



US 20090001360A1

(19) **United States**

(12) **Patent Application Publication**  
**Nakayama**

(10) **Pub. No.: US 2009/0001360 A1**  
(43) **Pub. Date: Jan. 1, 2009**

(54) **ORGANIC EL DISPLAY AND METHOD FOR PRODUCING THE SAME**

**Publication Classification**

(76) Inventor: **Masaya Nakayama**, Kanagawa (JP)

- (51) **Int. Cl.**  
*H01L 51/05* (2006.01)  
*H01L 27/12* (2006.01)  
*H01L 21/84* (2006.01)
- (52) **U.S. Cl. ....** 257/40; 257/43; 438/155; 257/E27.112; 257/E51.006; 257/E21.7

Correspondence Address:  
**BIRCH STEWART KOLASCH & BIRCH**  
**PO BOX 747**  
**FALLS CHURCH, VA 22040-0747 (US)**

(57) **ABSTRACT**

The present invention provides an organic electroluminescence display having an organic EL element and a thin film field-effect transistor formed on the organic EL element, wherein an electrically conductive etching protective layer which is electrically connected to an upper electrode is disposed between the upper electrode and the thin film field-effect transistor, a protective insulating layer is disposed between the electrically conductive etching protective layer and the thin film field-effect transistor, and a source electrode or a drain electrode of the thin film field-effect transistor and the electrically conductive etching protective layer are electrically connected through a contact hole formed in the protective insulating layer; and a method for producing thereof.

(21) Appl. No.: **12/146,355**

(22) Filed: **Jun. 25, 2008**

(30) **Foreign Application Priority Data**

Jun. 29, 2007 (JP) ..... 2007-172448  
May 30, 2008 (JP) ..... 2008-142748

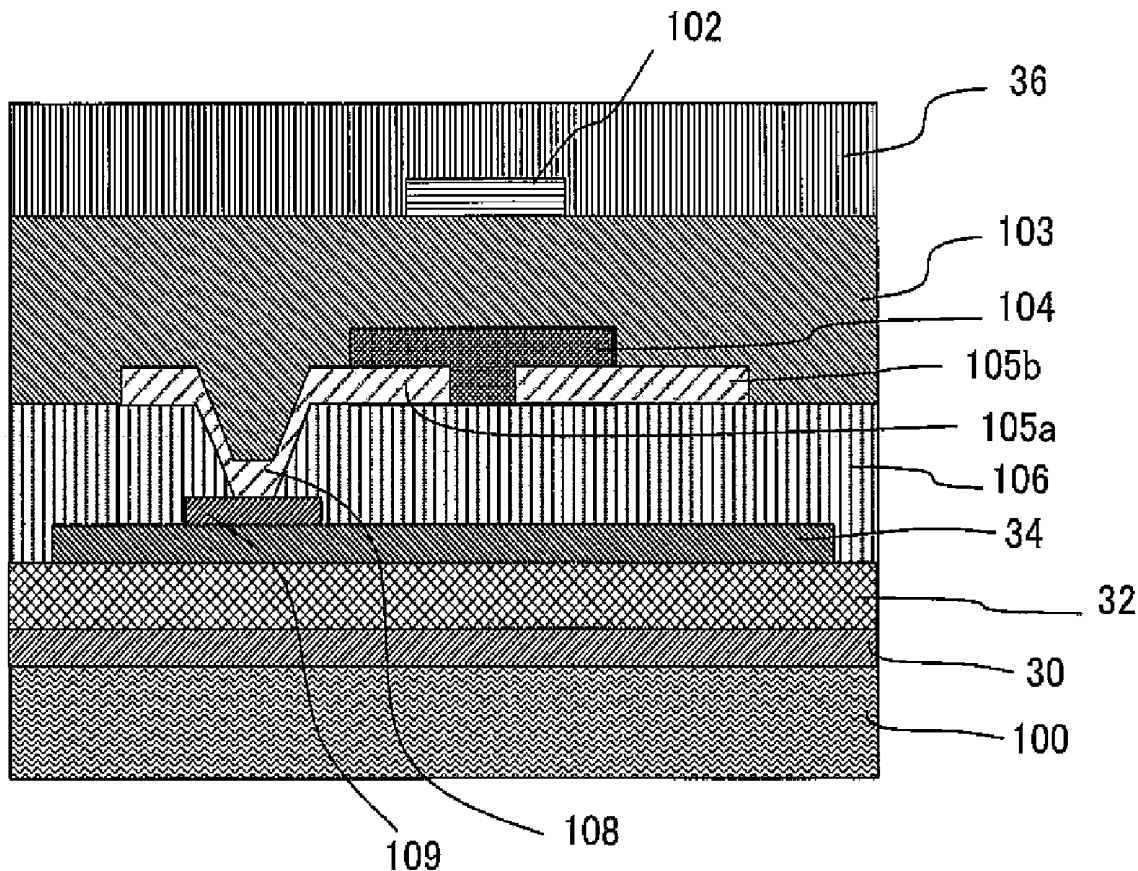


Fig. 1

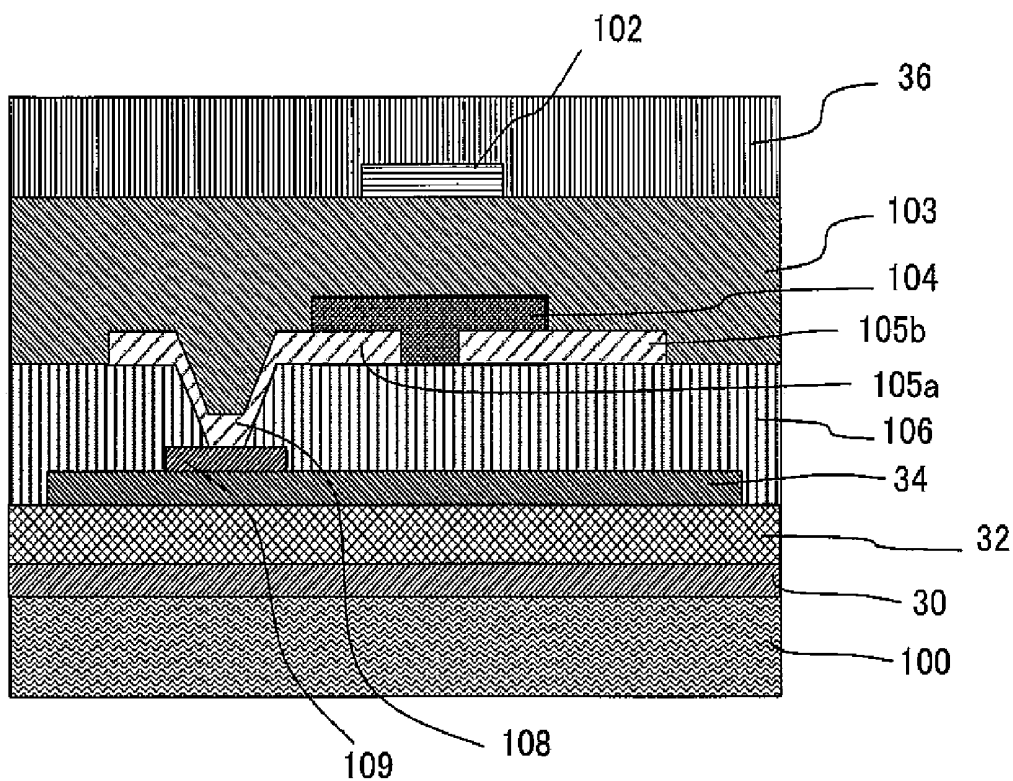


Fig. 2

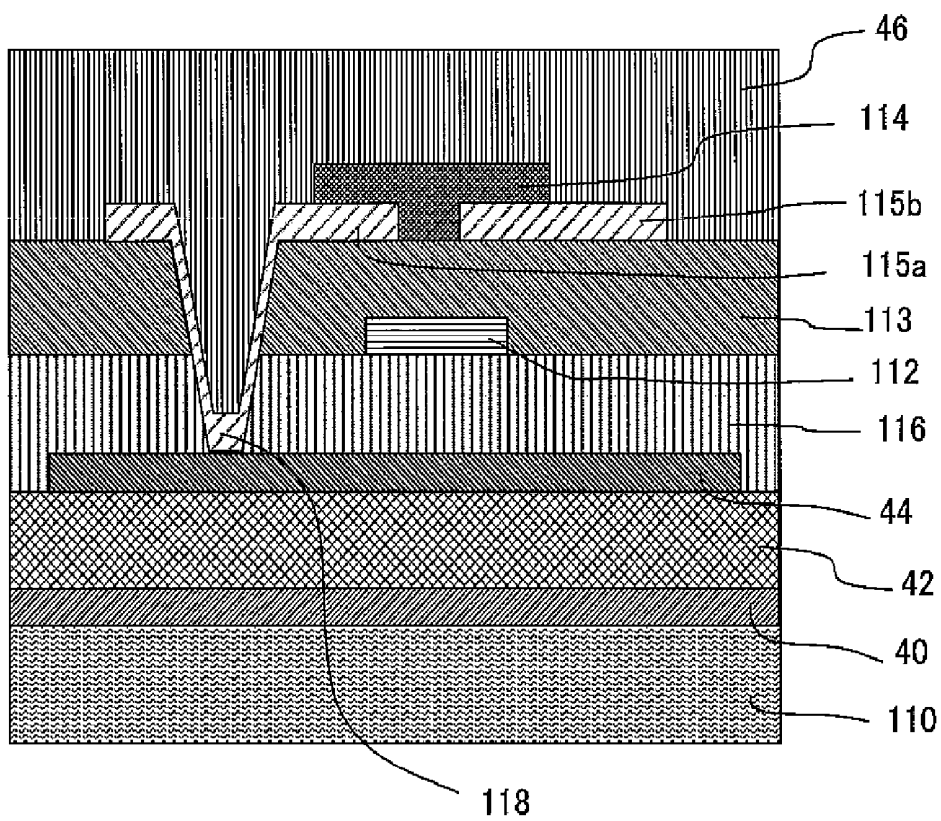


Fig. 3

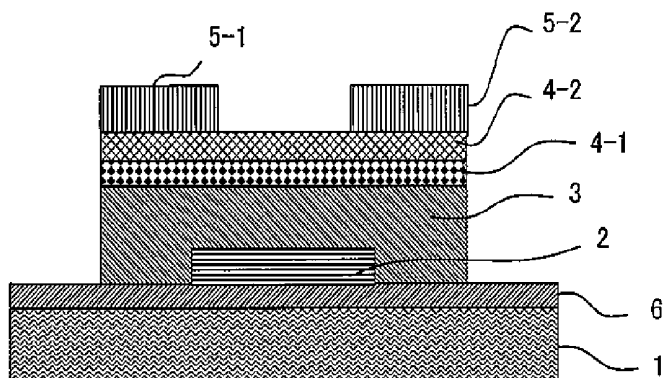


Fig. 4

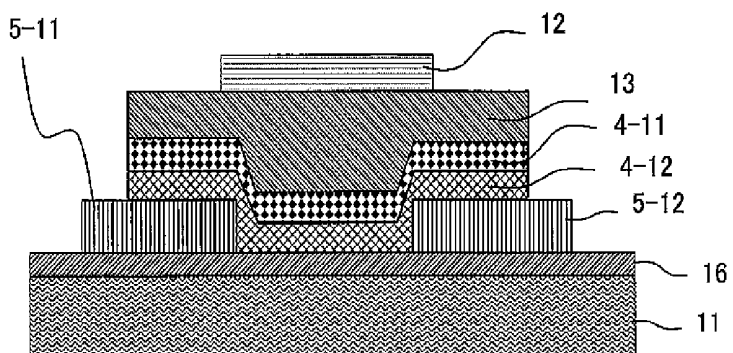
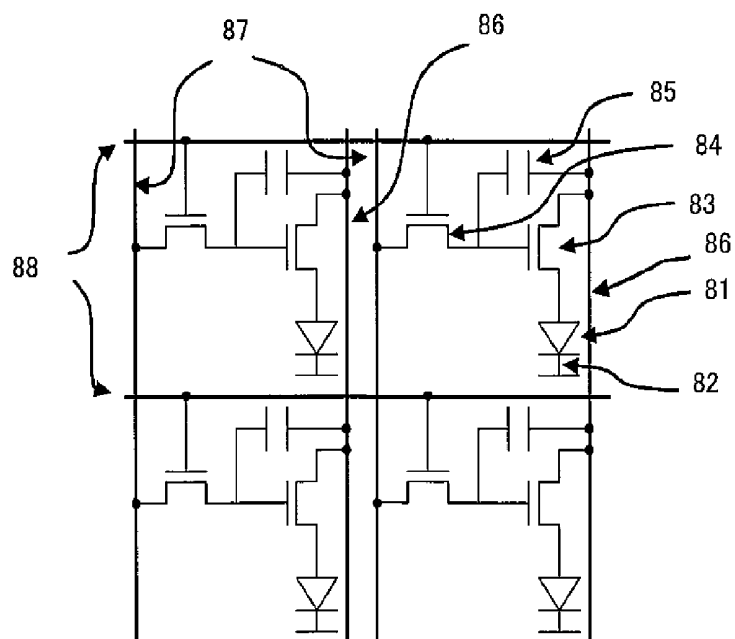


Fig. 5



## ORGANIC EL DISPLAY AND METHOD FOR PRODUCING THE SAME

### CROSS-REFERENCE TO RELATED APPLICATION

[0001] This application claims priority under 35 USC 119 from Japanese Patent Application Nos. 2007-172448 and 2008-142748, the disclosures of which are incorporated by reference herein.

### BACKGROUND OF THE INVENTION

[0002] 1. Field of the Invention

[0003] The present invention concerns an organic EL display in which a thin film field effect transistor for driving an organic EL element is disposed on the organic EL element, as well as a manufacturing method thereof. Particularly, it relates to an active-type organic EL display capable of obtaining a high aperture ratio, and having high definition, high brightness, high stability, high reliability and high durability, as well as a manufacturing method thereof.

[0004] 2. Description of the Related Art

[0005] In recent years, flat panel displays (FPDs) have been put to practical use, due to the progress made in liquid crystal and electroluminescence (EL) technologies, etc. In particular, an organic electroluminescence element (hereinafter referred to as an "organic EL element" in some cases) formed using a thin film material which emits light by excitation due to application of electric current can provide light emission of high brightness at a low voltage, and thus is expected to achieve reduction in device thickness, weight, and size, and power saving, etc. in wide ranging applications including mobile phone displays, personal digital assistants (PDA), computer displays, car information displays, TV monitors, and general illumination.

[0006] These FPDs are driven by an active matrix circuit including field effect-type thin film transistors each using, as an active layer, an amorphous silicon thin film or a polycrystalline silicon thin film provided on a glass substrate. (In the description below, a field effect-type thin film transistor is sometimes referred to as a "thin film transistor" or "TFT".)

[0007] On the other hand, for attaining even higher definition, higher brightness and higher durability in the active-type organic EL display, it has been known that a top-emission type is advantageous, due to being able to obtain a high aperture ratio. However, in the organic EL element having the top-emission structure, since it is difficult to form a transparent electrically conductive layer such as ITO directly on an organic layer without any damage, it is difficult to provide a practically useful element having high efficiency and high durability at present.

[0008] As another approach, Japanese Patent Application Laid-Open (JP-A) No. 2005-242028, for example, discloses forming TFT superposed above an organic EL element having a bottom-emission structure, wherein the TFT is constituted with an organic semiconductor. As film formation with an organic TFT using an organic semiconductor can be conducted at low temperature, the organic TFT can be formed on an organic EL element with no damage to the organic EL element. However, the organic TFT has a problem in view of drive stability and also has a problem in view of reliability such that strict sealing is necessary against the external atmosphere and moisture in order to enhance the storage stability. Further, since the organic TFT has low carrier mobility, a size

(channel width) of the TFT increases extremely in order to increase driving current. Therefore, it is difficult to provide an organic EL display having high definition and high brightness.

[0009] On the other hand, transistors using thin silicon films are favorable in view of stability and operational reliability, but as their manufacturing process requires a thermal treatment step at a relatively high temperature, it involves a problem in that damage is caused to an organic EL element in a case of forming the transistor above the organic EL element.

[0010] TFTs using, as a semiconductor thin film, a film of an amorphous oxide, such as an In—Ga—Zn—O-based amorphous oxide, which can be formed at a low temperature, have been disclosed in JP-A No. 2006-165529 and IDW/AD'05, pages 845-846 (Dec. 6, 2005). As the films for a TFT made with an amorphous oxide semiconductor can be formed at room temperature, the TFT can be prepared on a film (flexible substrate). Therefore, amorphous oxide semiconductors have been attracting attention as a material for active layers of film (flexible) TFTs lately. For example, it has been reported that a TFT formed using a-IGZO has a field effect mobility of about  $10 \text{ cm}^2/\text{Vs}$  even on a PEN substrate, which is higher than that of an a-Si TFT on glass, in *NATURE*, vol. 432, pages 488-492 (Nov. 25, 2004).

[0011] However, in the case of using, as for example, a drive circuit of a display, a TFT formed using a-IGZO, there are the problems that mobility ranges from  $1 \text{ cm}^2/\text{Vs}$  to  $10 \text{ cm}^2/\text{Vs}$ , which provide insufficient performance, the OFF current is high, and the ON-OFF ratio is low. Particularly, in order to apply such a TFT to a display incorporating organic EL devices, further increase in mobility and improvement in ON-OFF ratio are required.

[0012] Furthermore, in a case where a TFT is formed on an organic EL element having a bottom emission structure, to electrically connect the organic EL element and the TFT, a protective insulating layer is provided between the organic EL element and the TFT to electrically shield these from each other, a contact hole is formed by perforating the protective insulating layer at a portion requiring electric connection, and thereby electrical conduction between the organic EL element and the TFT is achieved through the contact hole. To perforate a contact hole in a protective insulating layer, an etching method, a transfer method utilizing a printing technique, an ink-jet method, a method of forming a shadow pattern by a mask and depositing an insulating layer, and the like are known.

[0013] For example, an organic EL display equipped with a TFT using an organic semiconductor on an upper electrode of an organic EL element via an organic protective layer such as parylene is disclosed. JP-A No. 2006-12785 proposes that an organic protective layer is formed by masking a contact hole portion and using a chemical vapor deposition method. However, it is difficult to form a high definition pattern according to the chemical vapor deposition method by masking. A TFT using an organic semiconductor involves a small driving current, and therefore it is difficult to form a high definition pixel in the first place. Consequently, the chemical vapor deposition method by masking is merely a formation means of a contact hole used in the combination with such a TFT, and is therefore not suitable as a contact hole formation means for a high definition organic EL display.

[0014] An etching method is preferred as a means for providing a contact hole with high definition. In particular, in producing a high definition and high brightness organic EL

display in which a TFT is formed on an organic EL element having a bottom emission structure, contact hole formation by an etching method is a preferred means, but an inherent problem in forming a TFT on the organic EL element has been clarified. The etching method includes wet etching, dry etching, laser abrasion or the like, but regardless of the method that is used, there is concern that the method may damage an upper electrode of an organic EL element. An upper electrode of an organic EL element is formed using vacuum deposition by thermal resistance heating which generally causes less damage to an organic layer. Although vacuum deposition by thermal resistance heating involves less damage during deposition, a resulting layer has crude density and contains many pinholes. As a result, an etching material affects the upper electrode to cause damage thereto through the pinholes in an etching step for producing a contact hole in a protective insulating layer. Furthermore, if the damage proceeds, it also causes damage to an organic layer, resulting in deterioration of properties of an organic EL element.

[0015] Therefore, an etching method that does not cause damage to an organic EL element including an upper electrode has been desired.

#### SUMMARY OF THE INVENTION

[0016] The present invention has been made in view of the above circumstances and provides an organic EL display and a method for producing the organic EL display with the following aspects.

[0017] A first aspect of the invention provides an organic EL display comprising:

[0018] an organic EL element comprising at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode, in this order, on a substrate; and

[0019] a thin film field-effect transistor which drives the organic EL element and is formed on the organic EL element,

[0020] wherein an electrically conductive etching protective layer which is electrically connected to the upper electrode is disposed between the upper electrode and the thin film field-effect transistor, a protective insulating layer is disposed between the electrically conductive etching protective layer and the thin film field-effect transistor, and a source electrode or a drain electrode of the thin film field-effect transistor and the electrically conductive etching protective layer are electrically connected through a contact hole formed in the protective insulating layer.

[0021] A second aspect of the invention provides a method for producing an organic EL display comprising at least:

[0022] forming an organic EL element part by sequentially forming at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode on a substrate;

[0023] forming an electrically conductive etching protective layer that is electrically connected to the upper electrode;

[0024] forming a protective insulating layer on the electrically conductive etching protective layer;

[0025] forming a contact hole in the protective insulating layer by causing an etching material to act; and

[0026] forming a thin film field-effect transistor on the protective insulating layer,

[0027] wherein a source electrode or a drain electrode of the thin film field-effect transistor and the electrically conductive etching protective layer are electrically connected through the contact hole.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0028] FIG. 1 is a schematic diagram showing the structure of an organic EL display according to the invention.

[0029] FIG. 2 is a schematic diagram showing the structure of an organic EL display for comparison.

[0030] FIG. 3 is a schematic diagram showing the structure of a TFT used in the display according to the invention.

[0031] FIG. 4 is a schematic diagram showing the structure of a TFT used in the display according to another embodiment of the invention.

[0032] FIG. 5 is a schematic diagram of a pixel-circuit of an organic EL display according to the invention.

#### DETAILED DESCRIPTION OF THE INVENTION

[0033] An object of the invention is to provide an organic EL display comprising an organic EL element having provided thereon a TFT for driving the organic EL element, and a method for producing the same. In particular, an object of the invention is to provide an active-type organic EL display, wherein a high aperture ratio is obtained, and which has high definition, high brightness and high durability, and a method for producing the same. More particularly, an object of the invention is to provide an active-type organic EL display of high definition, high brightness and high durability by reducing damage to an organic EL element during forming of a contact hole in a protective insulating layer on the organic EL element, and a method for producing the same.

[0034] The organic EL display of the invention comprises:

[0035] an organic EL element comprising at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode, in this order, on a substrate; and

[0036] a TFT which drives the organic EL element and is formed on the organic EL element,

[0037] wherein an electrically conductive etching protective layer which is electrically connected to the upper electrode is disposed between the upper electrode and the TFT, a protective insulating layer is disposed between the electrically conductive etching protective layer and the TFT, and a source electrode or a drain electrode of the TFT and the electrically conductive etching protective layer are electrically connected through a contact hole formed in the protective insulating layer.

[0038] Preferably, the contact hole is formed by etching with an etching material, in which an etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the upper electrode by the etching material.

[0039] Preferably, the contact hole is formed by etching with an etching material, in which an etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the protective insulating layer by the etching material.

[0040] Preferably, the upper electrode is formed by an electric resistance heating deposition method, and the electrically conductive etching protective layer is formed by a sputtering method, an ion plating method or a CVD (chemical vapor deposition) method.

[0041] Preferably, an active layer of the TFT contains a semiconductor material selected from the group consisting of an oxide semiconductor, an organic semiconductor and a carbon nanotube. More preferably, the active layer contains an oxide semiconductor. Preferably, the oxide semiconductor is an amorphous oxide semiconductor.

[0042] Preferably, the lower electrode is a light transmitting electrode, and the upper electrode is a light reflective electrode. Preferably, the lower electrode is an anode, and the upper electrode is a cathode.

[0043] Preferably, the TFT has n-type polarity.

[0044] Preferably, an electric resistance layer is provided so as to be electrically connected between the active layer and at least one of the source electrode or the drain electrode.

[0045] Preferably, the electric resistance layer and the active layer are in a layered state, the active layer is in contact with a gate insulating layer of the TFT, and the electric resistance layer is contacted with at least one of the source electrode or the drain electrode. Preferably, the electric resistance layer is thicker than the active layer.

[0046] Preferably, an electric conductivity changes continuously between the electric resistance layer and the active layer.

[0047] Preferably, the active layer and the electric resistance layer comprise an oxide semiconductor. Preferably, the oxide semiconductor comprises at least one selected from the group consisting of In, Ga and Zn, or a composite oxide thereof. More preferably, the oxide semiconductor comprises In and Zn, and a composition ratio between Zn and In in the electric resistance layer (expressed by a ratio of Zn to In, Zn/In) is larger than a composition ratio Zn/In in the active layer.

[0048] Preferably, the active layer has an oxygen concentration that is lower than that of the electric resistance layer.

[0049] Preferably, the active layer has an electric conductivity of  $10^{-4}$  Scm<sup>-1</sup> or more, and less than  $10^2$  Scm<sup>-1</sup>, and more preferably of  $10^{-1}$  Scm<sup>-1</sup> or more, and less than  $10^2$  Scm<sup>-1</sup>.

[0050] Preferably, a ratio of an electric conductivity of the active layer to an electric conductivity of the electric resistance layer (electric conductivity of active layer/electric conductivity of electric resistance layer) is from 101 to  $10^{10}$ , and more preferably from  $10^2$  to  $10^8$ .

[0051] Preferably, the substrate is a flexible resin substrate.

[0052] The method for producing an organic EL display of the present invention includes at least:

[0053] forming an organic EL element part by sequentially forming at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode on a substrate;

[0054] forming an electrically conductive etching protective layer that is electrically connected to the upper electrode;

[0055] forming a protective insulating layer on the electrically conductive etching protective layer;

[0056] forming a contact hole in the protective insulating layer by causing an etching material to act; and

[0057] forming a thin film field-effect transistor on the protective insulating layer,

[0058] wherein a source electrode or a drain electrode of the thin film field-effect transistor and the electrically conductive etching protective layer are electrically connected through the contact hole.

[0059] Preferably, an etching rate of the electrically conductive etching protective layer by the etching material is

slower than an etching rate of the upper electrode by the etching material. More preferably, an etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the protective insulating layer by the etching material.

[0060] Preferably, the upper electrode is formed by an electric resistance heating deposition method, and the electrically conductive etching protective layer is formed by a sputtering method, an ion plating method or a chemical vapor deposition method (CVD method).

[0061] A TFT using an amorphous oxide semiconductor can undergo film formation at room temperature, and therefore can be provided on an organic EL element without causing damage to the organic EL element. In particular, when an In—Ga—Zn—O-based oxide is used as an active layer, a TFT having performance in which field-effect mobility is  $10$  cm<sup>2</sup>/Vs, and an ON/OFF ratio exceeds  $10^3$  can be prepared. Furthermore, when a TFT has a constitution which has an active layer and an electric resistance layer each containing an amorphous oxide semiconductor, wherein the active layer is in contact with a gate insulating layer, and the electric resistance layer is electrically connected between the active layer and at least one of the source electrode or the drain electrode, a TFT which achieves both excellent OFF characteristics and high mobility can be formed. In particular, it has been found that a constitution in which at least the electric resistance layer and the active layer are provided in a layered form on the substrate, the active layer is in contact with the gate insulating layer, and the electric resistance layer is in contact with at least one of the source electrode or the drain electrode is an effective means. In the following description of the invention, the term "semiconductor layer" is sometimes used to refer to a layer including an active layer and an electric resistance layer.

[0062] According to the invention, there can be provided an active-type organic EL display of high definition, high brightness and high durability, comprising an organic EL element and a TFT which is formed on the organic EL element, for driving the organic EL element, by which a high aperture ratio is obtained, and a method for producing the same. In particular, according to the invention, a contact hole which electrically connects an organic EL element and a TFT can be prepared without causing damage to the organic EL element, and therefore, an active-type organic EL display having high brightness and high durability, and a method for producing the same can be provided.

## 1. Organic EL Display

[0063] The organic EL display of the invention is an organic EL display having an organic EL element comprising at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode, in this order, on a substrate, and a TFT which drives the organic EL element, formed on the organic EL element, wherein an electrically conductive etching protective layer which is electrically connected to the upper electrode is disposed between the upper electrode and the TFT, a protective insulating layer is disposed between the electrically conductive etching protective layer and the TFT, and a source electrode or a drain electrode of the TFT and the electrically conductive etching protective layer are electrically connected via contact hole formed in the protective insulating layer. The TFT is provided on the back side with respect to a light emitting surface of the organic EL element. Therefore, an opening of the organic EL element

from which emitted light is extracted can be formed widely. Preferably, the lower electrode is a light-transmitting electrode, and the upper electrode is a light reflective electrode.

**[0064]** 1) Electrically Conductive Etching Protective Layer

**[0065]** In the organic EL display of the invention, the electrically conductive etching protective layer is formed on the upper electrode of the organic EL element. The upper electrode of the organic EL element and a source electrode or a drain electrode of the driving TFT are electrically connected via the electrically conductive etching protective layer.

**[0066]** The electrically conductive etching protective layer has a function to prevent and reduce damage to the upper electrode or the organic layer of the organic EL element during forming a contact hole in the protective insulating layer formed on the organic EL element, by etching. Therefore, it is preferred that an etching rate of the electrically conductive etching protective layer by an etching material for forming a contact hole in the protective insulating layer is slower than an etching rate of the upper electrode of the organic EL element by the etching material. More preferably, the etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the protective insulating layer by the etching material. Even more preferably, the etching rate of the electrically conductive etching protective layer is  $\frac{1}{10}$  or less with respect to the etching rate of the protective insulating layer. Furthermore, the upper electrode of the organic EL element and the source electrode or the drain electrode of the driving TFT are electrically connected via the electrically conductive etching protective layer. Therefore, in order to connect electrically the organic EL element and the driving TFT, the electrically conductive etching protective layer is required to have an electric conductivity.

**[0067]** Preferred materials for the electrically conductive etching protective layer include metals such as Al, Ag, Mo, Cr, Ti, Au, Pt, Ir, In and Sn, or alloys thereof; electrically conductive oxides such as  $\text{SnO}_2$ ,  $\text{In}_2\text{O}_3$ , ITO and IZO; and electrically conductive semiconductors having impurities added thereto in a large amount, such as P-doped Si and B-doped Si. In particular,  $\text{SnO}_2$  has good etching resistance, and is therefore suitable for an electrically conductive etching protective layer.

**[0068]** The electrically conductive etching protective layer has preferably a thickness of from 10 nm to 10  $\mu\text{m}$ . When the thickness is less than 10 nm, the function as the etching protective layer deteriorates. When the thickness is more than 10  $\mu\text{m}$ , process temperature elevates during a film formation process of the electrically conductive etching protective layer, and there is a possibility of giving damage to the organic EL element.

**[0069]** As a method for forming the electrically conductive etching protective layer, it is suitable to adopt the following methods by which a tight and thin layer can be obtained as compared with vacuum deposition by thermal resistance heating, an electron beam deposition method, sputtering method, reactive sputtering method, MBE (molecular beam epitaxy) method, cluster ion beam method, ion plating method, plasma polymerization method (high frequency exciton ion plating method), plasma CVD method, laser CVD method, thermal CVD method, gas source CVD method, and the like. In particular, a sputtering method, ion plating method or CVD method is preferred from the standpoint of manufacturability such as possibility of large area.

**[0070]** 2) Protective Insulating Layer

**[0071]** In the organic EL display of the invention, the whole organic EL element is protected with the protective insulating layer. The protective insulating layer has a function to reduce damage to the organic EL element during production of TFT on the organic EL element, and a function to electrically insulate the organic EL element and the TFT. It is further preferred for the protective insulating layer to have a function to prevent penetration of substances which accelerate deterioration of the element, such as moisture, oxygen and the like, into the element.

**[0072]** Specific examples of materials for the protective insulating layer include metal oxides such as  $\text{MgO}$ ,  $\text{SiO}$ ,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{GeO}$ ,  $\text{NiO}$ ,  $\text{CaO}$ ,  $\text{BaO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{TiO}_2$  and the like; metal nitrides such as  $\text{SiN}_x$ ,  $\text{SiN}_x\text{O}_y$ , and the like; metal fluorides such as  $\text{MgF}_2$ ,  $\text{LiF}$ ,  $\text{AlF}_3$ ,  $\text{CaF}_2$  and the like; polyethylene; polypropylene; polymethyl methacrylate; polyimide; polyurea; polytetrafluoroethylene; polychlorotrifluoroethylene; polydichlorodifluoroethylene; a copolymer of chlorotrifluoroethylene and dichlorodifluoroethylene; copolymers obtained by copolymerizing a monomer mixture containing tetrafluoroethylene and at least one comonomer; fluorine-containing copolymers each having a cyclic structure in the copolymerization main chain; water-absorbing materials each having a coefficient of water absorption of 1% or more; moisture permeation preventive substances each having a coefficient of water absorption of 0.1% or less; and the like.

**[0073]** There is no particular limitation as to a method for forming the protective insulating layer. For instance, a vacuum deposition method, a sputtering method, a reactive sputtering method, an MBE (molecular beam epitaxial) method, a cluster ion beam method, an ion plating method, a plasma polymerization method (high-frequency excitation ion plating method), a plasma CVD method, a laser CVD method, a thermal CVD method, a gas source CVD method, a coating method, a printing method, or a transfer method may be applied.

**[0074]** The protective insulating layer has preferably a thickness of from 10 nm to 10  $\mu\text{m}$ . When the thickness exceeds 10  $\mu\text{m}$ , it is difficult to form a contact hole for connecting the organic EL element and the TFT, and when the thickness is less than 10 nm, the function as a protective film is lost, which are not preferred.

**[0075]** The upper electrode of the organic EL element and the source electrode or the drain electrode of the driving TFT are required to be electrically connected, and it is therefore necessary to form a contact hole in the protective insulating layer. A method of forming a contact hole includes a wet etching method using an etchant, a dry etching method using plasma and an etching method using laser.

**[0076]** 3) Structure

**[0077]** The organic EL display of the invention is described in detail below by reference to the accompanying drawings.

**[0078]** FIG. 1 is a conceptual sectional view showing a constitution of the organic EL display of the invention.

**[0079]** On a substrate 100, an organic EL element part comprising a lower electrode 30, an organic layer 32 comprising at least a light-emitting layer, and an upper electrode 34 in this order; an electrically conductive etching protective layer 109; a protective insulating layer 106; and a TFT part comprising at least a source electrode 105a, a drain electrode 105b, an active layer 104, a gate insulating layer 103 and a gate electrode 102 are provided in this order. The electrically

conductive etching protective layer **109** is provided in a patterned form at a position where a contact hole **108** is formed. One of the source electrode **105a** or the drain electrode **105b**, and the electrically conductive etching protective layer **109** are electrically connected by the contact hole **108** formed in the protective insulating layer **106**. In other words, in this constitution, one of the source electrode or the drain electrode, and the upper electrode are not directly electrically connected, but are connected via the electrically conductive etching protective layer. The electrically conductive etching protective layer is formed by a sputtering method, an ion plating method or a CVD method. The electrically conductive etching protective layer is a tight layer, and therefore acts as a barrier to permeation of an etching material during the formation of a contact hole, thereby preventing the upper electrode and the organic layer from suffering damage.

**[0080]** In this constitution, the substrate and the lower electrode are transparent, and the upper electrode is light-reflective. Therefore, light generated upon light emission is extracted through the substrate to the outside.

**[0081]** FIG. 2 is a conceptual sectional view showing a constitution of a comparative organic EL display.

**[0082]** The constitution of the organic EL display shown in FIG. 2 differs from that shown in FIG. 1 in the point that the display does not have an electrically conductive etching protective layer. One of a source electrode **115a** or a drain electrode **115b**, and an upper electrode **44** are directly electrically connected. The upper electrode **44** is formed by an electric resistance heating deposition method, because there is no damage to an organic layer. However, the resulting layer has crude density, and many pinholes are present therein. Consequently, the layer does not act as a barrier to permeation of an etching material during the formation of a contact hole **118**, and the etching material permeates through the upper electrode **44** to an organic layer **42** and gives damage thereto, resulting in deterioration of the organic EL display.

**[0083]** In the invention, the TFT is disposed at a back surface of the organic EL element on a side opposite to a light-extracting surface. Since the TFT used in the invention is excellent in the ON/OFF characteristic and capable of supplying a large current, it enables downsizing of TFT so as to also sufficiently applicable to a high compact arrangement of organic EL elements, and thereby an opening portion of the organic EL element can be disposed widely.

**[0084]** Accordingly, an organic EL display having high reliability, high definition, high brightness and high durability is provided.

## 2. TFT

**[0085]** The TFT used in the invention is an active-type element having at least a gate electrode, a gate insulating layer, an active layer, a source electrode, and a drain electrode in this order, and having a function of switching a current between the source electrode and the drain electrode by applying a voltage to the gate electrode and controlling the current flowing to the active layer. An oxide semiconductor is used for an active layer of the TFT in the invention. The oxide semiconductor can be layered by low temperature and thereby can be formed with less damages to the organic EL element. Further, compared with an organic semiconductor such as pentacene, it is excellent not only in mobility but also excellent in view of driving stability and storage stability. Particularly, an amorphous oxide semiconductor is preferred for the active layer of the TFT in view of the uniformity of

TFT characteristics and stability of characteristics. With respect to the TFT structure, any of a stagger structure or an inverted stagger structure can be formed.

**[0086]** Preferably, the TFT has N-type polarity.

**[0087]** An organic EL element usually has a structure having a transparent anode using ITO for a lower electrode and a light reflective cathode using Al for an upper electrode. A source or a drain electrode of a driving TFT is preferably connected with the upper electrode, that is, the cathode of the organic EL element in view of manufacturing process or constitution thereof. For example, in a case of constituting a pixel circuit as a simple 2-transistor-1-capacitor (2Tr-1C), particularly excellent performance is obtained in driving characteristics by connecting the drain electrode of TFT with the cathode of the organic EL element, grounding the anode of the organic EL element and using a N-type TFT. This is because stable driving is possible since the gate voltage of the driving TFT is free from influences by driving voltage for the organic EL element. Accordingly, an existent compensation circuit such as 4Tr for stabilization is no more necessary, which enables down-sizing of the TFT portion and facilitates the design for an organic EL display of higher definition, higher brightness, and longer durability.

**[0088]** As the active layer in the present invention is formed on the organic EL element, the active layer preferably contains an organic semiconductor, an oxide semiconductor, a carbon nanotube or the like which can be formed at a lower temperature. In particular, in viewpoint of mobility and drive stability, the active layer preferably contains an oxide semiconductor, and more preferably contains an amorphous oxide semiconductor.

**[0089]** Preferably, the TFT in the present invention includes at least a gate electrode, a gate insulating layer, a semiconductor layer containing an active layer and an electric resistance layer, a source electrode and a drain electrode in this order, wherein an electric conductivity of the electric resistance layer is lower than that of the active layer, and the active layer is closer to the gate insulating layer, and the electric resistance layer is disposed with electrical contact between the active layer and at least one of the source electrode and the drain electrode. Preferably, the electric resistance layer in the present invention also contains an oxide semiconductor. Hereinafter, it may refer as a "semiconductor layer" as a definition to mean a term including an active layer and an electric resistance layer.

**[0090]** More preferably, at least an active layer and an electric resistance layer are formed to be layered, and the active layer is in contact with the gate insulating layer, and the electric resistance layer is contact with at least one of the source electrode and the drain electrode.

**[0091]** Preferably, a thickness of the electric resistance layer is larger than a thickness of the active layer in view of drive stability.

**[0092]** Further, as another embodiment, an embodiment in which the electric conductivity between the electric resistance layer and the active layer changes continuously. In the structure, there is no distinct boundary between the electric resistance layer and the active layer. With respect to a total thickness of a semiconductor layer including the electric resistance layer and the active layer, a 10% region adjacent with the gate insulating layer is defined as an active layer, and a 10% region for the thickness of the semiconductor layer adjacent with the source electrode or the drain electrode is defined as an electric resistance layer.

[0093] An oxygen concentration of the active layer is preferably lower than an oxygen concentration of the electric resistance layer.

[0094] An oxide semiconductor preferably includes at least one member selected from the group consisting of In, Ga, and Zn, or a composite oxide thereof. More preferably, the oxide semiconductor contains In and Zn, and a composition ratio between Zn and In in the electric resistance layer (represented by ratio of Zn to In, Zn/In) is larger than a composition ratio of Zn/In in the active layer. The Zn/In ratio in the electric resistance layer is preferably greater by 3% or more and, and more preferably by 10% or more than the Zn/In ratio in the active layer.

[0095] Preferably, a ratio of the electric conductivity of the active layer to the electric conductivity of the electric resistance layer (electric conductivity of active layer/electric conductivity of electric resistance layer) is from  $10^1$  to  $10^{10}$ , and more preferably from  $10^2$  to  $10^8$ .

[0096] Preferably, the substrate is a flexible resin substrate.

[0097] The electric conductivity of the active layer is preferably  $10^{-4}$  Scm $^{-1}$  or more, and less than  $10^2$  Scm $^{-1}$ , and more preferably  $10^{-1}$  Scm $^{-1}$  or more, and less than  $10^2$  Scm $^{-1}$ . The electric conductivity of the electric resistance layer is preferably  $10^{-2}$  Scm $^{-1}$  or lower, and more preferably  $10^{-9}$  Scm $^{-1}$  or more, and less than  $10^{-3}$  Scm $^{-1}$ , which is lower than the electric conductivity of the active layer.

[0098] In the case where the electric conductivity of the active layer is less than  $10^{-4}$  Scm $^{-1}$ , no high mobility is obtained as the field effect mobility. In the case where the electric conductivity of the active layer is  $10^2$  Scm $^{-1}$  or more, an OFF current increases and thereby no favorable ON/OFF ratio can be obtained, which is not preferred.

[0099] Preferably, a thickness of the electric resistance layer is larger than a thickness of the active layer in a view of drive stability. Preferably, a ratio of the thickness of the electric resistance layer to the thickness of the active layer (thickness of the electric resistance layer/thickness of the active layer) is more than 1 and 100 or less, and more preferably more than 1 and 10 or less.

[0100] Next, the structure of the TFT more preferably used in the invention will be described in detail with reference to the drawings.

[0101] 1) Structure

[0102] FIG. 3 is a schematic diagram showing an example of the reversed stagger structure of the thin film field-effect transistor of the invention. In the case where a substrate 1 is composed of a flexible substrate such as a plastic film or the like, the thin film field-effect transistor has an insulating layer 6 disposed on one surface of the substrate 1, and on the insulating layer 6, a gate electrode 2, a gate insulating layer 3, an active layer 4-1, and an electric resistance layer 4-2 are stacked. On the surface of the structure thus constructed, a source electrode 5-1 and a drain electrode 5-2 are disposed. The active layer 4-1 borders on the gate insulating layer 3, and the electric resistance layer 4-2 borders on the source electrode 5-1 and the drain electrode 5-2. The compositions of the active layer 4-1 and electric resistance layer 4-2 are determined so that the electric conductivity of the active layer 4-1 is higher than that of the electric resistance layer 4-2 when no voltage is applied to the gate electrode. Incidentally, for the active layer, oxide semiconductors disclosed in JP-A No. 2006-165529, e.g., In—Ga—Zn—O-based oxide semiconductors, are used. It is known that in these oxide semiconductors, the higher the concentration of electron carriers is, the

higher the electron mobility is. In other words, the higher the electric conductivity is, the higher the electron mobility is.

[0103] According to this structure of the invention, when the thin film field-effect transistor in the ON state under the condition where voltage is applied to the gate electrode, the active layer which becomes a channel has high electric conductivity. As a result, the field effect mobility of the transistor is increased and a large ON current can be obtained. On the other hand, in the OFF state, the electric resistance layer has a high resistance because of its low electric conductivity, and the OFF current is kept low. Thus, the ON-OFF ratio is remarkably improved.

[0104] Although it is not shown in the drawing, the point of the invention is to provide a semiconductor layer whose electric conductivity closer to the gate insulating layer is higher than that closer to the source electrode and the drain electrode. As long as this condition is achieved, the means for achieving this is not limited to providing a plurality of semiconductor layers as shown in FIG. 3. The electric conductivity may be changed continuously.

[0105] FIG. 4 is a schematic diagram showing an example of the top gate structure of the thin film field-effect transistor according to the invention. In the case where a substrate 11 is composed of a flexible substrate such as a plastic film or the like, the thin film field-effect transistor has an insulating layer 16 disposed on one surface of the substrate 11, a source electrode 5-11 and a drain electrode 5-12 are provided on the insulating layer, an electric resistance layer 4-12 and an active layer 4-11 are stacked, and then a gate insulating layer 13 and a gate electrode 12 are provided. Similar to the case of the reversed stagger structure, the active layer 4-11 (which is a high-electric conductivity layer) borders on the gate insulating layer 13, and the electric resistance layer 4-12 (which is a low-electric conductivity layer) borders on the source electrode 5-11 and the drain electrode 5-12. The compositions of the active layer 4-11 and electric resistance layer 4-12 are determined so that the electric conductivity of the active layer 4-11 is higher than that of the electric resistance layer 4-12 when no voltage is applied to the gate electrode.

[0106] 2) Electric Conductivity

[0107] Now, the electric conductivity of the active layer and the electric resistance layer in association with the invention will be explained.

[0108] The electric conductivity is a physical property which indicates how much electricity a substance can conduct. When a carrier concentration of a substance is denoted by  $n$ , a carrier mobility is denoted by  $\mu$ , and an electric elementary quantity is denoted by  $e$ , the electric conductivity  $\sigma$  of the substance is expressed as follows.

$$\sigma = ne\mu$$

[0109] When the active layer or the electric resistance layer is composed of an n-type semiconductor, the carrier is an electron. In this case, the carrier concentration refers to the concentration of electron carriers, and the carrier mobility refers to the electron mobility. Conversely, when the active layer or the electric resistance layer is a p-type semiconductor, the carrier is a hole. In this case, the carrier concentration refers to the concentration of hole carriers, and the carrier mobility refers to the hole mobility. Further, the carrier concentration and carrier mobility of a substance can be determined by Hall measurements.

<Method of Determining Electric Conductivity>

[0110] The electric conductivity of a film can be determined by measuring the sheet resistance of the film, provided

that the thickness of the film is known. The electric conductivity of a semiconductor changes depending on the temperature, and the electric conductivity cited herein refers to the electric conductivity at room temperature (20° C.).

**[0111]** 3) Gate Insulating Layer

**[0112]** For the gate insulating layer, an insulator such as SiO<sub>2</sub>, SiN<sub>x</sub>, SiON, Al<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, Ta<sub>2</sub>O<sub>5</sub>, HfO<sub>2</sub> and the like, or a mixed crystal compound containing at least two of these is used. Also, a polymeric insulator such as polyimide may be used for the gate insulating layer.

**[0113]** It is preferable that the gate insulating layer has a thickness from 10 nm to 10 μm. To reduce the leak current and raise the voltage resistance, it is required to make the gate insulating layer thicker to a certain extent. However, an increase in the thickness of the gate insulating layer results in a rise in the voltage needed for driving the TFT. Therefore, it is preferable that the thickness of the gate insulating layer is from 50 nm to 1000 nm for an inorganic insulator, and from 0.5 μm to 5 μm for a polymeric insulator. Especially, it is particularly preferable to use an insulator with a high dielectric constant, such as HfO<sub>2</sub>, for the gate insulating layer, because then the TFT can be driven with low voltage even when it is made thicker.

**[0114]** 4) Active Layer and Electric Resistance Layer

**[0115]** For the semiconductor layer including the active layer and the electric resistance layer in this invention, it is preferable to use an oxide semiconductor. Particularly, an amorphous oxide semiconductor is preferable. Films of oxide semiconductors, particularly amorphous oxide semiconductors, can be formed at a low temperature, and so can be prepared on a flexible substrate made of a resin such as plastic. Satisfactory amorphous oxide semiconductors which can be prepared at a low temperature include an oxide containing In, an oxide containing In and Zn, and an oxide containing In, Ga and Zn, as disclosed in JP-A No. 2006-165529. Considering their compositional structures, it is known that amorphous oxide semiconductors of InGaO<sub>3</sub>(ZnO)<sub>m</sub> (m is a natural number less than 6) are preferable. These oxide semiconductors are n-type semiconductors, in which electrons serve as carriers. Of course, p-type oxide semiconductors such as ZnO/Rh<sub>2</sub>O<sub>3</sub>, CuGaO<sub>2</sub>, and SrCu<sub>2</sub>O<sub>2</sub> may be used for the active layer and the electric resistance layer.

**[0116]** Specifically, an amorphous oxide semiconductor according to the invention preferably has a constitution including In—Ga—Zn—O. The amorphous oxide semiconductor is preferably an amorphous oxide semiconductor with a composition of InGaO<sub>3</sub>(ZnO)<sub>m</sub> (m is a natural number less than 6) in a crystalline state. Particularly, InGaZnO<sub>4</sub> is more preferable. An amorphous oxide semiconductor of such composition has a feature that the electron mobility tends to increase with an increase in the electric conductivity. In addition, as to the control of the electric conductivity, it is disclosed in JP-A No. 2006-165529 that the electric conductivity can be controlled by controlling the partial pressure of oxygen during the film formation.

**[0117]** As a matter of course, not only oxide semiconductors, but also inorganic semiconductors such as Si and Ge, compound semiconductors such as GaAs, and organic semiconductor materials such as pentacene, polythiophene, and the like can be used for the active layer and the electric resistance layer.

**[0118]** <Electric Conductivity of Active Layer and Electric Resistance Layer>

**[0119]** The active layer of the invention is characterized in that it is closer to the gate insulating layer, and the electric

conductivity thereof is higher than that of the electric resistance layer which is closer to the source electrode and the drain electrode.

**[0120]** The ratio of the electric conductivity of the active layer to the electric conductivity of the electric resistance layer (i.e., the electric conductivity of the active layer divided by the electric conductivity of the electric resistance layer) is preferably from 10<sup>1</sup> to 10<sup>10</sup>, and more preferably from 10<sup>2</sup> to 10<sup>8</sup>. The electric conductivity of the active layer is preferably 10<sup>-4</sup> Scm<sup>-1</sup> or more, and less than 10<sup>2</sup> Scm<sup>-1</sup>, and more preferably 10<sup>-1</sup> Scm<sup>-1</sup> or more, and less than 10<sup>2</sup> Scm<sup>-1</sup>. The electric conductivity of the electric resistance layer is preferably 10<sup>-1</sup> Scm<sup>-1</sup> or less, and more preferably 10<sup>-9</sup> Scm<sup>-1</sup> or more, and less than 10<sup>-3</sup> Scm<sup>-1</sup>.

**[0121]** <Thickness of Active Layer and Electric Resistance Layer>

**[0122]** It is preferable that the electric resistance layer is thicker than the active layer. More preferably, the ratio of a thickness of the electric resistance layer to that of the active layer is more than 1 and 100 or less, and even more preferably the ratio is more than 1 and 10 or less.

**[0123]** Preferably, the thickness of the active layer is from 1 nm to 100 nm, and more preferably, from 2.5 nm to 30 nm. Preferably, the thickness of the electric resistance layer is from 5 nm to 500 nm, and more preferably, from 10 nm to 100 nm.

**[0124]** Use of the active layer and the electric resistance layer arranged as described above achieves a TFT characterized by an ON-OFF ratio of 10<sup>6</sup> or higher and high mobility of 10 cm<sup>2</sup>/V/sec or higher.

**[0125]** <Means for Adjusting Electric Conductivity>

**[0126]** The electric conductivity of the semiconductor layer according to the present invention is adjusted so that, as described above, the electric conductivity of a part of the semiconductor layer closer to the gate insulating layer (active layer) becomes larger than the electric conductivity of a part of the semiconductor layer closer to the source electrode or the drain electrode (electric insulating layer).

**[0127]** In the case where the active layer and the electric resistance layer are composed of an oxide semiconductor, the means for adjusting the electric conductivity are what are described in the following items (1) to (4).

**[0128]** (1) Adjustment by Oxygen Defect

**[0129]** It is known that when an oxygen vacancy is made in oxide semiconductors, a carrier electron is generated, which results in an increase in electric conductivity. Hence, the electric conductivity of an oxide semiconductor can be controlled by adjusting the quantity of oxygen vacancies. Specifically, means for controlling the quantity of oxygen vacancies include adjusting the partial pressure of oxygen during the time of film formation, and oxygen concentration and treatment time of an after-treatment after the film formation. Specifically, examples of this after-treatment include heat treatment at a temperature of 100° C. or higher, processing by oxygen plasma, and UV ozone treatment. Among these, the method involving controlling the partial pressure of oxygen during the time of film formation is preferable in view of its productivity. It has been disclosed in JP-A No. 2006-165529 that the electric conductivity of an oxide semiconductor can be controlled by adjusting the partial pressure of oxygen during the time of film formation, and therefore this method is usable.

**[0130]** (2) Adjustment by Composition Ratio

**[0131]** It has been known that the electric conductivity can be changed by changing the composition ratio of metals of an oxide semiconductor. For instance, it has been disclosed in JP-A No. 2006-165529 that in the case of  $\text{InGaZn}_{1-x}\text{Mg}_x\text{O}_4$ , the electric conductivity lowers with an increase in the percentage of Mg. In addition, it has been reported that the electric conductivity of oxides of  $(\text{In}_2\text{O}_3)_{1-x}(\text{ZnO})_x$  lowers with an increase in the percentage of Zn when the Zn/In ratio is 10% or higher ("*TOMEI DOUDENMAKU NO SINTEN-KAI II (Developments of Transparent Conductive Films II)*", pages 34-35, CMC Publishing CO., LTD.). Specifically, means for changing the composition ratio for example in the case of a method of forming a film by sputtering include a means using targets with different composition ratios. Alternatively, multiple targets may be co-sputtered, changing the composition ratio of the resultant film by individually adjusting the sputtering rates for the targets.

**[0132]** (3) Adjustment by Impurities

**[0133]** It has been disclosed in JP-A No. 2006-165529 that when elements such as La, Na, Mn, Ni, Pd, Cu, Cd, C, N, and P are selectively added to an oxide semiconductor as an impurity, the concentration of electron carriers can be reduced, and therefore the electric conductivity can be made lower. Means for adding an impurity include co-vapor deposition of the oxide semiconductor and the impurity, and ion-doping of an oxide semiconductor film which has already been formed with ions of the impurity element.

**[0134]** (4) Adjustment by Oxide Semiconductor Material

**[0135]** While in the above items (1) to (3), the methods of adjusting the electric conductivity of the same oxide semiconductor system have been described, the electric conductivity can be changed by changing the oxide semiconductor material. It is known that the electric conductivity of  $\text{SnO}_2$ -based oxide semiconductors is lower than  $\text{In}_2\text{O}_3$ -based oxide semiconductors. In this way, the electric conductivity can be adjusted by changing the oxide semiconductor material. In particular, as the oxide materials having low electric conductivity, oxide insulator materials such as  $\text{Al}_2\text{O}_3$ ,  $\text{Ga}_2\text{O}_3$ ,  $\text{ZrO}_2$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{Ta}_2\text{O}_5$ ,  $\text{MgO}$ ,  $\text{HfO}_3$ , and the like are known, and it is possible to use these materials.

**[0136]** As the means for adjusting the electric conductivity, the means stated in the above (1) to (4) may be used independently or in combination.

**[0137]** <Method of Forming Active Layer and Electric Resistance Layer>

**[0138]** As the means for forming a film of the active layer and the electric resistance layer, it is suitable to adopt a vapor-phase film forming method using, as a target, a polycrystalline sintered compact of an oxide semiconductor. Among the vapor-phase film forming methods, sputtering method and pulsed laser deposition method (PLD method) are adequate. For mass production, sputtering method is preferable.

**[0139]** For instance, by an RF magnetron sputtering deposition method, a film can be formed while controlling the vacuum level and flow rate of oxygen. The higher the flow rate of oxygen is, the lower the electric conductivity can be made.

**[0140]** It can be verified by conventional X-ray diffraction that the resultant film is an amorphous film.

**[0141]** The thickness of the film can be determined by contact stylus-type surface profile measurement. The composition ratio can be determined by RBS analysis (Rutherford Backscattering Spectrometry).

**[0142]** 5) Gate Electrode

**[0143]** According to the invention, the following materials are among those which are preferable for the gate electrode: a metal such as Al, Mo, Cr, Ta, Ti, Au or Ag, an alloy such as Al—Nd or APC; a metal oxide conductive film of e.g., tin oxide, zinc oxide, indium oxide, indium-tin oxide (ITO), or indium-zinc oxide (IZO); an organic conductive compound such as polyaniline, polythiophene, or polypyrrole; or a mixture thereof.

**[0144]** The thickness of the gate electrode is preferably from 10 nm to 1000 nm.

**[0145]** The method of forming the electrode is not particularly limited. The film can be formed on the substrate according to a method which is appropriately selected from among wet methods such as a printing method and a coating method, physical methods such as a vacuum deposition method, a sputtering method and an ion plating method, chemical methods such as a CVD and plasma CVD method, and the like in consideration of the suitability with the material described above. For example, when ITO is selected, the film can be formed according to a DC or RF sputtering method, a vacuum deposition method, or an ion plating method. Further, in the case where an organic conductive compound is selected as the material of the gate electrode, the film formation can be performed according to a wet film-forming method.

**[0146]** 6) Source Electrode and Drain Electrode

**[0147]** According to the invention, the following are suitable for the material of the source electrode and the drain electrode: metals such as Al, Mo, Cr, Ta, Ti, Au and Ag; alloys such as Al—Nd and APC; metal oxide conductive films of, for example, tin oxide, zinc oxide, indium oxide, indium-tin oxide (ITO) and indium-zinc oxide (IZO); and organic conductive compounds such as polyaniline, polythiophene and polypyrrole, and mixtures thereof.

**[0148]** The thickness of the source electrode and the drain electrode is preferably from 10 nm to 1000 nm.

**[0149]** The method of forming the electrodes is not particularly limited. The films can be formed on the substrate according to a method which is appropriately selected from among wet methods such as a printing method and a coating method, a physical methods such as a vacuum deposition method, a sputtering method and an ion plating method, a chemical methods such as a CVD and plasma CVD method and the like in consideration of the suitability with the material described above. For example, when ITO is selected, the films can be formed according to a DC or RF sputtering method, a vacuum deposition method, an ion plating method, etc. Further, in the case where an organic conductive compound is selected as the material of the source electrode and the drain electrode, the film formation can be performed according to a wet film-forming method.

**[0150]** 7) Insulating Layer

**[0151]** If necessary, an insulating layer may be provided on TFT.

**[0152]** The insulating layer has a function to protect semiconductor layer (active layer and resistance layer) from deterioration by air, and to insulate a device formed on TFT from TFT.

**[0153]** Specific examples of materials for the insulating layer include metal oxides such as  $\text{MgO}$ ,  $\text{SiO}$ ,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{GeO}$ ,  $\text{NiO}$ ,  $\text{CaO}$ ,  $\text{BaO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{TiO}_2$  and the like; metal nitrides such as  $\text{SiN}_x$ ,  $\text{SiN}_x\text{O}_y$  and the like; metal fluorides such as  $\text{MgF}_2$ ,  $\text{LiF}$ ,  $\text{AlF}_3$ ,  $\text{CaF}_2$  and the like; polyethylene; polypropylene; polymethyl methacrylate; polyimide; poly-

urea; polytetrafluoroethylene; polychlorotrifluoroethylene; polydichlorodifluoroethylene; a copolymer of chlorotrifluoroethylene and dichlorodifluoroethylene; copolymers obtained by copolymerizing a monomer mixture containing tetrafluoroethylene and at least one co-monomer; fluorine-containing copolymers each having a cyclic structure in the copolymerization main chain; water-absorbing materials each having a coefficient of water absorption of 1% or more; moisture permeation preventive substances each having a coefficient of water absorption of 0.1% or less; and the like.

[0154] There is no particular limitation as to a method for forming the insulating layer. For instance, a vacuum deposition method, a sputtering method, a reactive sputtering method, an MBE (molecular beam epitaxial) method, a cluster ion beam method, an ion plating method, a plasma polymerization method (high-frequency excitation ion plating method), a plasma CVD method, a laser CVD method, a thermal CVD method, a gas source CVD method, a coating method, a printing method, or a transfer method may be applied.

[0155] 8) After Treatment

[0156] If necessary, heating treatment may be conducted as an after treatment for TFT. The heating treatment is performed under air or nitrogen environment at 100° C. or higher. The heating treatment may be conducted after film forming or at a last step of TFT fabrication steps. The heating treatment has results in that a fluctuation of TFT properties within a set of TFT is prevented, and drive stability is improved.

### 3. Organic Electroluminescence Element

[0157] The light-emitting element of the invention has a lower electrode and an upper electrode on a substrate, and an organic compound layer containing an organic light-emitting layer (hereinafter, sometimes simply referred to as a "light-emitting layer") between the electrodes. The upper electrode is connected with a source electrode or a drain electrode of a drive transistor. From the nature of a light-emitting element, at least one electrode of the lower electrode and the upper electrode is preferably transparent. One of the electrodes is an anode, and the other is a cathode. Generally, the lower electrode is an anode and the upper is a cathode.

[0158] As an integration pattern of the organic compound layers according to the present invention, it is preferred that the layers are integrated in the order of a hole transport layer, a light-emitting layer, and an electron transport layer from the anode side. Moreover, a charge-blocking layer or the like may be provided between the hole transport layer and the light-emitting layer or between the light-emitting layer and the electron transport layer. In addition, a hole injection layer may be provided between the anode and the hole transport layer, and similarly, an electron-injection layer may be provided between the cathode and the electron transport layer. Further, each of the layers may be composed of plural secondary layers.

[0159] The respective layers constituting the organic compound layer can be suitably formed in accordance with any of a dry film-forming method such as a vapor deposition method or a sputtering method; a wet film-forming method; a transfer method; a printing method; an ink-jet method; or the like.

[0160] Next, the components constituting the light-emitting material of the present invention will be described in detail.

(Substrate)

[0161] The substrate to be applied in the invention is preferably one which does not scatter or attenuate light emitted from the organic compound layer. Specific examples of materials for the substrate include zirconia-stabilized yttrium (YSZ); inorganic materials such as glass; polyesters such as polyethylene terephthalate, polybutylene phthalate, and polyethylene naphthalate; and organic materials such as polystyrene, polycarbonate, polyethersulfone, polyarylate, polyimide, polycycloolefin, norbornene resin, polychlorotrifluoroethylene, and the like.

[0162] For instance, when glass is used as the substrate, non-alkali glass is preferably used with respect to the quality of material in order to decrease ions eluted from the glass. In the case of employing soda-lime glass, it is preferred to use glass on which a barrier coat of silica or the like has been applied. In the case of employing an organic material, it is preferred to use a material excellent in heat resistance, dimension stability, solvent resistance, electric insulation, and workability.

[0163] There is no particular limitation as to the shape, the structure, the size or the like of the substrate, but it may be suitably selected according to the application, purposes and the like of the light-emitting element. In general, a plate-like substrate is preferred as the shape of the substrate. A structure of the substrate may be a monolayer structure or a laminated structure. Furthermore, the substrate may be formed from a single member or two or more members.

[0164] Although the substrate may be transparent and colorless, or transparent and colored, it is preferred that the substrate is transparent and colorless from the viewpoint that the substrate does not scatter or attenuate light emitted from the organic light-emitting layer.

[0165] A moisture permeation preventive layer (gas barrier layer) may be provided on the front surface or the back surface of the substrate.

[0166] For a material of the moisture permeation preventive layer (gas barrier layer), inorganic substances such as silicon nitride and silicon oxide may be preferably applied. The moisture permeation preventive layer (gas barrier layer) may be formed in accordance with, for example, a high-frequency sputtering method or the like.

[0167] In the case of applying a thermoplastic substrate, a hard-coat layer or an under-coat layer may be further provided as needed.

(Anode)

[0168] The anode may generally be any material as long as it has a function as an electrode for supplying holes to the organic compound layer, and there is no particular limitation as to the shape, the structure, the size or the like. However, it may be suitably selected from among well-known electrode materials according to the application and purpose of the light-emitting element. As mentioned above, the anode is usually provided as a transparent anode.

[0169] Materials for the anode preferably include, for example, metals, alloys, metal oxides, electrically conductive compounds, and mixtures thereof. Specific examples of the anode materials include electrically conductive metal oxides such as tin oxides doped with antimony, fluorine or the like (ATO and FTO), tin oxide, zinc oxide, indium oxide, indium tin oxide (ITO), and indium zinc oxide (IZO); metals such as gold, silver, chromium, and nickel; mixtures or laminates of

these metals and the electrically conductive metal oxides; inorganic electrically conductive materials such as copper iodide and copper sulfide; organic electrically conductive materials such as polyaniline, polythiophene, and polypyrrole; and laminates of these inorganic or organic electron-conductive materials with ITO. Among these, the electrically conductive metal oxides are preferred, and particularly, ITO is preferable in view of productivity, high electric conductivity, transparency and the like.

**[0170]** The anode may be formed on the substrate in accordance with a method which is appropriately selected from among wet methods such as printing methods, coating methods and the like; physical methods such as vacuum deposition methods, sputtering methods, ion plating methods and the like; and chemical methods such as CVD and plasma CVD methods and the like, in consideration of the suitability to a material constituting the anode. For instance, when ITO is selected as a material for the anode, the anode may be formed in accordance with a DC or high-frequency sputtering method, a vacuum deposition method, an ion plating method or the like.

**[0171]** In the organic electroluminescence element of the present invention, a position at which the anode is to be formed is not particularly limited, but it may be suitably selected according to the application and purpose of the light-emitting element. However, the anode is preferably formed on the substrate. The anode may be formed on either the whole surface or a part of the surface on either side of the substrate.

**[0172]** For patterning to form the anode, a chemical etching method such as photolithography, a physical etching method such as etching by laser, a method of vacuum deposition or sputtering through superposing masks, or a lift-off method or a printing method may be applied.

**[0173]** A thickness of the anode may be suitably selected according to the material constituting the anode and is therefore not definitely decided, but it is usually in a range of from 10 nm to 50  $\mu\text{m}$ , and preferably from 50 nm to 20  $\mu\text{m}$ .

**[0174]** A value of resistance of the anode is preferably  $10^3 \Omega/\square$  or less, and more preferably  $10^2 \Omega/\square$  or less. In the case where the anode is transparent, it may be either transparent and colorless, or transparent and colored. For extracting luminescence from the transparent anode side, it is preferred that a light transmittance of the anode is 60% or higher, and more preferably 70% or higher.

**[0175]** Concerning transparent anodes, there is a detailed description in "TOUMEI DENNKYOKU-MAKU NO SHINTENKAI (Novel Developments in Transparent Electrode Films)" edited by Yutaka Sawada, published by C.M.C. in 1999, the contents of which are incorporated by reference herein. In the case where a plastic substrate having a low heat resistance is applied, it is preferred that ITO or IZO is used to obtain a transparent anode prepared by forming the film at a low temperature of 150° C. or lower.

(Cathode)

**[0176]** The cathode may generally be any material as long as it has a function as an electrode for injecting electrons to the organic compound layer, and there is no particular limitation as to the shape, the structure, the size or the like. However it may be suitably selected from among well-known electrode materials according to the application and purpose of the light-emitting element.

**[0177]** Materials constituting the cathode include, for example, metals, alloys, metal oxides, electrically conductive compounds, and mixtures thereof. Specific examples thereof include alkali metals (e.g., Li, Na, K, Cs or the like), alkaline earth metals (e.g., Mg, Ca or the like), gold, silver, lead, aluminum, sodium-potassium alloys, lithium-aluminum alloys, magnesium-silver alloys, rare earth metals such as indium, and ytterbium, and the like. They may be used alone, but it is preferred that two or more of them are used in combination from the viewpoint of satisfying both stability and electron inject-ability.

**[0178]** Among these, as the materials for constituting the cathode, alkaline metals or alkaline earth metals are preferred in view of electron inject-ability, and materials containing aluminum as a major component are preferred in view of excellent preservation stability.

**[0179]** The term "material containing aluminum as a major component" refers to a material constituted by aluminum alone; alloys comprising aluminum and 0.01% by weight to 10% by weight of an alkaline metal or an alkaline earth metal; or the mixtures thereof (e.g., lithium-aluminum alloys, magnesium-aluminum alloys and the like).

**[0180]** Regarding materials for the cathode, they are described in detail in JP-A Nos. 2-15595 and 5-121172, the contents of which are incorporated by reference herein.

**[0181]** A method for forming the cathode is not particularly limited, but it may be formed in accordance with a well-known method.

**[0182]** For instance, the cathode may be formed in accordance with a method which is appropriately selected from among wet methods such as printing methods, coating methods and the like; physical methods such as vacuum deposition methods, sputtering methods, ion plating methods and the like; and chemical methods such as CVD and plasma CVD methods and the like, in consideration of the suitability to a material constituting the cathode. For example, when a metal (or metals) is (are) selected as a material (or materials) for the cathode, one or two or more of them may be applied at the same time or sequentially in accordance with a sputtering method or the like.

**[0183]** For patterning to form the cathode, a chemical etching method such as photolithography, a physical etching method such as etching by laser, a method of vacuum deposition or sputtering through superposing masks, or a lift-off method or a printing method may be applied.

**[0184]** In the present invention, a position at which the cathode is to be formed is not particularly limited, and it may be formed on either the whole or a part of the organic compound layer.

**[0185]** Furthermore, a dielectric material layer made of fluorides, oxides or the like of an alkaline metal or an alkaline earth metal may be inserted between the cathode and the organic compound layer with a thickness of 0.1 nm to 5 nm. The dielectric layer may be considered to be a kind of electron-injection layer. The dielectric material layer may be formed in accordance with, for example, a vacuum deposition method, a sputtering method, an ion plating method or the like.

**[0186]** A thickness of the cathode may be suitably selected according to materials for constituting the cathode and is therefore not definitely decided, but it is usually in a range of from 10 nm to 5  $\mu\text{m}$ , and preferably from 50 nm to 1  $\mu\text{m}$ .

**[0187]** Moreover, the cathode may be transparent or opaque. The transparent cathode may be formed by preparing

a material for the cathode with a small thickness of 1 nm to 10 nm, and further laminating a transparent electrically conductive material such as ITO or IZO thereon.

(Organic Compound Layer)

[0188] The organic electroluminescence element according to the present invention is to be described.

[0189] The organic EL element according to the present invention has at least one organic compound layer including a light-emitting layer. An organic compound layer apart from the light-emitting layer comprises a hole transport layer, an electron transport layer, a hole blocking layer, an electron blocking layer, a hole injection layer, an electron-injection layer and the like as described above.

[0190] In the organic EL element of the present invention, the respective layers constituting the organic compound layer can be suitably formed in accordance with any of a dry film-forming method such as a vapor deposition method, or a sputtering method; a wet film-forming method; a transfer method; a printing method; an ink-jet method; or the like.

(Light-Emitting Layer)

[0191] The light-emitting layer is a layer having functions of receiving holes from the anode, the hole injection layer, or the hole transport layer, and receiving electrons from the cathode, the electron-injection layer, or the electron transport layer, and providing a field for recombination of the holes with the electrons to emit a light, when an electric field is applied the layer.

[0192] The light-emitting layer according to the present invention may contain only a light-emitting material, but preferably is a mixture layer containing a light-emitting material and a host material. The light-emitting material may be a fluorescent light-emitting material or a phosphorescent light-emitting material, and may be a plurality of those compounds. Preferably, the host material is a charge-transporting material. The host material may be one or a plurality of compounds. For example, a mixture of a hole-transporting host material and an electron-transporting host material is preferable. Further, a material which does not emit light nor transport any charge may be contained in the light-emitting layer.

[0193] The light-emitting layer may be a single layer or a plurality of layers, wherein the layers may emit light with respectively different colors.

[0194] In the present invention, any of a fluorescent light-emitting material and a phosphorescent light-emitting material may be used as a light-emitting dopant.

[0195] The light-emitting layer of the present invention may contain two or more types of light-emitting dopants for improving color purity and expanding the luminescent wavelength region. It is preferred that the light-emitting dopant in the present invention is one satisfying a relationship between the above-described host material and the light-emitting dopant of  $1.2 \text{ eV} > \text{the difference of } I_p \text{ between host material and light-emitting dopant } (\Delta I_p) > 0.2 \text{ eV}$  and/or  $1.2 \text{ eV} > \text{the difference of } E_a \text{ between host material and light-emitting dopant } (\Delta E_a) > 0.2 \text{ eV}$  in view of drive durability.

[0196] <<Phosphorescent Light-Emitting Dopant>>

[0197] Examples of fluorescent light-emitting materials or dopants usable in the present invention include generally complexes containing a transition metal atom or a lanthanoid atom.

[0198] For instance, although the transition metal atom is not limited, it is preferably ruthenium, rhodium, palladium, tungsten, rhenium, osmium, iridium, or platinum; and more preferably rhenium, iridium, or platinum. In the present invention, an organic metal complex having platinum as a central metal is most preferable.

[0199] Examples of the lanthanoid atom include lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, terbium, dysprosium, holmium, erbium, thulium, ytterbium, and lutetium, and among these lanthanoid atoms, neodymium, europium, and gadolinium are preferred.

[0200] Examples of ligands in the complex include the ligands described, for example, in "Comprehensive Coordination Chemistry" authored by G. Wilkinson et al., published by Pergamon Press Company in 1987; "Photochemistry and Photophysics of Coordination compounds" authored by H. Yersin, published by Springer-Verlag Company in 1987; and "YUHKI KINZOKU KAGAKU—KISO TO OUYOU—(Metalorganic Chemistry—Fundamental and Application—)" authored by Akio Yamamoto, published by Shokabo Publishing Co., Ltd. in 1982.

[0201] Specific examples of ligand include halogen ligand (preferably, chlorine ligand), aliphatic carbon ring ligand (for example, preferably 5 to 30 carbon atoms, more preferably 6 to 30 carbon atoms, further preferably 6 to 20 carbon atoms, and particularly preferably 6 to 12 carbon atoms, such as cyclopentadienyl anion, benzene anion, naphthyl anion, or the like), nitrogen-containing hetero cyclic ligand (for example, preferably 5 to 30 atoms, more preferably 6 to 30 carbon atoms, further preferably 6 to 20 carbon atoms, and particularly preferably 6 to 12 carbon atoms, for example, phenyl pyridine, benzoquinoline, quinolinol, bipyridyl, phenanthroline, or the like), diketone ligand (for example, acetyl acetone, or the like), carboxylic acid ligand (for example, preferably 2 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, and further preferably 2 to 16 carbon atoms, such as acetic acid ligand, or the like), alcoholate ligand (for example, preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, and further preferably 6 to 20 carbon atoms, such as phenolate ligand, or the like), silyloxy ligand (for example, preferably 3 to 40 carbon atoms, more preferably 3 to 30 carbon atoms, and further preferably 3 to 20 carbon atoms, such as trimethyl silyloxy ligand, dimethyl tert-butyl silyloxy ligand, triphenyl silyloxy ligand, or the like), carbon monoxide ligand, isonitrile ligand, cyano ligand, phosphorus ligand (preferably 3 to 40 carbon atoms, more preferably 3 to 30 carbon atoms, further preferably 3 to 20 carbon atoms, and particularly preferably, 6 to 20 carbon atoms, such as triphenyl phosphine ligand, or the like), thiolate ligand (preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, and further preferably 6 to 20 carbon atoms, such as phenyl thiolate ligand, or the like), and phosphine oxide ligand (preferably 3 to 30 carbon atoms, more preferably 8 to 30 carbon atoms, and particularly preferably 18 to 30 carbon atoms, for example, triphenyl phosphine oxide ligand, or the like), and preferably, nitrogen-containing hetero cyclic ligand.

[0202] The above-described complexes may be either a complex containing one transition metal atom in the compound, or a so-called polynuclear complex containing two or more transition metal atoms wherein different metal atoms may be contained at the same time.

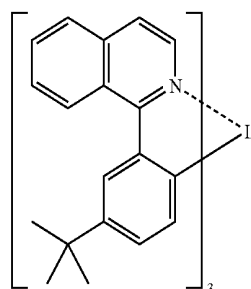
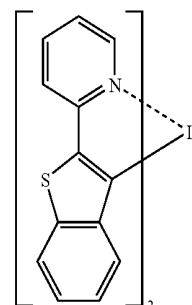
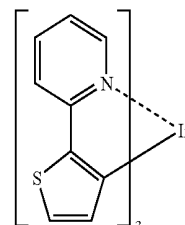
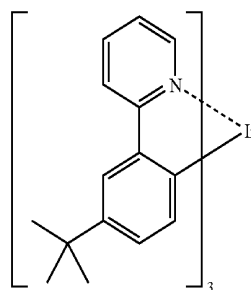
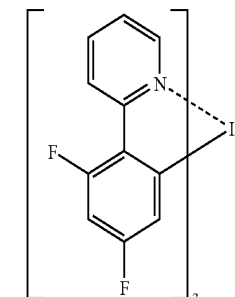
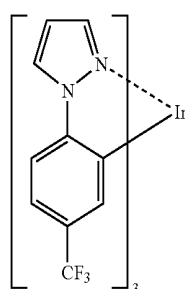
[0203] Among these, specific examples of the light-emitting dopants include phosphorescence luminescent compounds described in patent documents such as U.S. Pat. No. 6,303,238B1, U.S. Pat. No. 6,097,147, WO00/57676, WO00/70655, WO01/08230, WO01/39234A2, WO01/41512A1, WO02/02714A2, WO02/15645A1, WO02/44189A1, JP-A No. 2001-247859, Japanese Patent Application No. 2000-33561, JP-A Nos. 2002-117978, 2002-225352, and 2002-235076, Japanese Patent Application No. 2001-239281, JP-A No. 2002-170684, EP1211257, JP-A Nos. 2002-226495, 2002-234894, 2001-247859, 2001-298470, 2002-173674, 2002-203678, 2002-203679, and 2004-357791, Japanese Patent Application Nos. 2005-75340 and 2005-75341, etc. Among these, more preferable examples of the luminescent dopants include Ir complexes, Pt complexes, Cu complexes, Re complexes, W complexes, Rh complexes, Ru complexes, Pd complexes, Os complexes, Eu complexes, Tb complexes, Gd complexes, Dy complexes, and Cc complexes; particularly preferable are Ir complexes, Pt complexes, and Re complexes; and among these, Ir complexes, Pt complexes, and Re complexes each containing at least one coordination mode of metal-carbon bonds, metal-nitrogen bonds, metal-oxygen bonds, and metal-sulfur bonds are preferred. Particularly preferably, Ir complexes, Pt complexes, and Re complexes each containing a tri-dentate or higher poly-dentate ligand are preferred in view of light-emission efficiency, drive durability, color purity and the like.

[0204] <<Fluorescence Luminescent Dopant>>

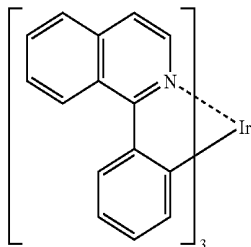
[0205] Examples of the above-described fluorescence luminescent dopants generally include benzoxazole, benzimidazole, benzothiazole, styrylbenzene, polyphenyl, diphenylbutadiene, tetraphenylbutadiene, naphthalimide, coumarin, pyran, perinone, oxadiazole, aldazine, pyralidine, cyclopentadiene, bis-styrylanthracene, quinacridone, pyrrolopyridine, thiadiazolopyridine, cyclopentadiene, styrylamine, aromatic dimethylidene compounds, condensed polyaromatic compounds (anthracene, phenanthroline, pyrene, perylene, rubrene, pentacene and the like), a variety of metal complexes represented by metal complexes of 8-quinolynol, pyromethene complexes or rare-earth complexes, polymer compounds such as polythiophene, polyphenylene or polyphenylenevinylene, organic silanes, and derivatives thereof.

[0206] Among these, specific examples of the luminescent dopants include the following compounds, but it should be noted that the present invention is not limited thereto.

-continued

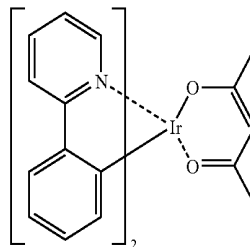


-continued

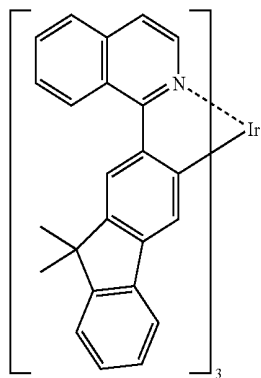


D-7

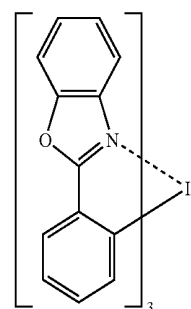
-continued



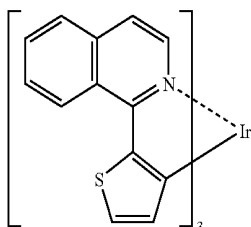
D-12



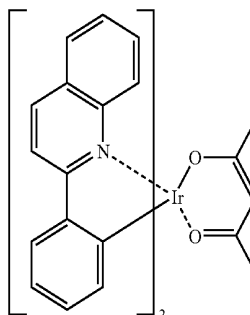
D-8



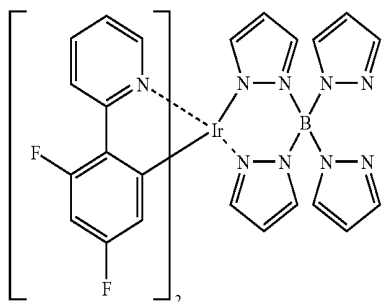
D-13



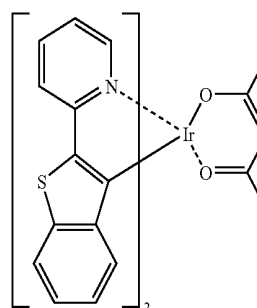
D-9



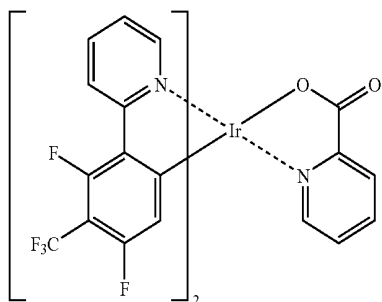
D-14



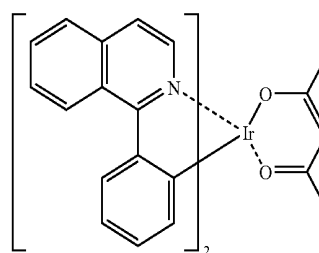
D-10



D-15

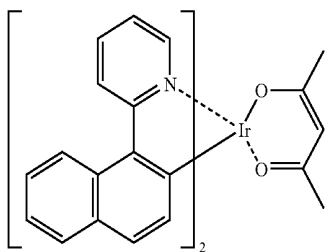


D-11



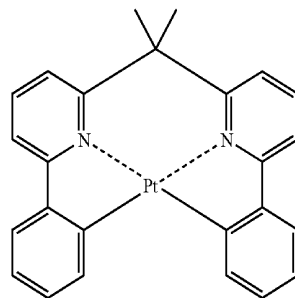
D-16

-continued

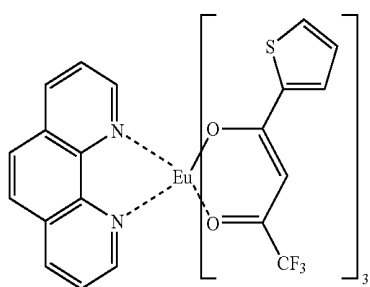


D-17

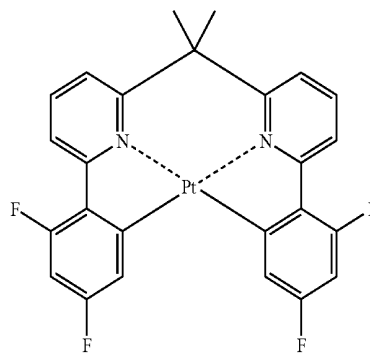
-continued



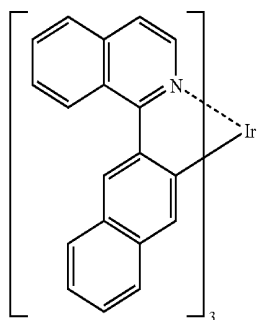
D-22



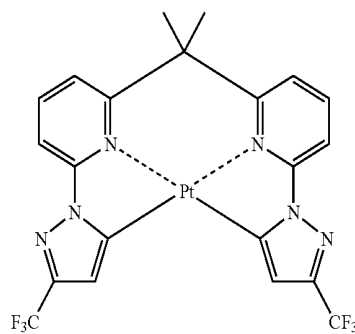
D-18



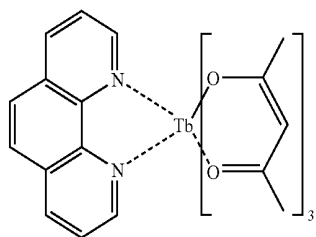
D-23



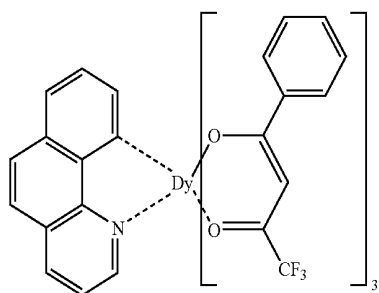
D-19



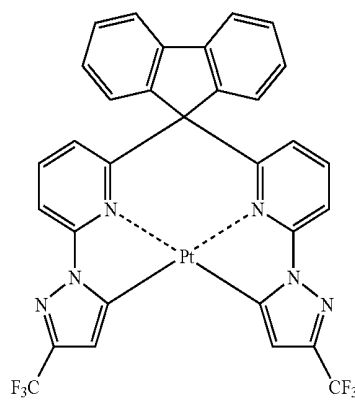
D-24



D-20

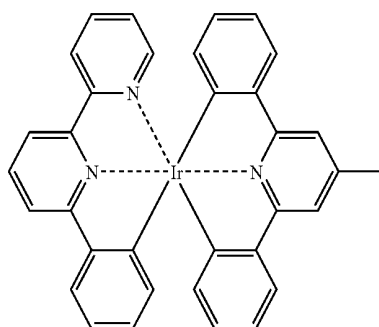
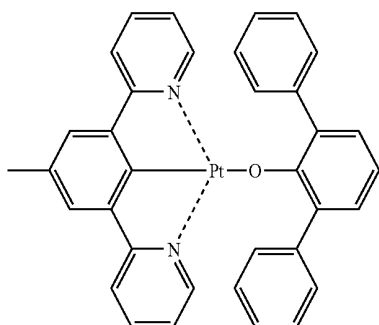
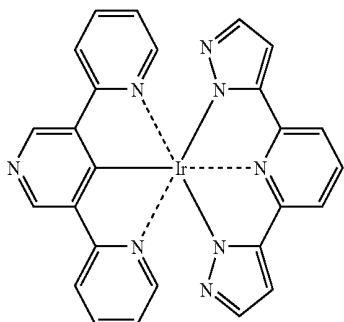
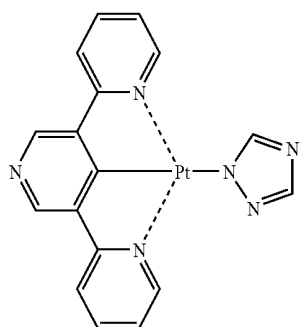
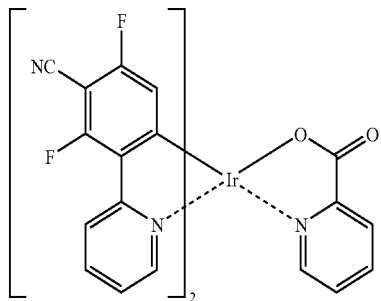


D-21



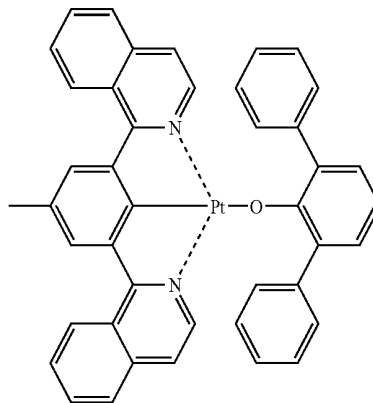
D-25

-continued



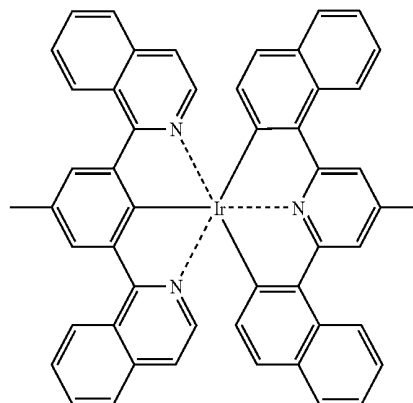
-continued

D-26



D-31

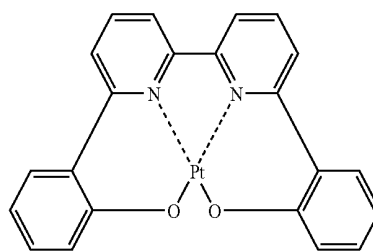
D-27



D-32

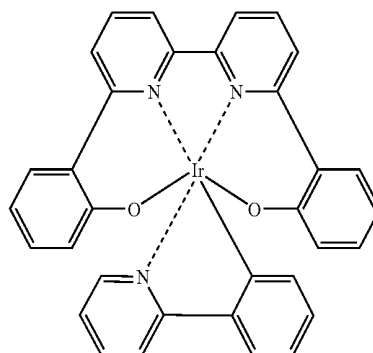
D-28

D-29



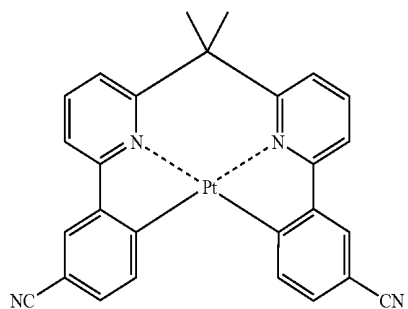
D-33

D-30



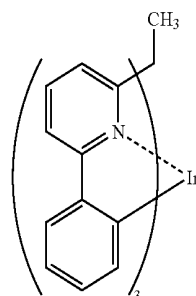
D-34

-continued

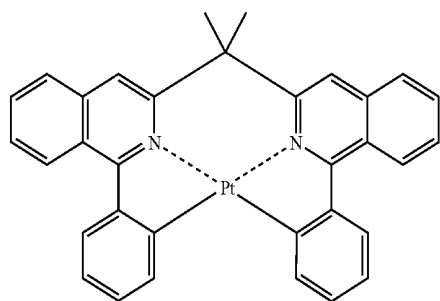


D-35

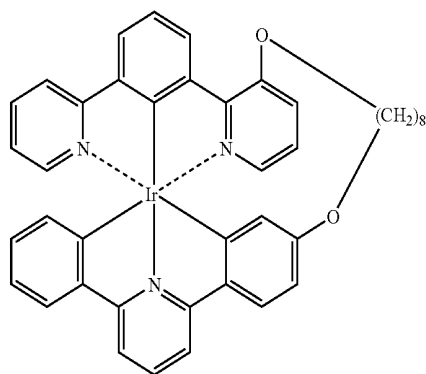
-continued



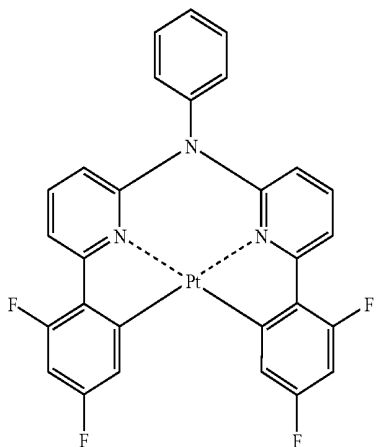
D-39



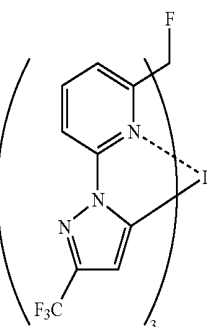
D-36



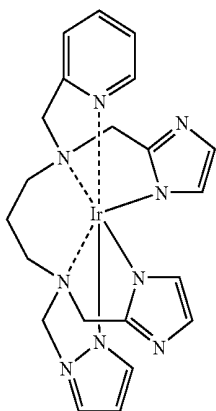
D-40



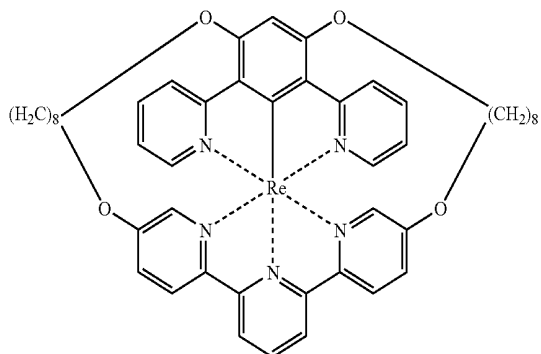
D-37



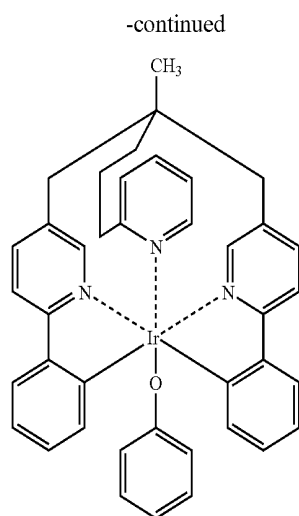
D-41



D-38



D-42



D-43

**[0207]** The light-emitting dopant or light-emitting material in a light-emitting layer is contained in an amount of 0.1% by mass to 50% by weight with respect to the total amount of the compounds generally forming the light-emitting layer, but it is preferably contained in an amount of 1% by weight to 50% by weight, and more preferably in an amount of 2% by weight to 40% by weight in view of drive durability and external light-emitting efficiency.

**[0208]** Although a thickness of the light-emitting layer is not particularly limited, 2 nm to 500 nm is usually preferred, and within this range, 3 nm to 200 nm is more preferable, and 5 nm to 100 nm is further preferred in view of external light-emitting efficiency.

**[0209]** <Host Material>

**[0210]** As the host materials to be used according to the present invention, hole transporting host materials excellent in hole transporting property (referred to as a "hole transporting host" in some cases) and electron transporting host compounds excellent in electron transporting property (referred to as an "electron transporting host" in some cases) may be used.

**[0211]** <<Hole Transporting Host>>

**[0212]** Specific examples of such hole transporting hosts used in the present invention include pyrrole, indole, carbazole, azaindole, azacarbazole, triazole, oxazole, oxadiazole, pyrazole, imidazole, thiophene, polyaryllalkane, pyrazoline, pyrazolone, phenylenediamine, arylamine, amino-substituted chalcone, styrylanthracene, fluorenone, hydrazone, stilbene, silazane, aromatic tertiary amine compounds, styrylamine compounds, aromatic dimethylidene compounds, porphyrin compounds, polysilane compounds, poly(N-vinyl-carbazole), aniline copolymers, electrically conductive high-molecular oligomers such as thiophene oligomers, polythiophenes and the like, organic silanes, carbon films, derivatives thereof, and the like.

**[0213]** Among these, indole derivatives, carbazole derivatives, aromatic tertiary amine compounds, and thiophene derivatives are preferable, and particularly, compounds containing a carbazole group in a molecule, and particularly preferably compounds containing t-butyl substituted carbazole group are preferred.

**[0214]** <<Electron Transporting Host>>

**[0215]** As the electron transporting host used in the present invention, it is preferred that an electron affinity  $E_a$  of the host is 2.5 eV to 3.5 eV, more preferably 2.6 eV to 3.4 eV, and further preferably 2.8 eV to 3.3 eV in view of improvements

in drive durability and decrease in drive voltage. Furthermore, it is preferred that an ionization potential  $I_p$  of the host is 5.7 eV to 7.5 eV, more preferably 5.8 eV to 7.0 eV, and further preferably 5.9 eV to 6.5 eV in view of improvements in drive durability and decrease in drive voltage.

**[0216]** Specific examples of such electron transporting hosts as mentioned above include pyridine, pyrimidine, triazine, imidazole, pyrazole, triazole, oxazole, oxadiazole, fluorenone, anthraquinonedimethane, anthrone, diphenylquinone, thiopyrandioxide, carbodiimide, fluorenylidene-methane, distyrylpyradine, fluorine-substituted aromatic compounds, heterocyclic tetracarboxylic anhydrides such as naphthalene, perylene and the like, phthalocyanine, derivatives thereof (which may form a condensed ring with another ring), and a variety of metal complexes represented by metal complexes of 8-quinolynol derivatives, metal phthalocyanine, and metal complexes having benzoxazole or benzothiazole as the ligand.

**[0217]** Preferable electron transporting hosts are metal complexes, azole derivatives (benzimidazole derivatives, imidazopyridine derivatives and the like), and azine derivatives (pyridine derivatives, pyrimidine derivatives, triazine derivatives and the like). Among these, metal complexes are preferred according to the present invention in view of durability. As the metal complex compound, a metal complex containing a ligand having at least one nitrogen atom, oxygen atom, or sulfur atom to be coordinated with the metal is more preferable.

**[0218]** Although a metal ion in the metal complex is not particularly limited, a beryllium ion, a magnesium ion, an aluminum ion, a gallium ion, a zinc ion, an indium ion, a tin ion, a platinum ion, or a palladium ion is preferred; more preferable is a beryllium ion, an aluminum ion, a gallium ion, a zinc ion, a platinum ion, or a palladium ion; and further preferable is an aluminum ion, a zinc ion, or a palladium ion.

**[0219]** Although there are a variety of well-known ligands to be contained in the above-described metal complexes, examples thereof include ligands described in "Photochemistry and Photophysics of Coordination Compounds" authored by H. Yersin, published by Springer-Verlag Company in 1987; "YUHKI KINZOKU KAGAKU—KISO TO OUYOU— (Metalorganic Chemistry—Fundamental and Application—)" authored by Akio Yamamoto, published by Shokabo Publishing Co., Ltd. in 1982, and the like.

**[0220]** The ligands are preferably nitrogen-containing heterocyclic ligands (having preferably 1 to 30 carbon atoms, more preferably 2 to 20 carbon atoms, and particularly preferably 3 to 15 carbon atoms); and they may be a unidentate ligand or a bi- or higher-dentate ligand. Preferable are bi- to hexa-dentate ligands, and mixed ligands of bi- to hexa-dentate ligands with a unidentate ligand are also preferable.

**[0221]** Examples of the ligands include azine ligands (e.g. pyridine ligands, bipyridyl ligands, terpyridine ligands and the like); hydroxyphenylazole ligands (e.g. hydroxyphenylbenzoimidazole ligands, hydroxyphenylbenzoxazole ligands, hydroxyphenylimidazole ligands, hydroxyphenylimidazopyridine ligands and the like); alkoxy ligands (those having preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, and particularly preferably 1 to 10 carbon atoms, examples of which include methoxy, ethoxy, butoxy, 2-ethylhexyloxy and the like); aryloxy ligands (those having preferably 6 to 30 carbon atoms, more preferably 6 to 20 carbon atoms, and particularly preferably 6 to 12 carbon atoms, examples of which include phenyloxy, 1-naphthyloxy,

2-naphthoxy, 2,4,6-trimethylphenoxy, 4-biphenyloxy and the like); heteroaryloxy ligands (those having preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, and particularly preferably 1 to 12 carbon atoms, examples of which include pyridyloxy, pyrazolyloxy, pyrimidyloxy, quinolyloxy and the like); alkylthio ligands (those having preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, and particularly preferably 1 to 12 carbon atoms, examples of which include methylthio, ethylthio and the like); arylthio ligands (those having preferably 6 to 30 carbon atoms, more preferably 6 to 20 carbon atoms, and particularly preferably 6 to 12 carbon atoms, examples of which include phenylthio and the like); heteroarylthio ligands (those having preferably 1 to 30 carbon atoms, more preferably 1 to 20 carbon atoms, and particularly preferably 1 to 12 carbon atoms, examples of which include pyridylthio, 2-benzimidazolylthio, benzoxazolylthio, 2-benzothiazolylthio and the like); siloxy ligands (those having preferably 1 to 30 carbon atoms, more preferably 3 to 25 carbon atoms, and particularly preferably 6 to 20 carbon atoms, examples of which include a triphenylsiloxy group, a triethoxysiloxy group, a triisopropylsiloxy group and the like); aromatic hydrocarbon anion ligands (those having preferably 6 to 30 carbon atoms, more preferably 6 to 25 carbon atoms, and particularly preferably 6 to 20 carbon atoms, examples of which include a phenyl anion, a naphthyl anion, an anthranyl anion and the like anion); aromatic heterocyclic anion ligands (those having preferably 1 to 30 carbon atoms, more preferably 2 to 25 carbon atoms, and particularly preferably 2 to 20 carbon atoms, examples of which include a pyrrole anion, a pyrazole anion, a triazole anion, an oxazole anion, a benzoxazole anion, a thiazole anion, a benzothiazole anion, a thiophene anion, a benzothiophene anion and the like); indolenine anion ligands and the like. Among these, nitrogen-containing heterocyclic ligands, aryloxy ligands, heteroaryloxy groups, aromatic hydrocarbon anion ligands, aromatic heterocyclic anion ligands or siloxy ligands are preferable, and nitrogen-containing heterocyclic ligands, aryloxy ligands, siloxy ligands, aromatic hydrocarbon anion ligands, or aromatic heterocyclic anion ligands are more preferable.

**[0222]** Examples of the metal complex electron transporting hosts include compounds described, for example, in Japanese Patent Application Laid-Open Nos. 2002-235076, 2004-214179, 2004-221062, 2004-221065, 2004-221068, 2004-327313 and the like.

**[0223]** In the light-emitting layer of the present invention, it is preferred that the lowest triplet excitation energy T1 of the host material is higher than T1 of the phosphorescence light-emitting material in view of color purity, light-emitting efficiency, and drive durability.

**[0224]** Although a content of the host compounds according to the present invention is not particularly limited, it is preferably 15% by weight to 95% by weight with respect to the total amount of the compounds forming the light-emitting layer in view of light emitting efficiency and drive voltage.

(Hole Injection Layer and Hole Transport Layer)

**[0225]** The hole injection layer and hole transport layer correspond to layers functioning to receive holes from an anode or from an anode side and to transport the holes to a cathode side. Materials to be introduced into a hole injection layer or a hole transport layer is not particularly limited, but either of a low molecular compound or a high molecular compound may be used.

**[0226]** As a material for the hole injection layer and the hole transport layer, it is preferred to contain specifically pyrrole derivatives, carbazole derivatives, imidazole derivatives, polyaryllalkane derivatives, pyrazoline derivatives, pyrazolone derivatives, phenylenediamine derivatives, arylamine derivatives, amino-substituted chalcone derivatives, styrylanthracene derivatives, fluorenone derivatives, hydrazone derivatives, stilbene derivatives, silazane derivatives, aromatic tertiary amine compounds, styrylamine compounds, aromatic dimethylidene compounds, phthalocyanine compounds, porphyrin compounds, thiophene derivatives, organosilane derivatives, carbon, or the like.

**[0227]** An electron-accepting dopant may be introduced into a hole injection layer or a hole transport layer in the organic electroluminescence element of the present invention. As the electron-accepting dopant to be introduced into a hole injection layer or a hole transport layer, either of an inorganic compound or an organic compound may be used as long as the compound has electron accepting property and a function for oxidizing an organic compound.

**[0228]** Specifically, the inorganic compound includes metal halides, such as ferric chloride, aluminum chloride, gallium chloride, indium chloride and antimony pentachloride and the like, and metal oxides, such as vanadium pentoxide, molybdenum trioxide and the like.

**[0229]** In case of the organic compounds, compounds having a substituent such as a nitro group, a halogen, a cyano group, a trifluoromethyl group or the like; quinone compounds; acid anhydride compounds; fullerenes; and the like may be preferably applied.

**[0230]** Specific examples thereof other than those above include compounds described in patent documents such as JP-A Nos. 6-212153, 11-111463, 11-251067, 2000-196140, 2000-286054, 2000-315580, 2001-102175, 2001-160493, 2002-252085, 2002-56985, 2003-157981, 2003-217862, 2003-229278, 2004-342614, 2005-72012, 2005-166637, 2005-209643 and the like.

**[0231]** Among these, hexacyanobutadiene, hexacyanobenzene, tetracyanoethylene, tetracyanoquinodimethane, tetrafluorotetracyanoquinodimethane, p-fluoranil, p-chloranil, p-bromanil, p-benzoquinone, 2,6-dichlorobenzoquinone, 2,5-dichlorobenzoquinone, 1,2,4,5-tetracyanobenzene, 1,4-dicyanotetrafluorobenzene, 2,3-dichloro-5,6-dicyanobenzoquinone, p-dinitrobenzene, m-dinitrobenzene, o-dinitrobenzene, 1,4-naphthoquinone, 2,3-dichloronaphthoquinone, 1,3-dinitronaphthalene, 1,5-dinitronaphthalene, 9,10-anthraquinone, 1,3,6,8-tetranitrocarbazole, 2,4,7-trinitro-9-fluorenone, 2,3,5,6-tetracyanopyridine, or fullerene C60 is preferable. Hexacyanobutadiene, hexacyanobenzene, tetracyanoethylene, tetracyanoquinodimethane, tetrafluorotetracyanoquinodimethane, p-fluoranil, p-chloranil, p-bromanil, 2,6-dichlorobenzoquinone, 2,5-dichlorobenzoquinone, 2,3-dichloronaphthoquinone, 1,2,4,5-tetracyanobenzene, 2,3-dichloro-5,6-dicyanobenzoquinone, or 2,3,5,6-tetracyanopyridine is more preferred, and tetrafluorotetracyanoquinodimethane is particularly preferred.

**[0232]** These electron-accepting dopants may be used alone or in a combination of two or more of them. Although an applied amount of these electron-accepting dopants depends on the type of material, 0.01% by weight to 50% by weight of a dopant is preferred with respect to a hole-transport layer

material, 0.05% by weight to 20% by weight is more preferable, and 0.1% by weight to 10% by weight is particularly preferred.

[0233] A thickness of the hole-injection layer and the hole-transport layer is preferably 500 nm or less in view of decrease in driving voltage.

[0234] A thickness of the hole-transport layer is preferably 1 nm to 500 nm, more preferably 5 nm to 200 nm, and 10 nm to 100 nm is particularly preferred. A thickness of the hole-injection layer is preferably 0.1 nm to 200 nm, more preferably 0.5 nm to 100 nm, and 1 nm to 100 nm is particularly preferred.

[0235] The hole-injection layer and the hole-transport layer may be composed of a monolayered structure comprising one or two or more of the above-mentioned materials, or a multilayer structure composed of plural layers of a homogeneous composition or heterogeneous compositions.

(Electron-Injection Layer and Electron-Transport Layer)

[0236] An electron-injection layer and an electron-transport layer are layers having any of functions for injecting electrons from the cathode, transporting electrons, and becoming a barrier to holes which could be injected from the anode. An electron injection material or an electron-transport material to be introduced therein is not particularly limited, but either of a low molecular compound or a high molecular compound may be used.

[0237] Specific examples of the materials include pyridine derivatives, quinoline derivatives, pyrimidine derivatives, pyrazine derivatives, phthalazine derivatives, phenanthroline derivatives, triazine derivatives, triazole derivatives, oxazole derivatives, oxadiazole derivatives, imidazole derivatives, fluorenone derivatives, anthraquinodimethane derivatives, anthrone derivatives, diphenylquinone derivatives, thiopyrandioxide derivatives, carbodiimide derivatives, fluorenylidene methane derivatives, distyrylpyridine derivatives, aryl tetracarboxylic anhydrides such as perylene or naphthalene, phthalocyanine derivatives, metal complexes represented by metal complexes of 8-quinolinol derivatives, metal phthalocyanine, and metal complexes containing benzoxazole, or benzothiazole as the ligand, organic silane derivatives represented by silole, and the like.

[0238] The electron-injection layer or the electron-transport layer may contain an electron donating dopant. As the electron donating dopant introduced in the electron-injection layer or the electron-transport layer, any material may be used as long as it has an electron-donating property and a property for reducing an organic compound, and alkaline metals such as Li, alkaline earth metals such as Mg, transition metals including rare-earth metals, and reducing organic compounds are preferably used. Particularly, metals having a work function of 4.2 V or less are preferably applied, and specific examples thereof include Li, Na, K, Be, Mg, Ca, Sr, Ba, Y, Cs, La, Sm, Gd, Yb, and the like. Specific examples of the reducing organic compounds include nitrogen-containing compounds, sulfur-containing compounds, phosphorus-containing compounds, and the like.

[0239] In addition, materials described in JP-A Nos. 6-212153, 2000-196140, 2003-68468, 2003-229278 and 2004-342614 may be used.

[0240] These electron donating dopants may be used alone or in a combination of two or more of them. An applied amount of the electron donating dopants differs dependent on the types of the materials, but it is preferably from 0.1% by

weight to 99% by weight with respect to an electron transport layer material, more preferably from 1.0% by weight to 80% by weight, and particularly preferably from 2.0% by weight to 70% by weight.

[0241] A thickness of the electron-injection layer and the electron-transport layer is preferably 500 nm or less in view of decrease in driving voltage.

[0242] A thickness of the electron-transport layer is preferably 1 nm to 500 nm, more preferably 5 nm to 200 nm, and 10 nm to 100 nm is particularly preferred. A thickness of the electron-injection layer is preferably 0.1 nm to 200 nm, more preferably 0.2 nm to 100 nm, and 0.5 nm to 100 nm is particularly preferred.

[0243] The electron-injection layer and the electron-transport may be composed of a monolayered structure comprising one or two or more of the above-mentioned materials, or a multilayer structure composed of plural layers of a homogeneous composition or heterogeneous compositions.

(Hole Blocking Layer)

[0244] A hole blocking layer is a layer having a function to prevent the holes transported from the anode side to the light-emitting layer from passing through to the cathode side. According to the present invention, a hole blocking layer may be provided as an organic compound layer adjacent to the light-emitting layer on the cathode side.

[0245] Examples of the compound constituting the hole blocking layer include an aluminum complex such as BA1q, a triazole derivative, a phenanthroline derivative such as BCP, or the like.

[0246] A thickness of the hole blocking layer is preferably from 1 nm to 500 nm, more preferably from 5 nm to 200 nm, and even more preferably from 10 nm to 100 nm.

[0247] The hole blocking layer may have either a monolayer structure comprising one or two or more of the above-mentioned materials, or a multilayer structure composed of plural layers of a homogeneous composition or a heterogeneous composition.

(Electron Blocking Layer)

[0248] An electron blocking layer is a layer having a function to prevent the electron transported from the cathode side to the light-emitting layer from passing through to the anode side. According to the present invention, an electron blocking layer may be provided as an organic compound layer adjacent to the light-emitting layer on the anode side.

[0249] Specific examples of the compound constituting the electron blocking layer include compounds explained above as a hole-transporting material.

[0250] A thickness of the electron blocking layer is preferably from 1 nm to 500 nm, more preferably from 5 nm to 200 nm, and even more preferably from 10 nm to 100 nm.

[0251] The electron blocking layer may have either a monolayer structure comprising one or two or more of the above-mentioned materials, or a multilayer structure composed of plural layers of a homogeneous composition or a heterogeneous composition.

(Protective Layer)

[0252] According to the present invention, the whole organic EL element may be protected by a protective layer.

[0253] A material contained in the protective layer may be one having a function to prevent penetration of substances

such as moisture and oxygen, which accelerate deterioration of the element, into the element.

[0254] Specific examples thereof include metals such as In, Sn, Pb, Au, Cu, Ag, Al, Ti, Ni and the like; metal oxides such as MgO, SiO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, GeO, NiO, CaO, BaO, Fe<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and the like; metal nitrides such as SiN<sub>x</sub>, SiN<sub>x</sub>O<sub>y</sub>, and the like; metal fluorides such as MgF<sub>2</sub>, LiF, AlF<sub>3</sub>, CaF<sub>2</sub> and the like; polyethylene; polypropylene; polymethylmethacrylate; polyimide; polyurea; polytetrafluoroethylene; polychlorotrifluoroethylene; polydichlorodifluoroethylene; a copolymer of chlorotrifluoroethylene and dichlorodifluoroethylene; copolymers obtained by copolymerizing a monomer mixture containing tetrafluoroethylene and at least one comonomer; fluorine-containing copolymers each having a cyclic structure in the copolymerization main chain; water-absorbing materials each having a coefficient of water absorption of 1% or more; moisture permeation preventive substances each having a coefficient of water absorption of 0.1% or less; and the like.

[0255] There is no particular limitation as to a method for forming the protective layer. For instance, a vacuum deposition method, a sputtering method, a reactive sputtering method, an MBE (molecular beam epitaxial) method, a cluster ion beam method, an ion plating method, a plasma polymerization method (high-frequency excitation ion plating method), a plasma CVD method, a laser CVD method, a thermal CVD method, a gas source CVD method, a coating method, a printing method, or a transfer method may be applied.

(Sealing)

[0256] The whole organic electroluminescence element of the present invention may be sealed with a sealing cap.

[0257] Furthermore, a moisture absorbent or an inert liquid may be used to seal a space defined between the sealing cap and the light-emitting element.

[0258] Although the moisture absorbent is not particularly limited, specific examples thereof include barium oxide, sodium oxide, potassium oxide, calcium oxide, sodium sulfate, calcium sulfate, magnesium sulfate, phosphorus pentoxide, calcium chloride, magnesium chloride, copper chloride, cesium fluoride, niobium fluoride, calcium bromide, vanadium bromide, molecular sieve, zeolite, magnesium oxide and the like. Although the inert liquid is not particularly limited, specific examples thereof include paraffins; liquid paraffins; fluorine-based solvents such as perfluoroalkanes, perfluoroamines, perfluoroethers and the like; chlorine-based solvents; silicone oils; and the like.

[0259] A sealing method by a resin sealing layer is also preferably applied.

[0260] (Resin Sealing Layer)

[0261] A functional element of the present invention is preferably prevented by the resin sealing layer from an intrusion of water content or oxygen to result in deterioration in performances of the element.

[0262] <Material>

[0263] The resin material for the resin seal layer is not particularly restricted and an acrylic resin, epoxy resin, a fluorocarbon resin, a silicone resin, a rubber resin, or an ester resins can be used. Among them, the epoxy resin is preferred with a view point of water content preventive function. In the epoxy resin, thermosetting epoxy resin, or photo-curable epoxy resin is preferred.

[0264] <Manufacturing Method>

[0265] The manufacturing method of the resin seal layer is not particularly restricted and includes, for example, a method of coating a resin solution, a method of press bonding or hot press bonding a resin sheet or a method of dry polymerization by vapor deposition or sputtering, etc.

[0266] <Film Thickness>

[0267] The thickness of the resin seal layer is 1 μm or more and, preferably, 1 mm or less. It is more preferably 5 μm or more and 100 μm or less and, most preferably, 10 μm or more and 50 μm or less. In a case where the thickness is smaller, the inorganic film may possibly be damaged upon mounting of the second substrate. Further, in a case where the thickness is larger, the thickness of the electroluminescence device per se increases to damage the thin film property as a feature of the organic electroluminescence device.

[0268] (Sealing Adhesive)

[0269] The sealing adhesive used in the invention has a function of preventing intrusion of water content or oxygen from the edge.

[0270] <Material>

[0271] As the material for the sealing adhesive, those identical with the materials used in the resin sealing layer can be used. Among all, an epoxy type adhesive is preferred with a view point of preventing water content and, particularly, a photo-curable epoxy type adhesive is preferred.

[0272] Further, addition of a filler to the materials described above is also preferred. The filler added to the sealant is preferably inorganic materials such as SiO<sub>2</sub>, SiO (silicon oxide), SiON (silicon oxynitride), or SiN (silicon nitride). The addition of the filler increases the viscosity of the sealant to improve the producibility and improve the humidity resistance.

[0273] <Drying Agent>

[0274] The sealing adhesive may also contain a drying agent, the drying agent is preferably barium oxide, calcium oxide, or strontium oxide.

[0275] The addition amount of the drying agent to the sealing adhesive is, preferably, 0.01% by weight or more and 20% by weight or less and, more preferably, 0.05% by weight or more and 15% by weight or less. The addition effect of the drying agent is reduced in a case where the amount is smaller. Further, it is difficult to uniformly disperse the drying agent in the sealing adhesive in a case where the amount is larger, which is not preferred.

[0276] <Formulation of Sealing Adhesive>

[0277] Polymer Composition, Concentration

[0278] The sealing adhesive is not particularly restricted and those described above can be used. For example, the photo-curable epoxy adhesive includes XNR5516 manufactured by Nagase Chemtech Co. and the drying agent may be added to and dispersed therein.

[0279] Thickness

[0280] The coating thickness of the sealing adhesive is preferably 1 μm or more and 1 mm or less. In a case where the thickness is smaller, the sealing adhesive can not be coated uniformly, which is not preferred. Further, in a case where the thickness is larger, water content intrusion paths are increased, which is not preferred.

[0281] <Sealing Method>

[0282] In the invention, the sealing adhesive incorporated with the drying agent is coated in an optional amount by a

dispenser or the like, a second substrate is stacked after coating, and they can be cured to obtain a functional device.

(Driving)

**[0283]** In the organic EL element of the present invention, when a DC (AC components may be contained as needed) voltage (usually 2 volts to 15 volts) or DC is applied across the anode and the cathode, luminescence can be obtained.

**[0284]** For the driving method of the organic EL element of the present invention, driving methods described in JP-A Nos. 2-148687, 6-301355, 5-29080, 7-134558, 8-234685, and 8-241047; Japanese Patent No. 2784615, U.S. Pat. Nos. 5,828,429 and 6,023,308 are applicable.

**[0285]** In the organic EL element of the present invention, the light-extraction efficiency can be improved by various known methods. It is possible to elevate the light-extraction efficiency and to improve the external quantum efficiency, for example, by modifying the surface shape of the substrate (for example by forming fine irregularity pattern), by controlling the refractive index of the substrate, the ITO layer and/or the organic layer, or by controlling the thickness of the substrate, the ITO layer and/or the organic layer.

**[0286]** The organic EL element of the present invention may have a so-called top-emission configuration in which the light emission is extracted from the anode side.

**[0287]** The organic EL element of the present invention may have a configuration of having an electric charge-generating layer provided between a plurality of the light-emitting layers for a purpose to enhance luminescent efficiency.

**[0288]** The electric charge-generating layer has a function of generating electric charges (holes or electrons) during an application of an electric field as well as a function of injecting the generated electric charges into a layer adjacent to the electric charge-generating layer.

**[0289]** The electric charge-generating layer is formed by any material as long as it satisfies for the aforementioned functions and may be formed by a single compound or a plurality of compounds.

**[0290]** Specific examples of the materials for the electric charge-generating layer include electrically conductive materials, semi-conductive materials such as doped organic compounds, electric insulating materials, and materials disclosed in JP-A Nos. 11-329748, 2003-272860 and 2004-39617.

**[0291]** More specific examples thereof include transparent electrically conductive materials such as indium tin oxide (ITO) and indium zinc oxide (IZO); fullerenes such as C<sub>60</sub>; electrically conductive organic substances such as thiophene oligomers; electrically conductive organic substances such as metal phthalocyanines, non-metal phthalocyanines, metal porphyrins and non-metal porphyrins; metal materials such as Ca, Ag, Al, Mg—Ag alloy and Al—Li alloy; hole conductive materials; electrically conductive materials, and mixtures thereof.

**[0292]** Specific examples of the hole conductive material include hole transfer organic materials such as 2-TNATA or NPD doped with oxidants having electron attracting properties such as F4-TCNQ, TCNQ, or FeCl<sub>3</sub>, P-type electrically conductive polymers, and P-type semiconductors. Specific examples of the electrically conductive materials include the electron transport organic materials doped with metals or metal compounds having a work function of less than 4.0 eV, N-type electrically conductive polymers, and an N-type semiconductors. Specific examples of the N-type semiconductors include N-type Si, N-type CdS, N-type ZnS and the like.

Specific examples of the P-type semiconductors include P-type Si, P-type CdTe, P-type CuO and the like.

**[0293]** Further, the electric charge-generating layer may use an electric insulating material such as V<sub>2</sub>O<sub>5</sub>.

**[0294]** The electric charge-generating layer can be formed by a single layer or a lamination of a plurality of layers. Specific examples of the laminations of the plurality of layers include a lamination of an electrically conductive material such as a transparent conductive material or a metal material and a hole conductive material, a lamination of electrically conductive materials, and a lamination of the hole conductive material and the electrically conductive material described above, and the like.

**[0295]** Preferably, a film thickness or a material in the electric charge-generating layer can be selected such that a transmittance of visible light has 50% or more. Further, the film thickness is not particularly limited; however, it is preferably 0.5 to 200 nm, more preferably 1 to 100 nm, further preferably 3 to 50 nm and most preferably 5 to 30 nm.

**[0296]** The method of preparing the electric charge-generating layer is not particularly limited, and the above-described method of preparing organic compound layers can also be used.

**[0297]** The electric charge-generating layer is formed between the two or more light-emitting layers. However, a material having a function of injecting an electric charge into layers adjacent thereto may be contained in a region of an anode side or in a cathode side of the electric charge-generating layer. In order to increase injection properties of electrons into layers adjacent at the anode side thereof, electron injection compounds such as BaO, SrO, Li<sub>2</sub>O, LiCl, LiF, MgF<sub>2</sub>, MgO, and CaF<sub>2</sub> may be laminated on the electric charge-generating layer at the anode side thereof.

**[0298]** Other than the materials according to the contents herein, materials for forming the electron charge generating layer may be selected on the basis of the descriptions in JP-A No. 2003-45676, and U.S. Pat. Nos. 6,337,492, 6,107,734, and 6872472.

**[0299]** The organic EL element in the invention preferably may have a resonator structure.

**[0300]** For example, on a transparent substrate, a multi-layered film mirror comprising a plurality of stacked films of different reflective indexes, a transparent or semi-transparent electrode, a light-emitting layer, and a metal electrode stacked to each other. The light generated in the light-emitting layer repeats reflection and conducts oscillation between the multi-layered film mirror and the metal electrode as reflection plates.

**[0301]** In another preferred embodiment of the resonator structure, a transparent or semi-transparent electrode and a metal electrode function respectively as reflection plates on a transparent substrate in which light generated in the light-emitting layer repeats reflection and conducts oscillation therebetween.

**[0302]** For forming the resonance structure, an optical channel length determined based on the effective refractive index of two reflection plates, and the refractive index and the thickness for each of the layers between the reflection plates are controlled to optimal values for obtaining a desired resonance wavelength.

**[0303]** A calculation formula in a case of the first embodiment is described in the specification of JP-A No. 9-180883

and the calculation formula in a case of the second embodiment is described in the specification of JP-A No. 2004-127795.

[0304] As a method for forming a full color-type organic EL display, there are known, for example, as described in *Monthly Display*, September 2000, pages 33 to 37, a tricolor light emission method which arranges organic EL elements emitting light corresponding to three primary colors (blue color (B), green color (G), and red color (R)) on a substrate respectively; a white color method which separates white light emitted by an organic EL element for white color emission into three primary colors through a color filter; and a color conversion method which converts a blue light emitted by an organic EL element for blue light emission into red color (R), and green color (G) through a fluorescent dye layer.

[0305] Further, by combining a plurality of organic EL elements of different light emission colors obtained by the methods described above, planar type light sources of desired emission colors can be obtained. For example, they include a white color light source obtained by combination of blue color and yellow color light emission elements, and a white color light source obtained by combination of blue, green and red light emitting elements.

#### 4. Pixel-Circuit of Organic EL Display

[0306] FIG. 5 is a schematic diagram of a pixel-circuit of an active matrix type organic EL display which uses TFT devices according to the invention. In FIG. 5, an organic EL element 81, a drive TFT 83, a switching TFT 84, and a capacitor 85 are wired with a scanning wire 88, signal wire 87, and common wire 86.

[0307] The circuit of the display according to the invention is not particularly limited to that shown in FIG. 5. A circuit which is conventionally known in the art may be applied as-is.

[0308] (Applications)

[0309] An organic EL display according to the invention has such wide ranging applications as a mobile phone display, a personal digital assistant (PDA), a computer display, a car information display, a TV monitor, and general illumination.

[0310] All publications, patent applications, and technical standards mentioned in this specification are herein incorporated by reference to the same extent as if each individual publication, patent application, or technical standard was specifically and individually indicated to be incorporated by reference.

#### EXAMPLES

[0311] In the following, the organic EL display of the invention will be described based on the examples, but it should be noted that the invention is not limited to these examples.

##### Example 1

#### 1. Preparation of Organic EL Display

##### 1-1. Preparation of Organic EL Display No. 1

[0312] An organic EL display having a configuration shown in FIG. 1 was prepared.

[0313] (Preparation of Organic EL Element Part)

[0314] 1) Formation of Lower Electrode

[0315] On a glass substrate (#1737, manufactured by Corning), indium tin oxide (which is referred to hereinafter as ITO) was deposited at a thickness of 150 nm to form an anode.

[0316] 2) Formation of Organic Layer

[0317] After cleaning, a hole injection layer, a hole transport layer, a light-emitting layer, a hole-blocking layer, an electron transport layer and an electron injection layer were disposed in this order.

[0318] The composition of each layer is as follows. Each layer was provided by resistance heating vacuum deposition.

[0319] Hole injection layer: a layer containing 4,4'-tris(2-naphthylphenylamino)triphenylamine (which is referred to as 2-TNATA) and 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (which is referred to as F4-TCNQ), wherein an amount of F4-TCNQ was 1% by weight with respect to 2-TNATA; at a thickness of 160 nm.

[0320] Hole transport layer: N,N'-dinaphthyl-N,N'-diphenyl-[1,1'-biphenyl]-4,4'-diamine (which is referred to as  $\alpha$ -NPD), at a thickness of 10 nm.

[0321] Light-emitting layer: a layer containing 1,3-bis(carbazol-9-yl)benzene (which is referred to as mCP) and platinum complex Pt-1 in an amount of 13% by weight with respect to mCP, at a thickness of 60 nm.

[0322] Hole-blocking layer: aluminium (III) bis(2-methyl-8-quinolinato)-4-phenylphenolate (which is referred to as BAq), at a thickness of 40 nm.

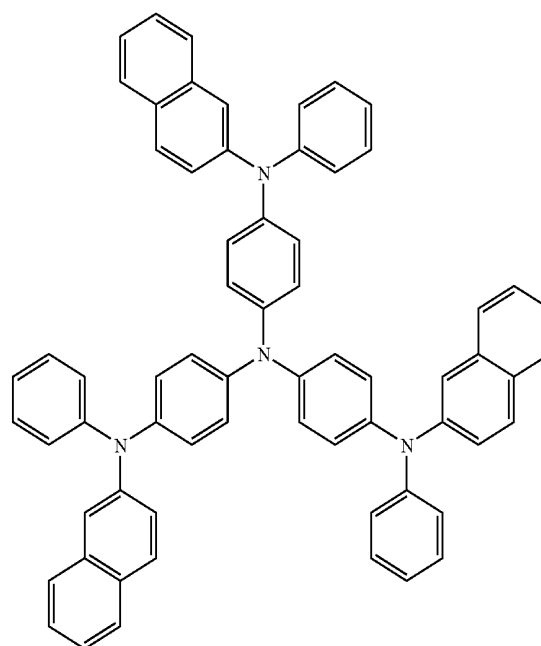
[0323] Electron transport layer: tris(8-hydroxyquinolato)aluminum (which is referred to as Alq3), at a thickness of 10 nm.

[0324] Electron injection layer: lithium fluoride (LiF), a thickness of 1 nm.

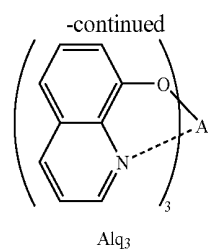
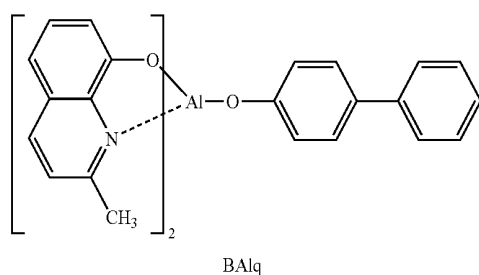
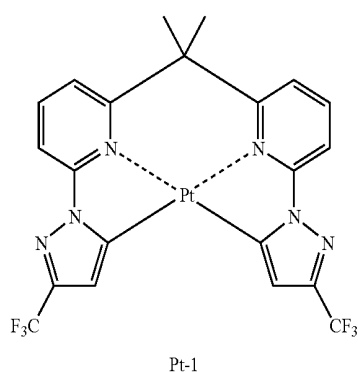
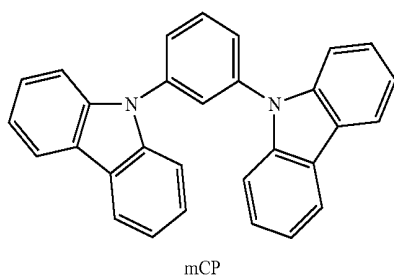
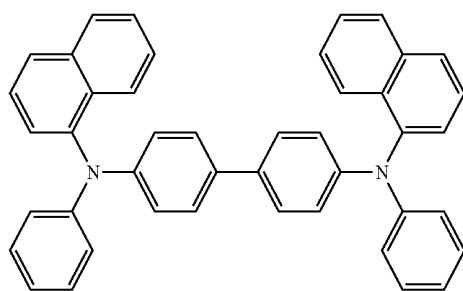
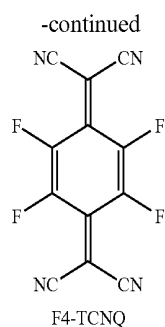
[0325] 3) Upper Electrode

[0326] Patterning was performed using a shadow mask so that the size of the element became 1 mm $\times$ 1 mm, and aluminum metal (Al) was deposited at a thickness of 100 nm to form a cathode.

[0327] Chemical structures of the compounds used in Examples are shown below.



2-TNATA



[0328] (Electrically Conductive Etching Protective Layer)

[0329] On the upper electrode, an SnO<sub>2</sub> layer with a thickness of 150 nm was formed as an electrically conductive etching protective layer by RF magnetron sputtering vacuum deposition. Patterning was performed using a shadow mask, and thereby, an electrically conductive etching protective layer having a size of 0.5 mm×0.5 mm was formed on the aluminum (Al) of cathode.

[0330] (Protective Insulating Layer)

[0331] An SiON layer with a thickness of 500 nm was formed as a protective insulating layer by an ion plating method.

[0332] (Formation of Contact Hole)

[0333] On the protective insulating layer, resist was coated and then, mask exposure was conducted so that resist was formed at the place other than the place where a contact hole was to be formed.

[0334] After forming the resist, the protective insulating layer was subjected to etching using buffered hydrofluoric acid to form a contact hole. Thereafter, the resist was removed.

[0335] (Preparation of TFT Part)

[0336] 1) Source Electrode and Drain Electrode

[0337] As a source electrode and a drain electrode, molybdenum (Mo) with a thickness of 40 nm and ITO with a thickness of 40 nm were deposited by RF magnetron sputtering vacuum deposition. Patterning of the source electrode and drain electrode was performed using a lift-off method. In this process, a gap between the source electrode and the drain electrode was formed to give a channel length (L) of 50  $\mu$ m and a channel width (W) of 250  $\mu$ m. The electrodes were formed to have a configuration in which the drain electrode and the electrically conductive etching protective layer connected with the upper electrode (cathode) of the organic EL element are electrically connected through the contact hole.

[0338] 2) Active Layer

[0339] Using a polycrystalline sintered body having a composition of InGaZnO<sub>4</sub> as a target, an RF magnetron sputtering vacuum deposition method was performed to form a deposition layer of IGZO at a thickness of 50 nm. The electric conductivity of the active layer was  $5.7 \times 10^{-3} \text{ Scm}^{-1}$ . Patterning was performed using a shadow mask.

[0340] 3) Gate Insulating Layer

[0341] A gate insulating layer was provided by performing RF magnetron sputtering vacuum deposition of SiO<sub>2</sub> at a thickness of 200 nm. Patterning was performed using a shadow mask.

[0342] 4) Gate Electrode

[0343] A gate electrode was provided by performing deposition of Mo at a thickness of 100 nm. Patterning was performed by a photolithography etching method.

1-2. Preparation of Organic EL Display A for Comparison

[0344] Preparation of Organic EL Display a for Comparison was Conducted in a similar manner to the process in the

preparation of the organic EL display No. 1, except that the electrically conductive etching protective layer was removed.

### 1-3. Preparation of Organic EL Display B

**[0345]** Preparation of Organic EL Display B was Conducted in a Similar Manner to the process in the preparation of the organic EL display No. 1, except that the active layer of the TFT was changed to a layer provided by performing deposition of pentacene, which is an organic semiconductor, to give a thickness of 50 nm.

### 2. Performance Evaluation

**[0346]** The gate voltage and source/drain voltage of the driving TFT were controlled so that an electric current of 1.0 mA/cm<sup>2</sup> was applied to the organic EL element portion of the organic EL display No. 1 and the organic EL display A for comparison. As a result, the organic EL display No. 1 and the organic EL display A for comparison both exhibited an excellent blue-light emission. Concerning the organic EL display No. 1, an emission brightness of 160 cd/m<sup>2</sup> was obtained. However, concerning the organic EL display A for comparison, only an emission brightness of 85 cd/m<sup>2</sup> was obtained. As for the organic EL display B, an emission brightness of 150 cd/m<sup>2</sup>, which is similar to the case of the organic EL display No. 1, was obtained.

### Example 2

#### 1. Preparation of Organic EL Display No. 2

**[0347]** Preparation of organic EL display No. 2 of the invention was conducted in a similar manner to the process in the preparation of the organic EL display No. 1 of Example 1, except that the active layer was changed to have a bilayer configuration including an active layer and an electric resistance layer disclosed below. The layer closer to the source electrode and the drain electrode is positioned to be the electric resistance layer, and the layer closer to the gate insulating layer is positioned to be the active layer.

**[0348]** Electric resistance layer: IGZO was deposited to give a thickness of 40 nm by an RF magnetron sputtering vacuum deposition method. The flow rates of argon (Ar) and oxygen (O<sub>2</sub>) were controlled, and the electric conductivity of the electric resistance layer was 1.0×10<sup>-4</sup> Scm<sup>-1</sup>.

**[0349]** Active layer: IGZO was deposited to give a thickness of 10 nm by an RF magnetron sputtering vacuum deposition method. The flow rates of Ar and O<sub>2</sub> were controlled, and the electric conductivity of the active layer was 2.6×10<sup>-1</sup> Scm<sup>-1</sup>.

### 2. Performance Evaluation

**[0350]** Similar to Example 1, when the gate voltage and source/drain voltage of the driving TFT were controlled so that an electric current of 1.0 mA/cm<sup>2</sup> was applied to the organic EL element portion, the organic EL display No. 2 gave an emission brightness of 160 cd/m<sup>2</sup>, that is similar to the organic EL display No. 1. When 14 volts was applied to the gate electrode of the driving TFT and 20 volts was applied to the anode of the organic EL element, an emission brightness of 230 cd/m<sup>2</sup> was obtained, which is unexpectedly remarkably higher than the brightness of 175 cd/m<sup>2</sup> obtained by the organic EL display No. 1.

### Example 3

#### 1. Preparation of Organic EL Display No. 3

**[0351]** Preparation of organic EL display No. 3 of the invention was conducted in a similar manner to the process in the preparation of the organic EL display No. 2 of Example 2, except that the glass substrate was changed to a PEN substrate which has 40 nm-thick SiON as a barrier layer on both sides thereof.

### 2. Performance Evaluation

**[0352]** When the gate voltage and source/drain voltage of the driving TFT were controlled so that an electric current of 1.0 mA/cm<sup>2</sup> was applied to the organic EL element portion, the organic EL display No. 3 gave an emission brightness of 150 cd/m<sup>2</sup>. When 14 volts was applied to the gate electrode of the driving TFT and 20 volts was applied to the anode of the organic EL element, an emission brightness of 210 cd/cm<sup>2</sup> was obtained. Even on a flexible substrate, an emission brightness similar to that obtained by the organic EL display No. 2 was obtained.

### Example 4

#### 1. Preparation of Organic EL Display Nos. 4 to 6

**[0353]** Preparation of organic EL display Nos. 4-6 was Conducted in a similar manner to the process in the preparation of the organic EL display Nos. 1-3 of Example Nos. 1-3, except that the electrically conductive etching protective layer was changed to the following composition.

**[0354]** (Electrically Conductive Etching Protective Layer)

**[0355]** An ITO layer with a thickness of 150 nm was formed as an electrically conductive etching protective layer by an ion-plating method. Patterning was performed using a shadow mask, and thereby, an electrically conductive etching protective layer having a size of 0.5 mm×0.5 mm was formed on the cathode Al.

### 2. Performance Evaluation

**[0356]** When the gate voltage and source/drain voltage of the driving TFT were controlled so that an electric current of 1.0 mA/cm<sup>2</sup> was applied to the organic EL element portion, both the organic EL display Nos. 4 and 5 gave an emission brightness of 150 cd/m<sup>2</sup>, and the organic EL display No. 6 gave an emission brightness of 130 cd/m<sup>2</sup>. These brightness values obtained were higher than that of the organic EL display A for comparison.

Reference numerals used in Figures of the invention are explained below.

- [0357]** 1, 11, 100, 110: Substrate
- [0358]** 6, 16: Insulating layer
- [0359]** 2, 12, 102, 112: Gate electrode
- [0360]** 3, 13, 103, 113: Gate insulating layer
- [0361]** 4, 104, 114: Active layer
- [0362]** 4-2, 4-12: Electric resistance layer
- [0363]** 4-1, 4-11: Active layer
- [0364]** 5-1, 5-11, 105a, 115a: Source electrode
- [0365]** 5-2, 5-12, 105b, 115b: Drain electrode
- [0366]** 30, 40: Lower electrode
- [0367]** 32, 42: Organic layer
- [0368]** 34, 44: Upper electrode
- [0369]** 106, 116: Protective insulating layer
- [0370]** 36, 46: Insulating film

- [0371] 108, 118: Contact hole  
 [0372] 109: Electrically conductive etching protective layer  
 [0373] 81: Organic EL element  
 [0374] 82: Cathode  
 [0375] 83: Driving TFT  
 [0376] 84: Switching TFT  
 [0377] 85: Capacitor  
 [0378] 86: Common wire  
 [0379] 87: Signal wire  
 [0380] 88: Scanning wire

What is claimed is:

1. An organic EL display comprising:
  - an organic EL element comprising at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode, in this order, on a substrate; and
  - a thin film field-effect transistor which drives the organic EL element and is formed on the organic EL element, wherein an electrically conductive etching protective layer which is electrically connected to the upper electrode is disposed between the upper electrode and the thin film field-effect transistor, a protective insulating layer is disposed between the electrically conductive etching protective layer and the thin film field-effect transistor, and a source electrode or a drain electrode of the thin film field-effect transistor and the electrically conductive etching protective layer are electrically connected through a contact hole formed in the protective insulating layer.
2. The organic EL display according to claim 1, wherein the contact hole is formed by etching with an etching material, in which an etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the upper electrode by the etching material.
3. The organic EL display according to claim 1, wherein the contact hole is formed by etching with an etching material, in which an etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the protective insulating layer by the etching material.
4. The organic EL display according to claim 1, wherein the upper electrode is formed by an electric resistance heating deposition method, and the electrically conductive etching protective layer is formed by a sputtering method, an ion plating method or a chemical vapor deposition method (CVD method).
5. The organic EL display according to claim 1, wherein an active layer of the thin film field-effect transistor comprises a semiconductor material selected from the group consisting of an oxide semiconductor, an organic semiconductor and a carbon nanotube.
6. The organic EL display according to claim 5, wherein the active layer of the thin film field-effect transistor comprises an oxide semiconductor.
7. The organic EL display according to claim 6, wherein the oxide semiconductor is an amorphous oxide semiconductor.
8. The organic EL display according to claim 1, wherein the lower electrode is a light transmitting electrode.
9. The organic EL display according to claim 8, wherein the upper electrode is a light reflective electrode.
10. The organic EL display according to claim 1, wherein the lower electrode is an anode, and the upper electrode is a cathode.
11. The organic EL display according to claim 10, wherein the thin film field-effect transistor has n-type polarity.
12. The organic EL display according to claim 1, wherein an electric resistance layer is electrically connected between an active layer of the thin film field-effect transistor and at least one of the source electrode or the drain electrode.
13. The organic EL display according to claim 12, wherein the electric resistance layer and the active layer are layered, the active layer is in contact with a gate insulating layer, and the electric resistance layer is in contact with at least one of the source electrode or the drain electrode.
14. The organic EL display according to claim 13, wherein the electric resistance layer is thicker than the active layer.
15. The organic EL display according to claim 12, wherein an electric conductivity changes continuously between the electric resistance layer and the active layer.
16. The organic EL display according to claim 12, wherein the active layer and the electric resistance layer comprise an oxide semiconductor.
17. The organic EL display according to claim 16, wherein the oxide semiconductor comprises at least one selected from the group consisting of In, Ga and Zn, or a composite oxide thereof.
18. The organic EL display according to claim 17, wherein the oxide semiconductor comprises In and Zn, and a composition ratio between Zn and In in the electric resistance layer (represented by a ratio of Zn to In, Zn/In) is larger than a composition ratio Zn/In in the active layer.
19. The organic EL display according to claim 16, wherein the active layer has an oxygen concentration that is lower than that of the electric resistance layer.
20. The organic EL display according to claim 12, wherein the active layer has an electric conductivity of  $10^{-4}$  Scm<sup>-1</sup> or more, and less than  $10^2$  Scm<sup>-1</sup>.
21. The organic EL display according to claim 20, wherein the active layer has an electric conductivity of  $10^{-1}$  Scm<sup>-1</sup> or more, and less than  $10^2$  Scm<sup>-1</sup>.
22. The organic EL display according to claim 12, wherein a ratio of an electric conductivity of the active layer to an electric conductivity of the electric resistance layer (electric conductivity of active layer/electric conductivity of electric resistance layer) is from  $10^1$  to  $10^{10}$ .
23. The organic EL display according to claim 22, wherein the ratio of an electric conductivity of the active layer to an electric conductivity of the electric resistance layer (electric conductivity of active layer/electric conductivity of electric resistance layer) is from  $10^2$  to  $10^8$ .
24. The organic EL display according to claim 1, wherein the substrate is a flexible resin substrate.
25. A method for producing an organic EL display, comprising at least:
  - forming an organic EL element part by sequentially forming at least a lower electrode, an organic layer comprising at least a light-emitting layer, and an upper electrode on a substrate;
  - forming an electrically conductive etching protective layer that is electrically connected to the upper electrode;
  - forming a protective insulating layer on the electrically conductive etching protective layer;

forming a contact hole in the protective insulating layer by causing an etching material to act; and forming a thin film field-effect transistor on the protective insulating layer,

wherein a source electrode or a drain electrode of the thin film field-effect transistor and the electrically conductive etching protective layer are electrically connected through the contact hole.

**26.** The method for producing an organic EL display according to claim **25**, wherein an etching rate of the electrically conductive etching protective layer by the etching material is slower than an etching rate of the upper electrode by the etching material.

**27.** The method for producing an organic EL display according to claim **25**, wherein an etching rate of the electrically conductive etching protective layer by the etching material is slower than all etching rate of the protective insulating layer by the etching material.

**28.** The method for producing an organic EL display according to claim **25**, wherein the upper electrode is formed by an electric resistance heating deposition method, and the electrically conductive etching protective layer is formed by a sputtering method, an ion plating method or a chemical vapor deposition method (CVD method).

\* \* \* \* \*

专利名称(译)	有机EL显示器及其制造方法		
公开(公告)号	<a href="#">US20090001360A1</a>	公开(公告)日	2009-01-01
申请号	US12/146355	申请日	2008-06-25
[标]申请(专利权)人(译)	NAKAYAMA MASAYA		
申请(专利权)人(译)	NAKAYAMA MASAYA		
当前申请(专利权)人(译)	NAKAYAMA MASAYA		
[标]发明人	NAKAYAMA MASAYA		
发明人	NAKAYAMA, MASAYA		
IPC分类号	H01L51/05 H01L27/12 H01L21/84		
CPC分类号	H01L27/1225 H01L27/3248 H01L27/1248 H01L27/3262 H01L27/124 H01L27/3258		
优先权	2007172448 2007-06-29 JP 2008142748 2008-05-30 JP		
外部链接	<a href="#">Espacenet</a> <a href="#">USPTO</a>		

摘要(译)

本发明提供一种有机电致发光显示器，其具有有机EL元件和形成在有机EL元件上的薄膜场效应晶体管，其中电连接到上电极的导电蚀刻保护层设置在上电极和上电极之间。薄膜场效应晶体管，保护绝缘层设置在导电蚀刻保护层和薄膜场效应晶体管之间，以及薄膜场效应晶体管的源电极或漏电极和导电蚀刻保护层通过形成在保护绝缘层中的接触孔电连接；及其制备方法。

