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(54) LIGHT-EMITTING MATERIAL FOR ORGANIC ELECTROLUMINESCENT DEVICE, ORGANIC ELECTROLUMINESCENT DEVICE USING SAME, AND MATERIAL FOR ORGANIC ELECTROLUMINESCENT DEVICE

LICHTEMITTIERENDES MATERIAL FÜR ORGANISCHE ELEKTROLUMINESZIERENDE VORRICHTUNG, DARAUF BASIERENDE ORGANISCHE ELEKTROLUMINESZIERENDE VORRICHTUNG UND MATERIAL FÜR ORGANISCHE ELEKTROLUMINESZIERENDE VORRICHTUNG

MATERIAU ELECTROLUMINESCENT POUR DISPOSITIF ORGANIQUE ELECTROLUMINESCENT ET DISPOSITIF ORGANIQUE ELECTROLUMINESCENT UTILISANT LEDIT MATERIAU

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(56) References cited:

EP-A- 1 009 044 EP-A- 1 496 041
JP-A- 11 228 951 JP-A- 2001 097 897
JP-A- 2003 086 380 JP-A- 2003 146 951
JP-A- 2003 313 156 JP-A- 2004 059 535
US-B1- 6 582 837

 DATABASE WPI Week 200173 Thomson Scientific, London, GB; AN 2001-628873 XP002507806 & JP 2001 097897 A (IDEMITSU KOSAN CO LTD) 10 April 2001 (2001-04-10) & JP 2001 097897 A (IDEMITSU KOSAN CO) 10 April 2001 (2001-04-10)

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Description

TECHNICAL FIELD

[0001] The present invention relates to a light emitting material for an organic electroluminescence device, an organic electroluminescent device using the material and a material for an organic electroluminescent device, in particular, to an organic electroluminescence device exhibiting high current efficiency and having a long lifetime, and also to a light-emitting material for an organic electroluminescence device and a material for an organic electroluminescent device for realizing the organic EL device.

BACKGROUND ART

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[0002] An organic electroluminescence ("electroluminescence" will be occasionally referred to as "EL", hereinafter) device is a spontaneous light emitting device which utilizes the principle that a fluorescent substance emits light by energy of recombination of holes injected from an anode and electrons injected from a cathode when an electric field is applied. Since an organic EL device of the laminate type driven under a low electric voltage was reported by C. W. Tang et al. of Eastman Kodak Company (C. W Tang and S. A. Vanslyke, Applied Physics Letters, Volume 51, Pages 913, 1987), many studies have been conducted on organic EL devices using organic materials as the constituting materials. [0003] Tang et al. used a laminate structure using tris(8-hydroxyquinolinol aluminum) for the light emitting layer and a triphenyldiamine derivative for the hole transporting layer. Advantages of the laminate structure are that the efficiency of hole injection into the light emitting layer can be increased, that the efficiency of forming excited particles which are formed by blocking and recombining electrons injected from the cathode can be increased, and that excited particles formed among the light emitting layer can be enclosed

[0004] As the structure of the organic EL device, a two-layered structure having a hole transporting (injecting) layer and an electron transporting and light emitting layer and a three-layered structure having a hole transporting (injecting) layer, a light emitting layer and an electron transporting (injecting) layer are well known. To increase the efficiency of recombination of injected holes and electrons in the devices of the laminate type, the structure of the device and the process for forming the device have been studied. As the light emitting material of the organic EL device, chelate complexes such as tris(8-quinolinolato)aluminum, coumarine derivatives, tetraphenylbutadiene derivatives, bisstyrylarylene derivatives and oxadiazole derivatives are known. It is reported that light in the visible region ranging from blue light to red light can be obtained by using these light emitting materials, and development of a device exhibiting color images is expected (refer to, for example, Patent literatures 1, 2 and 3).

[0005] In addition, Patent literature 4 discloses a device having phenylanthracene derivatives as a light emitting material. Although the anthracene derivatives were used as a blue light emitting material, it has been required to extend a lifetime of the device. A material having naphthyl groups at 9th and 10th positions of anthracene is disclosed in Patent literature 5 and a material having fluoranthene at 9th and 10th positions for a device is disclosed in Patent literature 6. Although these anthracene derivatives were used as a blue light emitting material, it has been required to improve a lifetime of the device.

[0006] Further, Patent literature 7 discloses various types of anthracene derivatives to be used for a hole transporting material. However, these have not yet been evaluated as a light emitting material.

[Patent literature 1] Japanese Patent Application Laid-Open No. Heisei 8(1996)-239655

[Patent literature 2] Japanese Patent Application Laid-Open No. Heisei 7(1995)-138561

[Patent literature 3] Japanese Patent Application Laid-Open No. Heisei 3(1991)-200289

[Patent literature 4] Japanese Patent Application Laid-Open No. Heisei 8(1996)-012600

[Patent literature 5] Japanese Patent Application Laid-Open No. Heisei 11(1999)-3782

[Patent literature 6] Japanese Patent Application Laid-Open No. 2001-257074

[Patent literature 7] Japanese Patent Application Laid-Open No. 2000-182776

Patent Literature 8: EP 1 496 041 A1

Patent Literature 9: JP 2001 097897 A

Patent Literature 10: US 6,582,837 B1

Patent Literature 11: EP 1 009 044 A2

Patent Literature 12: JP 11-323323 A

Patent Literature 13: JP 2003-146951 A

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Patent Literature 8 discloses devices containing at least one layer with an anthracene derivative. Patent Literature 9 to 11 disclose asymmetrical anthracene derivatives containing two anthryl groups or a 1-naphthyl group bonded to the anthryl group in the 9-position. The asymmetrical compounds which are disclosed in Patent Literature 12 and 13 contain

identical groups as substituents in the positions 9 and 10 of anthracene.

DISCLOSURE OF THE INVENTION

[0007] The present invention has been made to overcome the above problems and has an objective of providing an organic electroluminescence device exhibiting a great efficiency of light emission and having a long lifetime, and also to a light emitting material for realizing the organic EL device and a material for an organic electroluminescent device for realizing the organic EL device.

[0008] As a result of intensive researches and studies to achieve the above objective by the present inventors, it was found that employing a compound having a specific asymmetric anthracene structure represented by the following general formula (1) as a light emitting material of an organic EL device enables to provide the organic EL device exhibiting a great efficiency of light emission and having a long lifetime.

[0009] In other word, the present invention provides a material for an organic EL device comprising an asymmetric anthracene derivative represented by the following general formula (1), and an organic EL device comprising at least one of organic thin film layers including a light emitting layer interposed between a pair of electrode consisting of an anode and a cathode, wherein the organic thin film layer contains at least one selected from the aforementioned asymmetric anthracene derivatives represented the general formula (1) singly or as a component of mixture thereof.

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[0010] In the general formula (1),

A¹ and A² each independently represents a substituted or unsubstituted condensed aromatic hydrocarbon ring group selected from the group consisting of 2-naphthyl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-pyrenyl group, 4-pyrenyl group, 3-methyl-2-naphthyl group and 4-methyl-1-naphthyl group;

Ar¹ and Ar² each independently represents a hydrogen atom, or a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50;

R¹ to R³ each independently represents a hydrogen atom, a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50, a substituted or unsubstituted aromatic hetero ring group having ring atoms of 5 to 50, a substituted or unsubstituted alkyl group having carbon atoms of 1 to 50, a substituted or unsubstituted cycloalkyl group having carbon atoms of 3 to 50, a substituted alkoxy group having carbon atoms of 1 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted alkoxycarbonyl group having carbon atoms of 1 to 50, a substituted or unsubstituted silyl group, a carboxyl group, a halogen atom, a cyano group, a nitro group or a hydroxyl group;

R⁹ to R¹⁰ each independently represents a hydrogen atom, a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50, a substituted or unsubstituted alkyl group having carbon atoms of 1 to 50, a substituted or unsubstituted or unsubstituted alkoxy group having carbon atoms of 1 to 50, a substituted or unsubstituted aralkyl group having carbon atoms of 6 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted alkoxycarbonyl group having carbon atoms of 1 to 50, a substituted

or unsubstituted silyl group, a carboxyl group, a halogen atom, a cyano group, a nitro group or a hydroxyl group, and none of R⁹ and R¹⁰ is an alkenyl group;

Ar¹, Ar², R⁹ and R¹⁰ each may be a plural number, and two neighboring groups thereof may form a saturated or unsaturated ring structure; and wherein

(I) A¹ is different from A²,

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(II) when A¹ is the same with A², (II-i) Ar¹ is different from Ar², or (II-ii) R⁹ is different from R¹⁰, and

(II-iii) when Ar¹ and Ar² are the same with each other, and R⁹ and R¹⁰ are the same with each other,

(II-iii-1) the bonding position of A^1 to 9th position of the anthracene ring is different from the bonding position of A^2 to 10th position of the anthracene ring,

(II-iii-2) when both Ar^1 and Ar^2 are not hydrogen atoms, the bonding position of Ar^1 to A^1 is different from the bonding position of Ar^2 to A^2 , and

(II-iii-3) when both R^9 and R^2 are not hydrogen atoms, the bonding position of R^9 to A^1 is different from the bonding position of R^{10} to A^2 .

[0011] The light emitting material for an organic EL device or the organic EL device employed the asymmetric anthracene derivatives of the present invention exhibits high current efficiency and has a long lifetime.

INDUSTRIAL APPLICABILITY

[0012] As explained above in details, the light emitting material for an organic EL device or the organic EL device employed the asymmetric anthracene derivatives of the present invention represented by the general formula (1) exhibits high current efficiency and has a long lifetime. Therefore, the organic EL device is valuable for potential continuous use for a prolonged period.

THE PREFERRED EMBODIMENT TO CARRY OUT THE INVENTION

[0013] The light emitting material for an organic EL device comprises the asymmetric anthracene derivative represented by the general formula (1). It is excluded a case where the groups at 9th and 10th positions of anthracene at the core are symmetrical at x-y axis of symmetry and bond each other in the asymmetric derivatives represented by the general formula (1), which means that the general formula (1) has the following structures:

- (I) A¹ is different from A².
- (II) when A^1 is the same with A^2 , (11-i) Ar^1 is different from Ar^2 , or (II-ii) R^9 is different from R^{10} , and

(II-iii) when Ar¹ and Ar² are the same with each other, and R⁹ and R¹⁰ are the same with each other,

(II-iii-1) the bonding position of A^1 to 9th position of the anthracene ring is different from the bonding position of A^2 to 10th position of the anthracene ring,

(II-iii-2) when both Ar^1 and Ar^2 are not hydrogen atoms, the bonding position of Ar^1 to A^1 is different from the bonding position of Ar^2 to A^2 , and

(II-iii-3) when both R^9 and R^2 are not hydrogen atoms, the bonding position of R^9 to A^1 is different from the bonding position of R^{10} to A^2 .

[0014] In the general formula (1), A¹ and A² each independently is a substituted or unsubstituted condensed aromatic hydrocarbon ring having ring carbon atoms of 10 to 20, preferably 10 to 16, namely a 2-naphthyl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-pyrenyl group, 4-pyrenyl group, 3-methyl-2-naphthyl group or 4-methyl-1-naphthyl group. Among those, 2-naphthyl group and 9-phenanthryl group are preferred.

In the general formula (1), Ar¹ and Ar² each independently represents a hydrogen atom or a substituted or unsubstituted aromatic hydrocarbon ring having ring carbon atoms of 6 to 50, preferably 6 to 16.

[0015] Examples of the substituted or unsubstituted aromatic hydrocarbon ring group represented by Ar^1 and Ar^2 include phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 9-phenanthryl group, 9-phenanthryl group, 2-pyrenyl group, 2-pyrenyl group, 2-biphenylyl group, 3-pyrenyl group, 3-pyrenyl

[0016] p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-tolyl group, p-tolyl group, p-tolyl group, p-tolyl group, d-methyl-2-naphthyl group, 4-methyl-2-naphthyl group, d-methyl-2-naphthyl group,

biphenylyl group, 4-biphenylyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group,

1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenylyl group, 4"-t- butyl-p-terphenyl-4-yl group and the like. [0017] Among those, phenyl group, 1-naphthyl group, 2-naphthyl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenylyl group, 3biphenylyl group, 4-biphenylyl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group are preferred. [0018] In the general formula (1), R¹ to R⁸ each independently represents a hydrogen atom, a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50, a substituted or unsubstituted aromatic hetero ring group having ring atoms of 5 to 50, a substituted or unsubstituted alkyl group having carbon atoms of 1 to 50, a substituted or unsubstituted cycloalkyl group having carbon atoms of 3 to 50, a substituted or unsubstituted alkoxy group having carbon atoms of 1 to 50, a substituted or unsubstituted aralkyl group having carbon atoms of 6 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted arylthio group having carbon atoms of 5 to 50, a substituted or unsubstituted alkoxycarbonyl group having carbon atoms of 1 to 50, a substituted or unsubstituted silyl group, a caboxyl group, a halogen atom, a cyano group, a nitro group or a hydroxyl group. [0019] R⁹ to R¹⁰ each independently represents a hydrogen atom, a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50, a substituted or unsubstituted alkyl group having carbon atoms of 1 to 50, a substituted or unsubstituted cycloalkyl group having carbon atoms of 3 to 50, a substituted or unsubstituted alkoxy group having carbon atoms of 1 to 50, a substituted or unsubstituted aralkyl group having carbon atoms of 6 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted arylthio group having carbon atoms of 5 to 50, a substituted or unsubstituted alkoxycarbonyl group having carbon atoms of 1 to 50, a substituted or unsubstituted silyl group, a caboxyl group, a halogen atom, a cyano group, a nitro group or a hydroxyl group, and none of R9 and R10 is alkenyl group. Examples of a substituted or unsubstituted aromatic hydrocarbon ring $group\ represented\ by\ R^1\ to\ R^{10}\ include\ phenyl\ group,\ 1-naphthyl\ group,\ 2-naphthyl\ group,\ 1-anthryl\ group,\ 2-anthryl\ group,\$ group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenylyl group, 3-biphenylyl group, 4-biphenylyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, mtolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl) phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'-methylbiphenylyl group, 4"-t- butyl-p-terphenyl-4-yl group and the like. [0020] Examples of a substituted or unsubstituted aromatic heteroring group represented by R¹ to R⁸ include 1-pyrrolyl group, 2-pyrrolyl group, 3-pyrrolyl group, pyradinyl group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 1-indolyl group, 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group, 2-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5-isoindolyl group, 6-isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, 4-benzofuranyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group, quinolyl group, 3-quinolyl group, 4quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxanyl group, 5-quinoxanyl group, 6-quinoxanyl group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 9-carbazolyl group, 1-phenanthridinyl group, 2-phenanthridinyl group, 3-phenanthridinyl group, 4-phenanthridinyl group, 6-phenanthridinyl group, 7-phenanthridinyl group, 8-phenanthridinyl group, 9-phenanthridinyl group, 10-phenanthridinyl group, 1-acridinyl group, 2-acridinyl group, 3-acridinyl group, 4-acridinyl group, 9-acridinyl group, 1,7-phenanthrolin-2-yl group, 1,7-phenanthrolin-3-yl group, 1,7-phenanthrolin-4-yl group, 1,7-phenanthrolin-5-yl group, 1,7-phenanthro anthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10-yl group, 1,8phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4-yl group, 1,8-phenanthrolin-5-yl group, 1,8-phenan phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenan phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrolin-4-yl group, 2,9-phenanthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-8-yl group, 2,9-phenanthrolin-10yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8-phenanthrolin-4-yl group, 2,8-phenanthrolin-5yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8-phenanthrolin-9-yl group, 2,8-phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phenanthrolin-4-yl group, 2,7-phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenanthrolin-10-yl group, 1-phenazinyl group, 2-phenazinyl group, 1-phenothiazinyl group, 2-phenothiazinyl group, 3-phenothiazinyl group, 4-phenothiazinyl group, 10-phenothiazinyl group, 1-phenoxazinyl group, 2-phenoxazinyl group, 3-phenoxazinyl group, 4-phenoxazinyl group, 10-phenoxazinyl group, 2-oxazolyl group, 4-oxazolyl group, 5-oxazolyl group, 2-oxadiazolyl group, 5-oxadiazolyl group, 3-furazanyl group, 2-thienyl group, 3-thienyl group, 2-methylpyrrol-1-yl group, 2-methylpyrrol-3-yl group, 2-methylpyrrol-4-yl group, 2-methylpyrrole-5-yl group, 3-methylpyrrole-1-yl group, 3-methylpyrrole-2-yl group,

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3-methylpyrrole-4-yl group, 3- methylpyrrole-5-yl group, 2-t-butylpyrrole-4-yl group, 3-(2-phenylpropyl)pyrrole-1-yl group, 2-carbinyl-1-indolyl group, 4-methyl-1-indolyl group, 2-methyl-3-indolyl group, 4-methyl-3-indolyl group, 2-t-butyl-1-indolyl group, 4-t-butyl-3-indolyl group, 4-t-but

[0021] Examples of a substituted or unsubstituted alkyl group represented by R¹ to R¹0 includes methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-pentyl group, n-hexyl group, n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 1,2-dihydroxyethyl group, 1,3-dihydroxy-isopropyl group, 2,3-dihydroxy-t-butyl group, 1,2-dichloroethyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-dibromo-t-butyl group, 1,2-di-diodoethyl group, 1,2-diiodoethyl group, 1,3-diiodoisopropyl group, iodomethyl group, 1-iodoethyl group, 2-iodoethyl group, 2-iodoisobutyl group, 1-aminoethyl group, 2-aminoethyl group, 2-aminoethyl group, 2-aminoethyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diiodo-t-butyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diiodo-t-butyl group, 1,2-diaminoethyl group, 2-cyanoethyl group, 2-cyanoethyl group, 1,2-dicyanoethyl group, 1,2-dicyanoethyl group, 1,2-dicyanoethyl group, 1,2-diodoethyl group, 2-cyanoethyl group, 2-cyanoethyl group, 1,2-diiodoethyl group, 1,2-diiodoethyl group, 2-cyanoethyl group, 2-cyanoethyl group, 2-cyanoethyl group, 1,2-dicyanoethyl group, 1,2-diiodoethyl group, 1,2-diiodoethyl group, 1,2-diiodoethyl group, 1,2-diiodoethyl group, 2-cyanoethyl group, 2-cyanoethyl group, 2-cyanoethyl group, 1,2-diiodoethyl group, 1,2-d

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[0022] Examples of s substituted or unsubstituted cycloalkyl group represented by R¹ to R¹⁰ include cyclopropyl group, cyclobutyl group, cyclopentyl group, d-methylcyclohexyl group, 1-adamanthyl group, 2-adamanthyl group, 1-norbornyl group, 2-norbornyl group and the like.

[0023] A substituted or unsubstituted alkoxy group represented by R¹ to R¹⁰ is represented by -OY. and examples of Y include methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-pentyl group, n-hexyl group, n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 2-hydroxyisobutyl group, 1,2-dihydroxyethyl group, 1,3-dihydroxy-isopropyl group, 2,3-dihydroxy-t-butyl group, 1,2-3-trihydroxypropyl group, chloromethyl group, 1-chloroethyl group, 2-chloroethyl group, 2-chloroisobutyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2-bromoethyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-dibromo-t-butyl group, 1,2-diiodoethyl group, 1,3-diiodoisopropyl group, 2,3-diiodo-t-butyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2-diiomoethyl group, 2-aminoisobutyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2-dicyanoethyl group, 1,3-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2-dicyano-t-butyl group, 1,2-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2-dicyano-t-butyl group, 1,2-dicyano-t-but

[0024] 1-nitroethyl group, 2-nitroethyl group, 2-nitroisobutyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 2,3-dinitro-t-butyl group, 1,2,3-trinitropropyl group and the like.

[0025] Examples of a substituted or unsubstituted aralkyl group represented by R1 to R10 includes benzyl group, 1phenylethyl group, 2-phenylethyl group, 1-phenyl-isopropyl group, 2-phenylisopropyl group, phenyl-t-butyl group, αnaphthylmethyl group, $1-\alpha$ -naphthylethyl group, $2-\alpha$ -naphthylethyl group, $1-\alpha$ -naphthylisopropyl group, $2-\alpha$ -naphthylisopropyl group, $1-\alpha$ -naphthylis propyl group, β-naphthylmethyl group, 1-β-naphthylethyl group, 2-6-naphthylethyl group, 1-β-naphthylisopropyl group, 2-β-naphthylisopropyl group, 1-pyrrolylmethyl group, 2-(1-pyrrolyl)ethyl group, p-methylbenzyl group, m-methylbenzyl group, o-methylbenzyl group, p-chlorobenzyl group, m-chlorobenzyl group, o-chlorobenzyl group, p-bromobenzyl group, m-bromobenzyl group, o-bromobenzyl group, p-iodobenzyl group, m-iodobenzyl group, o-iodobenzyl group, p-hydroxybenzyl group, m-hydroxybenzyl group, o-hydroxybenzyl group, p-aminobenzyl group, m-aminobenzyl group, o-aminobenzyl group, p-nitrobenzyl group, m-nitrobenzyl group, o-nitrobenzyl group, p-cyanobenzyl group, m-cyanobenzyl group, o-cyanobenzyl group, 1-hydroxy-2-phenylisopropyl group, 1-chloro-2-phenylisopropyl group and the like. A substituted or unsubstituted aryloxy group represented by R1 to R10 is represented by -OY', and examples of Y' include phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenylyl group, 3-biphenylyl group, 4-biphenylyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, m-terphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-t-butylphenyl group, p-(2-phenylpropyl) phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'methylbiphenyl-yl group, 4"-t- butyl-p-terphenyl-4-yl group, 2-pyrrolyl group, 3-pyrrolyl group, pyradinyl group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 1-indolyl group, 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5-isoindolyl group, 6isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, furanyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group,

2-quinolyl group, 3-quinolyl group, 4-quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxanyl group, 5-quinoxanyl group, 6-quinoxanyl group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 1-phenanthridinyl group, 2-phenanthridinyl group, 3-phenanthridinyl group, 4phenanthridinyl group, 6-phenanthridinyl group, 7-phenanthridinyl group, 8-phenanthridinyl group, 9-phenanthridinyl group, 10-phenanthridinyl group, 1-acridinyl group, 2-acridinyl group, 3-acridinyl group, 4-acridinyl group, 9-acridinyl $group,\,1,7-phen anthrolin-2-yl\,group,\,1,7-phen anthrolin-3-yl\,group,\,1,7-phen anthrolin-4-yl\,group,\,1,7-phen anthrolin-5-yl\,group,\,1,7-phen anthrolin-5-y$ group, 1,7-phenanthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10yl group, 1,8-phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4-yl group, 1,8-phenanthrolin-5yl group, 1,8-phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9-phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrolin-4-yl group, 2,9-phenanthroli anthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-8-yl group, 2,9-phenanthro anthrolin-10-yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8-phenanthrolin-4-yl group, 2,8phenanthrolin-5-yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8-phenanthrolin-9-yl group, 2,8phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phenanthrolin-4yl group, 2,7phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenan phenanthrolin-10-yl group, 1-phenazinyl group, 2-phenazinyl group, 1-phenothiazinyl group, 2-phenothiazinyl group, 3phenothiazinyl group, 4-phenothiazinyl group, 1-phenoxazinyl group, 2-phenoxazinyl group, 3-phenoxazinyl group, 4phenoxazinyl group, 2-oxazolyl group, 4-oxazolyl group, 5-oxazolyl group, 2-oxadiazolyl group, 5-oxadiazolyl group, 3furazanyl group, 2-thienyl group, 3-thienyl group, 2-methylpyrrol-1-yl group, 2-methylpyrrol-3-yl group, 2-methylpyrrol-4-yl group, 2-methylpyrrole-5-yl group, 3-methylpyrrole-1-yl group, 3- methylpyrrole-2-yl group, 3-methylpyrrole-4-yl group, 3- methylpyrrole-5-yl group, 2-t-butylpyrrole-4-yl group, 3-(2-phenylpropyl)pyrrole-1-yl group, 2-methyl-1-indolyl group, 4-methyl-1-indolyl group, 2-methyl-3-indolyl group, 4-methyl-3-indolyl group, 2-t-butyl-1-indolyl group, 4-t-butyl-1-indolyl group, 2-t-butyl-3-indolyl group, 4-t-butyl-3-indolyl group and the like.

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[0026] A substituted or unsubstituted arylthio group represented by R¹ to R¹⁰ is represented by -SY", and examples of Y" include phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthryl group, 1phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 9-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 4-pyrenyl group, 2-biphenylyl group, 3-biphenylyl group, 4-biphenylyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, p-terphenyl-2-yl group, mterphenyl-4-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, o-tolyl group, m-tolyl group, p-tolyl group, p-tolyl group, p-tolyl group, p-tolyl group, m-terphenyl-3-yl group, m-terphenyl-3-yl group, m-terphenyl-3-yl group, m-terphenyl-3-yl group, m-terphenyl-3-yl group, m-tolyl group, m-tolyl group, p-tolyl group, m-tolyl gro butylphenyl group, p-(2-phenylpropyl) phenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl- $1-anthryl\,group, 4'-methylbiphenyl-yl\,group, 4"-t-\,butyl-p-terphenyl-4-yl\,group, 2-pyrrolyl\,group, 3-pyrrolyl\,group, pyradinyl\,group, 2-pyrrolyl\,group, 3-pyrrolyl\,group, 2-pyrrolyl\,group, 3-pyrrolyl\,group, 3$ group, 2-pyridinyl group, 3-pyridinyl group, 4-pyridinyl group, 1-indolyl group, 2-indolyl group, 3-indolyl group, 4-indolyl group, 5-indolyl group, 6-indolyl group, 7-indolyl group, 1-isoindolyl group, 3-isoindolyl group, 4-isoindolyl group, 5isoindolyl group, 6-isoindolyl group, 7-isoindolyl group, 2-furyl group, 3-furyl group, 2-benzofuranyl group, 3-benzofuranyl group, 4-benzofuranyl group, 5-benzofuranyl group, 6-benzofuranyl group, 7-benzofuranyl group, 1-isobenzofuranyl group, 3-isobenzofuranyl group, 4-isobenzofuranyl group, 5-isobenzofuranyl group, 6-isobenzofuranyl group, 7-isobenzofuranyl group, 2-quinolyl group, 3-quinolyl group, 4-quinolyl group, 5-quinolyl group, 6-quinolyl group, 7-quinolyl group, 8-quinolyl group, 1-isoquinolyl group, 3-isoquinolyl group, 4-isoquinolyl group, 5-isoquinolyl group, 6-isoquinolyl group, 7-isoquinolyl group, 8-isoquinolyl group, 2-quinoxanyl group, 5-quinoxanyl group, 6-quinoxanyl group, 1-carbazolyl group, 2-carbazolyl group, 3-carbazolyl group, 4-carbazolyl group, 1-phenanthridinyl group, 2-phenanthridinyl group, 3phenanthridinyl group, 4-phenanthridinyl group, 6-phenanthridinyl group, 7-phenanthridinyl group, 8-phenanthridinyl group, 9-phenanthridinyl group, 10-phenanthridinyl group, 1-acridinyl group, 2-acridinyl group, 3-acridinyl group, 4acridinyl group, 9-acridinyl group, 1,7-phenanthrolin-2-yl group, 1,7-phenanthrolin-3-yl group, 1,7-phenanthrolin-4-yl group, 1,7-phenanthrolin-5-yl group, 1,7-phenanthrolin-6-yl group, 1,7-phenanthrolin-8-yl group, 1,7-phenanthrolin-9-yl group, 1,7-phenanthrolin-10-yl group, 1,8-phenanthrolin-2-yl group, 1,8-phenanthrolin-3-yl group, 1,8-phenanthrolin-4yl group, 1,8-phenanthrolin-5-yl group, 1,8-phenanthrolin-6-yl group, 1,8-phenanthrolin-7-yl group, 1,8-phenanthrolin-9-yl group, 1,8-phenanthrolin-10-yl group, 1,9-phenanthrolin-2-yl group, 1,9-phenanthrolin-3-yl group, 1,9-phenanthrolin-4-yl group, 1,9-phenanthrolin-5-yl group, 1,9-phenanthrolin-6-yl group, 1,9-phenanthrolin-7-yl group, 1,9-phenanthrolin-8-yl group, 1,9-phenanthrolin-10-yl group, 1,10-phenanthrolin-2-yl group, 1,10-phenanthrolin-3-yl group, 1,10-phenanthrolinthrolin-4-yl group, 1,10-phenanthrolin-5-yl group, 2,9-phenanthrolin-1-yl group, 2,9-phenanthrolin-3-yl group, 2,9-phenanthrol anthrolin-4-yl group, 2,9-phenanthrolin-5-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthrolin-7-yl group, 2,9-phenanthrolin-6-yl group, 2,9-phenanthro anthrolin-8-yl group, 2,9-phenanthrolin-10-yl group, 2,8-phenanthrolin-1-yl group, 2,8-phenanthrolin-3-yl group, 2,8phenanthrohn-4-yl group, 2,8-phenanthrolin-5-yl group, 2,8-phenanthrolin-6-yl group, 2,8-phenanthrolin-7-yl group, 2,8phenanthrolin-9-yl group, 2,8-phenanthrolin-10-yl group, 2,7-phenanthrolin-1-yl group, 2,7-phenanthrolin-3-yl group, 2,7-phena

phenanthrolin-4-yl group, 2,7-phenanthrolin-5-yl group, 2,7-phenanthrolin-6-yl group, 2,7-phenanthrolin-8-yl group, 2,7-phenanthrolin-9-yl group, 2,7-phenanthrolin-10-yl group, 1-phenazinyl group, 2-phenazinyl group, 1-phenothiazinyl group, 2-phenothiazinyl group, 2-phenothiazinyl group, 2-phenoxazinyl grou

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[0027] A substituted or unsubstituted alkoxycarbonyl group for R¹ to R¹0 is represented by -COOZ, and examples of Z include methyl group, ethyl group, propyl group, isopropyl group, n-butyl group, s-butyl group, isobutyl group, t-butyl group, n-pentyl group, n-hexyl group, n-heptyl group, n-octyl group, hydroxymethyl group, 1-hydroxyethyl group, 2-hydroxyethyl group, 2-hydroxyisobutyl group, 1,2-dihydroxyethyl group, 1,3-dihydroxy-isopropyl group, 2,3-dihydroxy-t-butyl group, 1,2,3-trihydroxypropyl group, chloromethyl group, 1-chloroethyl group, 2-chloroethyl group, 2-chloroisobutyl group, 1,2-dichloroethyl group, 1,3-dichloroisopropyl group, 2,3-dichloro-t-butyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2-bromoethyl group, 2-bromoisobutyl group, 1,2-dibromoethyl group, 1,3-dibromoisopropyl group, 2,3-diiodoethyl group, 1,2-diiodoethyl group, 2-iodoethyl group, 2-iodoethyl group, 1,2-diiodoethyl group, 1,3-diiodoethyl group, 2-aminoisobutyl group, 1,2-diaminoethyl group, 1,3-diaminoisopropyl group, 2,3-diamino-t-butyl group, 1,2-dicyanoethyl group, cyanomethyl group, 1,2-dicyanoethyl group, 1,3-dicyanoisopropyl group, 2,3-dicyano-t-butyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 1,3-dinitro-t-butyl group, 2-nitroethyl group, 2-nitroisobutyl group, 1,2-dinitroethyl group, 1,3-dinitroisopropyl group, 2,3-dinitro-t-butyl group, 1,2-dinitropropyl group and the like.

[0028] Examples of a halogen atom for R¹ to R¹⁰ include fluorine atom, chlorine atom, bromine atom and the like [0029] A substituent for the Ar¹, Ar² and R¹ to R¹⁰ includes halogen atom, hydroxyl group, nitro group, cyano group, alkyl group, aryl group, cycloalkyl group, alkoxy group, aralkyl group, aryloxy group, arylthio group, alkoxycarbonyl group, carboxyl group or the like.

[0030] Ar¹, Ar², R⁹ and R¹⁰ each may be a plural number, and two neighboring groups thereof may form a saturated or unsaturated ring structure, and the ring structure includes, in addition to unsaturated 6 member ring such as benzene, saturated or unsaturated 5 member or 7 member ring structure and the like.

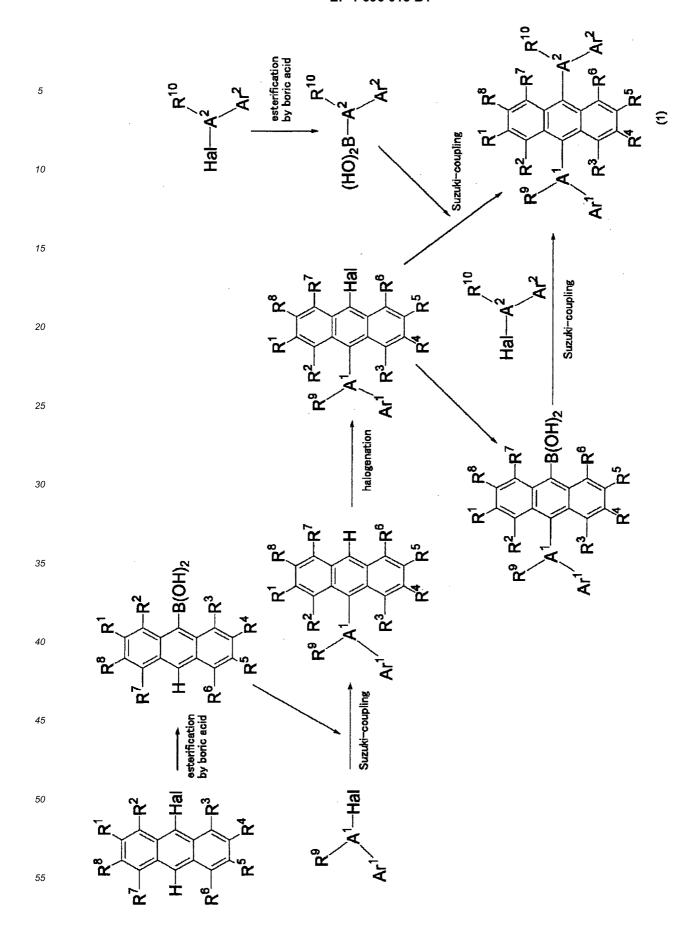
[0031] Further, in the present invention, it is preferable that the asymmetric anthracene derivatives represented by the aforementioned general formula (1) comprise a substituted or unsubstituted condensed aromatic hydrocarbon ring having ring carbon atoms of 12 to 20. Examples of the substituent includes the same with the substituent of the groups represented by Ar^{1} , Ar^{2} and R^{1} to R^{10}

[0032] Specific examples of the asymmetric anthracene of the present invention represented by the general formula (1) include as follows, but not limited thereto (compounds which are outside the range of the claims are distinguished with an asterisk):

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AN45 AN46

[0033] The asymmetric anthracene derivatives represented by the general formula (1) may be synthesized by using an aryl halide compound and an anthracene boronic acid compound as starting materials and applying appropriate combination of Suzuki-coupling reaction, halogenation reaction and esterification by boric acid through known methods. The following is a synthesis scheme:



[0034] Many reports have been made on the Suzuki-coupling reaction (Chem. Rev., Vol.95, No.7, 2457 (1995), etc.) and the reaction conditions described in the reports may be applied. The reaction may be carried out at normal pressure or as appropriate, under pressure in inert gas atmosphere such as nitrogen, argon and helium. The reaction temperature is in the range from 15 to 300 degC, preferably 30 to 200 degC. The reaction solvent includes water, aromatic hydrocarbon such as benzene, toluene and xylene, ether such as 1,2-dimethoxyethane, diethylether, methyl-t-butylether, tetrahydrofuran and dioxane, saturated hydrocarbon such as pentane, hexane, heptane, octane and cyclohexane, halide such as dichloromethane, chloroform, tetrachloromethane, 1,2-dichloroethane and 1,1,1-trichloroethane, nitrile such as acetonitrile and benzonitrile, ester such as ethylacetate, methylacetate and butylacetate, and amide such as N,N-dimethylformamide, N,N-dimethylacetoamide and N-methylpyrrolidone. These solvents may be used singly or as a component of mixture thereof. Among those, toluene, 1, 2-dimethoxyethane, dioxane and water are preferred. Amount of the solvent to be used is generally 3 to 50 fold by weight, preferably 4 to 20 fold by weight to aryl boronic acid or derivatives thereof. [0035] The base to be used for the reaction includes, for example, sodium carbonate, potassium carbonate, sodium hydroxide, potassium hydroxide, sodium bicarbonate, potassium bicarbonate, magnesium carbonate, lithium carbonate, potassium fluoride, cesium fluoride, cesium chloride, cesium bromide, cesium carbonate, potassium phosphate, sodium methoxide, potassium t-butoxide, sodium t-butoxide, lithium t-butoxide and the like, and sodium carbonate is preferable. Amount of the base to be used is generally 0.7 to 10 moles in equivalence, preferably 0.9 to 6 moles in equivalence to aryl boronic acid or derivatives thereof.

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[0036] The catalysts to be used for the reaction include, for example, palladium catalysts such as tetrakis(triphenylphosphine)palladium, dichlorobis(triphenylphosphine)palladium, dichloro[bis(diphenylphosphono)ethane]palladium, dichloro[bis(diphenylphosphono)butane]palladium, dichloro[bis(diphenylphosphono)butane]palladium, dichloro[bis(diphenylphosphono)butane]palladium, dichloro[bis(diphenylphosphono)ferrocene]palladium and the like, nickel catalysts such as tetrakis(triphenylphosphine)nickel, dichloro[bis(diphenylphosphono)ethane]nickel, dichloro[bis(diphenylphosphono)ferrocene]nickel dichloro[bis(diphenylphosphono)butane]nickel, dichloro[bis(diphenylphosphono)ferrocene]nickel and the like, and tetrakis(triphenylphosphine)palladium is preferable.

Amount of the catalyst to be used is generally 0.001 to 1 mole in equivalence, preferably 0.01 to 0.1 mole in equivalence to an aryl halide compound.

[0037] Halogen for the aryl halide compounds includes, for example, an iodine atom, a bromine atom, a chlorine atom and the like, and a iodine atom and a bromine atom are preferable. Although a halogenation agent for the halogenation reaction is not limited, for example, N-halogenated chlorosuccinimide is preferably used. Amount of the halogenation agent to be used is generally 0.8 to 10 moles in equivalence, preferably 1 to 5 moles in equivalence to an aryl compound. The reaction is generally carried out in an inert solvent under inert atmosphere such as nitrogen, argon, helium and the like. [0038] The inert solvent to be used includes, for example, N,N-dimethylformamide, N,N-dimethylacetoamide, N-methylpyrrolidone, dimethylsulfoxide, carbon tetrachloride, chlorobenzene, dichlorobenzene, nitrobenzene, toluene, xylene, methylcellosolve, ethylcellosolve, water and the like, and N,N-dimethylformamide and N-methylpyrrolidone are preferable. Amount of the solvent to be used is generally 3 to 50 fold by weight, preferably 5 to 20 fold by weight to an aryl compound. The reaction temperature is generally in the range from 0 to 200 degC, preferably 20 to 120 degC.

[0039] The esterification by boric acid may be carried out in accordance with known methods (Japan Chemical Society's editorial, The Experimental Chemistry Course No.4 edition, Vol 24, 61-90; J. Org. Chem., Vol.60, 7508 (1995), etc.) For example, by way of lithiation or Grignard reaction of an arylhalide compound, the esterification by boric acid is carried out generally under an inert atmosphere such as nitrogen, argon, helium and by using an inert solvent as a reaction solvent. The solvents include, for example, saturated hydrocarbon such as pentane, hexane, heptane, octane and cyclohexane, ether such as 1,2-dimethoxyethane, diethylether, methyl-t-butylether, tetrahydrofuran and dioxane, aromatic hydrocarbon such as benzene, toluene and xylene. These may be used singly or as mixture thereof, and dimethylether and toluene are preferred.

Amount of the solvent to be used is generally 3 to 50 fold by weight, preferably 4 to 20 fold by weight to an arylhalide compound.

[0040] The lithiation reagent to be used includes, for example, alkyl metal reagent suc as n-butyllithium, t-butyllithium, phenyllithium and methyllithium, amido-base such as lithium diisopropylamide and lithiumbistrimethylsilylamide, and n-butyllithium is preferred. Further, Grignard reagent may be prepared by reacting an arylhalide compound and a magnesium metal. Trialkyl borate to be used as esterification agent by boronic acid includes trimethyl borate, triisopropyl borate, triibutyl borate and the like, and trimethyl borate and triisopropyl borate are preferred. Each amount of the lithiation reagent and a magnesium metal to be used is generally from 1 to 10 moles in equivalence, preferably from 1 to 2 moles in equivalence respectively to an arylhalide compound. Amount of trialkyl borate to be used is generally from 1 to 10 moles in equivalence, preferably from 1 to 5 moles in equivalence to an arylhalide compound (or a pyrenylhalide compound). The reaction temperature is in the range from -100 to 50°C, preferably -75 to 10degC.

[0041] An organic EL device of the present invention which comprises at least one organic thin film layer, which contains at least a light emitting layer, interposed between a pair of electrode consisting of an anode and a cathode, wherein the organic thin film layer comprises at least one, singly or as a component of a mixture thereof. It is preferable

that the aforementioned light emitting layer comprises the aforementioned light emitting material for the organic EL device or the aforementioned material for the organic EL device singly or as a component of mixture thereof.

[0042] It is preferable that the aforementioned organic thin layer comprises the aforementioned light emitting material for the organic EL device or the aforementioned material for the organic EL device as a host material. In addition, the organic EL device of the present invention is preferred if the aforementioned light emitting layer contains further an arylamine compound and/or a styrylamine compound. The styrylamine compounds are shown by the following general formula (A):

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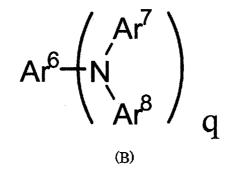
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$$Ar^3 + \left(N_{Ar^5}^{Ar^4}\right)$$

[0043] In the general formula above, Ar³ represents a group selected from a phenyl group, a biphenyl group, a terphenyl group, a stilbene group and a distyryl aryl group, and Ar⁴ and Ar⁵ each independently represents a hydrogen atom or an aromatic hydrocarbon ring group having 6 to 20 ring carbon atoms. Ar³, Ar⁴ and Ar⁵ each may be substituted, p represents an integer of 1 to 4, and more preferably, at least one of Ar⁴ and Ar⁵ is substituted with a styryl group.

[0044] In the preceding description, the aromatic hydrocarbon ring group having 6 to 20 carbon atoms includes preferably a phenyl group, a naphthyl group, an anthranil group, a phenanthryl group, a terphenyl group and the like. The preferred arylamine compounds are represented by the general formula (B):



[0045] In the general formula above, Ar⁶ to Ar⁸ each independently represents a substituted or unsubstituted aryl group having 5 to 40 ring carbon atoms. q represents an integer of 1 to 4.

[0046] In the preceding description, the aryl group having 5 to 40 ring carbon atoms include preferably a phenyl group, a naphthyl group, an anthranil group, a phenanthryl group, a pyrenyl group, a coronyl group, a biphenyl group, a terphenyl group, a pyrrolyl group, a furanyl group, a thiophenyl group, a benzthiophenyl group, an oxadiazolyl group, a diphenyl group, an indolyl group, a carbazolyl group, a pyridyl group, a benzoquinolyl group, a fluoranthenyl group, an acenaphthofluoranthenyl group, a stilbene group, a perylenyl group, a chrysenyl group, a picenyl group, a triphenyl group, a runicenyl group, a benzoanthracenyl group, a phenylanthranyl group, bisanthracenyl group or an aryl group represented by the following general formulae (C) and (D), and the like:

[0047] In the general formula (C), n represents an integer of 1 to 3. Additionally, preferable examples of the substituent for aforementioned aryl group include an alkyl group having 1 to 6 carbon atoms such as an ethyl group, a methyl group, an i-propyl group, a n-propyl group, a s-butyl group, a t-butyl group, a pentyl group, a hexyl group, a cyclopentyl group, a cyclohexyl group, etc., an alkoxy group having 1 to 6 carbon atoms such as an ethoxy group, a methoxy group, an ipropoxy group, a n-propoxy group, a s- butoxy group, a t-butoxy group, a pentoxy group, a hexyloxy group, a cyclopentoxy group, a cyclohexyloxy group, etc., an aryl group having 5 to 40 ring atoms, an amino group substituted with an aryl group having 5 to 40 ring atoms, an ester group which has an aryl group having 5 to 40 ring atoms, an ester group which has an alkyl group having 1 to 6 carbon atoms, a cyano group, a nitro group, a halogen atom and the like. Ar⁵ includes preferably a naphthyl group, an anthranyl group, a chrysenyl group, a pyrenyl group or an aryl group represented by the general formula (D), of which each may be substituted.

[0048] The following is a description of the construction of the organic EL device of the present invention. Typical examples of the construction of the organic EL device of the present invention include:

(1) an anode / a light emitting layer / a cathode;

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- (2) an anode / a hole injecting layer / a light emitting layer / a cathode;
- (3) an anode / a light emitting layer / an electron injecting layer / a cathode;
- (4) an anode / a hole injecting layer / a light emitting layer / an electron injecting layer / a cathode;
- (5) an anode / an organic semiconductor layer / a light emitting layer / a cathode;
- (6) an anode / an organic semiconductor layer / an electron barrier layer / a light emitting layer / a cathode;
- (7) an anode / an organic semiconductor layer / a light emitting layer / an adhesion improving layer / a cathode;
- (8) an anode / a hole injecting layer / a hole transporting layer / a light emitting layer / an electron injecting layer / a cathode;
- (9) an anode / an insulating layer / a light emitting layer / an insulating layer / a cathode;
- (10) an anode / an inorganic semiconductor layer / an insulating layer / a light emitting layer / an insulating layer / a cathode:
- (11) an anode / an organic semiconductor layer / an insulating layer / a light emitting layer / an insulating layer / a cathode:
- (12) an anode / an insulating layer / a hole injecting layer / a hole transporting layer / a light emitting layer / an insulating layer / a cathode; and
- (13) an anode / an insulating layer /a hole injecting layer / a hole transporting layer / a light emitting layer / an electron injecting layer / a cathode.

Among those, the construction (8) is generally employed in particular; however, the construction of the organic EL device is not limited to those shown above as the examples.

[0049] In general, the organic EL device is produced on a substrate which transmits light. It is preferable that the substrate which transmits light has a transmittance of light of 50% or greater in the visible region of 400 to 700 nm. It is also preferable that a flat and smooth substrate is employed.

[0050] As the substrate which transmits light, for example, glass sheet and synthetic resin sheet are advantageously employed. Specific examples of the glass sheet include soda ash glass, glass containing barium and strontium, lead glass, aluminosilicate glass, borosilicate glass, barium borosilicate glass, quartz and the like. In addition, specific examples of the synthetic resin sheet include sheet made of polycarbonate resins, acrylic resins, polyethylene terephthalate resins, polyether sulfide resins, polysulfone resins and the like.

[0051] The anode in the organic EL device of the present invention covers a role of injecting holes into a hole transport layer or into a light emitting layer, and it is effective that the anode has a work function of 4.5 eV or greater. Specific examples of the material for the anode include indium tin oxide alloy (ITO), tin oxide (NESA), gold, silver, platinum,

copper and the like.

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[0052] With regard to the cathode, its material preferably has a small work function with the aim of injecting electrons into an electron transport layer or into a light emitting layer. The anode can be prepared by forming a thin film of the electrode material described above in accordance with a process such as a vapor deposition process or a sputtering process. When the light emitted from the light emitting layer is observed through the anode, it is preferable that the anode has a transmittance of the emitted light greater than 10 %. It is also preferable that the sheet resistivity of the anode is several hundred Ω/\square or smaller. The thickness of the anode is, in general, selected in the range of from 10 nm to 1μ m and preferably in the range of from 10 to 200 nm depending on a kind of the materials.

[0053] In the organic EL device of the present invention, the light emitting layer has the following functions:

- (1) The injecting function: the function of injecting holes from the anode or the hole injecting layer and injecting electrons from the cathode or the electron injecting layer when an electric field is applied;
- (2) The transporting function: the function of transporting injected charges (electrons and holes) by the force of the electric field; and
- (3) The light emitting function: the function of providing the field for recombination of electrons and holes and leading the recombination to the emission of light.

As the process for forming the light emitting layer, a well known process such as the vapor deposition process, the spin coating process and the LB process can be employed.

[0054] It is preferable that a light emitting layer is a molecular sedimentation film particularly. Here, the molecular sedimentation film is defined as a thin film formed by sedimentation of a gas phase material compound or a thin film formed by condensation of a liquid phase material compound. The molecular sedimentation film may be differentiated from a thin film (a molecular build-up film) formed by the LB process, based on the differences between agglomeration structures and higher-order structures, and also the differences resulting from functionalities thereof. In addition, as shown in Japanese Patent Laid-open No. Showa57(1982)-51781, to form a light emitting layer, a thin film may be formed in accordance with the spin coating and the like of the solution to be prepared by dissolving a binder such as resin and a material compound in solvent.

[0055] In the present invention, any well known light emitting material other than a light emitting material consisting of an asymmetric pyrene derivative of the present invention may be optionally contained in the light emitting layer; or a light emitting layer containing other well known light emitting layer may be laminated with the light emitting layer containing the light emitting material of the present invention each in an extent of not obstructing to achieve the objective of the present invention respectively.

[0056] In the present invention, the hole injecting layer and the hole transporting layer are layers which assist injection of holes into the light emitting layer and transport the holes to the light emitting zone. The layers exhibit a great mobility of holes and, in general, have an ionization energy as small as 5.5 eV or smaller. For the hole injecting layer and the hole transporting layer, a material which transports holes to the light emitting layer at a small strength of the electric field is preferable. A material which exhibits, for example, a mobility of holes of at least 10⁻⁴ cm²/V • sec under application of an electric field of from 10⁴ to 10⁶ V/cm is preferable. As for such material, any arbitrary material selected from conventional material

[0057] commonly used as a charge transporting material for the holes in photoconducting materials and well known material employed for the hole injecting layer in the EL device is usable.

[0058] Further examples include triazole derivatives (refer to United States Patent No. 3,112,197, etc.), oxadiazole derivatives (refer to United States Patent No. 3,189,447, etc.), imidazole derivatives (refer to Japanese Examined Patent KOKOKU No. Shou 37-16096, etc.), poly arylalkane derivatives (refer to United States Patent Nos. 3,615,402, 3,820,989 and 3,542,544, Japanese Examined Patent KOKOKU Nos. Shou 45-555 and Shou 51-10983, Japanese Unexamined Patent Application Laid-Open Nos. Shou 51-93224, Shou 55-17105, Shou 56-4148, Shou 55-108667, Shou 55-156953, Shou 56-36656, etc.), pyrazoline derivatives and pyrazolone derivatives (refer to U.S Patent Nos. 3,180,729 and 4,278,746, Japanese Unexamined Application Patent Laid-Open Nos. Shou 55-88064, Shou 55-88065, Shou 49-105537, Shou 55-51086, Shou 56-80051, Shou 56-88141, Shou 57-45545, Shou 54-112637, Shou 55-74546, etc.), phenylenediamine derivatives (refer to U.S Patent No. 3,615,404, Japanese Examined Patent KOKOKU Nos. Shou 51-10105, Shou 46-3712 and Shou 47-25336, Japanese Unexamined Patent Application Laid-Open Nos. Shou 54-53435, Shou 54-110536, Shou 54-119925, etc.), arylamine derivatives (refer to U.S Patent Nos. 3,567,450, 3,180,703, 3,240,597, 3,658,520, 4,232,103, 4,175,961 and 4,012,376, Japanese Examined Patent KOKOKU Nos. Shou 49-35702 and Shou 39-27577, Japanese Unexamined Patent Application Laid-Open Nos. Shou 55-144250, Shou 56-119132 and Shou 56-22437, West German Patent No. 1,110,518, etc.), chalcone derivatives which is substituted with amino group (refer to U.S Patent No. 3,526,501, etc.), oxazole derivatives (disclosed in U.S Patent No. 3,257,203, etc.), styryl anthracene derivatives (refer to Japanese Unexamine Patent Application Laid-Open No. Shou 56-46234, etc.), fluorenone derivatives (refer to Japanese Unexamined Patent Application Laid-Open No. Shou 54-110837, etc.), hydrazone derivatives (refer

to U.S Patent Nos. 3,717,462, Japanese Unexamined Patent Application Laid-Open Nos. Shou 54-59143, Shou 55-52063, Shou 55-52064, Shou 55-46760, Shou 55-85495, Shou 57-11350, Shou 57-148749, Hei 2-311591, etc.), stilbene derivatives (refer to Japanese Unexamined Patent Application Laid-Open Nos. Shou 61-210363, Shou 61-228451, Shou 61-14642, Shou 61-72255, Shou 62-47646, Shou 62-36674, Shou 62-10652, Shou 62-30255, Shou 60-93455, Shou 60-94462, Shou 60-174749, Shou 60-175052, etc.), silazane derivatives (U.S Patent No. 4,950,950), polysilane-based copolymers (Japanese Unexamined Patent Application Laid-Open No. Hei 2-204996), aniline-based copolymers (Japanese Unexamined Patent Application Laid-Open No Hei 1-211399 (particularly, thiophene oligomer), etc.

[0059] With regard to the material of the hole injecting layer, the above materials are also employable, however, porphyrin compounds, aromatic tertiary amine compounds and styryl amine compounds (refer to U.S Patent No. 4,127,412, Japanese Unexamined Patent Application Laid-Open Nos. Shou 53-27033, Shou 54-58445, Shou 54-149634, Shou 54-64299, Shou 55-79450, Shou 55-144250, Shou 56-119132, Shou 61-295558, Shou 61-98353, Shou 63-295695, etc.) are preferable and the aromatic tertiary amine compounds are particularly preferable. Further examples include, for example, 4,4'-bis (N-(1-naphthyl) -N-phenylamino) biphenyl (abbreviated as NPD hereunder) having 2 fused aromatic rings in its molecular described in U.S Patent No. 5,061,569, 4,4',4"-tris (N-(3-methylphenyl) -N-phenylamino) triphenyl amine (abbreviated as MTDATA hereunder) made by connecting three triphenyl amine units to form a star burst type, etc. Further, in addition to the aforementioned aromatic dimethylidene based compounds described as a material for the light emitting layer, an inorganic compound such as p-type silicon, p-type silicon carbide or so is employable as the material for the hole injecting layer.

[0060] To form the hole injecting layer or the hole transporting layer, a thin film may be formed from the material for the hole injecting layer or the hole transporting layer, respectively, in accordance with a well known process such as the vacuum vapor deposition process, the spin coating process, the casting process and the LB process. Although the thickness of the hole injecting layer and the hole transporting layer is not particularly limited, the thickness is usually from 5 nm to 5 μ m. When a hole transportation zone comprises a compound of the present invention, the hole injecting / transporting layer may be constructed by a layer consisting of at least one of the aforementioned materials, and also the hole injecting / transporting layer may be laminated by a hole injecting / transporting layer consisting of a compound different from them.

[0061] In the organic EL device of the present invention, the organic semiconductor layer assists to inject the holes or to inject the electrons into the light emitting layer, and it is preferable for the organic semiconductor layer to have a electric conductivity of 10⁻¹⁰ S/cm or greater. With regard to a material for the organic semiconductor layer, electroconductive oligomers such as an oligomer having thiophene, an oligomer having arylamine disclosed in Japanese Laid-Open No.Heisei 8(1996)-193191 and so on, electroconductive dendrimers such as a dendrimer having an arylamine and so on are employable.

[0062] The electron injection / transporting layer in the organic EL device of the present invention is a layer which assists injection of electrons into the light emitting layer and exhibits a great mobility of electrons. Among the electron injecting layers, an adhesion improving layer is a layer made of a material exhibiting excellent adhesion with the cathode. As the material for the electron injecting layer, 8-hydroxyquinoline, metal complexes of derivatives thereof and oxadiazole derivatives are preferable. Examples of the 8-hydroxyquinoline and metal complexes of derivatives thereof include metal chelates of oxinoid compounds including chelates of oxine (in general, 8-quinolinol or 8-hydroxyquinoline). For example, tris(8-quinolinol)aluminum (Alq) can be employed as the electron injecting material. Further, examples of the oxadiazole derivatives include an electron transfer compound shown as the following general formulae:

$$Ar^{\frac{1}{2}} Ar^{\frac{1}{2}}$$

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$$Ar^{3}$$
 Ar^{4}
 Ar^{4}
 Ar^{5}

$$Ar^{\frac{6}{7}} Ar^{\frac{7}{7}}O - Ar^{\frac{8}{7}} Ar^{\frac{9}{7}}$$

[0063] In the general formulae above Ar^{1′}, Ar^{2′}, Ar^{3′}, Ar^{5′}, Ar^{6′} and Ar^{9′} each independently represents a substituted or unsubstituted aryl group, which may be the same with or different from each other; Ar^{4′}, Ar^{7′} and Ar^{8′} each independently represents a substituted or unsubstituted arylene group, which may be the same with or different from each other. Examples of the aryl group include a phenyl group, a biphenyl group, an anthranyl group, a perilenyl group and a pyrenyl group. Further, examples of the arylene group include a phenylene group, an anthranylene group, a perilenylene group, the pyrenylene group and the like. Furthermore, examples of the substituent include an alkyl group having 1 to 10 carbon atoms, an alkoxy group having 1 to 10 carbon atoms, a cyano group and the like. With regard to the electron transfer compound, those compounds having a thin film forming capability are preferable.

[0064] Specific examples of the electron transfer compounds are shown below:

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[0065] In the present invention, it is preferable that a reductive dopant is added in either the electron transporting zone or an interfacial zone between the cathode and the organic layer. The reductive dopant used in the present invention is defined as a substance which reduces the electron transporting compound. Examples of the reductive dopant include at least one compound selected from alkali metals, alkali metallic complexes, alkali metals compounds, rare earth metals, rare earth metallic complexes and rare earth metal compounds.

[0066] Examples of the preferable reductive dopant include at least one alkali metal selected from a group consisting of Na (the work function: 2.36 eV), K (the work function: 2.28 eV), Rb (the work function: 2.16 eV) and Cs (the work function: 1.95 eV) or at least one alkaline earth metals selected from a group consisting of Ca (the work function: 2.9eV), Sr (the work function: 2.0 to 2.5 eV) and Ba (the work function: 2.52eV); whose work function of 2.0 eV or less is particularly preferable. Among the above, the preferable reductive dopant include at least one alkali metal selected from a group consisting of K, Rb and Cs, the more preferred is Rb or Cs, and the most preferred is Cs. These alkali metals have particularly high ability of reduction so that improvement of an emission luminance and longer lasting of a lifetime of the organic EL device may be realized. In addition, a combination of two or more of alkali metals is also preferable as a reductive dopant having 2.9 eV or less of the work function. In particular, a combination of Cs, for example with Na, K or Rb, or Na and K is preferable. By a combination and containing Cs therein, the reduction ability can be demonstrated effectively, and improvement of an emission luminance and longer lasting of a lifetime of the organic EL device may be realized by adding it into an electron injecting area.

[0067] In the organic EL device of the present invention, an electron injecting layer formed with an insulating material or a semiconductor may be further interposed between the cathode and the organic thin film layer. The electron injecting layer effectively prevents leak in the electric current and improves the electron injecting capability. It is preferable that at least one metal compound selected from the group consisting of alkali metal chalcogenides, alkaline earth metal chalcogenides, alkali metal halides and alkaline earth metal halides is used as the insulating material. It is preferable that the electron injecting layer is constituted with the above alkali metal chalcogenides since the electron injecting property can be improved. Preferable examples of the alkali metal chalcogenides include Li₂O, LiO, Na₂S, Na₂Se and NaO. Preferable examples of the alkaline earth metal chalcogenide s include CaO, BaO, SrO, BeO, BaS and CaSe.

[0068] Preferable examples of the alkali metal halides include LiF, NaF, KF, LiCl, KCl, NaCl and the like. Preferable examples of the alkaline earth metal halides include fluorides such as CaF_2 , BaF_2 , SrF_2 , MgF_2 and BeF_2 and halides other than the fluorides.

[0069] Examples of the semiconductor constituting the electron transporting layer include oxides, nitrides and oxide nitrides containing at least one element selected from Ba, Ca, Sr, Yb, Al, Ga, In, Li, Na, Cd, Mg, Si, Ta, Sb and Zn, which are used singly or in combination of two or more. It is preferable that the inorganic compound constituting the electron transporting layer is in the form of a fine crystalline or amorphous insulating thin film. When the electron transporting layer is constituted with the above insulating thin film, a more uniform thin film can be formed and defective pixels such as dark spots can be decreased. Examples of the inorganic compound include the alkali metal chalcogenides, the alkali metal halides and the alkaline earth metal halides which are described above.

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[0070] As the cathode for the organic EL device of the present invention, an electrode substance such as metal, alloy, electroconductive compound and those mixture having a small work function (4 eV or smaller) is employed. Examples of the electrode substance include potassium, sodium- potassium alloy, magnesium, lithium, magnesium-silver alloy, aluminum / aluminum oxide, Al / Li₂O, Al / LiO₂, Al / LiF, aluminum-lithium alloy, indium, rare earth metal and the like. The cathode can be prepared by forming a thin film of the electrode material described above in accordance with a process such as the vapor deposition process and the sputtering process. When the light emitted from the light emitting layer is observed through the cathode, it is preferable that the cathode has a transmittance of the emitted light greater than 10 %. It is also preferable that the sheet resistivity of the cathode is several hundred Ω/\Box or smaller. The thickness of the cathode is, in general, selected in the range of from 10 nm to 1 μ m and preferably in the range of from 50 to 200 nm. [0071] In general, an organic EL device tends to form defects in pixels due to leak and short circuit since an electric field is applied to ultra-thin films. To prevent the formation of the defects, a layer of an insulating thin film may be inserted between the pair of electrodes. Examples of the material employed for the insulating layer include aluminum oxide, lithium fluoride, cesium fluoride, cesium oxide, magnesium fluoride, calcium oxide, calcium fluoride, aluminum nitride, titanium oxide, silicon oxide, germanium oxide, silicon nitride, boron nitride, molybdenum oxide, ruthenium oxide, vanadium oxide and the like. Mixtures and laminates of the above compounds can also be employed

[0072] To produce an organic EL device of the present invention, for example, a cathode, a light emitting layer and, where necessary, a hole injecting layer and an electron injecting layer are formed in accordance with the aforementioned process using the aforementioned materials, and the anode is formed in the last step. An organic EL device may be produced by forming the aforementioned layers in the order reverse to that described above, i.e., an anode being formed in the first step and a cathode in the last step. An embodiment of the process for producing an organic EL device having a construction in which a cathode, a hole injecting layer, a light emitting layer, an electron injecting layer and an anode are disposed sequentially on a light-transmitting substrate will be described in the following. On a suitable light-transmitting substrate, a thin film made of a material for the cathode is formed in accordance with the vapor deposition process or the sputtering process so that the thickness of the formed thin film is 1 μ m or smaller and preferably in the range of 10 to 200 nm. The formed thin film is employed as the cathode.

[0073] Then, a hole injecting layer is formed on the cathode. The hole injecting layer can be formed in accordance with the vacuum vapor deposition process, the spin coating process, the casting process or the LB process, as described above. The vacuum vapor deposition process is preferable since a uniform film can be easily obtained and the possibility of formation of pin holes is small. When the hole injecting layer is formed in accordance with the vacuum vapor deposition process, in general, it is preferable that the conditions in general are suitably selected in the following ranges: temperature of the deposition source: 50 to 450° C; vacuum level: 10^{-7} to 10^{-3} Torr; deposition rate: 0.01 to 50 nm/second; temperature of the substrate: -50 to 300° C; and film thickness: 5 nm to 5 μ m; although the conditions of the vacuum vapor deposition are different depending on the employed compound (the material for the hole injecting layer) and the crystal structure and the recombination structure of the hole injecting layer to be formed.

[0074] Subsequently, the light-emitting layer is formed on the hole-injecting layer formed above. Also the formation of the light emitting layer can be made by forming the light emitting material according to the present invention into a thin film in accordance with the vacuum vapor deposition process, the sputtering process, the spin coating process or the casting process. The vacuum vapor deposition process is preferable because a uniform film can be easily obtained and the possibility of formation of pinholes is small. When the light-emitting layer is formed in accordance with the vacuum

vapor deposition process, in general, the conditions of the vacuum vapor deposition process can be selected in the same ranges as those described for the vacuum vapor deposition of the hole-injecting layer although the conditions are different depending on the used compound. It is preferable that the thickness is in the range of from 10 to 40 nm.

[0075] Next, the electron-injecting layer is formed on the light-emitting layer formed above. Similarly to the hole injecting layer and the light-emitting layer, it is preferable that the electron-injecting layer is formed in accordance with the vacuum vapor deposition process since a uniform film must be obtained. The conditions of the vacuum vapor deposition can be selected in the same ranges as those for the hole injecting layer and the light-emitting layer.

[0076] In the last step, the anode is formed on the electron-injecting layer, and an organic EL device can be fabricated. The anode is made of a metal and can be formed in accordance with the vacuum vapor deposition process or the sputtering process. It is preferable that the vacuum vapor deposition process is employed in order to prevent the lower organic layers from damages during the formation of the film.

[0077] In the above production of the organic EL device, it is preferable that the above layers from the anode to the cathode are formed successively while the production system is kept in a vacuum after being evacuated. The process for forming the layers in the organic EL device of the present invention is not particularly limited. A conventional process such as the vacuum vapor deposition process and the spin coating process can be used.

[0078] The organic thin film layer comprising the compound having a spiro bond represented by the foregoing general formula (1) used in the organic EL device of the present invention can be formed in accordance with the vacuum vapor deposition process, the molecular beam epitaxy process (the MBE process) or, using a solution prepared by dissolving the compound into a solvent, in accordance with a conventional coating process such as the dipping process, the spin coating process, the casting process, the bar coating process and the roller coating process. The thickness of each layer in the organic thin film layer in the organic EL device of the present invention is not particularly limited, therefore, a thickness within the range of several nanometers to 1 μ m is preferable so as to reduce the defects such as pin holes and improve the efficiency.

[0079] When a direct voltage is applied on the organic EL device produced in the above manner, when a direct voltage of 5 to 40 V is applied in the condition that the cathode is connected to a positive electrode (+) and the anode is connected to a negative electrode (-), then a light emitting is observed. When the connection is reversed, no electric current is produced and no light is emitted at all. When an alternating voltage is applied on the organic EL device, the uniform light emission is observed only in the condition that the polarity of the cathode is positive and the polarity of the anode is negative. When an alternating voltage is applied on the organic EL device, any type of wave shape can be employed.

Example

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[0080] This invention will be described in further detail with reference to Examples, which does not limit the scope of this invention.

Synthesis Example 1 (Synthesis of 1-bromo-4-phenylnaphthalene)

[0081] 15 g of 1,4-dibromonaphthalene on the market, 7.7 g of phenylboronic acid and 1.8 g of tetrakis(triphenylphophine)palladium were mixed, followed by argon displacement. After adding 200 ml of toluene and 90 ml of 2M sodium carbonate aqueous solution thereto, it was refluxed on heating for 7 hours. After standing to cool, the organic layer was extracted by toluene, and then it was washed by water and saturated salt water. Subsequently the organic layer was dried by using sodium sulfate anhydride, and then the solvent was removed by distillation. The product was refined through a silica gel chromatography and then 8.9 g of 1-4-bromo-4-phenylnaphthalene of white crystal was obtained (Yield: 60%).

Synthesis Example 2 (Synthesis of 1-bromo-4-(naphthalene-2-yl) naphthalene)

[0082] The procedure of Synthesis Example 1 was repeated except that 2-naphthalene boronic acid in place of phenylboronic acid was used, and then 7.5 g of 1-bromo-4-(naphthalene-2-yl) naphthalene of white crystal was obtained (yield: 43%). Synthesis Example 3 (Synthesis of 2-(biphenyl-2-yl)-6-bromonaphthalene) 15 g of 2,6-dibromonaphthalene on the market, 12.5 g of 2-biphenylboronic acid and 1.8 g of tetrakis(triphenylphophine)palladium were mixed, followed by argon displacement. After adding 250 ml of toluene and 90 ml of 2M sodium carbonate aqueous solution thereto, it was refluxed on heating for 7 hours. After standing to cool, the organic layer was extracted by toluene, and then it was washed by water and saturated salt water. Subsequently the organic layer was dried by using sodium sulfate anhydride, and then the solvent was removed by distillation. The product was refined through a silica gel chromatography and then 10.9 g of 2-(biphenyl-2-yl)-6-bromonaphthalene of white crystal was obtained (yield: 58%).

Synthesis Example 4 (Synthesis of 9-(naphthalene-2-yl) anthracene)

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[0083] 22.5 g of 9-bromoanthracene, 15.8 g of 2-naphthalene boronic acid and 2.0 g of tetrakis(triphenylphophine)palladium were mixed, followed by argon displacement. After adding 150 ml of toluene and 140 ml of 2M sodiumcarbonate aqueous solution thereto, it was refluxed on heating for 7 hours. After standing to cool, the crystal precipitated was filtrated it was washed by ethanol and toluene. The crystal obtained was recrystallized in toluene, followed by filtering and drying, and then 23.1 g of 9-(naphthalene-2-yl)anthracene was obtained (yield: 87%).

Synthesis Example 5 (Synthesis of 9-bromo-10-(naphthalene-2-yl) anthracene)

[0084] 23.1 g of 9-(naphthalene-2-yl) anthracene was dispersed into 250 ml of DMF (dimethylformamide), and then 14.9 g of NBS (N-bromosuccinimide) in DMF solution (150 ml) was dropped therein. After stirred at room temperature for 7 hours, it was left over a night. 200 ml of water was added thereto, and then the crystal precipitated was separated by filtration. Subsequently it was washed by ethanol adequately, followed by drying, and 28.8 g of 9-bromo-10-(naphthalene-2-yl)anthracene of beige color crystal was obtained (Yield: 99%).

Synthesis Example 6 (Synthesis of 10-(naphthalene-2-yl) anthracene-9-boronic acid

[0085] Under argon atmosphere, 150 ml of dehydrated toluene and 150 ml of dehydrated ether were added to 28.8 g of 9-bromo-10-(naphthalene-2-yl)anthracene, and it was cooled to -63 degC. 58 ml of 1.58M-n-butyllithium / hexane solution was dropped therein. Subsequently, it was stirred at -63 degC. for 30 minutes, followed by heating up to -10degC..lt was cooled to -70degC. again, and 23.4 ml of trimethyl boronic acid ester / dehydrated ether solution was dropped therein stepwise. After it was stirred at -70 degC. for 2 hours, it was heated to room temperature steadily. After left over a night, it was acidified by 10 % hydrochloric aqueous solution, followed by two time extractions by toluene. The organic layer obtained was washed by saturated salt water and then dried by using sodium sulfate anhydride. After the solvent was removed by distillation, the crystal obtained was recrystallized in toluene / hexane, followed by drying, and then 17 g of 10-(naphthalene-2-yl)anthracene-9-boronic acid was obtained (yield: 65%).

Synthesis Example 7 (Synthesis of 9-phenanthrene boronic acid)

[0086] 80 ml of dehydrated toluene and 160 ml of dehydrated THF (tetrahydrofuran) were added to 38.6 g of 9-bromophenanthrene, and was cooled to -40 degC.. After 106 ml of 1.58M n-butyllithium hexane solution was dropped, it was stirred at -40 degC. for 30 minutes and heated to -10 degC.. It was cooled to -70degC. again, and 50.0 ml of trimethyl boronic acid ester / dehydrated ether solution was dropped therein stepwise. Subsequently, it was stirred at -70 degC. for 2 hours and then heated to room temperature steadily. After left over a night, it was acidified by 10 % hydrochloric aqueous solution, followed by two time extractions by toluene. The organic layer obtained was washed by saturated salt water and then dried by using sodium sulfate anhydride. After the solvent was removed by distillation, the crystal obtained was recrystallized in toluene / hexane, followed by drying, and then 21.5 g of 9-phenanthrene boronic acid of pale brown color crystal was obtained (yield: 64%).

Synthesis Example 8 (Synthesis of 10-(phenanthrene-9-yl) anthracene-9- boronic acid)

[0087] The procedures of Synthesis Example 4 to 6 were repeated, except that 9-phenanthrene boronic acid in place of 2-naphthalene boronic acid as the starting material was used, and then 10-(phenanthrene-9-yl)anthracene-9- boronic acid was obtained.

Production Example 1 (Synthesis of AN7)

[0088] 5.0 g of 1-bromo-4-phenylnaphthalene, 7.38 g of 10-(naphthalene-2-yl) anthracene-9-boronic acid and 0.61 g of tetrakis(triphenylphophine)palladium were mixed, and followed by argon displacement. After adding 100 ml of DME (dimethoxyethane) and 30 ml of 2M sodium carbonate aqueous solution thereto, it was refluxed on heating for 10 hours. [0089] After standing to cool, the crystal precipitated was filtrated, it was washed by water, methanol and then toluene. The crystal obtained was recrystallized in toluene, followed by filtering and drying, and then 6.37 g of AN7 of cream color crystal was obtained (yield: 71%).

[0090] The measurement result of the compound by FD-MS (Field Desorption Mass Spectrometry analysis) showed m/z (measured value) =506 to $C_{40}H_{26}$ = 506, therefore the objective compound (AN7) was confirmed.

Production Example 2 (Synthesis of AN8)

[0091] The procedure of Production Example 1 was repeated except that 1-bromo-4-(naphthalene-2-yl)naphthalene in place of 1-bromo-4-phenylnaphthalene was used, and then AN8 of cream color crystal was obtained (yield: 63%).

[0092] The measurement result of the compound by FD-MS showed m/z =556 to $C_{44}H_{28}$ = 556, therefore the objective compound (AN8) was confirmed.

Production Example 3 (Synthesis of AN11)

[0093] The procedure of Production Example 1 was repeated, except that 2-bromonaphthalene and 10-(phenanthrene-9-yl)anthracene-9-boronic acid in place of 1-bromo-4-phenylnaphthalene and 10-(naphthalene-2-yl)anthracene-9-boronic acid respectively were used, and then AN11 of cream color crystal was obtained (yield: 67%).

[0094] The measurement result of the compound by FD-MS showed m/z =480 to $C_{38}H_{24}$ = 480.

Production Example 4 (Synthesis of AN13)

[0095] The procedure of Production Example 1 was repeated except that 2-(biphenyl-2-yl)-6-bromonaphthalene and 10-(phenanthrene-9-yl)anthracene-9-boronic acid in place of 1-bromo-4-phenylnaphthelene and 10-(naphthalene-2-yl)anthracene-9-boronic acid respectively were used, and then AN13 of cream color crystal was obtained (yield: 67%). [0096] The measurement result of the compound by FD-MS showed m/z = 632 to $C_{50}H_{32} = 632$.

Production Example 5 (Synthesis of AN44)

[0097] The procedure of Production Example 1 was repeated except that 1-bromonaphthalene in place of 1-bromo-4-naphthalene was used, and then AN44 of cream color crystal was obtained (yield: 69%).

[0098] The measurement result of the compound by FD-MS showed m/z =430 to $C_{34}H_{22}$ = 430.

Production Example 6 (Synthesis of AN6)

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[0099] The procedure of Production Example 1 was repeated except that 2-bromo-6-phenylnaphthalene in place of 1-bromo-4-naphthalene was used, and then AN6 of cream color crystal was obtained (yield: 54%).

[0100] The measurement result of the compound by FD-MS showed m/z =506 to $C_{40}H_{26} = 506$.

Production Example 7 (Synthesis of AN12)

[0101] The procedure of Production Example 1 was repeated except that 2-bromo-6-phenylnaphthalene and 10-(phen-anthrene-9-yl)anthracene-9-boronic acid in place of 1-bromo-4-naphthalene and 10-(naphthalene-2-yl)anthracene-9-boronic acid respectively were used, and then AN13 of cream color crystal was obtained (yield: 60%).

[0102] The measurement result of the compound by FD-MS showed m/z =556 to $C_{44}H_{28}$ = 556.

Example 1 (Fabrication of an organic EL device)

[0103] A glass substrate (manufactured by GEOMATEC Company) of 25 mmx75 mm×1.1 mm thickness having an ITO transparent electrode was cleaned by application of ultrasonic wave in isopropyl alcohol for 5 minutes and then by exposure to ozone generated by ultraviolet light for 30 minutes. The cleaned glass substrate having an ITO transparent electrode line was fixed to a substrate holder of a vacuum deposition apparatus, and on the surface, where the ITO transparent electrode line was fixed, of the substrate, a film (hereinafter referred to as TPD232 film) having film thickness of 60 nm of the following N,N'-bis(N,N'-diphenyl-4-aminophenyl)-N,N-diphenyl-4,4'-diamino-1,1'-biphenyl was formed so as to cover the transparent electrode. The TPD232 film performs as a hole injecting layer.

[0104] Subsequently, a layer having layer thickness of 20 nm of the following N,N,N',N'-tetra(4-biphenyl)-diaminobiphenylene was formed (hereinafter referred to as TBDB layer). The layer performs as a hole transporting layer.

[0105] Subsequently, a film having a film thickness of 40 nm of the aforementioned compound AN-7 was formed as a hole transporting layer by a vapor deposition. Concurrently, as a light emitting material, the following amino compound D1 containing a styryl group was deposited at the ratio by weight between AN7 and D 1 of 40 : 3 by a vapor deposition. The film performs as a light emitting layer. On the film, a film having a Alq film thickness of 10 nm was formed. The film performs as an electron injecting layer.

[0106] Further, a film (film thickness: 10 nm) of Alq: Li (the source of lithium: manufactured by SAES GETTERS Company) as an electron injecting layer or a cathode was formed by binary vapor deposition of Li as a reductive dopant

and the following Alq. On the Alq: Li film, Al metal was deposited to form a metal cathode; therefore, an organic EL device was fabricated.

[0107] The current efficiency of the organic EL device was measured, and also the half lifetime thereof was measured at an initial luminance of 1,000 nits. The results are shown in Table 1.

35 Examples 2 to 7 (fabrication of organic EL devices)

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[0108] Organic EL devices were fabricated similar to the example 1 except that the compounds described in Table 1 were used in place of the compound AN7, and then the current efficiencies and half lifetimes of the devices obtained were tested by the similar method to Example 1. The results are shown in Table 1.

Example 8 (fabrication of organic EL device)

[0109] Organic EL device was fabricated similar to the example 1 except that the compound AN11 in place of the compound AN7 and the amine compound D2 in place of the amine compound D1 were used as the material for the light emitting layer, and then the current efficiency and half lifetime of the device obtained were tested by the similar method to Example 1. The results are shown in Table 1.

Me Me

Me

N

N

Me

D2

Me

Comparative Example 1

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[0110] Organic EL devices were fabricated similar to the example 1 except that an1 was used in place of the compound AN7, and then the current efficiency and half lifetime of the device obtained was tested by the similar methods to Example 1

[0111] The results are shown in Table 1.

an1

Table 1

Compound of Light-Emitting Layer Current Efficiency (cd/A) Half Lifetime (hours) Example 1* AN7 /D1 10.9 4,200 Example 2* AN8 /D1 1.3 4,200 Example 3 AN11/D1 11.0 5,800 AN13/D1 Example 4 10.8 3,700 Example 5* AN44 /D1 10.0 3,000 10.1 3,300 Example 6 AN6/D1 Example 7 AN12/D1 10.8 4,900 AN11 / D2 10.3 3,700 Example 8 Comparative Example 1 an1/D1 9.0 2,200 *: not part of the invention

[0112] As shown in Table 1, the organic EL devices of Examples 1 to 8 exhibited a higher current efficiency and a longer lifetime than those of the device of Comparative Example 1.

Claims

1. A light emitting material for an organic electroluminescent device comprising an asymmetric anthracene derivative represented by the following general formula (1);

wherein A¹ and A² each independently represents a substituted or unsubstituted condensed aromatic hydrocarbon ring group selected from the group consisting of 2-naphthyl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-pyrenyl group, 2-pyreriyl group, 4-pyrenyl group, 3-methyl-2-naphthyl group and 4-methyl-1-naphthyl group;

Ar¹ and Ar² each independently represents a hydrogen atom, or a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50; R¹ to R8 each independently represents a hydrogen atom, a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50, a substituted or unsubstituted aromatic hetero ring group having ring atoms of 5 to 50, a substituted or unsubstituted alkyl group having carbon atoms of 1 to 50, a substituted or unsubstituted or unsubstituted or unsubstituted aralkyl group having carbon atoms of 3 to 50, a substituted or unsubstituted aralkyl group having carbon atoms of 6 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted alkoxycarbonyl group having carbon atoms of 1 to 50, a substituted or unsubstituted alkoxycarbonyl group having carbon atoms of 1 to 50, a substituted or unsubstituted silyl group, a carboxyl group, a halogen atom, a cyano group, a nitro group or a hydroxyl group;

R⁹ to R¹⁰ each independently represents a hydrogen atom, a substituted or unsubstituted aromatic hydrocarbon ring group having ring carbon atoms of 6 to 50, a substituted or unsubstituted alkyl group having carbon atoms of 1 to 50, a substituted or unsubstituted or unsubstituted or unsubstituted alkoxy group having carbon atoms of 1 to 50, a substituted or unsubstituted aralkyl group having carbon atoms of 6 to 50, a substituted or unsubstituted aryloxy group having carbon atoms of 5 to 50, a substituted or unsubstituted arylthio group having carbon atoms of 5 to 50, a substituted or unsubstituted arylthio group having carbon atoms of 5 to 50, a substituted or unsubstituted aryloxy group, a carboxyl group, a halogen atom, a cyano group, a nitro group or a hydroxyl group, and none of R⁹ and R¹⁰ is an alkenyl group;

Ar¹, Ar², R⁹ and R¹⁰ each may be a plural number, and two neighboring groups thereof may form a saturated or unsaturated ring structure; and wherein

(I) A¹ is different from A²,

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(II) when A¹ is the same with A², (II-i) Ar¹ is different from Ar², or (II-ii) R⁹ is different from R¹⁰, and

(II-iii) when Ar^1 and Ar^2 are the same with each other, and R^9 and R^{10} are the same with each other, (II-iii-1) the bonding position of A^1 to 9th position of the anthracene ring is different from the bonding position of A^2 to 10th position of the anthracene ring,

(II-iii-2) when both Ar^1 and Ar^2 are not hydrogen atoms, the bonding position of Ar^1 to A^1 is different from the bonding position of Ar^2 to A^2 , and

(II-iii-3) when both R^9 and R^2 are not hydrogen atoms, the bonding position of R^9 to A^1 is different from the bonding position of R^{10} to A^2 .

- 2. The light emitting material for the organic electroluminescent device according to Claim 1, wherein, in the general formula (1), A¹ and A² each independently represents any one of 2-naphthyl group, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, and 3-methyl-2-naphthyl group.
- 3. The light emitting material for the organic electroluminescent device according to Claim 1, wherein, in the general

formula (1), A^1 and A^2 each independently represents a 2-naphthyl group or 9-phenanthryl group.

- **4.** The light emitting material for the organic electroluminescent device according to Claim 1, wherein, in the general formula (1), Ar¹ and Ar² each independently represents a hydrogen atom or an aromatic hydrocarbon ring having ring carbon atoms of 6 to 16.
- 5. The light emitting material for the organic electroluminescent device according to Claim 2, wherein, in the general formula (1), Ar¹ and Ar² each independently represents any one of a hydrogen atom, phenyl group, 1-naphthyl group, 2-naphthyl group, 1-anthryl group, 2-anthryl group, 9-anthrylgroup, 1-phenanthryl group, 2-phenanthryl group, 3-phenanthryl group, 4-phenanthryl group, 9-phenanthryl group, 1-naphthacenyl group, 2-naphthacenyl group, 1-pyrenyl group, 2-pyrenyl group, 2-biphenylyl group, 3-biphenylyl group, 4-biphenylyl group, p-terphenyl-4-yl group, p-terphenyl-3-yl group, m-terphenyl-4-yl group, p-terphenyl-2-yl group, m-terphenyl-3-yl group, m-terphenyl-2-yl group, m-terphenyl-2-yl group, m-terphenyl group, 3-methyl-2-naphthyl group, 4-methyl-1-naphthyl group, 4-methyl-1-anthryl group, 4'methylbiphenylyl group and 4"-t- butyl-p-terphenyl-4-yl group.
- **6.** The light emitting material for the organic electroluminescent device according to Claim 3, wherein, in the general formula (1), Ar¹ and Ar² each independently represents any one of a hydrogen atom, 1-naphthyl group, 2-naphthyl group and 9-phenanthryl group.
- 7. The light emitting material for an organic electroluminescent device according to claim 1, wherein the asymmetric anthracene derivative is selected from the compounds

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- 8. An organic electroluminescent device comprising at least one organic thin film layer, which contains at least a light emitting layer, which is interposed between a pair of electrodes consisting of an anode and a cathode, wherein a light emitting zone comprises the light emitting material for the organic electroluminescent device according to Claim 1 singly or as a component of a mixture thereof.

- **9.** The organic electroluminescent device according to Claim 8, wherein the light emitting layer contains the light emitting material for the organic electroluminescent device singly or as a component of a mixture thereof.
- 10. The organic electroluminescent device according to Claim 8, wherein the organic thin film layer contains the lightemitting material for the organic electroluminescent device.
 - **11.** The organic electroluminescent device according to any one of Claims 8 to 10, wherein, the light emitting layer contains additionally an arylamine compound.

12. The organic electroluminescent device according to any one of Claims 8 to 10, wherein the light emitting layer contains additionally a styrylamine compound.

5 Patentansprüche

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1. Licht emittierendes Material für eine organische Elektrolumineszenz-Vorrichtung, umfassend ein durch die folgende allgemeine Formel (1) dargestelltes asymmetrisches Anthracen-Derivat

wobei A¹ und A² jeweils unabhängig voneinander eine substituierte oder unsubstituierte kondensierte aromatische Kohlenwasserstoffringgruppe darstellen, ausgewählt aus der aus 2-Naphthylgruppe, 1-Phenanthrylgruppe, 2-Phenanthrylgruppe, 3-Phenanthrylgruppe, 4-Phenanthrylgruppe, 9-Phenanthrylgruppe, 1-Naphthacenylgruppe, 1-Pyrenylgruppe, 2-Pyrenylgruppe, 4-Pyrenylgruppe, 3-Methyl-2-naphthylgruppe und 4-Methyl-1-naphthylgruppe bestehenden Gruppe;

Ar¹ und Ar² jeweils unabhängig voneinander ein Wasserstoffatom oder eine substituierte oder unsubstituierte aromatische Kohlenwasserstoffringgruppe mit 6 bis 50 Ringkohlenstoffatomen darstellen;

R¹ bis R³ jeweils unabhängig voneinander ein Wasserstoffatom, eine substituierte oder unsubstituierte aromatische Kohlenwasserstoffringgruppe mit 6 bis 50 Ringkohlenstoffatomen, eine substituierte oder unsubstituierte aromatische Heteroringgruppe mit 5 bis 50 Ringatomen, eine substituierte oder unsubstituierte Alkylgruppe mit 1 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte Cycloalkylgruppe mit 3 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte oder unsubstituierte Aralkylgruppe mit 6 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte Arylthiogruppe mit 5 bis 50 Kohlenstoffatomen, eine substituierte Arylthiogruppe mit 5 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte oder unsubstituierte

R⁹ bis R¹⁰ jeweils unabhängig voneinander ein Wasserstoffatom, eine substituierte oder unsubstituierte aromatische Kohlenwasserstoffringgruppe mit 6 bis 60 Ringkohlenstoffatomen, eine substituierte oder unsubstituierte Alkylgruppe mit 1 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte Oger unsubstituierte oder unsubstituierte oder unsubstituierte oder unsubstituierte oder unsubstituierte oder unsubstituierte Aralkylgruppe mit 6 bis 50 Kohlenstoffatomen, eine substituierte Aryloxygruppe mit 5 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte Arylthiogruppe mit 5 bis 50 Kohlenstoffatomen, eine substituierte oder unsubstituierte Oger unsub

Ar¹, Ar², R⁹ und R¹⁰ jeweils mehrfach vorliegen können, und zwei benachbarte Gruppen davon eine gesättigte oder ungesättigte Ringstruktur bilden können; und wobei

- (I) A¹ von A² verschieden ist,
- (II) wenn A¹ gleich ist wie A², (II-i) Ar¹ verschieden ist von Ar²,
- oder (II-ii) R⁹ verschieden ist von R¹⁰ und
 - (II-iii) wenn Ar1 und Ar2 gleich sind, R9 und R10 gleich sind,
 - (II-iii-1) die Bindungsposition von A¹ zur 9^{ten} Position des Anthracenrings verschieden ist von der Bindungspo-

sition von A² zu 10^{ten} Position des Anthracenrings,

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(II-iii-2) wenn sowohl Ar^1 als auch Ar^2 keine Wasserstoffatome sind, die Bindungsposition von Ar^1 zu A^1 verschieden ist von der Bindungsposition von Ar^2 zu A^2 und

- (II-iii-3) wenn sowohl R^9 als auch R^2 keine Wasserstoffatome sind, die Bindungsposition von R^9 zu A^1 verschieden ist von der Bindungsposition von R^{10} zu A^2 .
- 2. Licht emittierendes Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 1, wobei in der allgemeinen Formel (1) A¹ und A² jeweils unabhängig einer von einer 2-Naphthylgruppe, 1-Phenanthrylgruppe, 2-Phenanthrylgruppe, 3-Phenanthrylgruppe, 4-Phenanthrylgruppe, 9-Phenanthrylgruppe und 3-Methyl-2-naphthylgruppe darstellen.
- 3. Licht emittierendes Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 1, wobei in der allgemeinen Formel (1) A¹ und A² jeweils unabhängig voneinander eine 2-Naphthylgruppe oder 9-Phenanthrylgruppe darstellen.
- **4.** Licht emittierendes Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 1, wobei in der allgemeinen Formel (1) Ar¹ und Ar² jeweils unabhängig voneinander ein Wasserstoffatom oder einen aromatischen Kohlenwasserstoffring mit 6 bis 16 Ringkohlenstoffatomen darstellen.
- 5. Licht emittierendes Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 2, wobei in der allgemeinen Formel (1) Ar¹ und Ar² jeweils unabhängig voneinander einen von Wasserstoffatom, Phenylgruppe, 1-Naphthylgruppe, 2-Naphthylgruppe, 1-Anthrylgruppe, 2-Anthrylgruppe, 9-Anthrylgruppe, 1-Phenanthrylgruppe, 2-Phenanthrylgruppe, 3-Phenanthrylgruppe, 4-Phenanthrylgruppe, 2-Pyrenylgruppe, 4-Pyrenylgruppe, 2-Biphenylylgruppe, 3-Biphenylylgruppe, 4-Biphenylylgruppe, p-Terphenyl-4-ylgruppe, p-Terphenyl-3-ylgruppe, m-Terphenyl-4-ylgruppe, m-Terphenyl-4-ylgruppe, m-Tolylgruppe, p-tolylgruppe, p-t-Butylphenylgruppe, p-(2-Phenylpropyl)phenylgruppe, 3-Methyl-2-naphthylgruppe, 4-Methyl-1-anthrylgruppe, 4'-Methylbiphenylylgruppe und 4"-t-Butyl-p-terphenyl-4-ylgruppe darstellt.
 - **6.** Licht emittierendes Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 3, wobei in der allgemeinen Formel (1) Ar¹ und Ar² jeweils unabhängig voneinander einen von Wasserstoffatom, 1-Naphthylgruppe, 2-Naphthylgruppe und 9-Phenanthrylgruppe darstellen.
- 7. Licht emittierendes Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 1, wobei das asymmetrische Anthracen-Derivat aus den Verbindungen

ausgewählt wird.

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- 8. Organische Elektrolumineszenz-Vorrichtung, umfassend mindestens eine organische Dünnfilmschicht, welche mindestens eine lichtemittierende Schicht enthält, welche zwischen einem Elektrodenpaar angeordnet ist, das aus einer Anode und einer Kathode besteht, wobei eine lichtemittierende Zone das lichtemittierende Material für die organische Elektrolumineszenz-Vorrichtung nach Anspruch 1 einzeln oder als Komponente einer Mischung davon umfasst.
- 9. Organische Elektrolumineszenz-Vorrichtung nach Anspruch 8, wobei die lichtemittierende Schicht das lichtemittierende Material für die organische Elektrolumineszenz-Vorrichtung einzeln oder als Komponente einer Mischung davon enthält.
- **10.** Organische Elektrolumineszenz-Vorrichtung nach Anspruch 8, wobei die organische Dünnfilmschicht das lichtemittierende Material für die organische Elektrolumineszenz-Vorrichtung enthält.

- 11. Organische Elektrolumineszenz-Vorrichtung nach einem der Ansprüche 8 bis 10, wobei die lichtemittierende Schicht zusätzlich eine Arylamin-Verbindung enthält.
- **12.** Organische Elektrolumineszenz-Vorrichtung nach einem der Ansprüche 8 bis 10, wobei die lichtemittierene Schicht zusätzlich eine Styrylamin-Verbindung enthält.

Revendications

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 Matériau émettant de la lumière pour dispositif organique électroluminescent comprenant un dérivé anthracène asymétrique représenté par la formule suivante (1);

dans laquelle A¹ et A² chacun indépendamment représente un groupe annulaire hydrocarbure aromatique condensé substitué ou non-substitué sélectionné parmi le groupe consistant en le groupe 2-naphtyle, le groupe 1-phenantryle, le groupe 2-phenantryle, le groupe 3-phenantryle, le groupe 4-phenantryle, le groupe 9-phenantryle, le groupe 1-naphtacényle, le groupe 2-naphtacényle, le groupe 2-pyrényle, le groupe 2-pyrényle, le groupe 3-méthyle-2-naphtyle, et le groupe 4-méthyle-1-naphtyle;

 Ar^1 et Ar^2 chacun indépendamment représente un atome d'hydrogène, ou un groupe annulaire hydrocarbure aromatique substitué ou non-substitué ayant des atomes de carbone annulaires de 6 à 50 ;

R¹ à R³ chacun indépendamment représente un atome d'hydrogène, un groupe annulaire hydrocarbure aromatique substitué ou non-substitué ayant des atomes de carbone annulaires de 6 à 50, un groupe hétéro annulaire aromatique substitué ou non-substitué ayant des atomes annulaires de 5 à 50, un groupe alkyle substitué ou non-substitué ayant des atomes de carbone de 1 à 50, un groupe cycloalkyle substitué ou non-substitué ayant des atomes de carbone de 1 à 50, un groupe aralkyle substitué ou non-substitué ayant des atomes de carbone de 1 à 50, un groupe aralkyle substitué ou non-substitué ayant des atomes de carbone de 6 à 50, un groupe aryloxy substitué ou non-substitué ayant des atomes de carbone de 5 à 50, un groupe alkoxycarbone de 5 à 50, un groupe atomes de carbone de 1 à 50, un groupe alkoxycarbonyle substitué ou non-substitué ayant des atomes de carbone de 1 à 50, un groupe silyle substitué ou non-substitué, un groupe carboxyle, un atome d'halogène, un groupe cyano, un groupe nitro ou un groupe hydroxyle;

 R^9 à R^{10} chacun indépendamment représente un atome d'hydrogène, un groupe annulaire hydrocarbure aromatique substitué ou non-substitué ayant des atomes de carbone annulaires de 6 à 50, un groupe alkyle substitué ou non-substitué ayant des atomes de carbone de 6 à 50, un groupe cycloalkyle substitué ou non-substitué ayant des atomes de carbone de 3 à 50, un groupe alkoxy substitué ou non-substitué ayant des atomes de carbone de 1 à 50, un groupe aralkyle substitué ou non-substitué ayant des atomes de carbone de 6 à 50, un groupe aryloxy substitué ou non-substitué ayant des atomes de carbone de 5 à 50, un groupe alkoxycarbonyle substitué ou non-substitué ayant des atomes de carbone de 1 à 50, un groupe silyle substitué ou non-substitué, un groupe carboxyle, un atome d'halogène, un groupe cyano, un groupe nitro ou un groupe hydroxyle, et aucun de R^9 et R^{10} est un groupe alkényle ;

Ar¹, Ar², R⁹ et R¹⁰ chacun peut être un nombre pluriel, et deux groupes voisins de ceux-ci peuvent former une structure annulaire saturée ou non-saturée ; et dans lequel

(I) A¹ est différent de A²,

(II) lorsque A¹ est égal à A², (II-i) A¹ est différent de A², ou (II-ii) R⁹ est différent de R¹⁰, et

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- (II-iii) lorsque Ar¹ et A² sont égaux l'un à l'autre, et R⁹ et R¹⁰ sont égaux l'un à l'autre,
- (II-iii-1) la position liante de A¹ à la neuvième position de l'anneau anthracène est différente de la position liante de A² à la dixième position de l'anneau anthracène,
- (II-iii-2) lorsque tous les deux de Ar^1 et Ar^2 ne sont pas des atomes d'hydrogène, la position liante de Ar^1 à A^1 est différente de la position liante de Ar^2 à A^2 , et
- (II-iii-3) lorsque tous les deux de R^9 et R^2 ne sont pas des atomes d'hydrogène, la position liante de R^9 à A^1 est différente de la position liante de R^{10} à A^2 .
- 2. Matériau émettant de la lumière pour dispositif organique électroluminescent selon la revendication 1, dans lequel dans la formule générale (1) A¹ et A² chacun indépendamment représente l'un quelconque de groupe 2-naphtyle, groupe 1-phenantryle, groupe 2-phenantryle, le groupe 3-phenantryle, le groupe 4-phenantryle, le groupe 9-phenantryle et le groupe 3-méthyle-2-naphtyle.
- **3.** Matériau émettant de la lumière pour dispositif organique électroluminescent selon la revendication 1, dans lequel dans la formule générale (1) A¹ et A² chacun indépendamment représente l'un quelconque de groupe 2-naphtyle, groupe 1-phenantryle, groupe 2-phenantryle, le groupe 3-phenantryle, le groupe 4-phenantryle, le groupe 9-phenantryle et le groupe 3-méthyle-2-naphtyle.
- **4.** Matériau émettant de la lumière pour dispositif organique électroluminescent selon la revendication 1, dans lequel dans la formule générale (1) Ar¹ et Ar² chacun indépendamment représente un atome d'hydrogène, ou un groupe annulaire hydrocarbure aromatique ayant des atomes de carbone annulaires de 6 à 16.
- 5. Matériau émettant de la lumière pour dispositif organique électroluminescent selon la revendication 2, dans lequel dans la formule générale (1) Ar¹ et Ar² chacun indépendamment représente l'un quelconque d'un atome d'hydrogène, un groupe phényle, un groupe 1-naphtyle, un groupe 2-naphtyle, un groupe 1-anthryle, un groupe 2-anthryle, un groupe 2-phenanthryle, un groupe 3-phenanthryle, un groupe 4-phenanthryle, un groupe 9-phenanthryle, un groupe 1-naphtacényle, un groupe 2-naphtacényle, un groupe 9-naphtacényle, un groupe 1-pyrényle, un groupe 2-pyrényle, un groupe 4-pyrényle, un groupe 2-biphénylyle, un groupe 3-biphénylyle, un groupe 4-biphénylyle, un groupe p-terphényle-4-yle, un groupe p-terphényle-3-yle, un groupe m-terphényle-2-yle, un groupe m-terphényle-3-yle, un groupe m-terphényle-2-yle, un groupe p-tolyle, un groupe p-tolyle, un groupe p-t-butylphényle, un groupe p-(2-phénylpropyle) phényle, un groupe 4-méthyle-1-naphtyle, un groupe 4-méthyle-1-anthryle, un groupe 4-méthylbiphényle et un groupe 4"-t-butyle-p-terphényle-4-yl.
 - **6.** Matériau émettant de la lumière pour dispositif organique électroluminescent selon la revendication 3, dans lequel dans la formule générale (1) Ar¹ et Ar² chacun indépendamment représente l'un quelconque d'un atome d'hydrogène, un groupe 1-naphtyle, un groupe 2-naphtyle et un groupe 9-phenanthryle.
 - 7. Matériau émettant de la lumière pour dispositif organique électroluminescent selon la revendication 1, dans lequel le dérivé anthracène asymétrique est sélectionné parmi les composés

8. Dispositif organique électroluminescent comprenant au moins une couche organique à film mince qui contient au moins une couche émettant de la lumière, qui est entreposée entre une paire d'électrodes consistant en une anode et une cathode, dans lequel une zone émettant de la lumière comprend le matériau émettant de la lumière pour le dispositif organique électroluminescent selon la revendication 1 seul ou en tant qu'un composant d'un mélange de ceux-ci.

- **9.** Dispositif organique électroluminescent selon la revendication 8, dans lequel la couche émettant de la lumière contient le matériau émettant de la lumière pour le dispositif organique électroluminescent seul ou en tant qu'un composant d'un mélange de ceux-ci.
- 10. Dispositif organique électroluminescent selon la revendication 8 dans lequel la couche organique à film mince contient le matériau émettant de la lumière pour le dispositif organique électroluminescent.
 - 11. Dispositif organique électroluminescent selon l'une des revendications 8 à 10, dans lequel la couche émettant de

la lumière contient en plus un composé arlyamine. 12. Dispositif organique électroluminescent selon l'une des revendications 8 à 10, dans lequel la couche émettant de la lumière contient en plus un composé styrylamine.

REFERENCES CITED IN THE DESCRIPTION

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Patent documents cited in the description

- JP HEISEI81996239655 B [0006]
- JP HEISEI71995138561 B [0006]
- JP HEISEI31991200289 B [0006]
- JP HEISEI81996012600 B [0006]
- JP HEISEI1119993782 B [0006]
- JP 2001257074 A [0006]
- JP 2000182776 A [0006]
- EP 1496041 A1 [0006]
- JP 2001097897 A [0006]
- US 6582837 B1 [0006]
- EP 1009044 A2 [0006]
- JP 11323323 A [0006]
- JP 2003146951 A [0006]
- JP SHOWA57198251781 B [0054]
- US 3112197 A [0058]
- US 3189447 A [0058]
- JP SHOU3716096 B [0058]
- JP 3615402 B [0058]
- JP 3820989 B [0058]
- JP 3542544 B [0058]
- JP SHOU45555 B [0058]
- JP SHOU5110983 B [0058]
- JP SHOU5193224 B [0058]
- JP SHOU5517105 B [0058]
- JP SHOU564148 B **[0058]**
- JP SHOU55108667 B [0058]
- JP SHOU55156953 B [0058]
 JP SHOU5636656 B [0058]
- JP 3HO03030030 B [0
- US 3180729 A [0058]
- US 4278746 A [0058]JP SHOU5588064 B [0058]
- JP SHOU5588065 B [0058]
- JP SHOU49105537 B [0058]
- JP SHOU5551086 B [0058]
- JP SHOU5680051 B [0058]
- JP SHOU5688141 B [0058]
- JP SHOU5745545 B [0058]
- JP SHOU54112637 B [0058]
- JP SHOU5574546 B [0058]
- US 3615404 A [0058]
- JP SHOU5110105 B [0058]
- JP SHOU463712 B [0058]
- JP SHOU4725336 B [0058]
- JP SHOU5453435 B [0058]
- JP SHOU54110536 B **[0058]**
- JP SHOU54119925 B [0058]
- US 3567450 A [0058]
- US 3180703 A [0058]
- US 3240597 A [0058]

- US 3658520 A [0058]
- US 4232103 A [0058]
- US 4175961 A [0058]
- US 4012376 A [0058]
- JP SHOU4935702 B [0058]
- JP SHOU3927577 B [0058]
- JP SHOU55144250 B [0058] [0059]
- JP SHOU56119132 B [0058] [0059]
- JP SHOU5622437 B [0058]
- DE 1110518 [0058]
- US 3526501 A [0058]
- US 3257203 A [0058]
- JP SHOU5646234 B [0058]
- JP SHOU54110837 B [0058]
- US 3717462 A [0058]
- JP SHOU5459143 B [0058]
- JP SHOU5552063 B [0058]
- JP SHOU5552064 B [0058]
- JP SHOU5546760 B [0058]
- JP SHOU5585495 B [0058]
- JP SHOU5711350 B **[0058]**
- JP SHOU57148749 B **[0058]**
- JP HEI2311591 B [0058]
- JP SHOU61210363 B [0058]
- JP SHOU61228451 B [0058]
- JP SHOU6114642 B [0058]
- JP SHOU6172255 B **[0058]**
- JP SHOU6247646 B [0058]
- JP SHOU6236674 B [0058]
- JP SHOU6210652 B [0058]JP SHOU6230255 B [0058]
- JP SHOU6093455 B [0058]
- JP SHOU6094462 B [0058]
- JP SHOU60174749 B [0058]
- JP SHOU60175052 B [0058]
- US 4950950 A [0058]
- JP HEI2204996 B [0058]
- JP HEI2282263 B [0058]
- JP HEI1211399 B [0058]
- UO 4407440 A FOOFOI
- US 4127412 A [0059]
- JP SHOU5327033 B [0059]
- JP SHOU5458445 B [0059]
- JP SHOU54149634 B [0059]
- JP SHOU5464299 B [0059]
- JP SHOU5579450 B **[0059]**
- JP SHOU61295558 B [0059]JP SHOU6198353 B [0059]
- JP SHOU63295695 B [0059]
- US 5061569 A [0059]

• JP HEISEI81996193191 B [0061]

Non-patent literature cited in the description

- C. W. TANG; C. W TANG; S. A. VANSLYKE et al. Applied Physics Letters. Eastman Kodak Company, 1987, vol. 51, 913 [0002]
- Chem. Rev., 1995, vol. 95 (7), 2457 [0034]
- The Experimental Chemistry Course. vol. 24, 61-90 [0039]
- J. Org. Chem., 1995, vol. 60, 7508 [0039]



专利名称(译)	用于有机电致发光器件的发光材料,使用该发光材料的有机电致发光器件和用于有机电致发光器件的材料			
公开(公告)号	EP1696015B1	公开(公告)日	2015-05-13	
申请号	EP2004807321	申请日	2004-12-13	
[标]申请(专利权)人(译)	出光兴产株式会社			
申请(专利权)人(译)	出光兴产股份有限公司.			
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IPC分类号	C09K11/06 H01L51/00 C07C15/28 C07C15/38 H01L51/50 H05B33/14 H05B33/22			
CPC分类号	H01L51/0058 C07C15/28 C07C15/38 C07C15/60 C07C2603/24 C07C2603/50 C09K11/06 C09K2211 /1007 C09K2211/1011 C09K2211/1014 H01L51/0059 H01L51/5012 H01L51/5056 H05B33/14			
优先权	2003423317 2003-12-19 JP			
其他公开文献	EP1696015A1 EP1696015A4			
外部链接	Espacenet			

摘要(译)

公开了一种用于有机电致发光(EL)器件的发光材料,其由特定结构的不对称蒽衍生物组成。还公开了用于有机EL器件的材料和有机EL器件,其中由包括至少一个发光层的一层或多层组成的有机薄膜层插入在阴极和阳极之间。由有机薄膜层构成的至少一层本身或作为混合物的组分包含有机EL器件的材料。因此,有机EL器件具有高效率和长寿命。还公开了用于有机EL器件的发光材料和用于有机器件的材料,其能够实现这种有机EL器件。