications of Japan 1996-2019

Published registered utility model applications of Japan 1994-2019

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

CAPLUS/REGISTRY (STN)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	WO 2017/183625 A1 (HODOGAYA CHEMICAL CO., LTD.) 26 October 2017, claims 1-5, 14-16, paragraphs [0070]-[0131], [0149]-[0186] & EP 3432688 A1, claims 1-5, 14-16, paragraphs [0054]-[0059], [0077]-[0111] & CN 109076658 A & KR 10-2018-0134331 A	1-4, 6-7, 10-15 5, 8-9
X A	KR 10-0861168 B1 (UNIVERSITY-INDUSTRY COOPERATION GROUP OF KYUNG HEE UNIVERSITY) 30 September 2008, <42> (Family: none)	15 1-14

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

☐ See patent family annex.

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Date of the actual completion of the international search
27.03.2019

Date of mailing of the international search report
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2019/000452

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X A	JP 2002-151268 A (KONICA CORPORATION) 24 May 2002, paragraph [0098] & JP 2010-118686 A	15 1-14
X A	JP 2003-68464 A (CANON INC.) 07 March 2003, paragraph [0048] (Family: none)	15 1-14
X A	JP 2013-72087 A (NITTO DENKO CORPORATION) 22 April 2013, examples 1, 11, 12, 17 & US 2013/0074927 A1, examples 1, 11, 12, 17 & WO 2013/049062 A2 & EP 2760941 A2 & TW 201323476 A & CN 103562323 A & KR 10-2014-0074257 A	15 1-14
A	KR10-2017-0116927 A (LG CHEMICAL LTD.) 20 October 2017, entire text, all drawings (Family: none)	1-15
A	JP 2017-191661 A (IDEMITSU KOSAN CO., LTD.) 19 October 2017, entire text, all drawings (Family: none)	1-15

Form PCT/ISA/210 (continuation of second sheet) (January 2015)

REFERENCES CITED IN THE DESCRIPTION

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