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(12) **United States Patent**
Toba(10) **Patent No.:** **US 8,404,362 B2**
(45) **Date of Patent:** ***Mar. 26, 2013**(54) **ORGANIC LIGHT-EMITTING ELEMENT**
USING TRIAZINE RING-CONTAINING
POLYMER COMPOUND(75) Inventor: **Masahiko Toba**, Chiba (JP)(73) Assignee: **Showa Denko K.K.**, Tokyo (JP)

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544/215(58) **Field of Classification Search** None
See application file for complete search history.(56) **References Cited**

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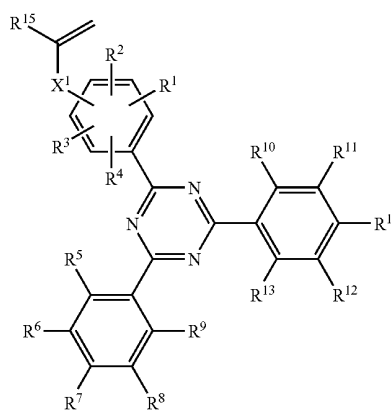
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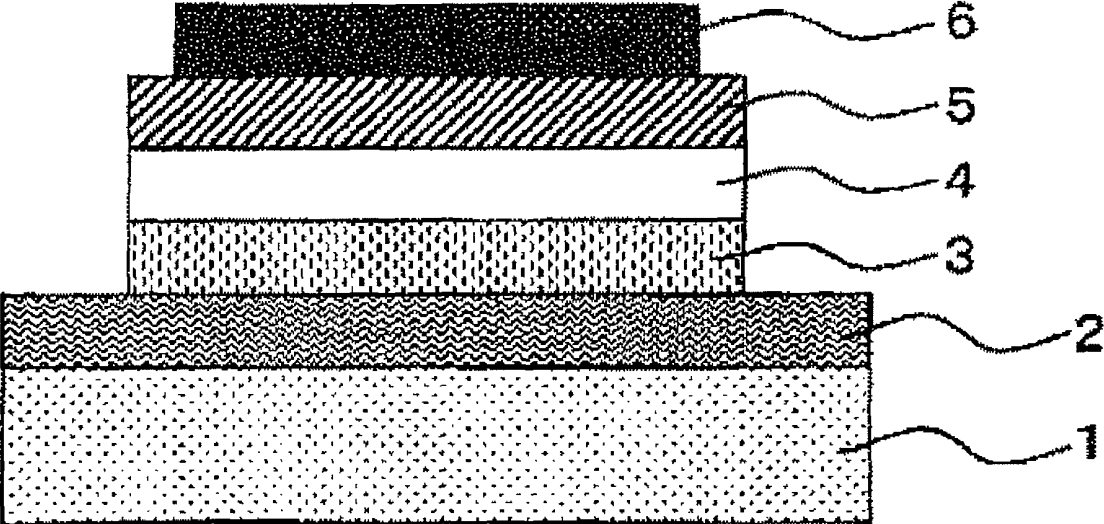
Primary Examiner — Dawn L. Garrett(74) *Attorney, Agent, or Firm* — Sughrue Mion, PLLC(57) **ABSTRACT**

The organic light-emitting element of the present invention is an organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, and is characterized in that the light-emitting layer contains a polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;



wherein R¹ to R¹⁴ are each independently a hydrogen atom, a halogen atom, a cyano group, an amino group, an alkyl group of 1 to 12 carbon atoms or the like, R¹⁵ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X¹ is a single bond or the like.

8 Claims, 1 Drawing Sheet



**ORGANIC LIGHT-EMITTING ELEMENT
USING TRIAZINE RING-CONTAINING
POLYMER COMPOUND**

TECHNICAL FIELD

The present invention relates to an organic light-emitting element using a triazine ring-containing polymer compound. More particularly, the invention relates to an organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, wherein the light-emitting layer contains a triazine ring-containing polymer compound.

BACKGROUND ART

As an organic light-emitting element, an element having a multi-layer structure wherein one or more organic layers are provided between an anode and a cathode is known. In a patent document 1, an organic light-emitting element containing a triazine derivative that is a low-molecular compound in an organic layer is disclosed.

When a layer is formed using a low-molecular compound, a vacuum deposition method is generally used. In this method, however, there is a problem such that vacuum equipment is necessary or the film thickness of the layer is liable to become uniform.

In contrast with this, use of a polymer compound has an advantage that the layer can be formed by a coating method such as spin coating. For example, in a patent document 2, a fluorescent conjugated polymer compound comprising a repeating unit containing a triazine ring is disclosed.

In a non-patent document 1, a non-conjugated polymer compound comprising a triazine skeleton, which can be utilized for organic light-emitting elements or field effect transistors, is disclosed. This polymer compound is specifically a polymer compound obtained by copolymerizing an electron transport triphenyltriazine derivative monomer and a hole transport triphenylamine derivative monomer.

In a non-patent document 2, a non-conjugated polymer compound having a constitutional unit derived from a triphenyltriazine derivative, a constitutional unit derived from a perylene derivative and a constitutional unit derived from an iridium complex, and an organic light-emitting element using the polymer compound are disclosed. In this polymer compound, moieties of electron transport property, hole transport property and phosphorescent property are each bonded to a polymer main chain through an amide bond.

Patent document 1: Japanese Patent Laid-Open Publication No. 173569/2006

Patent document 2: Japanese Patent Laid-Open Publication No. 129155/2002

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Non-patent document 2: Se Young Oh et al., "Characteristics of Polymer Light Emitting Diode Using a Phosphorescent Terpolymer Containing Perylene, Triazine and Ir(ppy) 3 Moieties in the Polymer Side Chain", *Molecular Crystals and Liquid Crystals*, 2006, 458, pp. 227-235.

DISCLOSURE OF THE INVENTION

Problem to be Solved by the Invention

In the organic light-emitting element using a polymer compound described in the non-patent document 2, however,

there is yet room for improvement in respects of driving voltage, luminous efficiency and maximum attainable brightness.

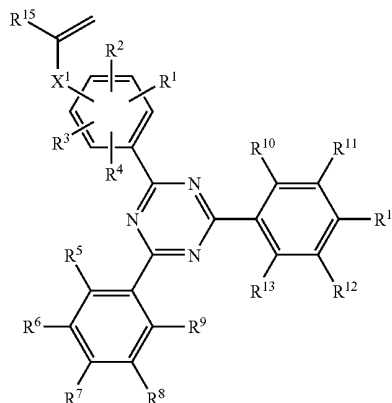
Accordingly, it is an object of the present invention to provide an organic light-emitting element using a polymer compound which has a low driving voltage and by which high luminous efficiency and high luminance are obtained.

Means to Solve the Problem

In order to solve the above problems, the present inventors have earnestly studied. As a result, they have found that an organic light-emitting element having a low driving voltage and having high luminous efficiency and high luminance is obtained by the use of a polymer compound containing constitutional units derived from specific polymerizable compounds, and they have accomplished the present invention.

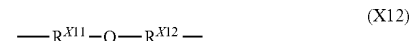
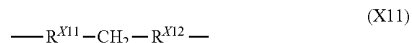
The present invention relates to, for example, the following matters [1] to [9].

[1] An organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, wherein the light-emitting layer contains a polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;



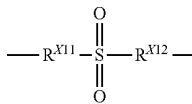
(1)

wherein R^1 to R^{14} are each independently a hydrogen atom, a halogen atom, a cyano group, an amino group, an alkyl group of 1 to 12 carbon atoms or an alkoxy group of 1 to 12 carbon atoms, in each of R^1 to R^4 , R^5 to R^9 , and R^{10} to R^{14} , two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring, R^{15} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X^1 is a single bond or a group represented by any one of the following formulas (X11) to (X14):



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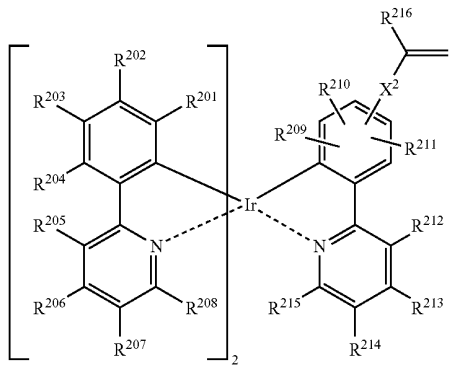


wherein R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group.

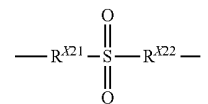
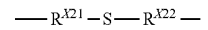
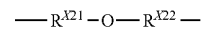
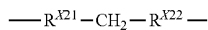
[2] The organic light-emitting element as stated in the above [1], wherein the polymer compound further comprises a constitutional unit derived from a hole transport polymerizable compound.

[3] The organic light-emitting element as stated in the above [1], wherein the light-emitting layer further contains a hole transport compound.

[4] The organic light-emitting element as stated in any one of the above [1] to [3], wherein the phosphorescent polymerizable compound is a complex represented by any one of the following formulas (2) to (4):



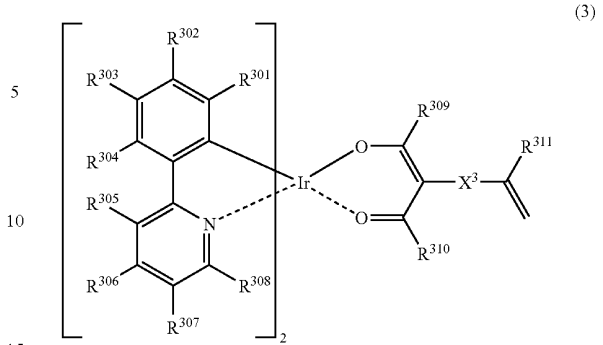
wherein R²⁰¹ to R²¹⁵ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, in each of R²⁰¹ to R²⁰⁴, R²⁰⁵ to R²⁰⁸, R²⁰⁹ to R²¹¹, and R²¹² to R²¹⁵, two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring, R²¹⁶ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X² is a single bond or a group represented by any one of the following formulas (X21) to (X24):



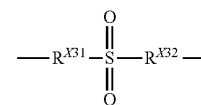
wherein R^{X21} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X22} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;

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(X14)



wherein R³⁰¹ to R³⁰⁸ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R³⁰⁹ to R³¹⁰ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, in each of R³⁰¹ to R³⁰⁴, and R³⁰⁵ to R³⁰⁸, two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring, R³¹¹ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X³ is a single bond or a group represented by any one of the following formulas (X31) to (X34):



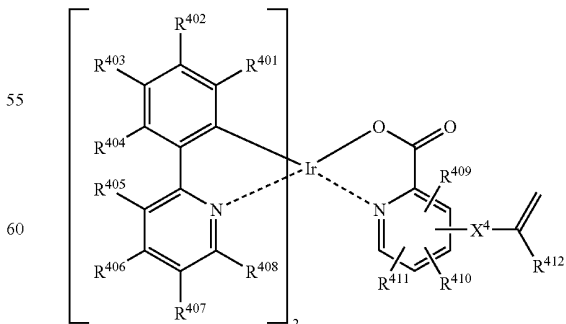
wherein R^{X31} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X32} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;

(X21)

(X22)

(X23)

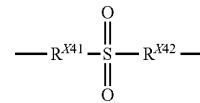
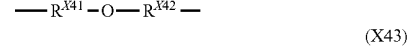
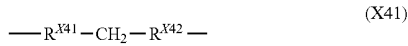
(X24)



wherein R⁴⁰¹ to R⁴¹¹ are each independently an atom or a substituent selected from the group consisting of a hydrogen

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atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, in each of R⁴⁰¹ to R⁴⁰⁴, R⁴⁰⁵ to R⁴⁰⁸, and R⁴⁰⁹ to R⁴¹¹, two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring, R⁴¹² is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X⁴ is a single bond or a group represented by any one of the following formulas (X41) to (X44):



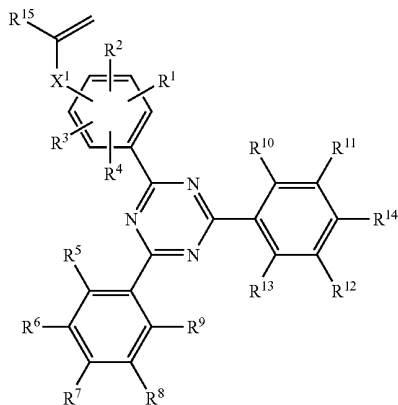
wherein R^{X41} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X42} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group.

[5] The organic light-emitting element as stated in the above [2], wherein the hole transport polymerizable compound is a carbazole derivative or a triarylamine derivative.

[6] A surface-emitting light source using the organic light-emitting element as stated in any one of the above [1] to [5].

[7] An image display using the organic light-emitting element as stated in any one of the above [1] to [5].

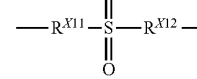
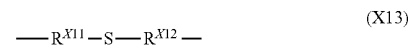
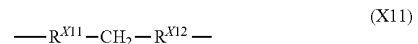
[8] A polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;



wherein R¹ to R¹⁴ are each independently a hydrogen atom, a halogen atom, a cyano group, an amino group, an alkyl group of 1 to 12 carbon atoms or an alkoxy group of 1 to 12 carbon atoms, in each of R¹ to R⁴, R⁵ to R⁹, and R¹⁰ to R¹⁴, two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring, R¹⁵ is a hydrogen atom or an alkyl group of 1 to

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12 carbon atoms, and X¹ is a single bond or a group represented by any one of the following formulas (X11) to (X14):



wherein R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group.

[9] The polymer compound as stated in the above [8], further comprising a constitutional unit derived from a hole transport polymerizable compound.

Effect of the Invention

According to the organic light-emitting element of the present invention, not only a low driving voltage but also high luminous efficiency and high luminance are obtained.

BRIEF DESCRIPTION OF THE DRAWING

FIG. 1 is a sectional view of an example of the organic light-emitting element according to the present invention.

DESCRIPTION OF NUMERICAL SYMBOLS

- 1: glass substrate
- 2: anode
- 3: hole transport layer
- 4: light-emitting layer
- 5: electron transport layer
- 6: cathode

BEST MODE FOR CARRYING OUT THE INVENTION

The present invention is described in detail hereinafter. In the present specification, the "electron transport property" and the "hole transport property" are together referred to also as "carrier transport property".

Organic Light-Emitting Element

The organic light-emitting element of the invention is an organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, and is characterized in that the light-emitting layer contains a specific polymer compound having a constitutional unit derived from an electron transport polymerizable compound represented by the aforesaid formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound. That is to say, in this polymer compound, the moiety containing an electron transport triazine ring is bonded to a polymer chain through a specific divalent group. According to the polymer compound, therefore, an organic light-emitting element having not only a low driving voltage but also high luminous efficiency and high luminance is obtained.

Specific embodiments of the invention are described below.

Embodiment 1 of the Invention

The organic light-emitting element (embodiment 1) of the invention is an organic light-emitting element constituted by arranging one light-emitting layer between an anode and a cathode, and the light-emitting layer contains a specific polymer compound (1) comprising a constitutional unit derived from an electron transport polymerizable compound represented by the aforesaid formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound and further comprising a constitutional unit derived from a hole transport polymerizable compound.

The polymer compound (1) for use in the embodiment 1 has the above constitutional units, and is obtained by copolymerizing an electron transport polymerizable compound represented by the aforesaid formula (1), a phosphorescent polymerizable compound and a hole transport polymerizable compound.

The electron transport polymerizable compound represented by the formula (1) has a carbon-carbon double bond that is a polymerizable functional group.

In the formula (1), R^1 to R^{14} are each independently a hydrogen atom, a halogen atom, a cyano group, an amino group, an alkyl group of 1 to 12 carbon atoms or an alkoxy group of 1 to 12 carbon atoms.

As the halogen atom, a fluorine atom, a chlorine atom, a bromine atom or an iodine atom can be mentioned.

Examples of the alkyl groups of 1 to 12 carbon atoms include methyl group, ethyl group, propyl group, isopropyl group, butyl group, isobutyl group, t-butyl group, amyl group, hexyl group, octyl group, decyl group, 2-ethylhexyl group and dodecyl group.

Examples of the alkoxy groups of 1 to 12 carbon atoms include methoxy group, ethoxy group, propoxy group, isopropoxy group, butoxy group, isobutoxy group, t-butoxy group, hexyloxy group, 2-ethylhexyloxy group, decyloxy group and dodecyloxy group.

From the viewpoints of enhancement of solubility and symmetry of molecule, it is preferable that R^7 and R^{14} are each an alkyl group of 1 to 12 carbon atoms or an alkoxy group of 1 to 12 carbon atoms and R^1 to R^6 and R^8 to R^{13} are each a hydrogen atom.

In each of R^1 to R^4 , R^5 to R^9 , and R^{10} to R^{14} , two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring.

R^{15} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms. Examples of the alkyl groups of 1 to 12 carbon atoms include the aforesaid alkyl groups.

Of these, a hydrogen atom is preferable as X^{15} from the viewpoint of carrier transport ability.

X^1 is a single bond or a group represented by any one of the aforesaid formulas (X11) to (X14). In the formulas, R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group. In the formula (1), it is preferable that R^{X11} is bonded to a benzene ring having R^1 to R^4 and R^{X12} is bonded to a vinyl group. According to such X^1 , an organic light-emitting element having not only a low driving voltage but also high luminous efficiency and high luminance is obtained.

Of these, a single bond or an alkylene group of 1 to 20 carbon atoms is preferable as X^1 , and a single bond is more

preferable. When no hetero atom is contained in X^1 as above, an organic light-emitting element having higher luminous efficiency is obtained.

The above electron transport polymerizable compounds may be used singly or may be used in combination of two or more kinds.

Such an electron transport polymerizable compound can be prepared by, for example, subjecting a benzonitrile derivative and bromobenzoyl chloride to cyclization reaction using Lewis acid and then coupling vinylboric acid by Suzuki coupling method.

The phosphorescent polymerizable compound has a substituent having a polymerizable functional group, and is not specifically restricted provided that it is a low-molecular compound from which light emission from a triplet excited state can be obtained at room temperature. Preferable are a palladium complex, an osmium complex, an iridium complex, a platinum complex and a gold complex each of which contains a substituent having a polymerizable functional group; more preferable are the iridium complex and the platinum complex; and most preferable is the iridium complex.

As such an iridium complex, a complex represented by any one of the aforesaid formulas (2) to (4) is preferably used. These polymerizable compounds have a carbon-carbon double bond that is a polymerizable functional group.

In the formula (2), R^{201} to R^{215} are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group.

As the halogen atom, a fluorine atom, a chlorine atom, a bromine atom or an iodine atom can be mentioned.

Examples of the alkyl groups of 1 to 10 carbon atoms include methyl group, ethyl group, propyl group, isopropyl group, butyl group, isobutyl group, t-butyl group, amyl group, hexyl group, octyl group and decyl group.

Examples of the aryl groups of 6 to 10 carbon atoms include phenyl group, tolyl group, xylyl group, mesityl group and naphthyl group.

Examples of the amino groups which may be substituted by an alkyl group of 1 to 10 carbon atoms include amino group, dimethylamino group, diethylamino group and dibutylamino group.

Examples of the alkoxy groups of 1 to 10 carbon atoms include methoxy group, ethoxy group, propoxy group, isopropoxy group, butoxy group, isobutoxy group, t-butoxy group, hexyloxy group, 2-ethylhexyloxy group and decyloxy group.

Examples of the silyl groups include trimethylsilyl group, triethylsilyl group, t-butyl dimethylsilyl group and trimethoxysilyl group.

From the viewpoint of carrier transport ability, it is preferable that R^{201} to R^{215} are each independently a hydrogen atom, a fluorine atom, a cyano group, a methyl group, a t-butyl group, a dimethylamino group, a butoxy group or a 2-ethylhexyloxy group, and it is more preferable that R^{202} and R^{204} are each a fluorine atom and R^{206} and R^{213} are each a t-butyl group. In this case, R^{201} to R^{215} which are none of polymerizable substituents, the above atoms and the above substituents are each a hydrogen atom.

In each of R^{201} to R^{204} , R^{205} to R^{208} , R^{209} to R^{211} , and R^{212} to R^{215} , two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring.

R^{216} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms. Examples of the alkyl groups of 1 to 12 carbon atoms include the aforesaid alkyl groups.

Of these, a hydrogen atom is preferable as R^{216} from the viewpoint of carrier transport ability.

X^2 is a single bond or a group represented by any one of the aforesaid formulas (X21) to (X24). In the formulas, R^{X21} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X22} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group. In the formula (2), it is preferable that R^{X21} is bonded to a benzene ring having R^{209} to R^{211} and R^{X22} is bonded to a vinyl group. When no hetero atom is contained in X^2 , an organic light-emitting element having higher luminous efficiency is obtained.

In the formula (3), R^{301} to R^{308} are each independently the same atom or substituent as that for R^{201} .

R^{309} to R^{310} are each independently the same atom or substituent as that for R^{201} , and a preferred range and the reason are the same as those for R^{201} (except a halogen atom).

From the viewpoint of carrier transport ability, it is preferable that R^{301} to R^{310} are each independently a hydrogen atom, a fluorine atom, a cyano group, a methyl group, a t-butyl group, a dimethylamino group, a butoxy group or a 2-ethylhexyloxy group, and it is more preferable that R^{302} and R^{304} are each a fluorine atom and R^{301} to R^{310} except R^{302} and R^{304} are each a hydrogen atom.

In each of R^{301} to R^{304} , and R^{305} to R^{308} , two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring.

R^{311} is the same atom or substituent as that for R^{216} , and a preferred range and the reason are the same as those for R^{216} .

X^3 is the same atom or substituent as that for X^2 , and a preferred range and the reason are the same as those for X^2 .

In the formula (4), R^{401} to R^{411} are each independently the same atom or substituent as that for R^{201} .

From the viewpoint of carrier transport ability, it is preferable that R^{401} to R^{411} are each independently a hydrogen atom, a fluorine atom, a cyano group, a methyl group, a t-butyl group, a dimethylamino group, a butoxy group or a 2-ethylhexyloxy group, and it is more preferable that R^{402} and R^{404} are each a fluorine atom and R^{401} to R^{411} except R^{402} and R^{404} are each a hydrogen atom.

In each of R^{401} to R^{404} , R^{405} to R^{408} , and R^{409} to R^{411} , two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring.

R^{412} is the same atom or substituent as that for R^{216} , and a preferred range and the reason are the same as those for R^{216} .

X^4 is the same atom or substituent as that for X^2 , and a preferred range and the reason are the same as those for X^2 .

The above phosphorescent polymerizable compounds may be used singly or may be used in combination of two or more kinds.

Such a phosphorescent polymerizable compound can be prepared by, for example, allowing iridium chloride and a phenylpyridine derivative to react with each other to form a binuclear complex of iridium and then allowing it to react with a ligand having a polymerizable functional group (ligand coordinated to the right-hand side of Ir in the formulas (2) to (4)).

As the hole transport polymerizable compound, a carbazole derivative or a triarylamine derivative, each containing a substituent having a polymerizable functional group, is preferably used.

Examples of such carbazole derivatives and triarylamine derivatives include

N,N,N' -diphenyl- N,N' -(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD),

N,N,N',N' -tetrakis(3-methylphenyl)-1,1'-(3,3'-dimethyl)biphenyl-4,4'-diamine (HMTPD),

4,4',4''-tris(3-methylphenylphenylamino)triphenylamine (m-MTDATA),

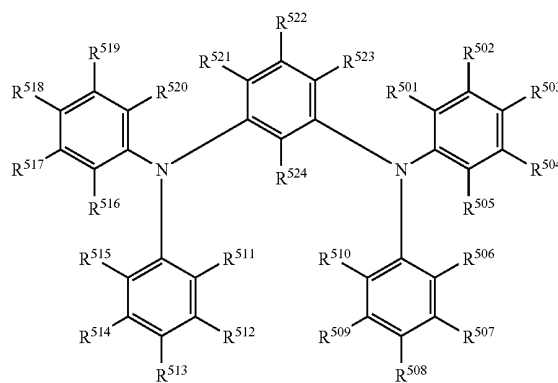
4,4'-biscarbazolylbiphenyl (CBP) and

4,4'-biscarbazolyl-2,2'-dimethylbiphenyl (CDBP) each of which contains a substituent having a polymerizable functional group.

The above hole transport polymerizable compounds may be used singly or may be used in combination of two or more kinds.

Hole transport polymerizable compounds represented by the following formulas (5) to (6) are also preferably used in the invention from the viewpoints of carrier transport ability and optical properties.

(5)

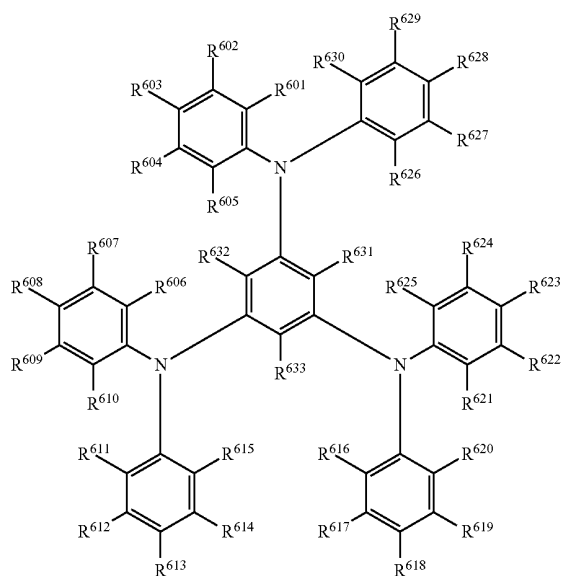


In the formula (5), at least one of R^{501} to R^{524} is a substituent having a polymerizable functional group, and R^{501} to R^{524} which are not the substituents having a polymerizable functional group are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms, a carbazole group and a silyl group.

Examples of the above atoms and substituents include the aforesaid atoms and substituents. The carbazole group may have a substituent, such as methyl group, ethyl group, t-butyl group or methoxy group.

In each of R^{501} to R^{505} , R^{506} to R^{510} , R^{511} to R^{515} , R^{516} to R^{520} , and R^{521} to R^{523} , two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring.

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In the formula (6), at least one of R⁶⁰¹ to R⁶³³ is a substituent having a polymerizable functional group, and R⁶⁰¹ to R⁶³³ which are not the substituents having a polymerizable functional group are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group.

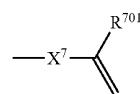
Examples of the above atoms and substituents include the aforesaid atoms and substituents.

In each of R⁶⁰¹ to R⁶⁰⁵, R⁶⁰⁶ to R⁶¹⁰, R⁶¹¹ to R⁶¹⁵, R⁶¹⁶ to R⁶²⁰, R⁶²¹ to R⁶²⁵, and R⁶²⁶ to R⁶³⁰, two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring.

With regard to the polymerizable compound represented by the formula (5) of the above compounds, at least one in each of R⁵⁰¹ to R⁵⁰⁵, R⁵⁰⁶ to R⁵¹⁰, R⁵¹¹ and R⁵¹⁵, and R⁵¹⁶ to R⁵²⁰ is preferably the above atom or the above substituent other than a hydrogen atom. In this case, R⁵⁰¹ to R⁵²⁴ which are none of polymerizable functional groups, the above atoms and the above substituents are each a hydrogen atom. With regard to the polymerizable compound represented by the formula (6), at least one in each of R⁶⁰¹ to R⁶⁰⁵, R⁶⁰⁶ to R⁶¹⁰, R⁶¹¹ to R⁶¹⁵, R⁶¹⁶ to R⁶²⁰, R⁶²¹ to R⁶²⁵, to R⁶³⁰ is preferably the above atom or the above substituent other than a hydrogen atom. In this case, R⁶⁰¹ to R⁶³³ which are none of polymerizable functional groups, the above atoms and the above substituents are each a hydrogen atom.

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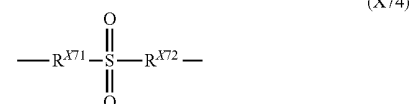
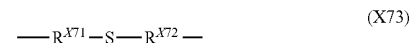
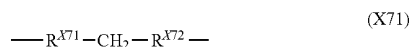
(6) As the substituent having a polymerizable functional group, a substituent represented by the following formula (7) is preferable.



In the formula (7), R⁷⁰¹ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms. Examples of the alkyl groups of 1 to 12 carbon atoms include the aforesaid alkyl groups.

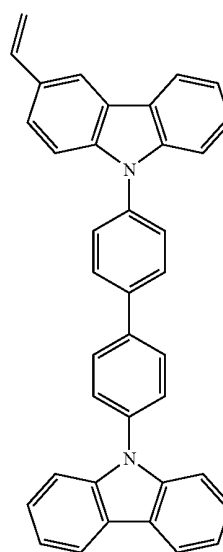
Of these, a hydrogen atom is preferable as R⁷⁰¹ from the viewpoint of carrier transport ability.

X⁷ is a group represented by any one of the following formulas (X71) to (X74).



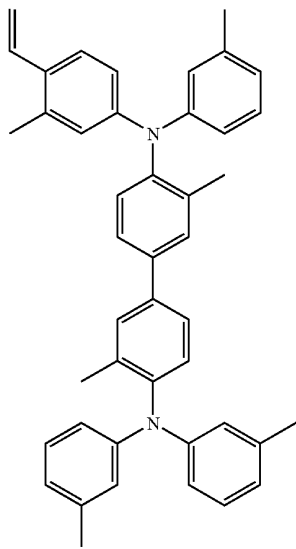
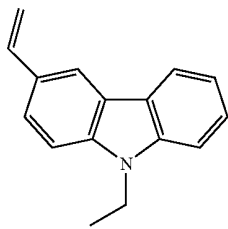
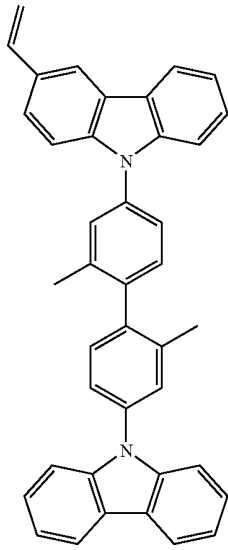
In the above formulas, R^{X71} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X72} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group. In the formulas (5) and (6), it is preferable that R^{X71} is bonded to a benzene ring and R^{X72} is bonded to a vinyl group. When no hetero atom is contained in X⁷, an organic light-emitting element having higher luminous efficiency is obtained.

As the hole transport polymerizable compounds, there can be more specifically mentioned compounds represented by the following formulas (8-1) to (8-10).



(8-1)

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-continued



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-continued

(8-2)

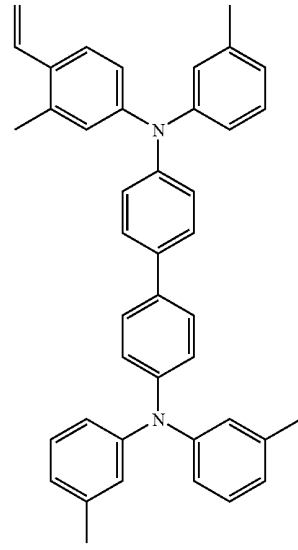
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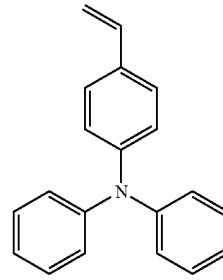
(8-5)

(8-3)

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(8-6)

(8-4)

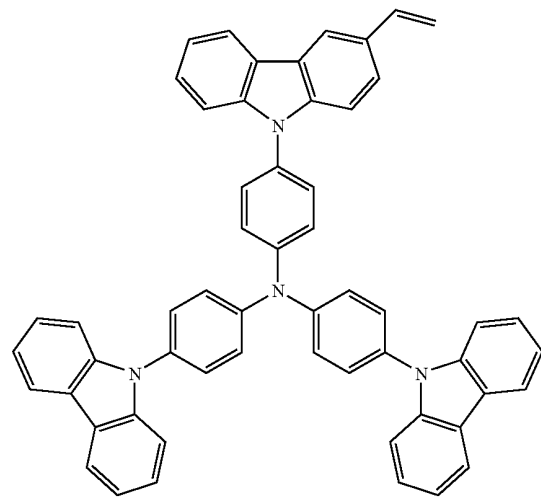
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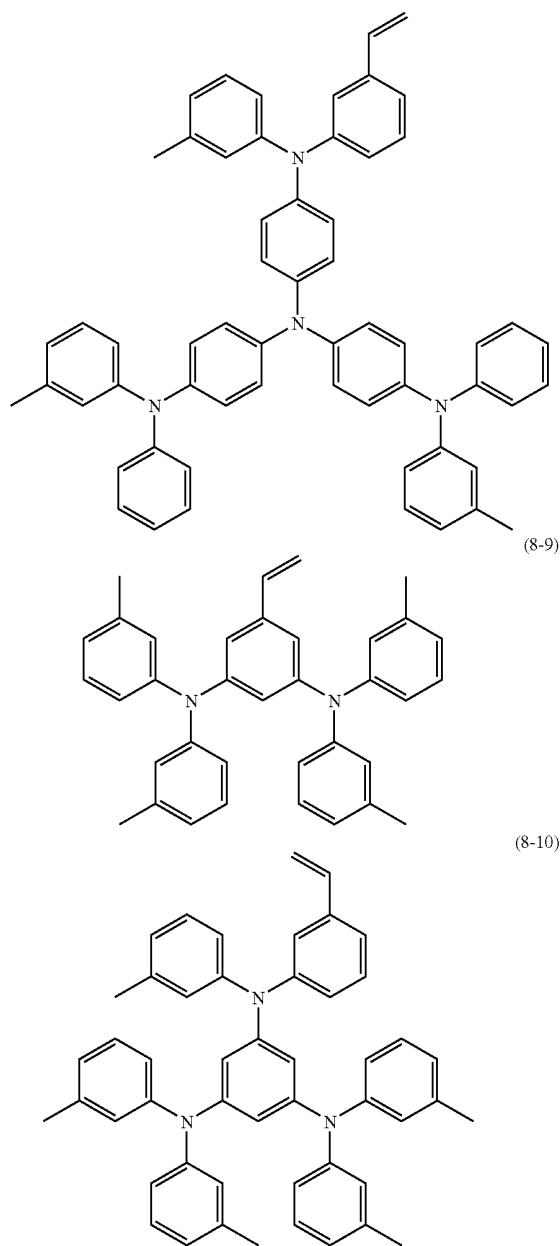
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(8-7)

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-continued



The above hole transport polymerizable compounds may be used singly or may be used in combination of two or more kinds.

The compound represented by the formula (5) can be prepared by, for example, palladium catalyst substitution reaction of a *m*-phenylenediamine derivative with halogenated aryl, or diarylamine with a *m*-dibromobenzene derivative. A specific method of the substitution reaction is described in, for example, *Tetrahedron Letters*, 1998, vol. 39, p. 2367. The compound represented by the formula (6) can be prepared by, for example, palladium catalyst substitution reaction of 1,3,5-triaminobenzene with halogenated aryl, or diarylamine with 1,3,5-trihalogenated benzene. A specific method of the substitution reaction is described in, for example, *Tetrahedron Letters*, 1998, vol. 39, p. 2367.

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In the preparation of the polymer compound (1), other polymerizable compounds may be further used. Examples of the other polymerizable compounds include compounds having no carrier transport property, e.g., alkyl(meth)acrylates, such as methyl acrylate and methyl methacrylate, styrene and derivatives thereof, but the other polymerizable compounds are not limited to these compounds. In the polymer compound (1), the content of structural units derived from other polymerizable compounds is preferably in the range of 0 to 50% by mol.

Although the preparation of the polymer compound (1) may be carried out by any of radical polymerization, cationic polymerization, anionic polymerization and addition polymerization using the aforesaid polymerizable compounds, it is preferable to carry out the preparation by radical polymerization.

The weight-average molecular weight of the polymer compound (1) is desired to be in the range of usually 1,000 to 2,000,000, preferably 5,000 to 500,000. When the weight-average molecular weight is in this range, the polymer compound (1) is soluble in organic solvents and a uniform thin film is obtained, so that such a weight-average molecular weight is preferable. The weight-average molecular weight is a value measured by gel permeation chromatography (GPC) at 40° C. using tetrahydrofuran as a solvent.

With regard to the solubility of the polymer compound (1) in organic solvents such as toluene and chloroform, it is preferable that 1 part by weight of the polymer compound (1) is dissolved in 1 to 200 parts by weight of an organic solvent, and it is more preferable that 1 part by weight of the polymer compound (1) is dissolved in 10 to 50 parts by weight of an organic solvent.

When the number of constitutional units derived from the phosphorescent polymerizable compound is designated by *m* and the number of constitutional units derived from the carrier transport polymerizable compound is designated by *n* (*m* and *n* are each an integer of not less than 1) in the polymer compound (1), the proportion of the number of constitutional units derived from the phosphorescent polymerizable compound to the number of all the constitutional units, namely a value of $m/(m+n)$, is in the range of preferably 0.001 to 0.5, more preferably 0.001 to 0.2. When the value of $m/(m+n)$ is in this range, an organic light-emitting element which has high carrier mobility, has low concentration quenching effect and has high luminous efficiency is obtained.

When the number of constitutional units derived from the hole transport polymerizable compound is designated by *x* and the number of constitutional units derived from the electron transport polymerizable compound is designated by *y* (*x* and *y* are each an integer of not less than 1) in the polymer compound (1), *x*, *y* and the above *n* have a relationship of $n=x+y$ between them. Optimum values of the proportion of the number of structural units derived from the hole transport polymerizable compound to the number of structural units derived from the carrier transport compound, i.e., x/n , and the proportion of the number of structural units derived from the electron transport polymerizable compound to the number of structural units derived from the carrier transport compound, i.e., y/n , are determined by electric charge transport ability, concentration, etc. of the structural units. In the case where the light-emitting layer of the organic light-emitting element is formed from only the polymer compound (1), values of x/n and y/n are each in the range of preferably 0.05 to 0.95, more preferably 0.20 to 0.80. Then, an expression $x/n+y/n=1$ is established. The proportions of the constitutional units in such a polymer compound as above are estimated by ICP elemental analysis and ¹³C-NMR measurement.

If polymerization is carried out by properly controlling the ratio between the electron transport polymerizable compound, the phosphorescent polymerizable compound and the hole transport polymerizable compound in the above range, a polymer compound (1) having a desired structure is obtained.

The polymer compound (1) may be any of a random copolymer, a block copolymer and an alternating copolymer.

In the organic light-emitting element of the embodiment 1, one light-emitting layer containing the specific polymer compound (1) is arranged between an anode and a cathode. The polymer compound (1) has all of electron transport property, phosphorescent property and hole transport property, and therefore, when the light-emitting layer is formed by using this, an organic light-emitting element having high luminous efficiency can be formed even if a layer of another organic material is not provided. In the embodiment 1, the light-emitting layer can be formed from only the polymer compound (1), so that this embodiment also has an advantage that the production process can be more simplified.

The light-emitting layer is formed on an anode provided on a substrate usually in the following manner. First, a solution in which the polymer compound (1) has been dissolved is prepared. The solvent used for preparing the solution is not specifically restricted, but for example, chlorine-based solvents, such as chloroform, methylene chloride and dichloroethane, ether-based solvents, such as tetrahydrofuran and anisole, aromatic hydrocarbon-based solvents, such as toluene and xylene, ketone-based solvents, such as acetone and methyl ethyl ketone, and ester-based solvents, such as ethyl acetate, butyl acetate and ethyl cellosolve acetate, are used. Subsequently, the solution thus prepared is applied onto a substrate by, for example, wet film-forming processes, such as spin coating, casting, microgravure coating, gravure coating, bar coating, roll coating, wire bar coating, dip coating, spray coating, screen printing, flexographic printing, offset printing and ink-jet printing. In the case of spin coating or dip coating, the solution preferably contains the solvent in an amount of 1000 to 20000 parts by weight based on 100 parts by weight of the polymer compound (1) though the amount of the solvent depends upon the compound used, the film-forming conditions, etc.

By providing a cathode on the light-emitting layer thus formed, an organic light-emitting element of the embodiment 1 is obtained.

As the substrate for use in the embodiment 1, an insulating substrate transparent to the emission wavelength of the above light-emitting material is preferably used, and specifically, glass, transparent plastics such as PET (polyethylene terephthalate) and polycarbonate, etc. are used.

As the anode materials for use in the embodiment 1, publicly known transparent conductive materials, e.g., ITO (indium tin oxide), tin oxide, zinc oxide and conductive polymers, such as polythiophene, polypyrrole and polyaniline, are preferably used. The surface resistance of the electrode formed by the transparent conductive material is preferably in the range of 1 to $50\Omega/\square$ (ohm/square). The thickness of the anode is preferably in the range of 50 to 300 nm.

As the cathode materials for use in the embodiment 1, publicly known cathode materials, e.g., alkali metals, such as Li, Na, K and Cs; alkaline earth metals, such as Mg, Ca and Ba; Al; Mg—Ag alloy; and alloys of Al and an alkali metal or an alkaline earth metal, such as Al—Li and Al—Ca, are preferably used. The thickness of the cathode is desired to be

in the range of preferably 10 nm to 1 μm , more preferably 50 to 500 nm. In the case where a metal having high activity, such as an alkali metal or an alkaline earth metal, is used, the thickness of the cathode is desired to be in the range of preferably 0.1 to 100 nm, more preferably 0.5 to 50 nm. In this case, a metal layer stable to the atmosphere is laminated on this cathode in order to protect the cathode metal. The metal to form the metal layer is, for example, Al, Ag, Au, Pt, Cu, Ni or Cr. The thickness of the metal layer is desired to be in the range of preferably 10 nm to 1 μm , more preferably 50 to 500 nm.

As the film-forming method using the above anode material, electron beam deposition, sputtering, chemical reaction, coating or the like is used, and as the film-forming method using the cathode material, resistance heating deposition, electron beam deposition, sputtering, ion plating or the like is used.

Embodiment 2 of the Invention

The organic light-emitting element (embodiment 2) of the invention is an organic light-emitting element constituted by arranging one light-emitting layer between an anode and a cathode, and the light-emitting layer contains a specific polymer compound (2) having a constitutional unit derived from an electron transport polymerizable compound represented by the aforesaid formula (1) and a constitutional unit derived from a light-emitting polymerizable compound and further contains a hole transport compound.

The polymer compound (2) for use in the embodiment (2) has the above constitutional units, and is obtained by copolymerizing an electron transport polymerizable compound represented by the aforesaid formula (1) and a light-emitting polymerizable compound.

The electron transport polymerizable compound represented by the formula (1) and the light-emitting polymerizable compound have the same meanings as those of the electron transport polymerizable compound and the light-emitting polymerizable compound for use in the embodiment 1, and a preferred range and the reason are the same as those in the embodiment (1).

Other polymerizable compounds which may be further used in the preparation of the polymer compound (2) are also the same as those in the embodiment (1).

Although the preparation of the polymer compound (2) may be carried out by any of radical polymerization, cationic polymerization, anionic polymerization and addition polymerization, it is preferable to carry out the preparation by radical polymerization.

The weight-average molecular weight of the polymer compound (2) is the same as that in the embodiment (1). The solubility of the polymer compound (2) in organic solvents is the same as that in the embodiment (1).

When the number of constitutional units derived from the phosphorescent polymerizable compound is designated by m and the number of constitutional units derived from the electron transport polymerizable compound is designated by n (m and n are each an integer of not less than 1) in the polymer compound (2), the proportion of the number of constitutional units derived from the phosphorescent polymerizable compound to the number of all the constitutional units, namely a value of $m/(m+n)$, is in the range of preferably 0.001 to 0.5, more preferably 0.001 to 0.2. When the value of $m/(m+n)$ is in this range, an organic light-emitting element which has high carrier mobility, has low concentration quenching effect and has high luminous efficiency is obtained. The proportions of

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the constitutional units in such a polymer compound as above are estimated by ICP elemental analysis and ^{13}C -NMR measurement.

If polymerization is carried out by properly controlling the ratio between the electron transport polymerizable compound and the phosphorescent polymerizable compound in the above range, a polymer compound (2) having a desired structure is obtained.

The polymer compound (2) may be any of a random copolymer, a block copolymer and an alternating copolymer.

As the hole transport compound for use in the embodiment 2, a carbazole derivative or a triarylamine derivative is preferably used.

Examples of the carbazole derivatives and the triarylamine derivatives include

N,N' -diphenyl- N,N' -(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD),

N,N,N',N' -tetrakis(3-methylphenyl)-1,1'-(3,3'-dimethyl)biphenyl-4,4'-diamine (HMTPD),

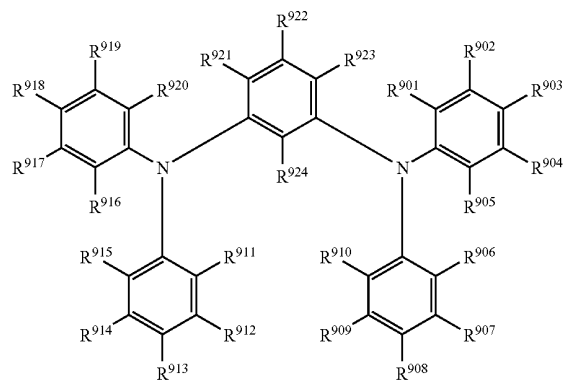
4,4',4''-tris(3-methylphenylphenylamino)triphenylamine (m-MTDATA),

4,4'-biscarbazolylbiphenyl (CBP) and

4,4'-biscarbazolyl-2,2'-dimethylbiphenyl (CDBP).

The above hole transport polymerizable compounds may be used singly or may be used in combination of two or more kinds.

Hole transport compounds represented by the following formulas (9) to (10) are also preferably used in the invention from the viewpoints of carrier transport ability and optical properties.



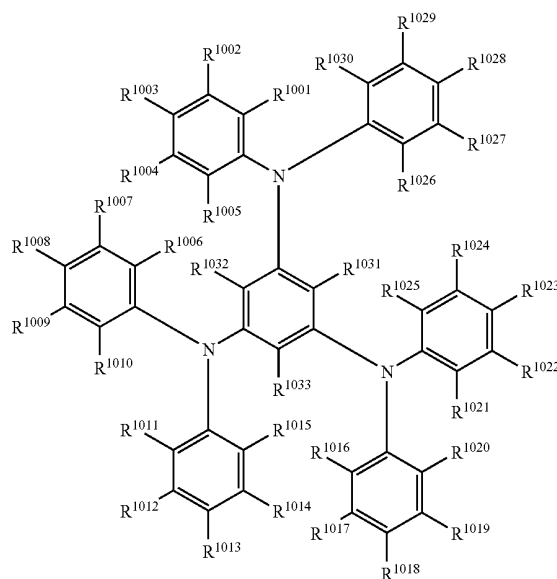
In the formula (9), R^{901} to R^{924} are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms, a carbazole group and a silyl group.

Examples of the above atoms and substituents are the same as those for R^{501} .

In each of R^{901} to R^{905} , R^{906} to R^{910} , R^{911} to R^{915} , R^{916} to R^{920} , and R^{921} to R^{923} , two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring.

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(10)



In the formula (10), R^{1001} to R^{1033} are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group.

Examples of the above atoms and substituents are the same as those for R^{601} .

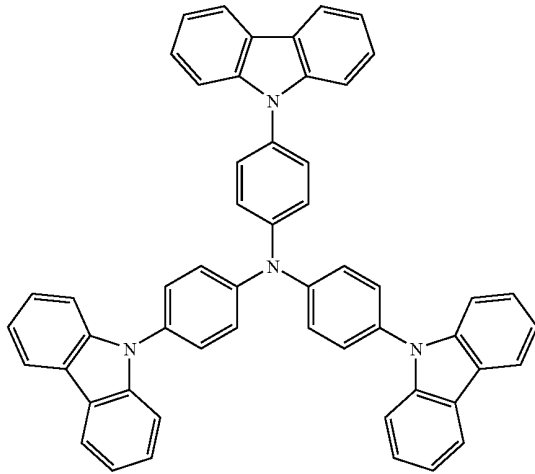
In each of R^{1001} to R^{1005} , R^{1006} to R^{1010} , R^{1011} to R^{1015} , R^{1016} to R^{1020} , R^{1021} to R^{1025} , and R^{1026} to R^{1030} , two groups bonded to carbon atoms adjacent to each other on a benzene ring may be bonded to each other to form a condensed ring.

With regard to the polymerizable compound represented by the formula (9) of the above compounds, at least one in each of R^{901} to R^{905} , R^{906} to R^{910} , R^{911} to R^{915} , and R^{916} to R^{920} is preferably the above atom or the above substituent other than a hydrogen atom. In this case, R^{901} to R^{924} which are neither the above atoms nor the above substituents are each a hydrogen atom. With regard to the polymerizable compound represented by the formula (10), at least one in each of R^{1001} to R^{1005} , R^{1006} to R^{1010} , R^{1011} to R^{1015} , R^{1016} to R^{1020} , R^{1021} to R^{1025} , and R^{1026} to R^{1030} is preferably the above atom or the above substituent other than a hydrogen atom. In this case, R^{1001} to R^{1033} which are neither the above atoms nor the above substituents are each a hydrogen atom.

As the hole transport compounds, there can be more specifically mentioned compounds represented by the following formulas (11-1) to (11-10).

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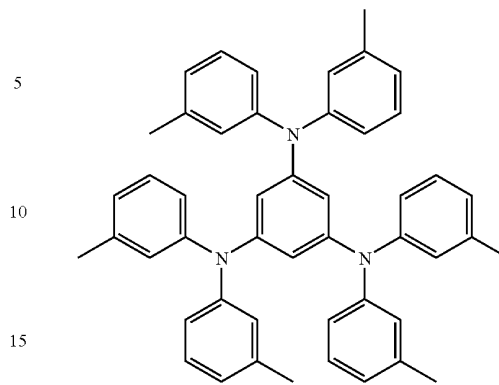
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(11-10)

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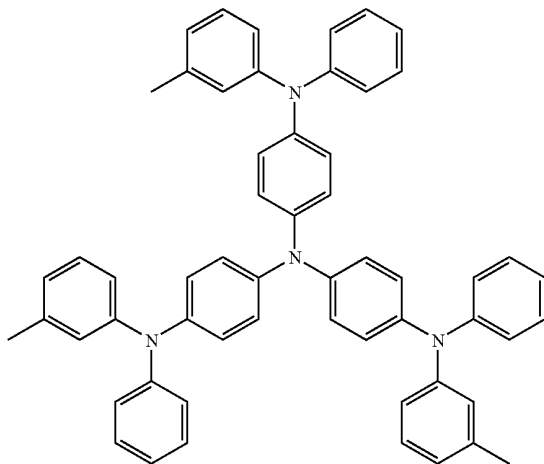
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The above hole transport compounds may be used singly or may be used in combination of two or more kinds.

The compound represented by the formula (9) can be prepared by, for example, palladium catalyst substitution reaction of a m-phenylenediamine derivative with halogenated aryl, or diarylamine with a m-dibromobenzene derivative. A specific method of the substitution reaction is described in, for example, *Tetrahedron Letters*, 1998, vol. 39, p. 2367. The compound represented by the formula (10) can be prepared by, for example, palladium catalyst substitution reaction of 1,3,5-triaminobenzene with halogenated aryl, or diarylamine with 1,3,5-trihalogenated benzene. A specific method of the substitution reaction is described in, for example, *Tetrahedron Letters*, 1998, vol. 39, p. 2367.

(11-8)



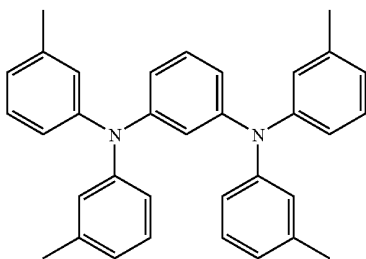
In the case where the hole transport compound is a low-molecular compound and is a compound which is unable to be applied, a polymer compound obtained by polymerizing the hole transport polymerizable compound described in the embodiment 1 may be used.

When the hole transport polymer compound is used, a preferred weight-average molecular weight and solubility in organic solvents are the same as those in the embodiment 1.

In the organic light-emitting element of the embodiment 2, one light-emitting layer containing the hole transport compound together with the specific polymer compound (2) is arranged between an anode and a cathode. In this case, the light-emitting layer desirably contains the hole transport compound in an amount of preferably 10 to 200 parts by weight, more preferably 50 to 150 parts by weight, based on 100 parts by weight of the polymer compound (2). When the light-emitting layer is formed by using the hole transport compound together with the polymer compound (2) having both of electron transport property and phosphorescent property, an organic light-emitting element having high luminous efficiency can be prepared even if a layer of another organic material is not provided.

The light-emitting layer is formed on an anode provided on a substrate usually in the following manner. First, a solution in which the polymer compound (2) and the hole transport compound have been dissolved is prepared. The solvent used for preparing the solution is the same as that in the embodiment 1. The film-forming method using the solution prepared is the same as that in the embodiment 1. In the case of spin coating or dip coating, the solution preferably contains the hole transport compound in an amount of 10 to 200 parts by weight and the solvent in an amount of 1000 to 20000 parts by weight, each amount being based on 100 parts by weight of the polymer compound (2), though the amounts of them depend upon the compounds used, the film-forming conditions, etc.

(11-9)



By providing a cathode on the light-emitting layer thus formed, an organic light-emitting element of the embodiment 2 is obtained.

The substrate, the anode material, the cathode material and the film-forming methods using the anode material and the cathode material are the same as those in the embodiment 1.

Embodiment 3 of the Invention

The organic light-emitting element according to the invention may be an organic light-emitting element constituted by arranging the light-emitting layer described in the embodiment 1 or 2 and other organic layers between an anode and a cathode (embodiment 3).

Examples of the other organic layers include a hole transport layer, an electron transport layer, a hole block layer and a buffer layer. By providing these layers, the luminous efficiency can be further enhanced.

One example of the constitution of the organic light-emitting element (embodiment 3) according to the invention is shown in FIG. 1. In FIG. 1, between an anode (2) provided on a transparent substrate (1) and a cathode (6), a hole transport layer (3), the light-emitting layer (4) described in the embodiment 1 or 2, and an electron transport layer (5) are provided in this order.

In the embodiment 3, between the anode (2) and the cathode (6) may be provided for example any one of [1] a hole transport layer/the above light-emitting layer and [2] the above light-emitting layer/an electron transport layer.

The above organic layers may be each formed by mixing a high-molecular material as a binder. Examples of the high-molecular materials include polymethyl methacrylate, polycarbonate, polyester, polysulfone and polyphenylene oxide.

A hole transport compound and an electron transport compound for use in the above hole transport layer and the above electron transport layer may be each used singly to form respective layers, or may be each mixed with a material having a different function to form respective layers.

Examples of the hole transport compounds to form the hole transport layer include TPD (N,N'-dimethyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine); α -NPD (4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl); a low-molecular triphenylamine derivative, such as m-MTDATA (4,4',4''-tris(3-methylphenylphenylamino)triphenylamine); polyvinylcarbazole; a polymer compound obtained by introducing a polymerizable functional group into the above triphenylamine derivative and polymerizing them; and fluorescence-emitting polymer compounds, such as polyparaphenylene vinylene and polydialkylfluorene. An example of the above polymer compound is a polymer compound having a triphenylamine skeleton disclosed in Japanese Patent Laid-Open Publication No. 157575/1996. The above hole transport compounds may be used singly or as a mixture of two or more kinds, or different hole transport compounds may be laminated upon each other. The thickness of the hole transport layer is desired to be in the range of preferably 1 nm to 5 μ m, more preferably 5 nm to 1 μ m, particularly preferably 10 nm to 500 nm, though it depends upon electrical conductivity of the hole transport layer, etc.

Examples of the electron transport compounds to form the electron transport layer include low-molecular compounds, specifically, quinolinol derivative metal complex such as

Alq3 (aluminum trisquinolinolate), oxadiazole derivative, triazole derivative, imidazole derivative, triazine derivative and triarylborane derivative; and polymer compounds obtained by introducing a polymerizable substituent into the above low-molecular compounds and polymerizing them. An example of the polymer compound is poly-PBD disclosed in Japanese Patent Laid-Open Publication No. 1665/1998. The above electron transport compounds may be used singly or as a mixture of two or more kinds, or different electron transport compounds may be laminated upon each other. The thickness of the electron transport layer is desired to be in the range of preferably 1 nm to 5 μ m, more preferably 5 nm to 1 μ m, particularly preferably 10 nm to 500 nm, though it depends upon electrical conductivity of the electron transport layer, etc.

For the purpose of inhibiting passing of holes through the light-emitting layer and thereby efficiently recombining holes with electrons in the light-emitting layer, a hole block layer may be provided adjacently to the cathode side of the light-emitting layer. For the formation of the hole block layer, publicly known materials, such as triazole derivative, oxadiazole derivative and phenanthroline derivative, are used.

Between the anode and the hole transport layer or between the anode and the organic layer laminated adjacently to the anode, a buffer layer may be further provided in order to lower injection barrier in the hole injection. For forming the buffer layer, publicly known materials, such as copper phthalocyanine and a mixture of polyethylene dioxythiophene and polystyrenesulfonic acid (PEDOT:PSS), are used.

Between the cathode and the electron transport layer or between the cathode and the organic layer laminated adjacently to the cathode, an insulating layer having a thickness of 0.1 to 10 nm may be further provided in order to enhance electron injection efficiency. For forming the insulating layer, publicly known materials, such as lithium fluoride, sodium fluoride, magnesium fluoride, magnesium oxide and alumina, are used.

As methods for forming the hole transport layer and the electron transport layer, there can be used for example dry film-forming methods, such as resistance heating deposition, electron beam deposition and sputtering, and wet film-forming methods, such as spin coating, casting, microgravure coating, gravure coating, bar coating, roll coating, wire bar coating, dip coating, spray coating, screen printing, flexographic printing, offset printing and ink-jet printing, can be used. In the case of a low-molecular compound, a dry film-forming method is preferably used, and in the case of a polymer compound, a wet film-forming method is preferably used.

Usage

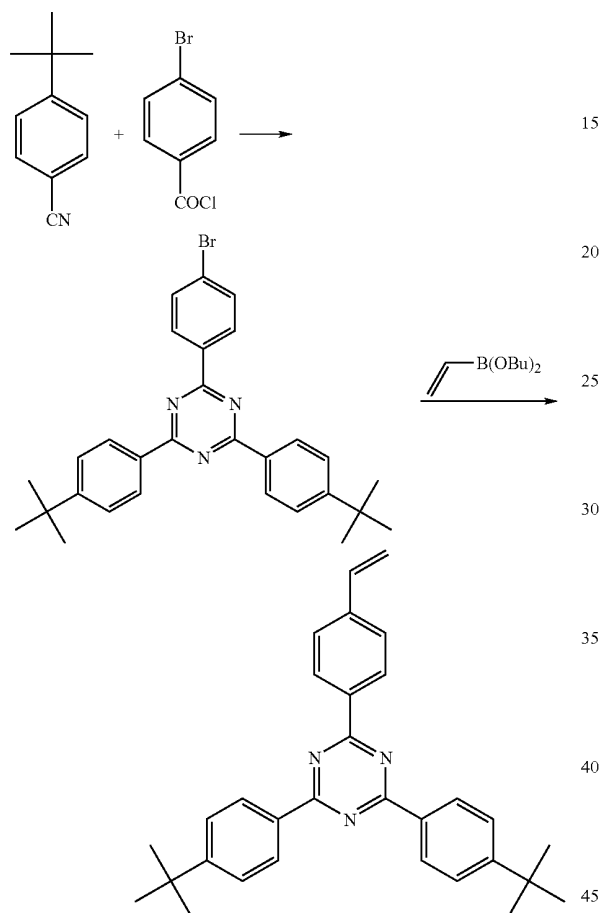
The organic light-emitting element according to the invention is preferably used as a pixel by matrix system or segment system in an image display in a publicly known manner. Moreover, the organic light-emitting element is preferably used also as a surface-emitting light source without forming a pixel.

The organic light-emitting element according to the invention is preferably used for, specifically, display, backlight, electrophotograph, illuminating light source, recording light source, exposure light source, reading light source, marker, advertizing display, interior goods, optical communication, etc.

The present invention is further described with reference to the following examples, but it should be construed that the invention is in no way limited to those examples.

Examples

Synthesis Example 1



In 50 ml of dichloroethane, 10 mmol of 4-bromobenzoyl chloride and 30 mmol of 4-tert-butylbenzonitrile were dissolved, and 10 mmol of aluminum chloride and 40 mmol of ammonium chloride were added, followed by refluxing by heating for 24 hours. After the resulting solution was cooled down to room temperature, the solution was poured into 10% hydrochloric acid, and they were stirred for 1 hour. Then, extraction was carried out using chloroform, and purification was carried out by column chromatography to obtain a halogenated triazine derivative.

In a 100 ml egg plant type flask, 3 mmol of the resulting halogenated triazine derivative, 3.3 mmol of dibutyl vinylborate and 1.5 mmol of tetrabutyl ammonium bromide were placed, and 45 ml of toluene and 30 ml of a 2M potassium carbonate aqueous solution were further added. A small amount of a polymerization inhibitor was added, and 0.15 mmol of tetrakis(triphenylphosphine)palladium was added, followed by refluxing by heating for 3 hours. After the resulting solution was cooled down to room temperature, extraction was carried out using ethyl acetate, and column chroma-

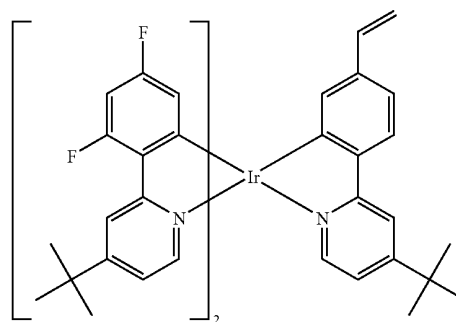
tography and recrystallization operation were carried out to obtain a white vinyl monomer of a triazine derivative (1.20 g, yield: 27%).

Example 1-1

Synthesis of Polymer Compound (1)

In a closed container, 95 mg of the compound synthesized in Synthesis Example 1, 10 mg of a compound (F) and 95 mg of the hole transport polymerizable compound (8-3) were placed, and 2.4 ml of dehydrated toluene was further added. Subsequently, a toluene solution (0.1 M, 39 μ l) of V-601 (available from Wako Pure Chemical Industries, Ltd.) was added, followed by five cycles of freezing and degassing. The container was closed up tight under vacuum, and the solution was stirred at 60° C. for 60 hours. After the reaction, the reaction solution was dropwise added to 100 ml of acetone to obtain a precipitate. Then, reprecipitation operation with toluene-acetone was further repeated twice, and thereafter, the precipitate was vacuum dried for one night at 50° C. to obtain a polymer compound (1). The polymer compound (1) had a weight-average molecular weight (Mw) of 82,000 and a molecular weight distribution index (Mw/Mn) of 1.80. The m/(m+n) value, the x/n value and the y/n value of the polymer compound estimated from the results of ICP elemental analysis and ¹³C-NMR measurement were 0.09, 0.51 and 0.49, respectively.

Compound (F)



Example 1-2

Element Using Polymer Compound (1)

A substrate with ITO (available from Nippo Electric Co, Ltd.) was used. This substrate was a substrate in which two ITO (indium tin oxide) electrodes (anodes) each having a width of 4 mm had been formed in the form of stripes on one surface of a glass substrate of 25 mm square.

In the first place, onto the substrate with ITO, poly(3,4-ethylenedioxythiophene)polystyrenesulfonic acid (available from Bayer Co., Ltd., trade name: Baytron P) was applied by spin coating under the conditions of a revolution number of 3500 rpm and a coating time of 40 seconds. Thereafter, the coating layer was dried under reduced pressure at 60° C. for 2 hours by a vacuum dryer to form an anode buffer layer. The thickness of the resulting anode buffer layer was about 50 nm. Next, 90 mg of the polymer compound (1) was dissolved in 2910 mg of toluene (available from Wako Pure Chemical Industries, Ltd., special grade), and this solution was filtered through a filter having a pore diameter of 0.2 μ m to prepare a coating solution. Subsequently, onto the anode buffer layer, the coating solution was applied by spin coating under the

conditions of a revolution number of 3000 rpm and a coating time of 30 seconds. After the application, the coating layer was dried at room temperature (25° C.) for 30 minutes to form a light-emitting layer. The thickness of the resulting light-emitting layer was about 100 nm.

Next, the substrate having the light-emitting layer thus formed was placed in a deposition apparatus. Then, barium and aluminum were co-deposited in a weight ratio of 1:10 to form two cathodes each having a width of 3 mm in the form of stripes so that the cathodes should be at right angles to the extending directions of the anodes. The film thickness of the resulting cathode was above 50 nm.

In the last place, a lead wire (wiring) was connected to the anode and the cathode in an argon atmosphere to prepare four organic EL elements of 4 mm (length)×3 mm (width). To the organic EL elements, a voltage was applied by the use of programmable direct voltage/current source (TR6143, manufactured by Advantest Corporation) to allow the elements to undergo light emission.

The light emission luminance was measured by the use of a luminance meter (BM-8, manufactured by Topcon Corporation). Maximum external quantum efficiency, maximum attainable brightness and driving voltage of the organic light-emitting elements prepared and brightness half life measured when the organic light-emitting elements were lighted up at an initial luminance of 100 cd/m² and subjected to constant-current driving are set forth in Table 1.

Example 2-1

Synthesis of Polymer Compound (2)

In a closed container, 160 mg of the compound synthesized in Synthesis Example 1 and 40 mg of the compound (F) were placed, and 2.4 ml of dehydrated toluene was further added. Subsequently, a toluene solution (0.1 M, 39 μl) of V-601 (available from Wako Pure Chemical Industries, Ltd.) was added, and freeze degassing operation was repeated 5 times. The container was closed up tight under vacuum, and the solution was stirred at 60° C. for 60 hours. After the reaction, the reaction solution was dropwise added to 100 ml of acetone to obtain a precipitate. Then, reprecipitation operation with toluene-acetone was further repeated twice, and thereafter, the precipitate was vacuum dried for one night at 50° C. to obtain a polymer compound (2). The polymer compound (2) had a weight-average molecular weight (Mw) of 85,000 and a molecular weight distribution index (Mw/Mn) of 1.62. The m/(m+n) value estimated from the results of ICP elemental analysis and ¹³C-NMR measurement was 0.16.

Example 2-2

Element Using Polymer Compound (2)

First, a polymer compound (4) was synthesized in the following manner. In a closed container, 95 mg of the hole transport polymerizable compound (8-3) was placed, and 2.2 ml of dehydrated toluene was further added. Subsequently, a toluene solution (0.1 M, 43 μl) of V-601 (available from Wako Pure Chemical Industries, Ltd.) was added, and freeze degassing operation was repeated 5 times. The container was closed up tight under vacuum, and the solution was stirred at 60° C. for 60 hours. After the reaction, the reaction solution was dropwise added to 100 ml of acetone to obtain a precipitate. Then, reprecipitation operation with toluene-acetone was further repeated twice, and thereafter, the precipitate was vacuum dried for one night at 50° C. to obtain a polymer compound (4).

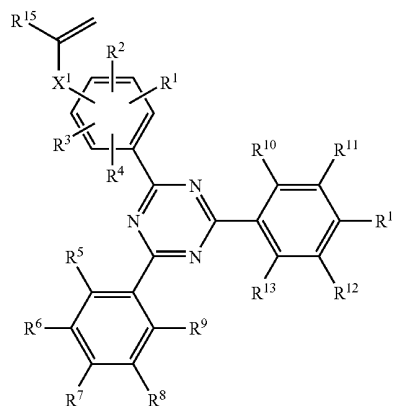
Next, organic light-emitting elements were prepared in the same manner as in Example 1-2, except that a coating solution for use in the formation of a light-emitting layer was prepared using 45 mg of the polymer compound (2), 45 mg of the polymer compound (4) and 2910 mg of toluene instead of 90 mg of the polymer compound (1) and 2910 mg of toluene. Maximum external quantum efficiency, maximum attainable brightness and driving voltage of the organic light-emitting elements prepared and luminance half life measured when the organic light-emitting elements were lighted up at an initial luminance of 100 cd/m² and subjected to constant-current driving are set forth in Table 1.

TABLE 1

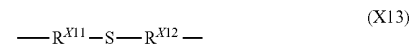
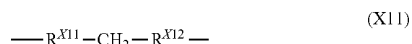
Ex.	Polymer compound	Maximum external quantum efficiency (%)	Maximum attainable brightness (cd/m ²)	Driving voltage (v)	Brightness half life (h)
1-2	(1)	7.1	37,000	3.2	1000
2-2	(2), (4)	5.9	31,000	4.4	760

The invention claimed is:

1. An organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, wherein the light-emitting layer contains a polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;

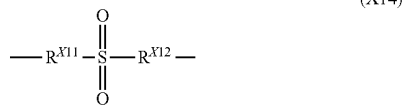


wherein in formula (1), R⁷ and R¹⁴ are each independently an alkyl group having 1 to 12 carbon atoms or an alkoxy group having 1 to 12 carbon atoms, and R¹ to R⁶ and R⁸ to R¹³ are hydrogen atoms, R¹⁵ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X¹ is a single bond or a group represented by any one of the following formulas (X11) to (X14):



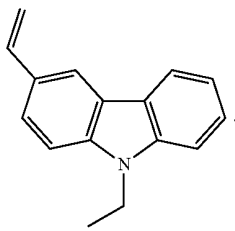
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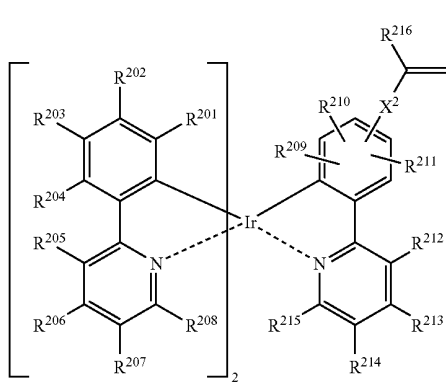


wherein R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group, and

wherein the polymer compound further comprises a constitutional unit derived from a hole transport polymerizable compound represented by formula (8-3):

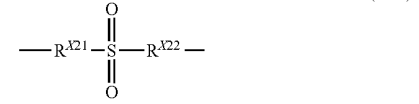
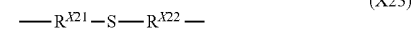


2. The organic light-emitting element as claimed in claim 1, wherein the phosphorescent polymerizable compound is a complex represented by any one of the following formulas (2) to (4):

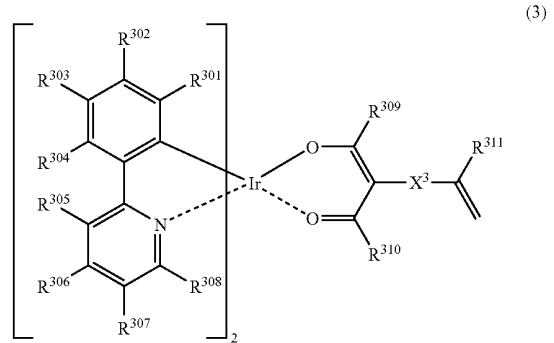


wherein R²⁰¹ to R²¹⁵ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, in each of R²⁰¹ to R²⁰⁴, R²⁰⁵ to R²⁰⁸, R²⁰⁹ to R²¹¹, and R²¹² to R²¹⁵, two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring, R²¹⁶ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X² is a single bond or a group represented by any one of the following formulas (X21) to (X24):

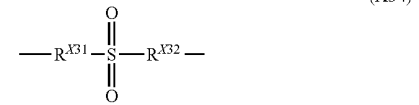
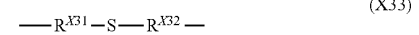
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wherein R^{X21} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X22} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;

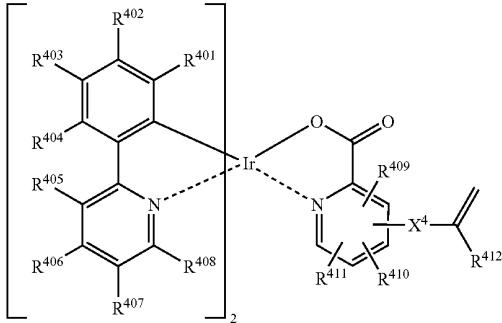


wherein R³⁰¹ to R³⁰⁸ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R³⁰⁹ to R³¹⁰ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, in each of R³⁰¹ to R³⁰⁴, and R³⁰⁵ to R³⁰⁸, two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring, R³¹¹ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X³ is a single bond or a group represented by any one of the following formulas (X31) to (X34):

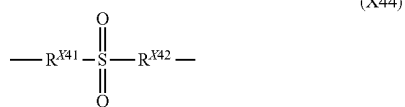
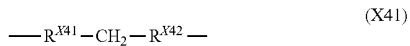


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wherein R^{X31} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X32} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;



wherein R^{401} to R^{411} are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, in each of R^{401} to R^{404} , R^{405} to R^{408} , and R^{409} to R^{411} , two groups bonded to carbon atoms adjacent to each other on a ring may be bonded to each other to form a condensed ring, R^{412} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X^4 is a single bond or a group represented by any one of the following formulas (X41) to (X44):



wherein R^{X41} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X42} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group.

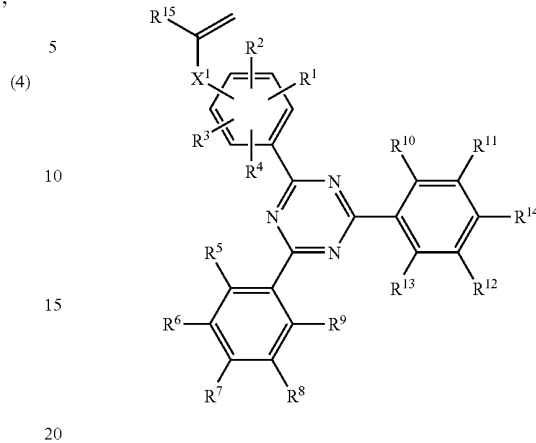
3. A surface-emitting light source comprising the organic light-emitting element as claimed in claim 1.

4. An image display comprising the organic light-emitting element as claimed in claim 1.

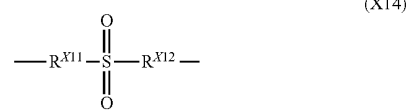
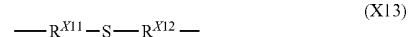
5. An organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, wherein the light-emitting layer contains a polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;

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(1)

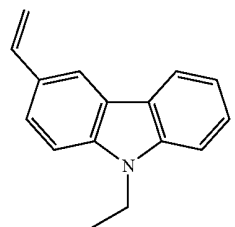


wherein R^7 and R^{14} are each independently an alkyl group having 1 to 12 carbon atoms or an alkoxy group having 1 to 12 carbon atoms, and R^1 to R^6 and R^8 to R^{13} are hydrogen atoms, R^{15} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X^1 is a single bond or a group represented by any one of the following formulas (X11) to (X14):



wherein R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group, and

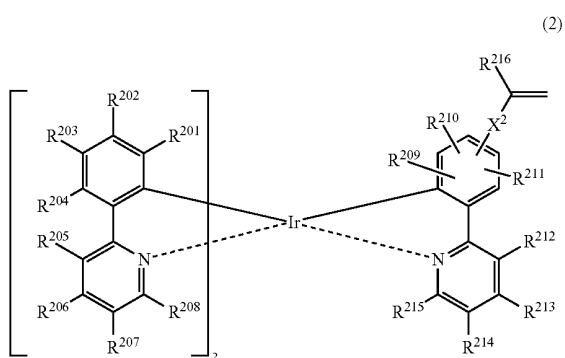
wherein the light-emitting layer further contains a hole transport polymer compound having a structural unit derived from a polymerizable compound represented by formula (8-3):



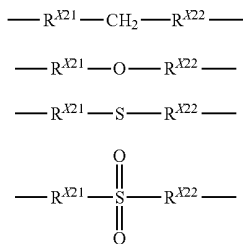
(8-3)

6. The organic light-emitting element as claimed in claim 5, wherein the phosphorescent polymerizable compound is a complex represented by any one of the following formulas (2) to (4):

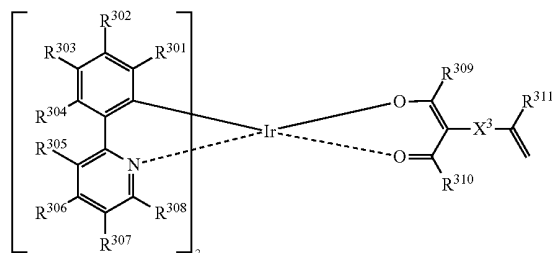
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wherein R²⁰² and R²⁰⁴ are fluorine atoms, R²⁰⁶ and R²¹³ are t-Bu groups, R²⁰¹, R²⁰³, R²⁰⁵, R²⁰⁷ to R²¹², R²¹⁴ and R²¹⁵ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R²¹⁶ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X² is a single bond or a group represented by any one of the following formulas (X21) to (X24):



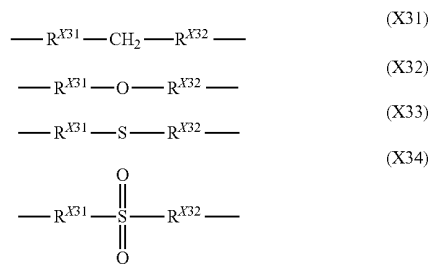
wherein R^{X21} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X22} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;



wherein R³⁰² and R³⁰⁴ are fluorine atoms, R³⁰⁶ is t-Bu group, R³⁰¹, R³⁰³, R³⁰⁵, R³⁰⁷ and R³⁰⁸ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10

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carbon atoms and a silyl group, R³⁰⁹ to R³¹⁰ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R³¹¹ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X³ is a single bond or a group represented by any one of the following formulas (X31) to (X34):



wherein R^{X31} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X32} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;

(X21)

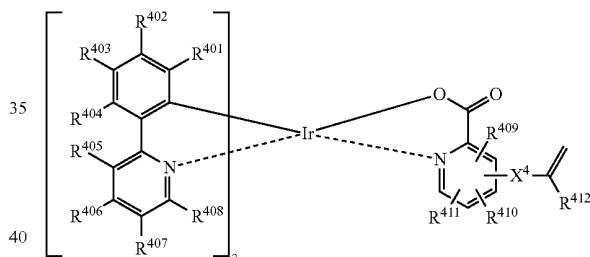
(X22)

(X23)

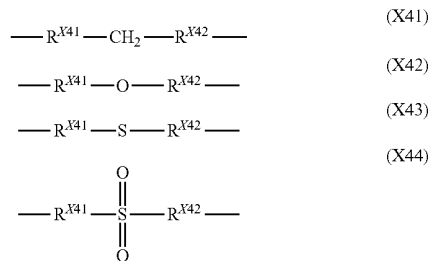
(X24)

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(4)



wherein R⁴⁰² and R⁴⁰⁴ are fluorine atoms, R⁴⁰⁶ is t-Bu group, R⁴⁰¹, R⁴⁰³, R⁴⁰⁵, and R⁴⁰⁷ to R⁴¹¹ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R⁴¹² is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X⁴ is a single bond or a group represented by any one of the following formulas (X41) to (X44):



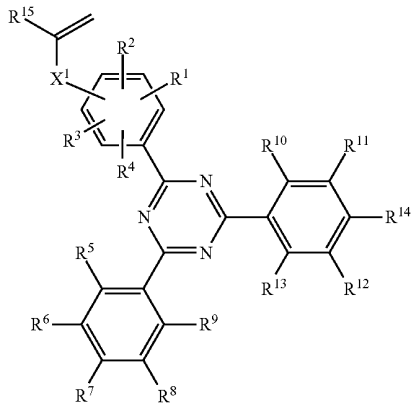
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wherein R^{X41} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X42} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group.

7. A polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;



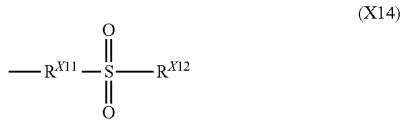
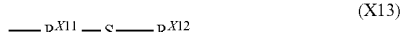
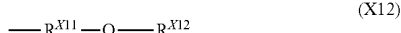
(1) 10

15

20

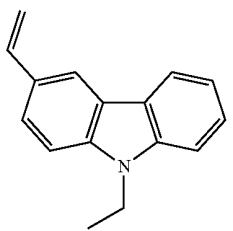
25

wherein R^7 and R^{14} are each independently an alkyl group having 1 to 12 carbon atoms or an alkoxy group having 1 to 12 carbon atoms, and R^1 and R^6 and R^8 and R^{13} are hydrogen atoms, R^{15} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X^1 is a single bond or a group represented by any one of the following formulas (X11) to (X14):



wherein R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group,

said polymer compound further comprising a constitutional unit derived from a hole transport polymerizable compound represented by formula (8-3):



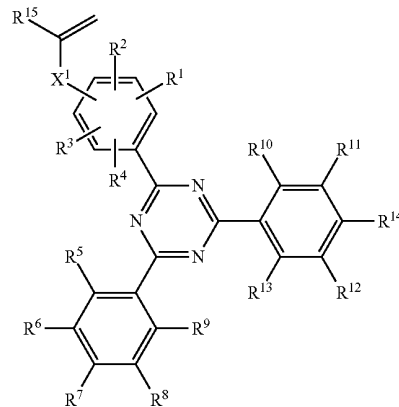
(8-3)

60

65

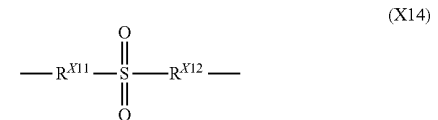
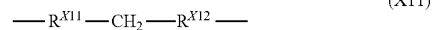
38

8. An organic light-emitting element constituted by arranging a light-emitting layer between an anode and a cathode, wherein the light-emitting layer contains a polymer compound comprising a constitutional unit derived from an electron transport polymerizable compound represented by the following formula (1) and a constitutional unit derived from a phosphorescent polymerizable compound;



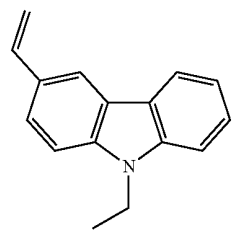
(1)

wherein formula (1), R^7 and R^{14} are each independently an alkyl group of 1 to 12 carbon atoms or an alkoxy group of 1 to 12 carbon atoms, and R^1 to R^6 and R^8 to R^{13} are hydrogen atoms, R^{15} is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X^1 is a single bond or a group represented by any one of the following formulas (X11) to (X14):



wherein R^{X11} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X12} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group,

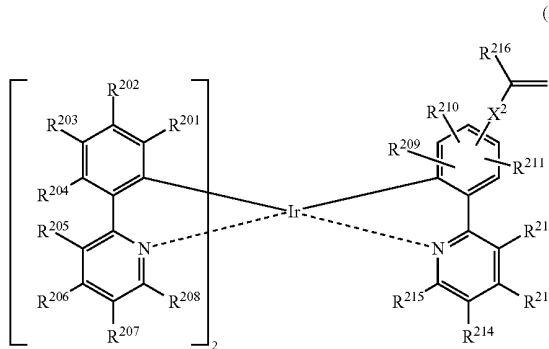
wherein the polymer compound further comprises a constitutional unit derived from a hole transport polymerizable compound represented by formula (8-3):



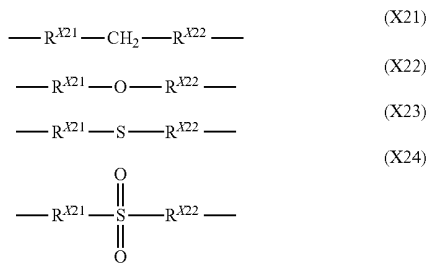
(8-3)

39

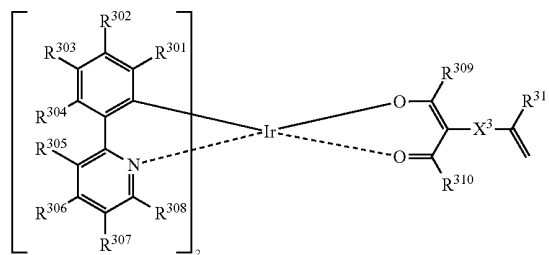
and wherein the phosphorescent polymerizable compound is a complex represented by any one of the following formulas (2) to (4):



wherein R²⁰² and R²⁰⁴ are fluorine atoms, R²⁰⁶ and R²¹³ are t-Bu groups, R²⁰¹, R²⁰³, R²⁰⁵, R²⁰⁷ to R²¹², R²¹⁴ and R²¹⁵ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R²¹⁶ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X² is a single bond or a group represented by any one of the following formulas (X21) to (X24):



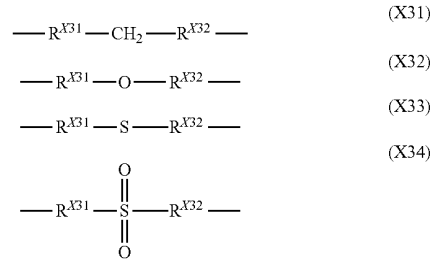
wherein R^{X21} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X22} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;



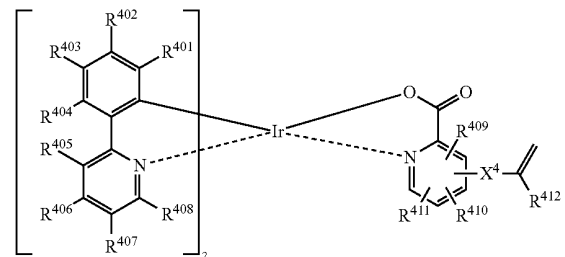
wherein R³⁰² and R³⁰⁴ are fluorine atoms, R³⁰⁶ is t-Bu group, R³⁰¹, R³⁰³, R³⁰⁵, R³⁰⁷ and R³⁰⁸ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10

40

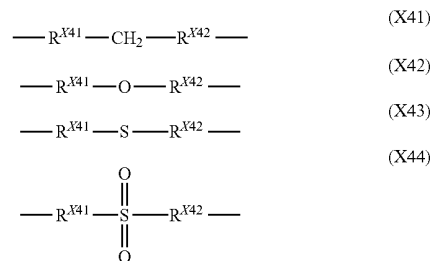
carbon atoms and a silyl group, R³⁰⁹ to R³¹⁰ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R³¹¹ is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X³ is a single bond or a group represented by any one of the following formulas (X31) to (X34):



wherein R^{X31} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X32} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group;



wherein R⁴⁰² and R⁴⁰⁴ are fluorine atoms, R⁴⁰⁶ is t-Bu group, R⁴⁰¹, R⁴⁰³, R⁴⁰⁵, and R⁴⁰⁷ to R⁴¹¹ are each independently an atom or a substituent selected from the group consisting of a hydrogen atom, a halogen atom, a cyano group, an alkyl group of 1 to 10 carbon atoms, an aryl group of 6 to 10 carbon atoms, an amino group which may be substituted by an alkyl group of 1 to 10 carbon atoms, an alkoxy group of 1 to 10 carbon atoms and a silyl group, R⁴¹² is a hydrogen atom or an alkyl group of 1 to 12 carbon atoms, and X⁴ is a single bond or a group represented by any one of the following formulas (X41) to (X44):



wherein R^{X41} is a single bond or an alkylene group of 1 to 12 carbon atoms, and R^{X42} is a single bond, an alkylene group of 1 to 12 carbon atoms or a phenylene group.

* * * * *

专利名称(译)	使用含三嗪环的高分子化合物的有机发光元件		
公开(公告)号	US8404362	公开(公告)日	2013-03-26
申请号	US12/668819	申请日	2008-07-10
[标]申请(专利权)人(译)	昭和电工株式会社		
申请(专利权)人(译)	昭和电工株式会社制		
当前申请(专利权)人(译)	昭和电工株式会社制		
[标]发明人	TOBA MASAHIKO		
发明人	TOBA, MASAHIKO		
IPC分类号	H01L51/54 C07D251/12		
CPC分类号	C08F226/06 C09K11/06 H01L51/0043 C09K2211/1007 C09K2211/1059 H01L51/5012 H01L51/0059 H01L51/0067 H01L51/0085		
代理机构(译)	萨格鲁MION, PLLC		
优先权	2007184402 2007-07-13 JP		
其他公开文献	US20100201258A1		
外部链接	Espacenet USPTO		

摘要(译)

本发明的有机发光元件是通过在阳极和阴极之间设置发光层而构成的有机发光元件，其特征在于，发光层含有包含结构单元的高分子化合物。衍生自由下式(1)表示的电子传输可聚合化合物和衍生自磷光可聚合化合物的结构单元；其中R1-R14各自独立地为氢原子，卤原子，氰基，氨基，1-12个碳原子的烷基等，R15为氢原子或1-12碳的烷基原子，X1是单键等。

