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(12) **United States Patent**  
**Watabe et al.**

(10) **Patent No.:** US 12,389,739 B2  
(45) **Date of Patent:** Aug. 12, 2025

(54) **LIGHT-EMITTING DEVICE, DISPLAY DEVICE, LIGHT-EMITTING APPARATUS, ELECTRONIC DEVICE, AND LIGHTING DEVICE**

(71) Applicant: **SEMICONDUCTOR ENERGY LABORATORY CO., LTD.**, Atsugi (JP)

(72) Inventors: **Takeyoshi Watabe**, Kanagawa (JP); **Airi Ueda**, Kanagawa (JP); **Yuta Kawano**, Kanagawa (JP); **Nobuharu Ohsawa**, Kanagawa (JP); **Satoshi Seo**, Kanagawa (JP)

(73) Assignee: **Semiconductor Energy Laboratory Co., Ltd.**, Atsugi (JP)

(\*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 774 days.

(21) Appl. No.: 17/672,799

(22) Filed: Feb. 16, 2022

(65) **Prior Publication Data**

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(30) **Foreign Application Priority Data**

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May 10, 2021 (JP) ..... 2021-079545  
Oct. 18, 2021 (JP) ..... 2021-170317

(51) **Int. Cl.**

H10K 50/11 (2023.01)  
H10K 50/13 (2023.01)

(Continued)

(52) **U.S. Cl.**

CPC ..... H10K 50/11 (2023.02); H10K 50/13 (2023.02); H10K 50/818 (2023.02); H10K 85/00 (2023.02);

(Continued)

(58) **Field of Classification Search**

CPC ..... H10K 50/13; H10K 50/11; H10K 50/818; H10K 2101/40; H10K 59/80518;  
(Continued)

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(Continued)

Primary Examiner — Jay C Chang

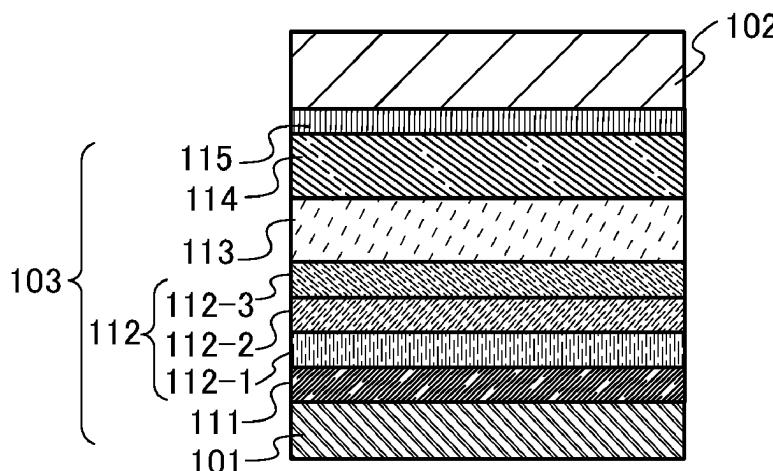
Assistant Examiner — Thaddeus J Kolb

(74) Attorney, Agent, or Firm — Fish & Richardson P.C.

(57) **ABSTRACT**

A light-emitting device with high emission efficiency is provided. The light-emitting device includes a first electrode, a second electrode, and an EL layer positioned between the first electrode and the second electrode. The EL layer includes at least a light-emitting layer, a first layer, a second layer, and a third layer. The first layer is positioned between the first electrode and the light-emitting layer. The third layer is positioned between the first layer and the light-emitting layer. The second layer is positioned between the first layer and the third layer. The first layer includes a first organic compound. The second layer includes a second organic compound. The third layer includes a third organic

(Continued)



compound. The ordinary refractive index of the second organic compound is higher than the ordinary refractive indices of the first organic compound and the third organic compound.

## 26 Claims, 50 Drawing Sheets

### (51) Int. Cl.

<b>H10K 50/818</b>	(2023.01)
<b>H10K 85/00</b>	(2023.01)
<b>H10K 59/80</b>	(2023.01)
<b>H10K 101/40</b>	(2023.01)

### (52) U.S. Cl.

CPC ..... *H10K 59/80518* (2023.02); *H10K 59/876* (2023.02); *H10K 59/879* (2023.02); *H10K 2101/40* (2023.02)

### (58) Field of Classification Search

CPC .... H10K 59/879; H10K 85/00; H10K 59/876; H10K 50/156; H10K 85/631; H10K 85/6572; H10K 85/6574; H10K 85/6576; H10K 2101/30; H10K 2102/351

See application file for complete search history.

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FIG. 1A

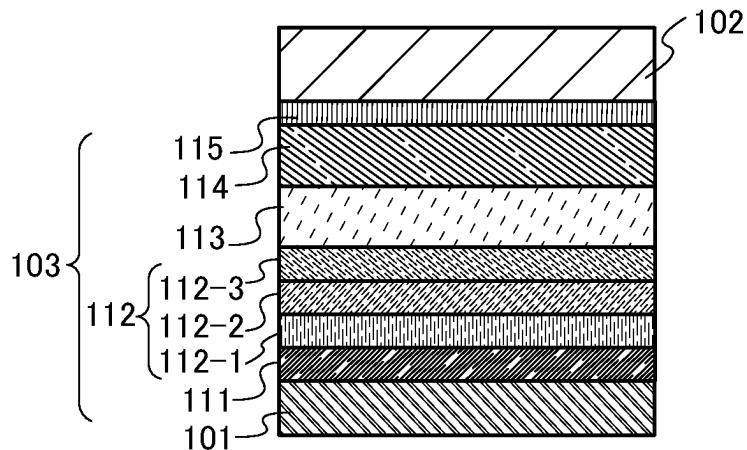


FIG. 1B

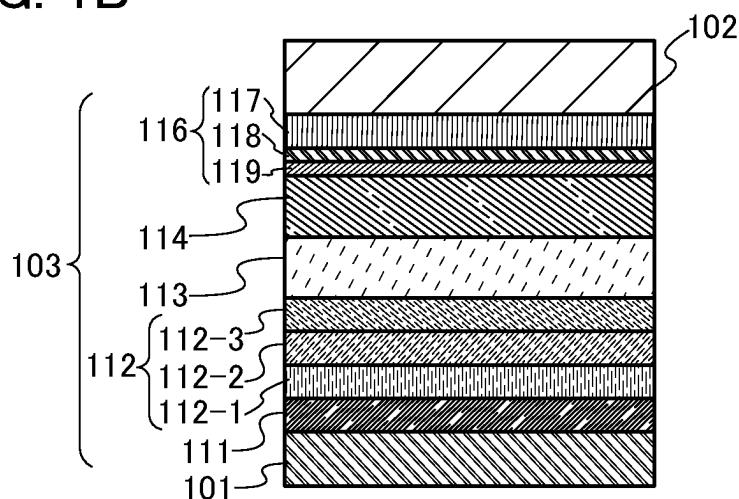


FIG. 1C

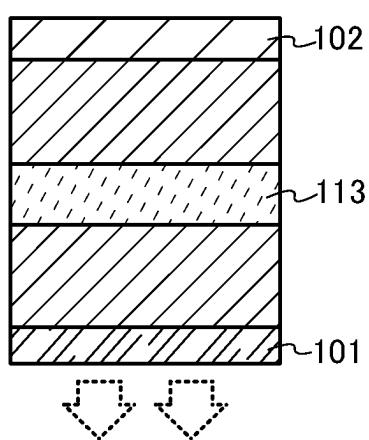
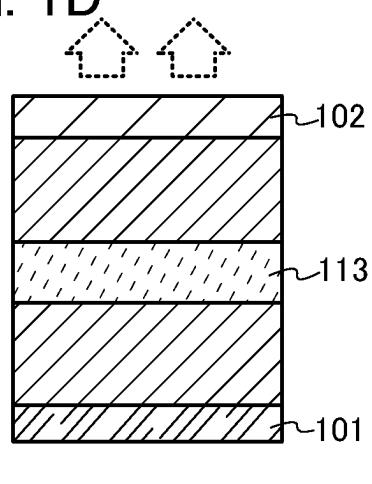
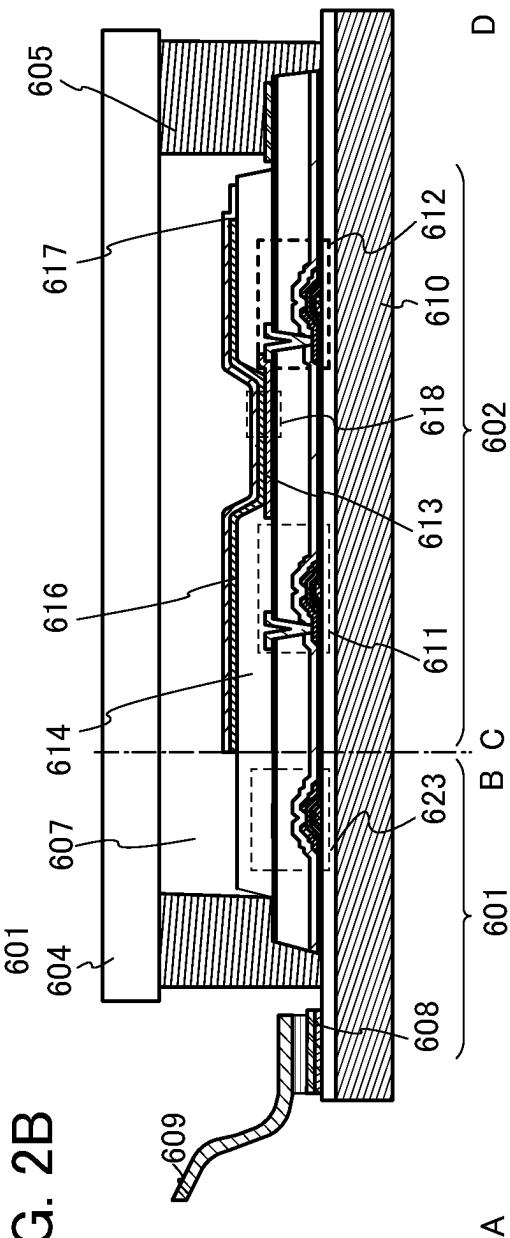
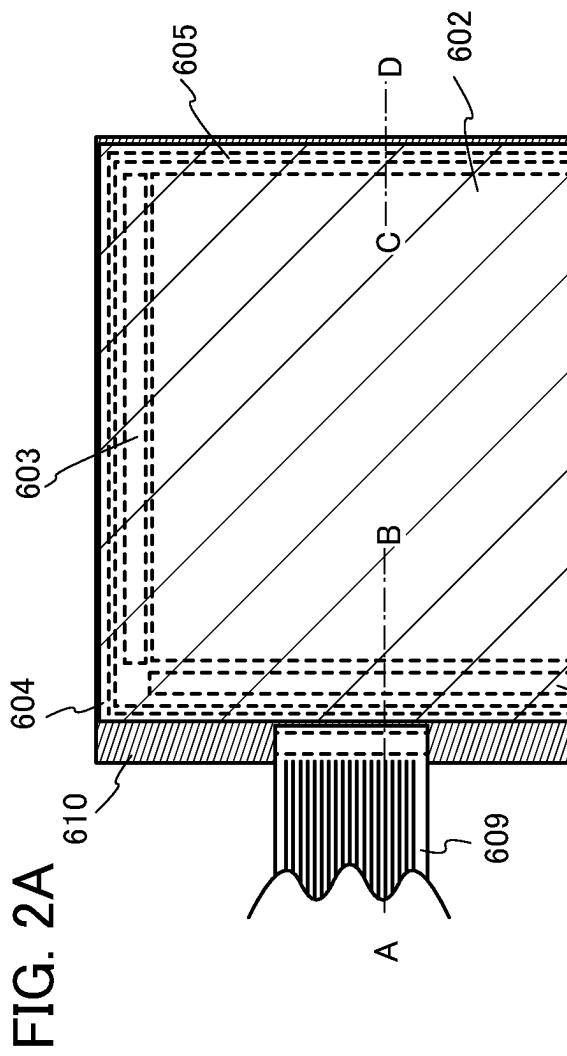
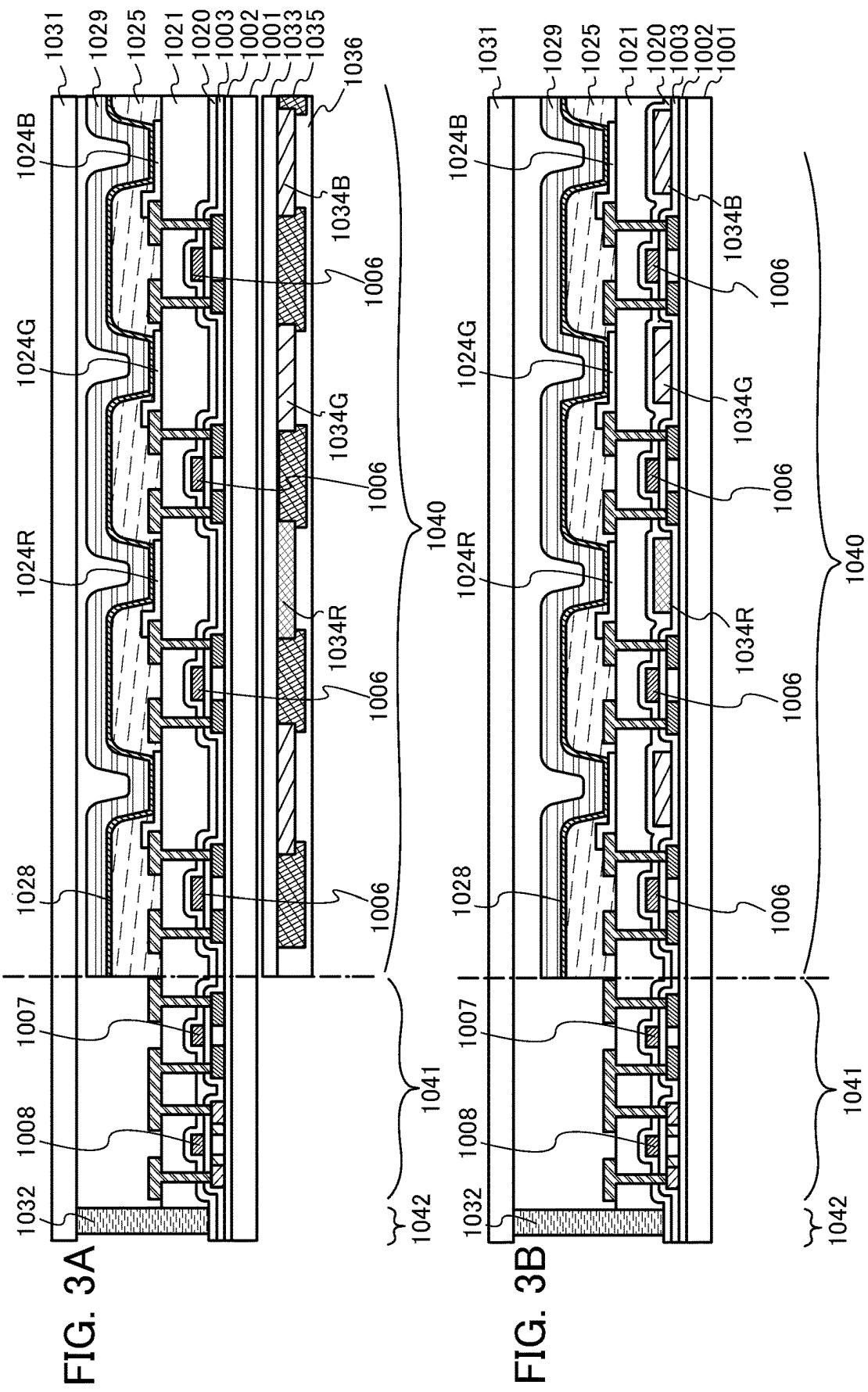


FIG. 1D







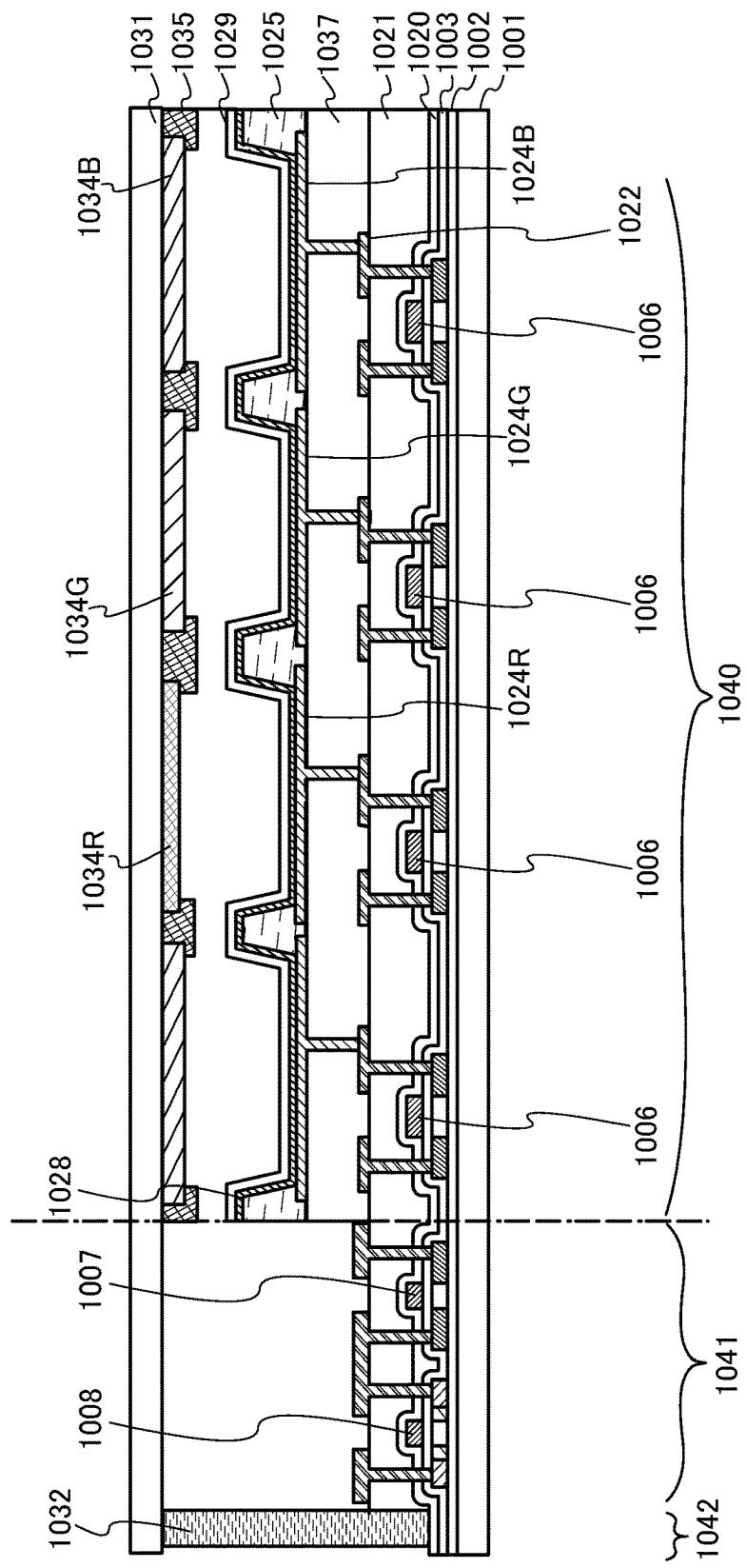


FIG. 4

FIG. 5A

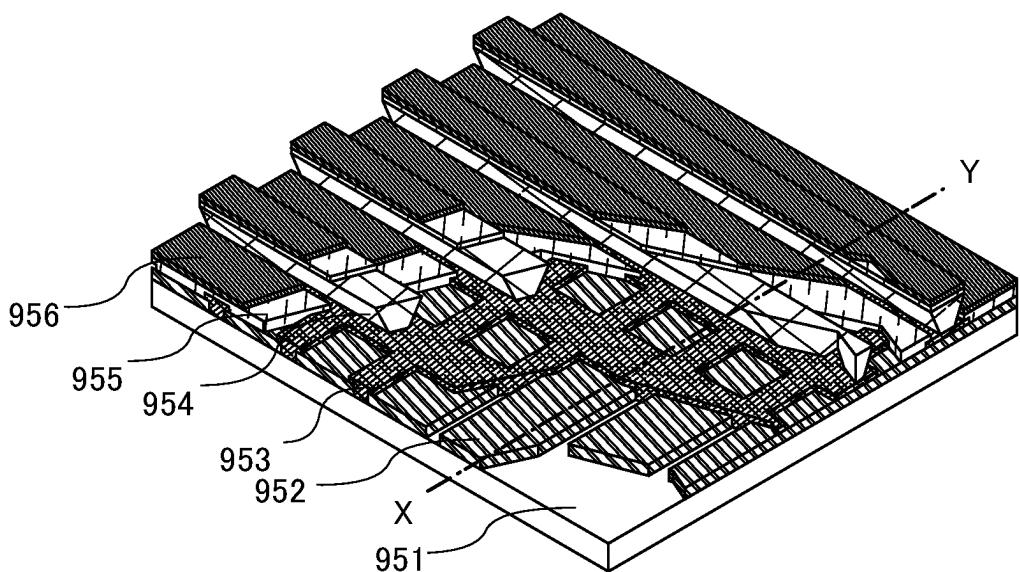
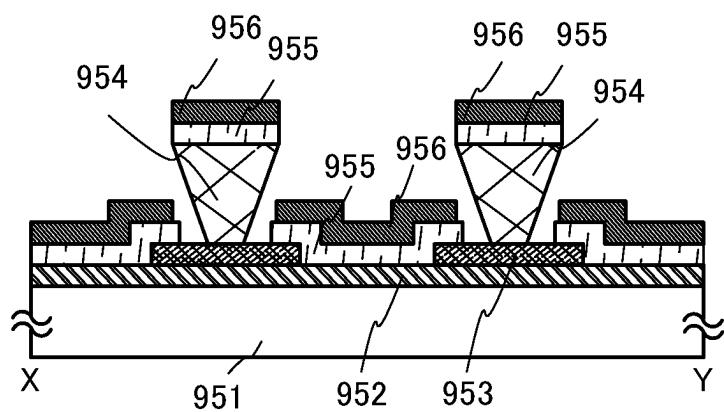


FIG. 5B



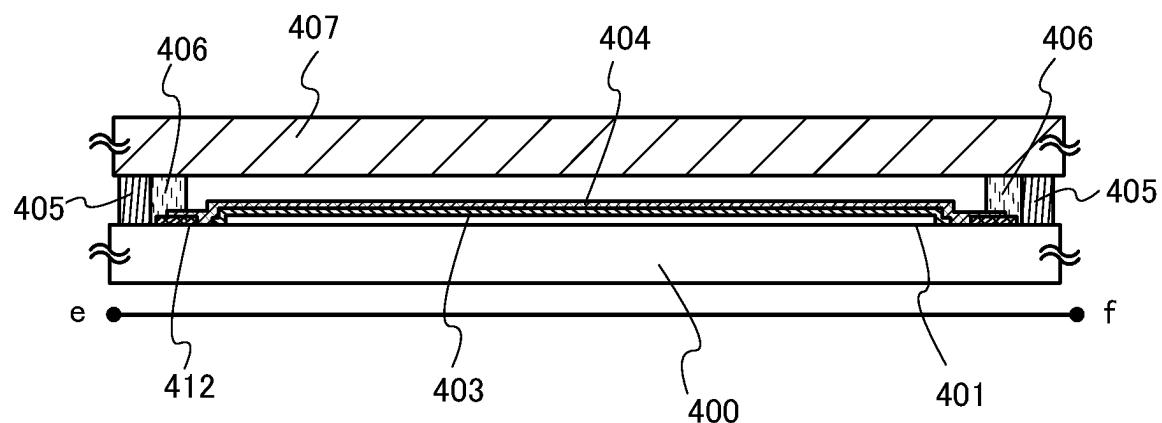
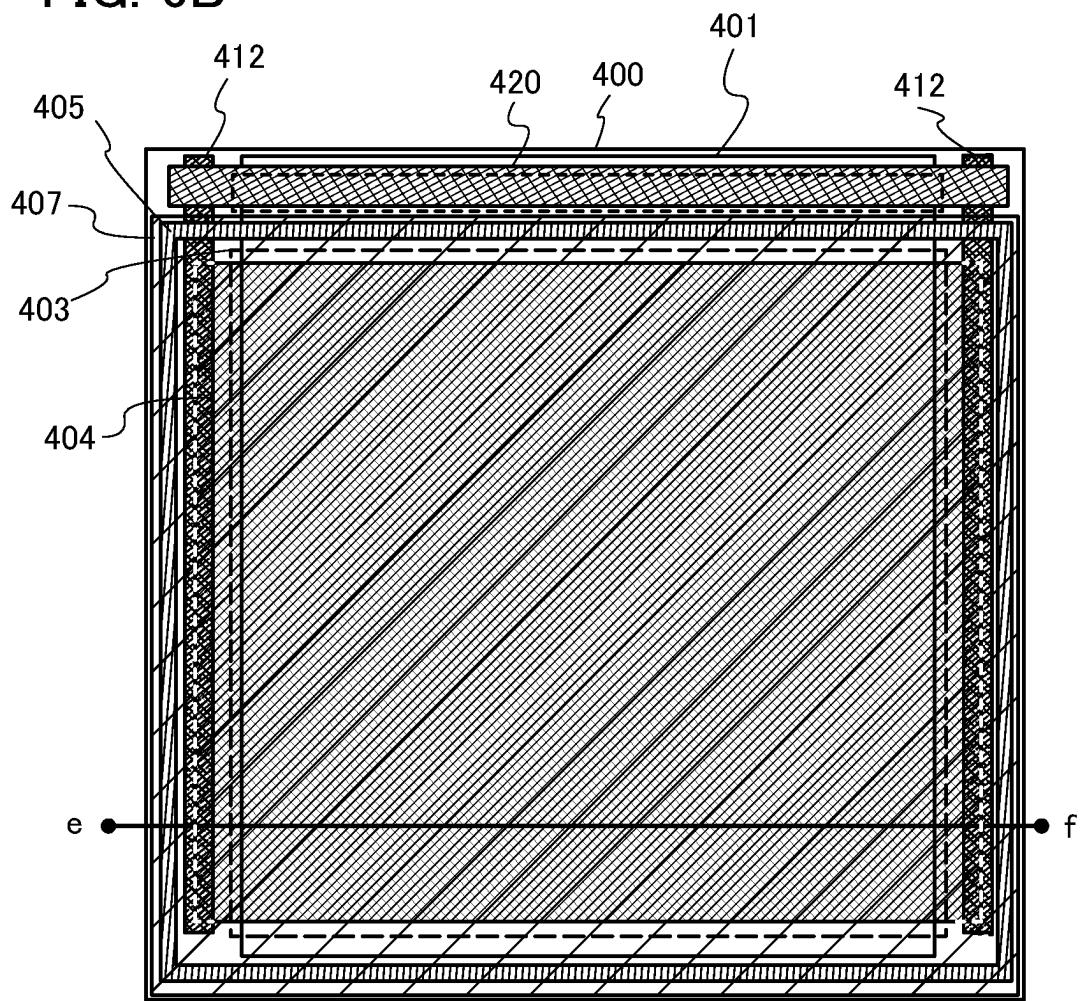
**FIG. 6A****FIG. 6B**

FIG. 7A

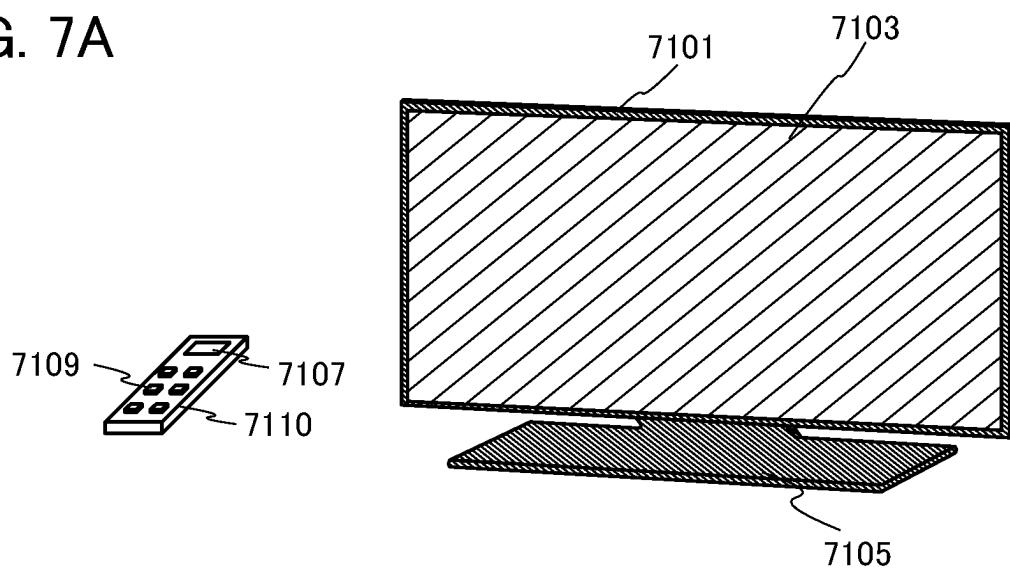


FIG. 7B1

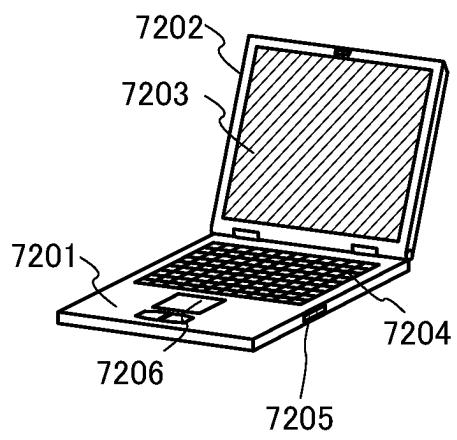


FIG. 7B2

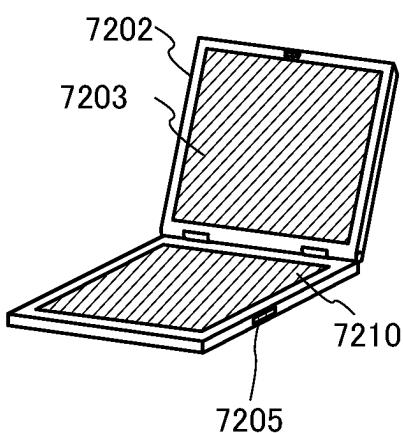
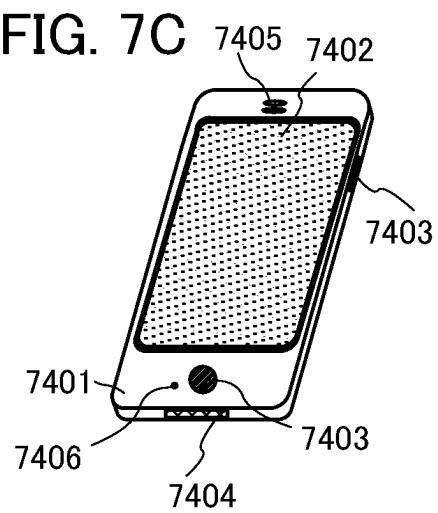


FIG. 7C



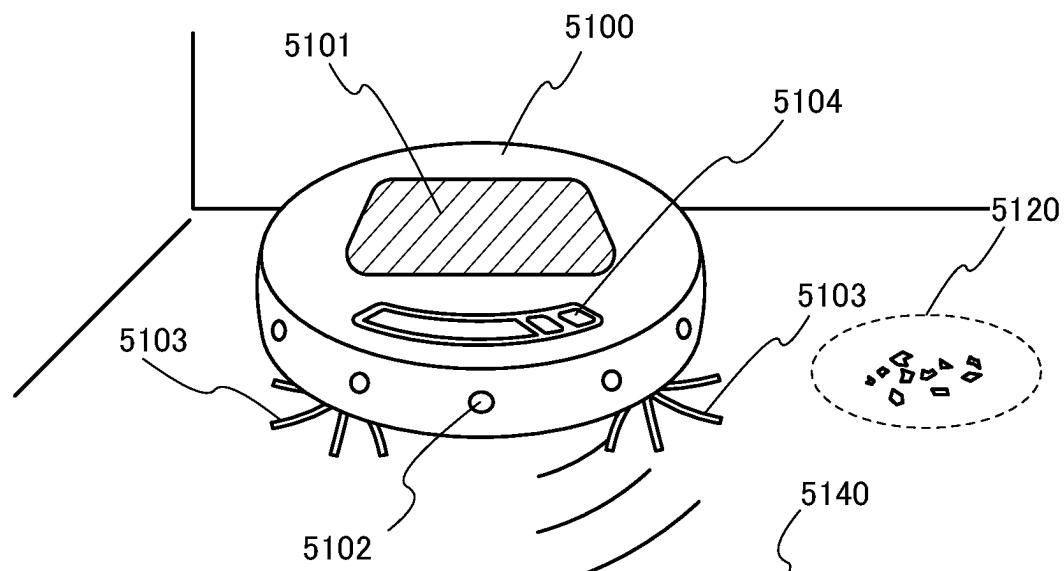
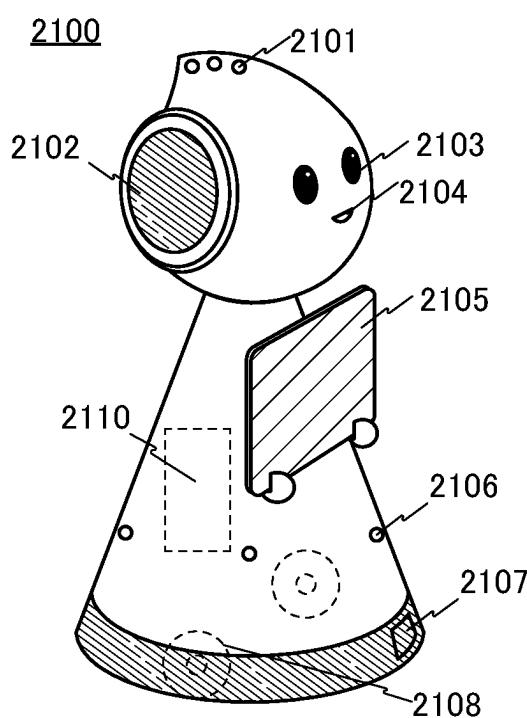
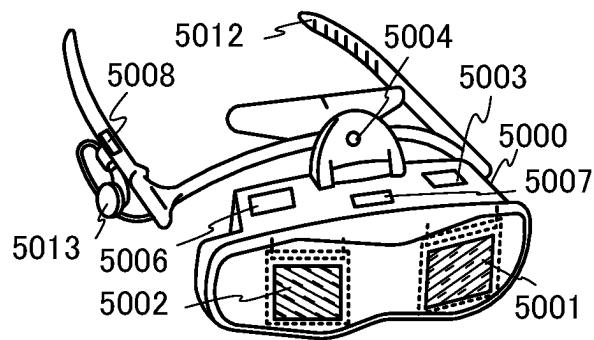
**FIG. 8A****FIG. 8B****FIG. 8C**

FIG. 9

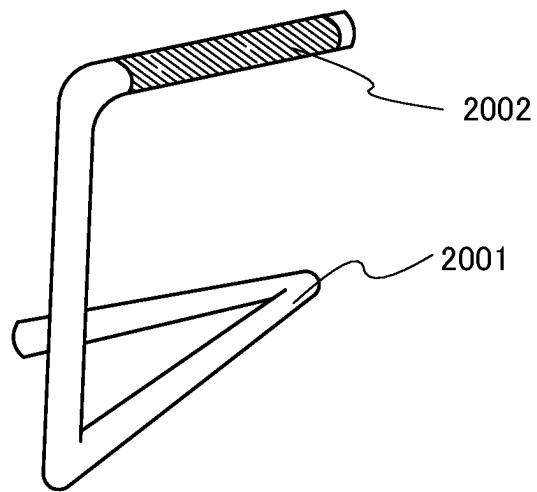
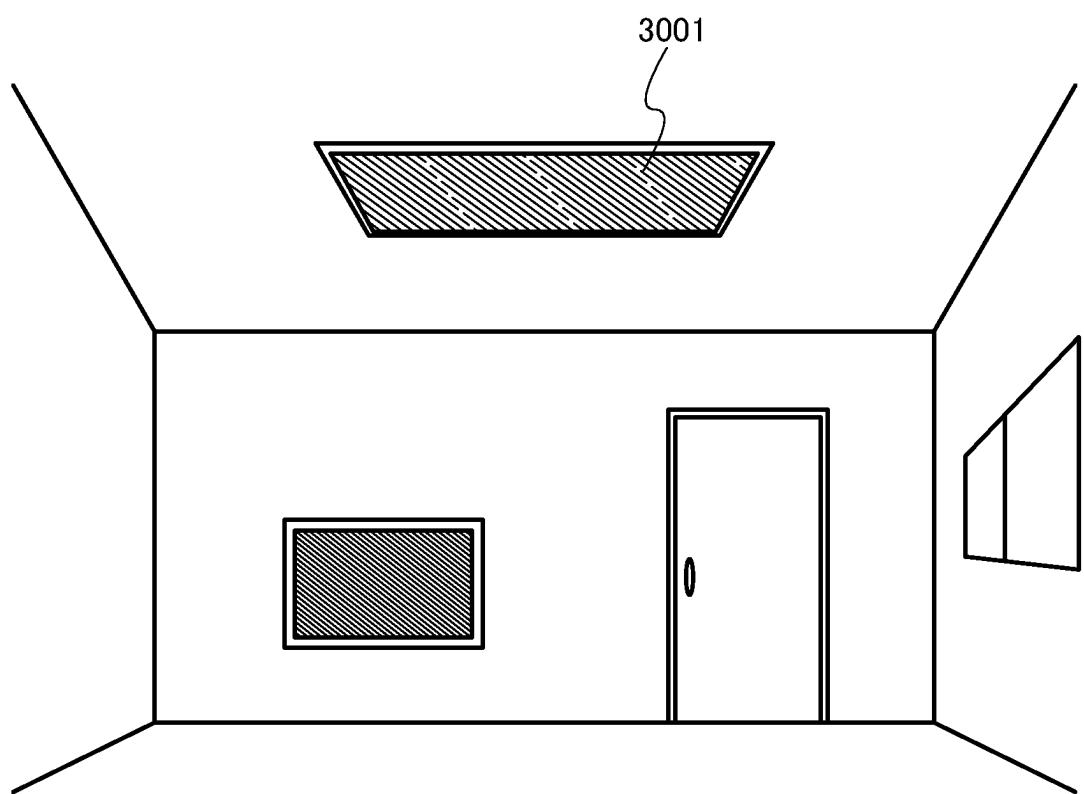
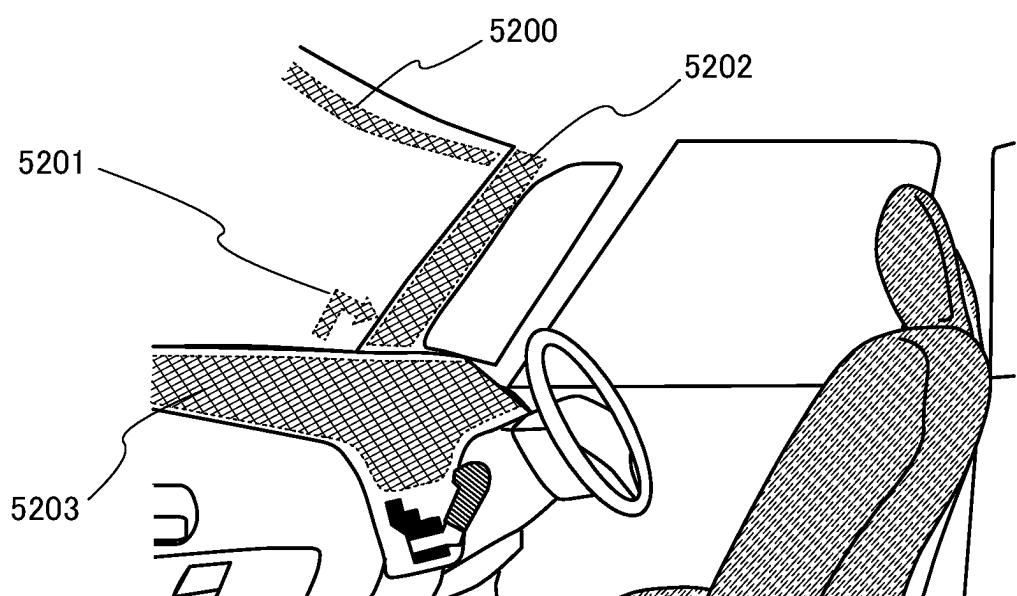
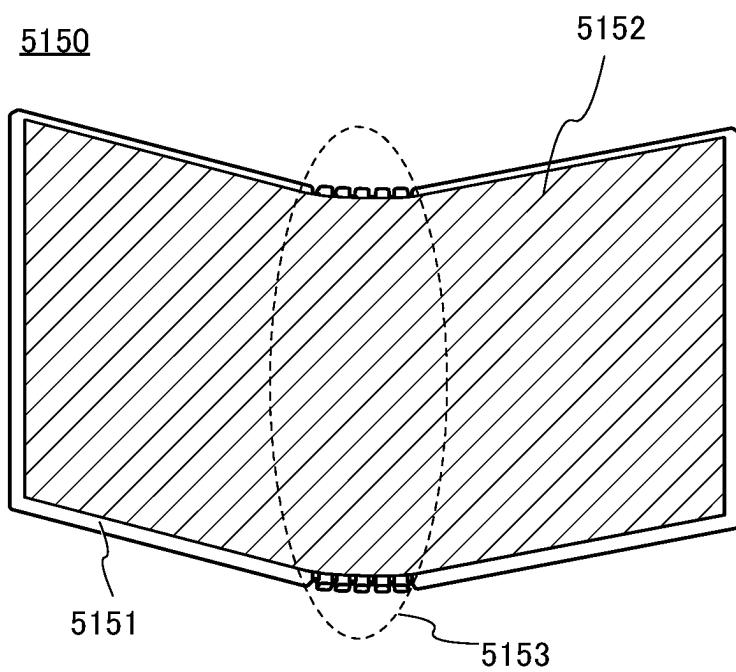
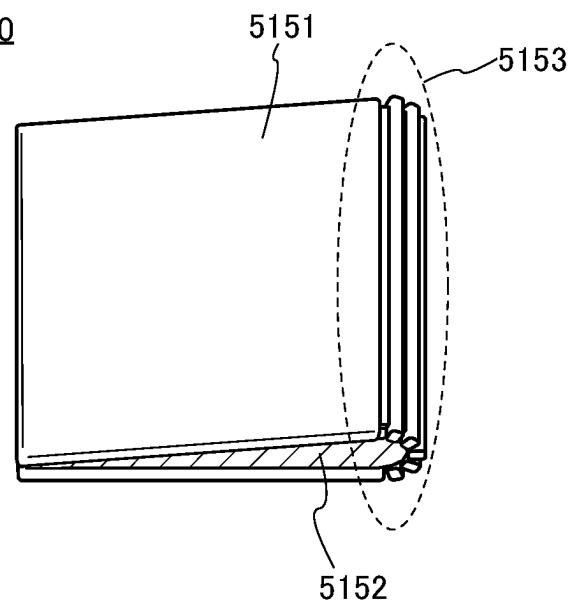
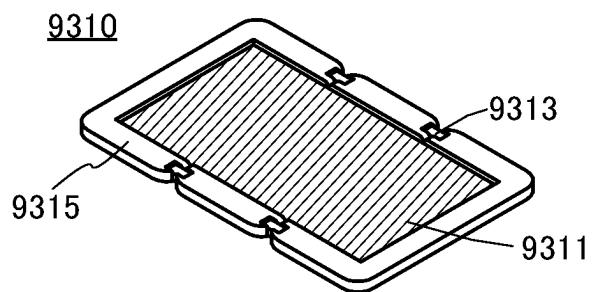
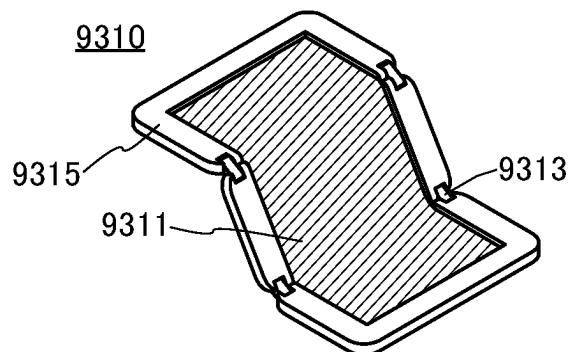
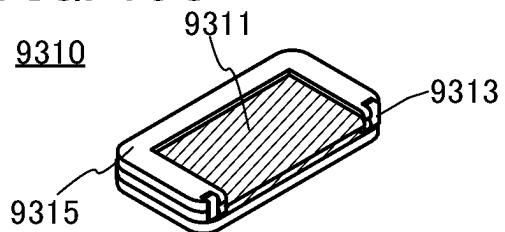


FIG. 10



**FIG. 11**

**FIG. 12A****FIG. 12B**

**FIG. 13A****FIG. 13B****FIG. 13C**

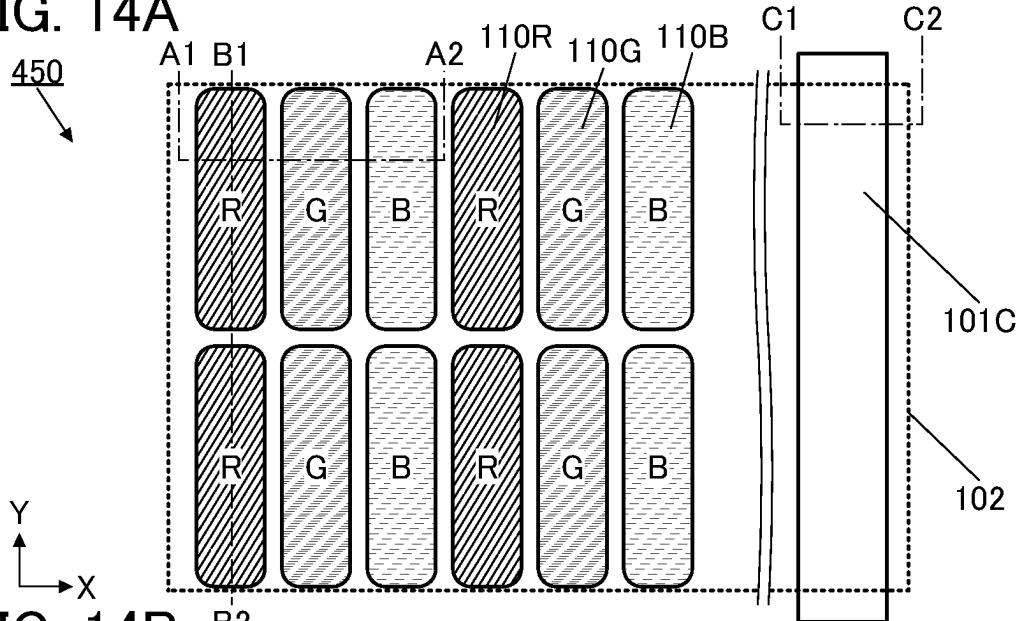
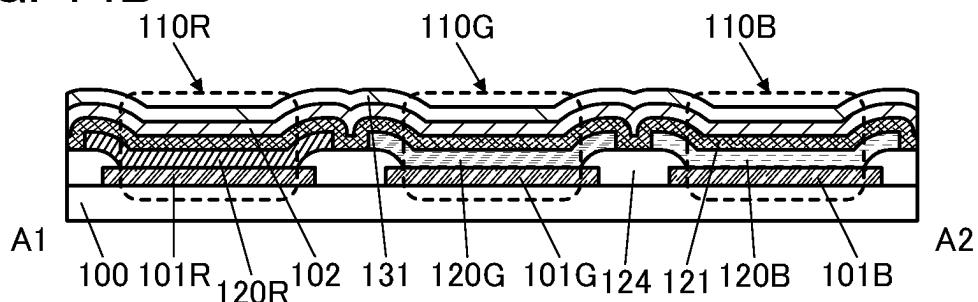
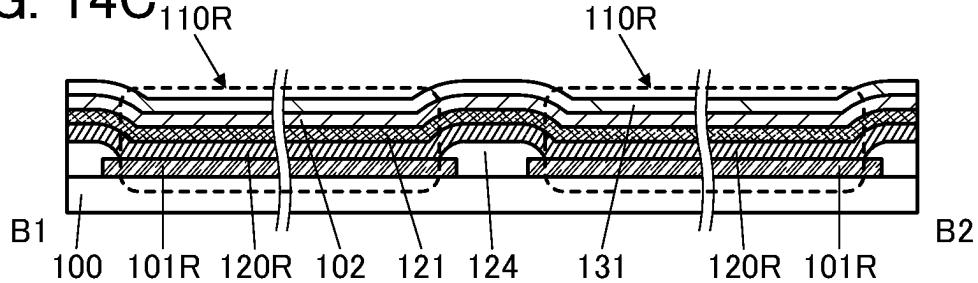
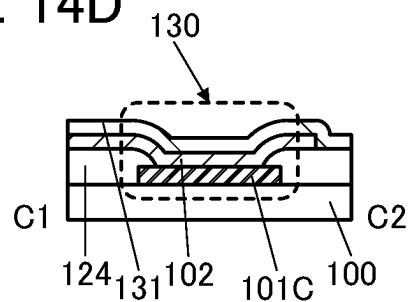
**FIG. 14A****FIG. 14B****FIG. 14C****FIG. 14D**

FIG. 15A

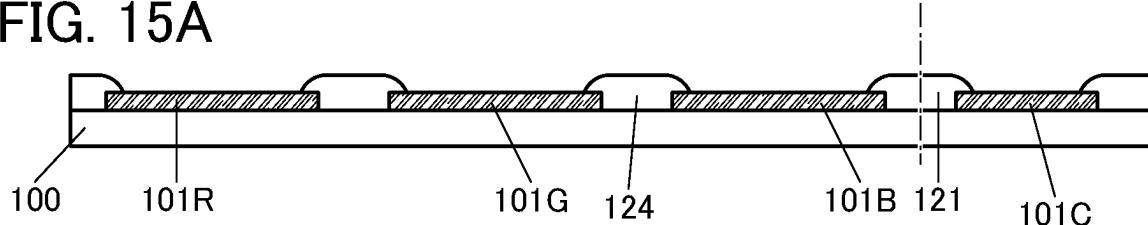


FIG. 15B

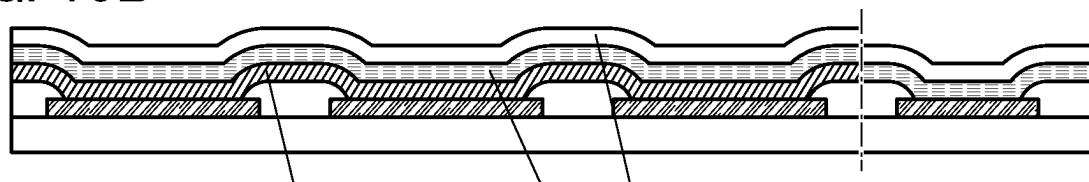


FIG. 15C

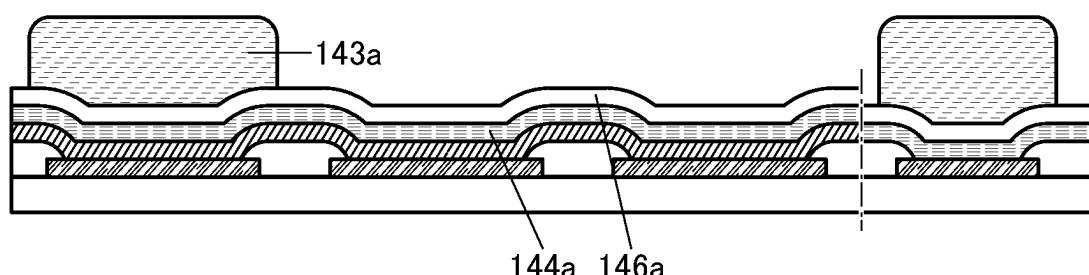


FIG. 15D

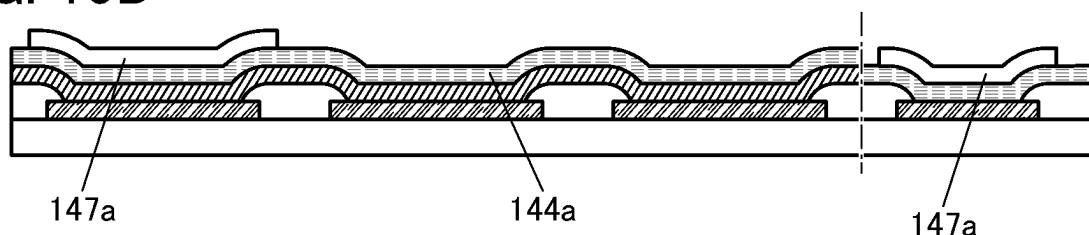


FIG. 15E

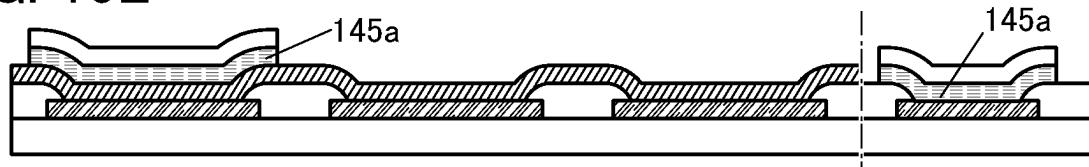


FIG. 15F

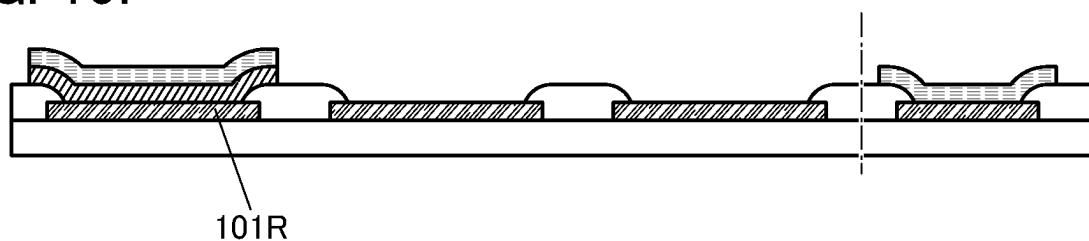


FIG. 16A

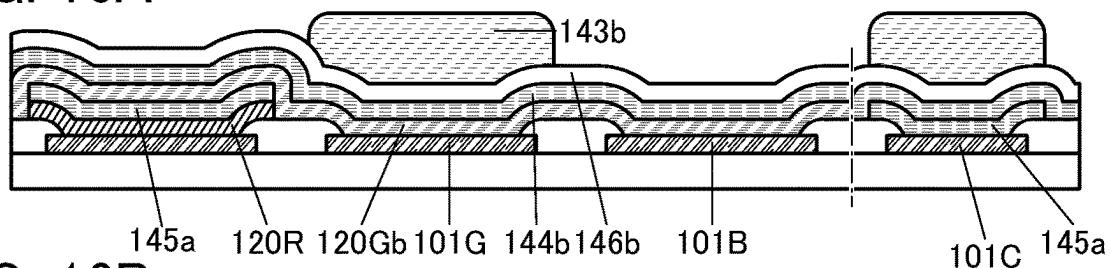


FIG. 16B

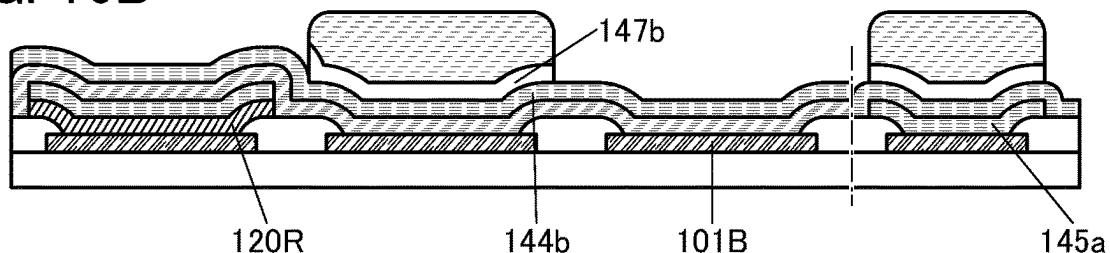


FIG. 16C

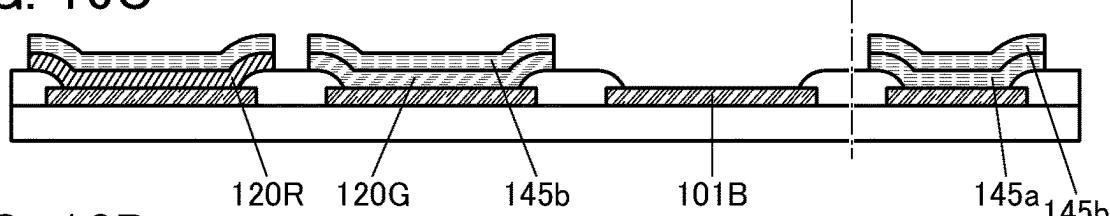


FIG. 16D

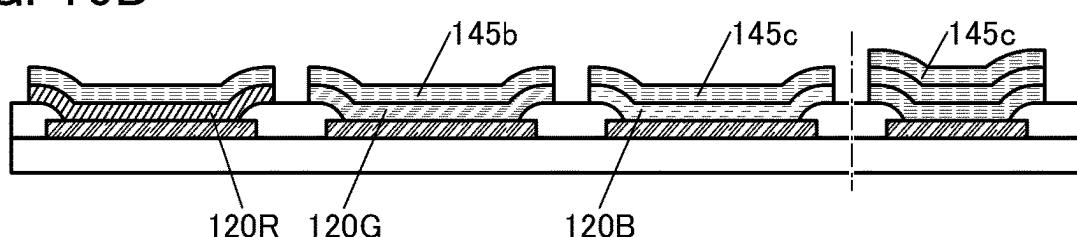


FIG. 16E

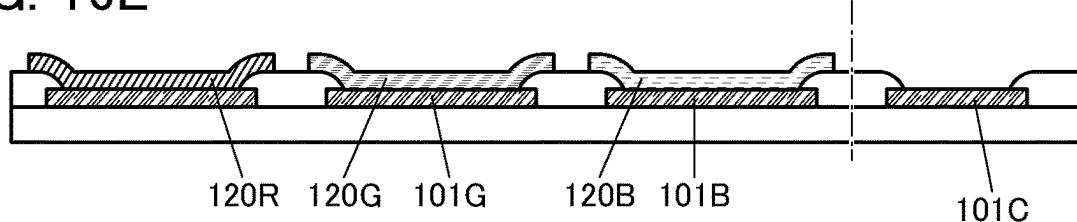


FIG. 16F

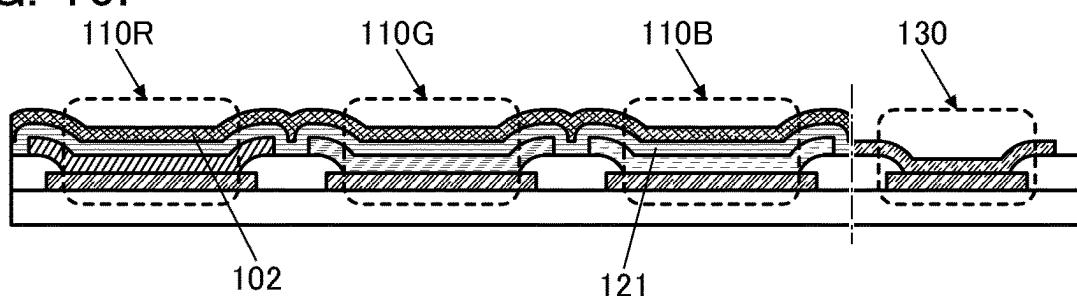


FIG. 17

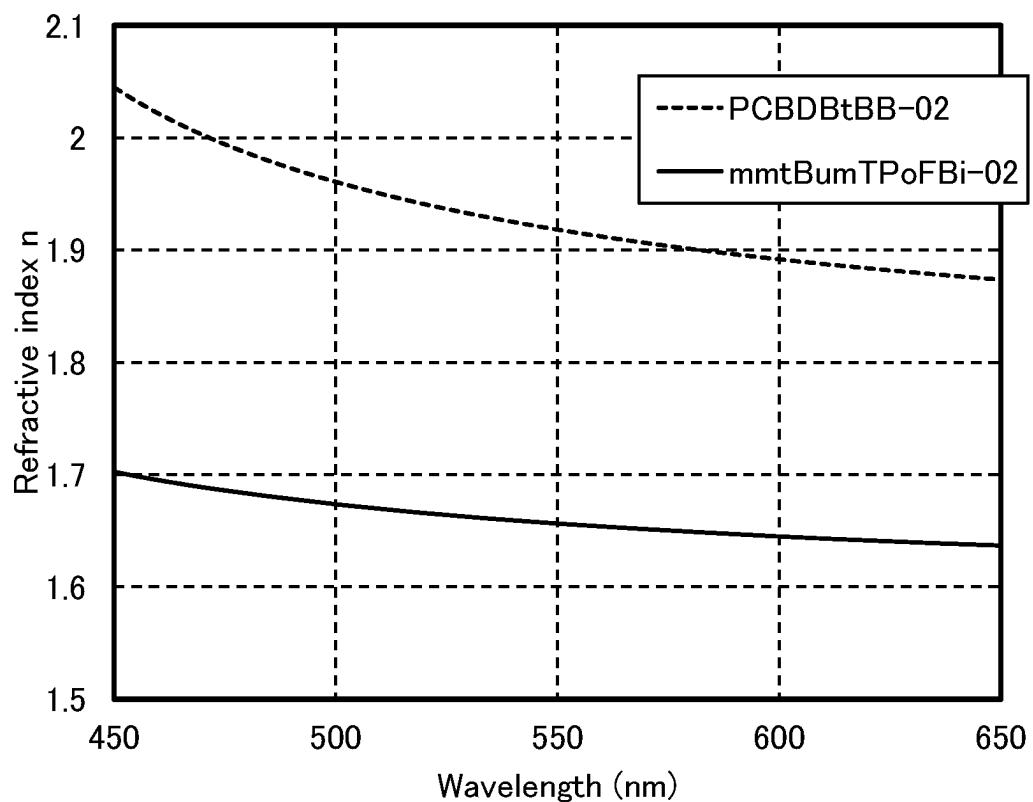


FIG. 18

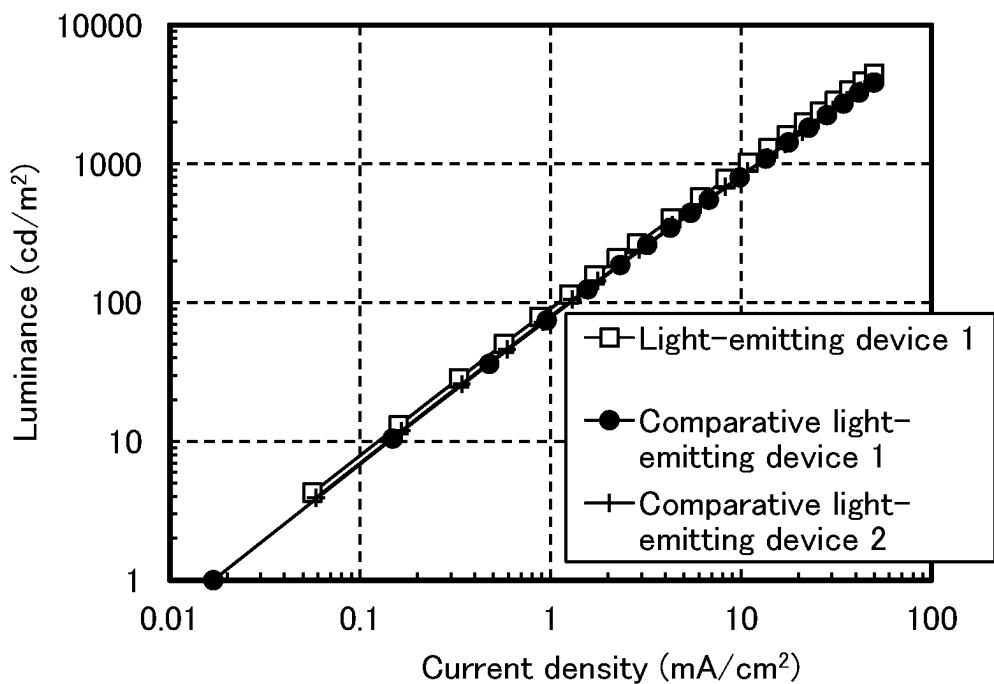


FIG. 19

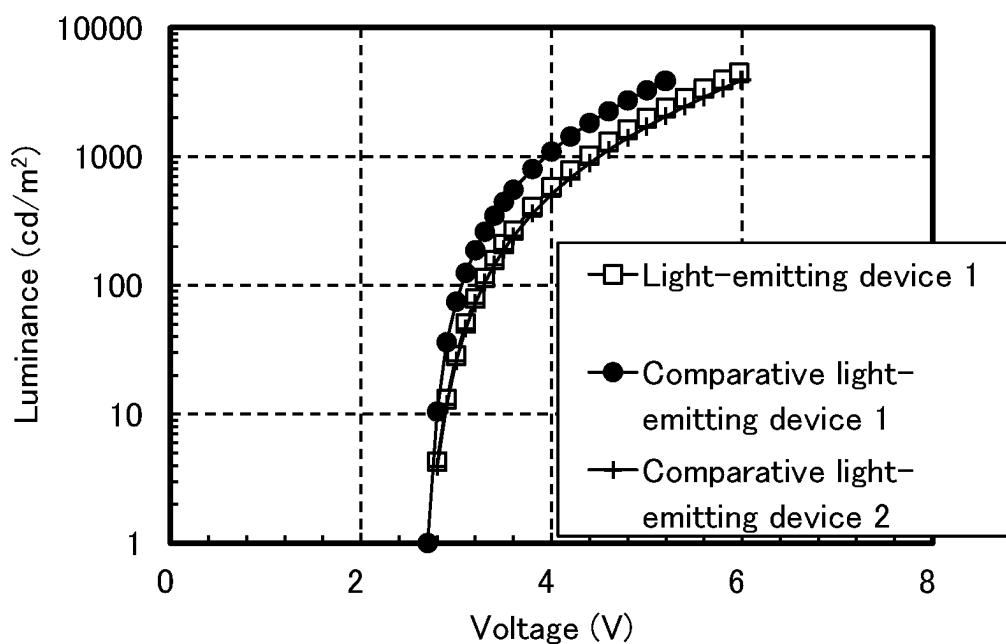


FIG. 20

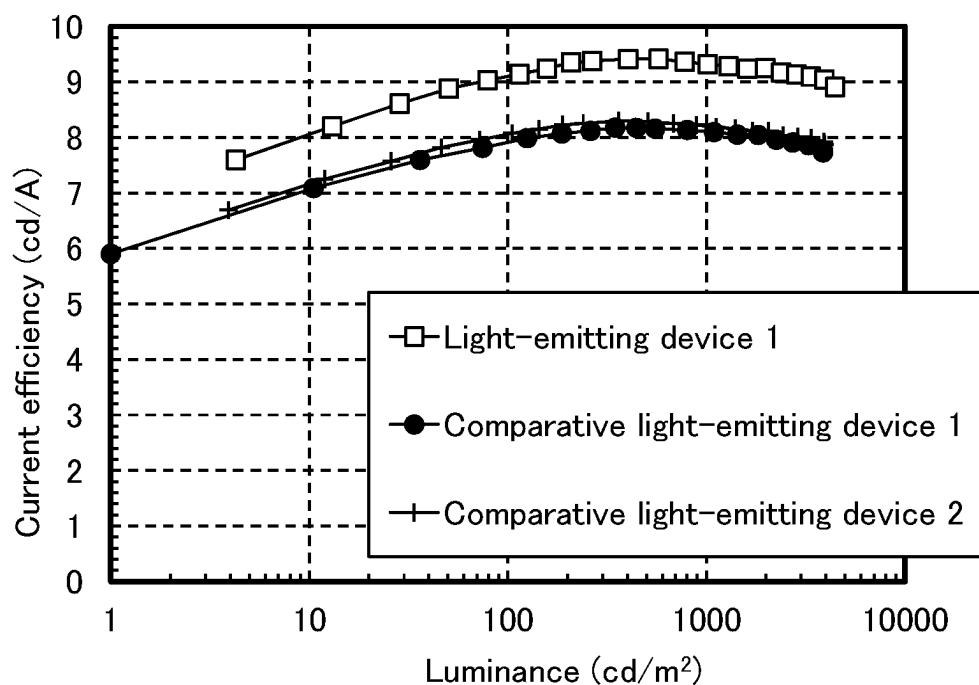


FIG. 21

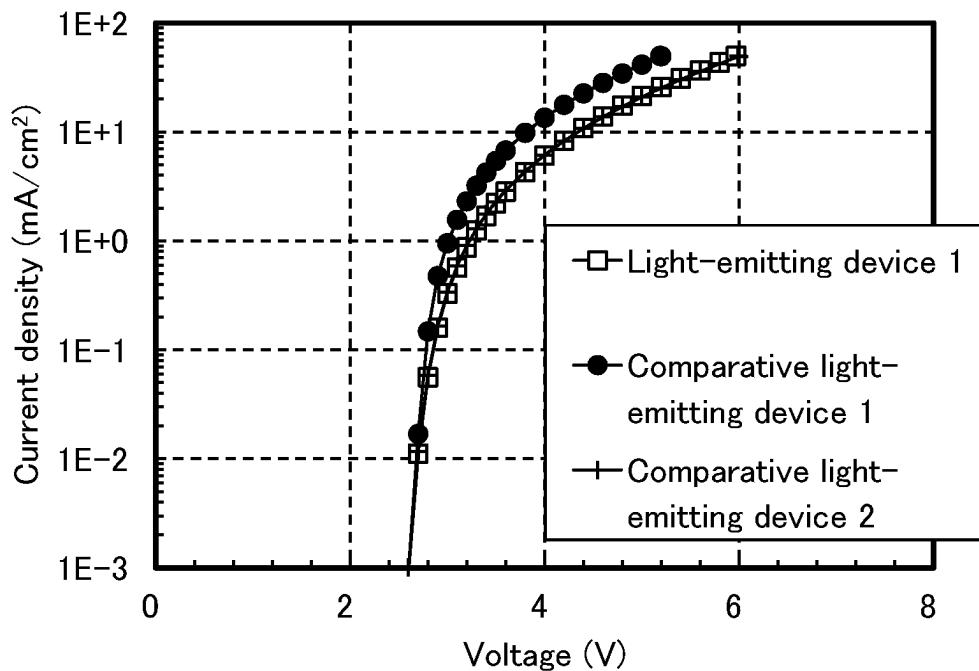


FIG. 22

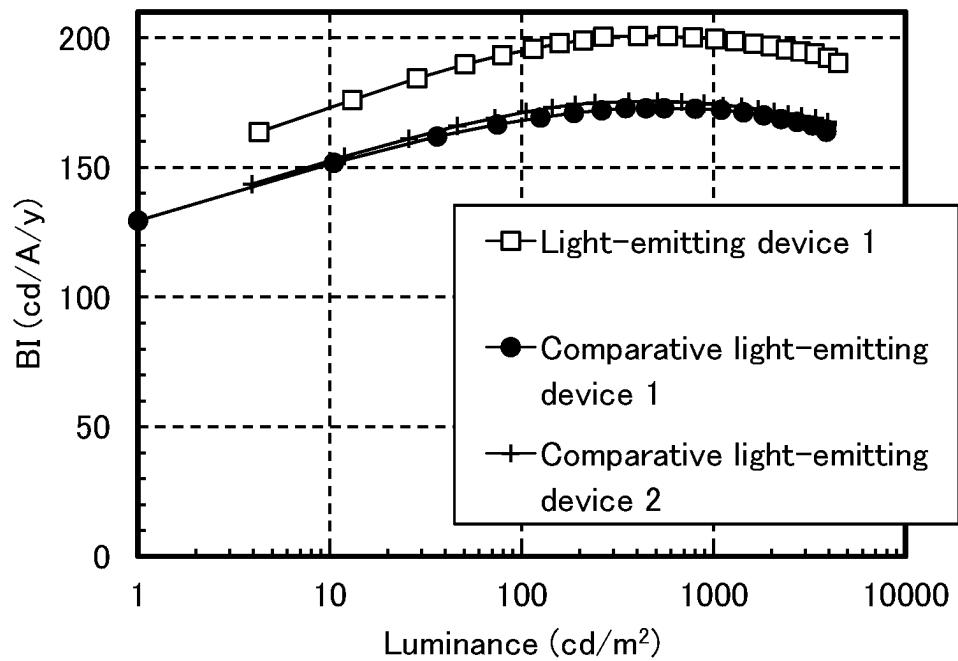


FIG. 23

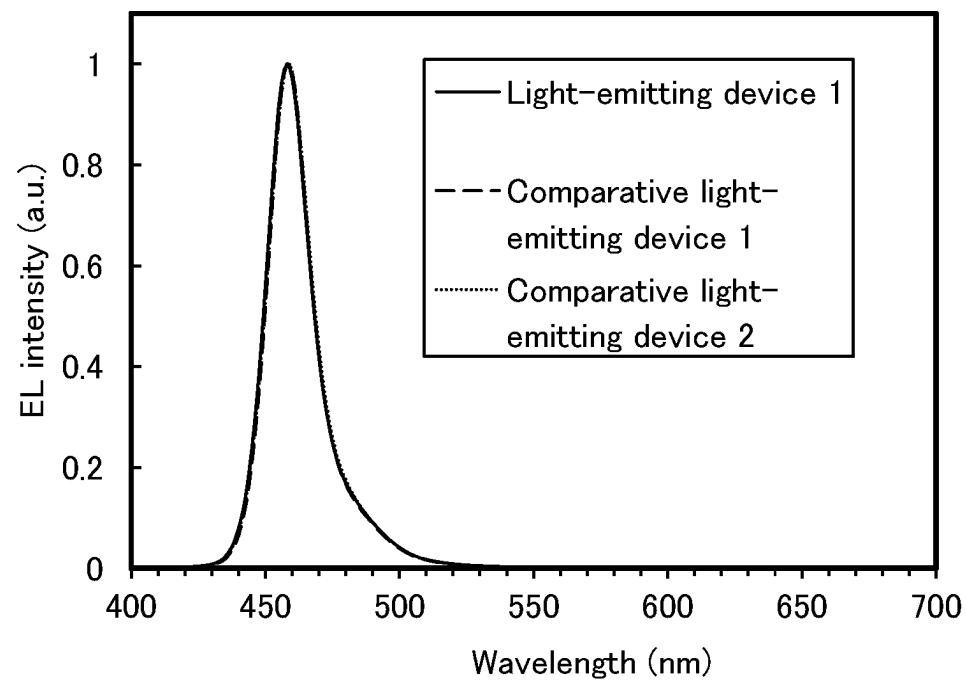


FIG. 24

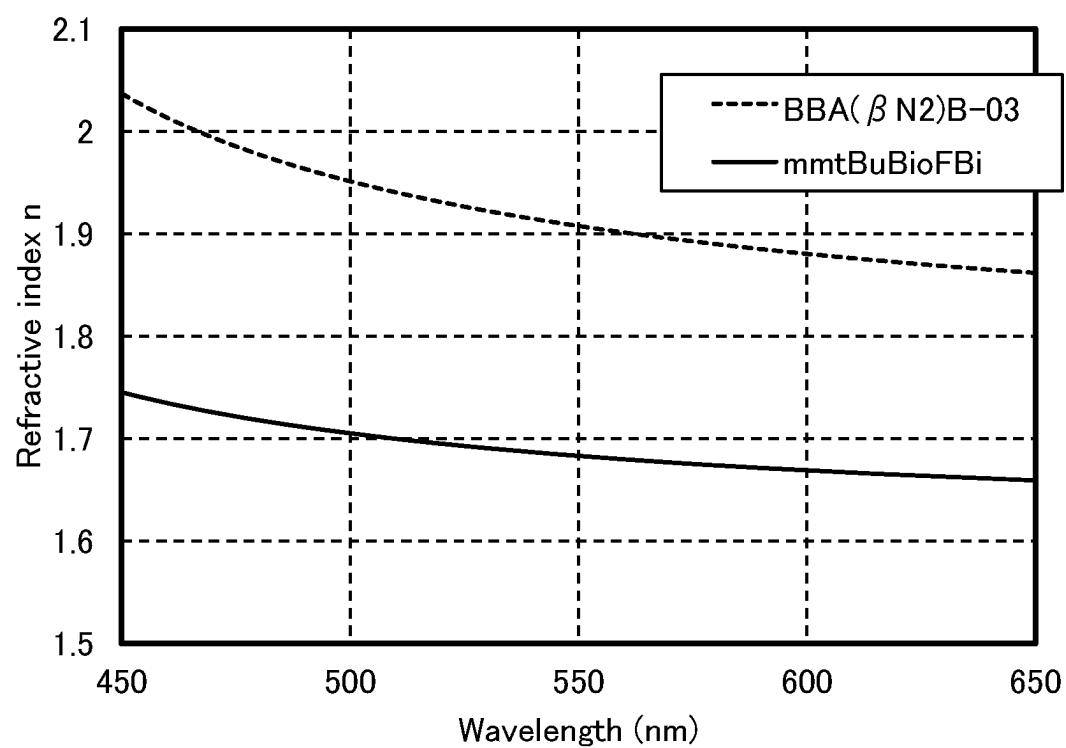


FIG. 25

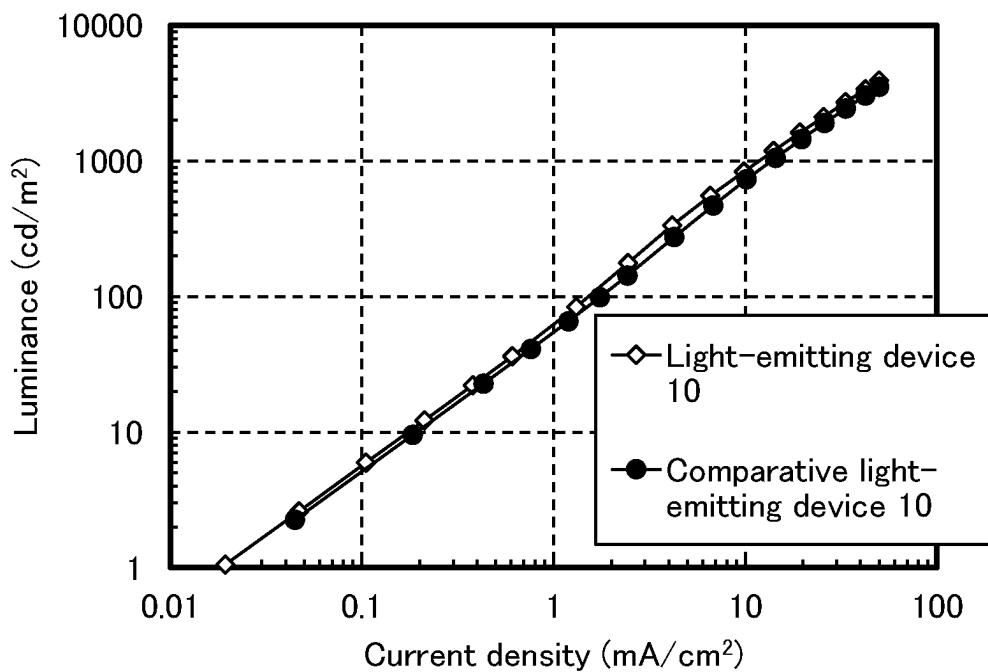


FIG. 26

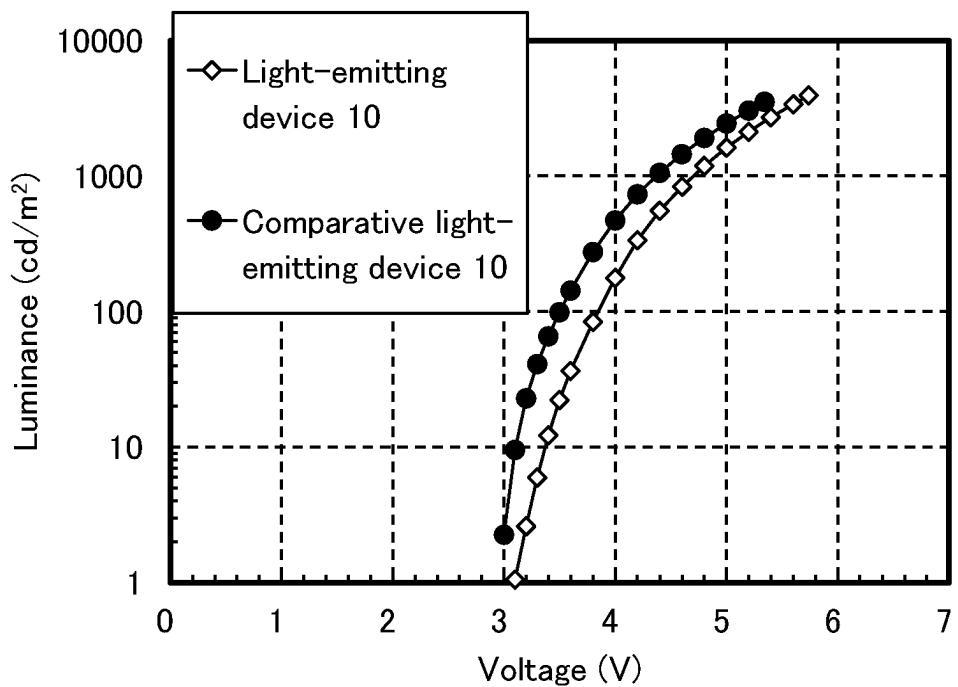


FIG. 27

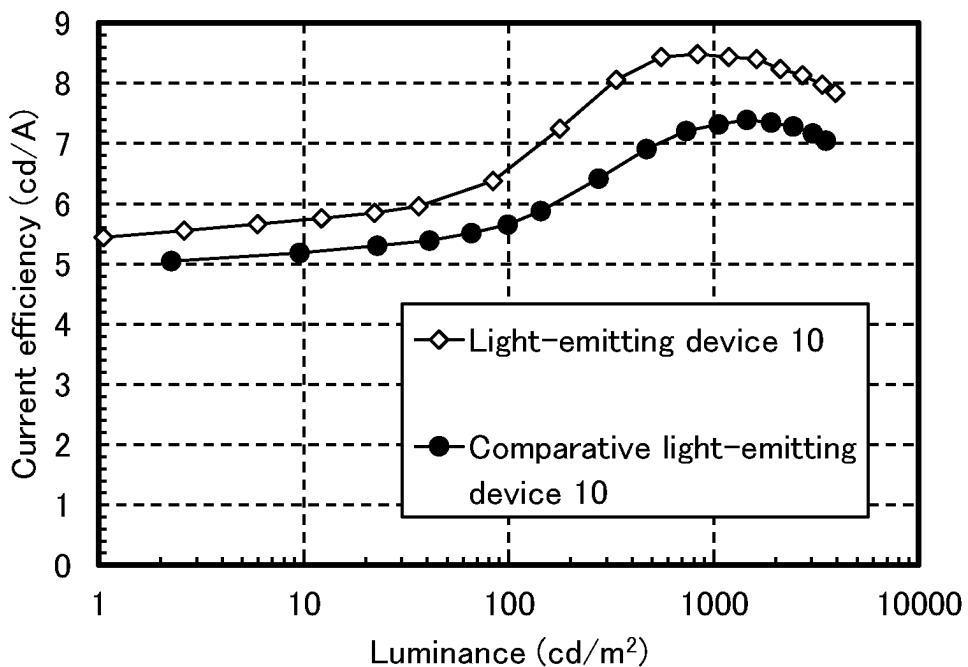


FIG. 28

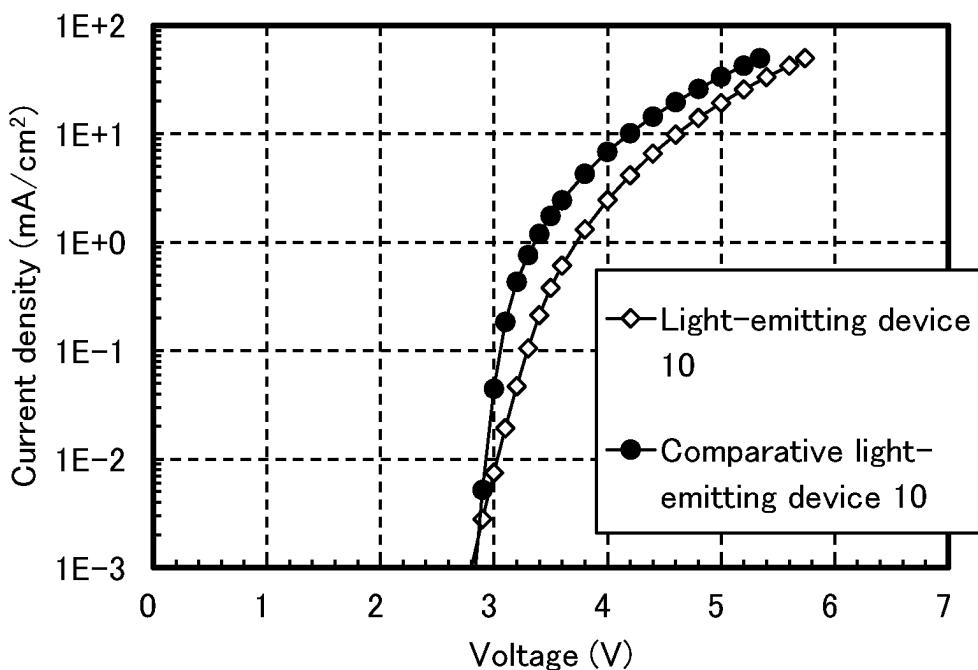


FIG. 29

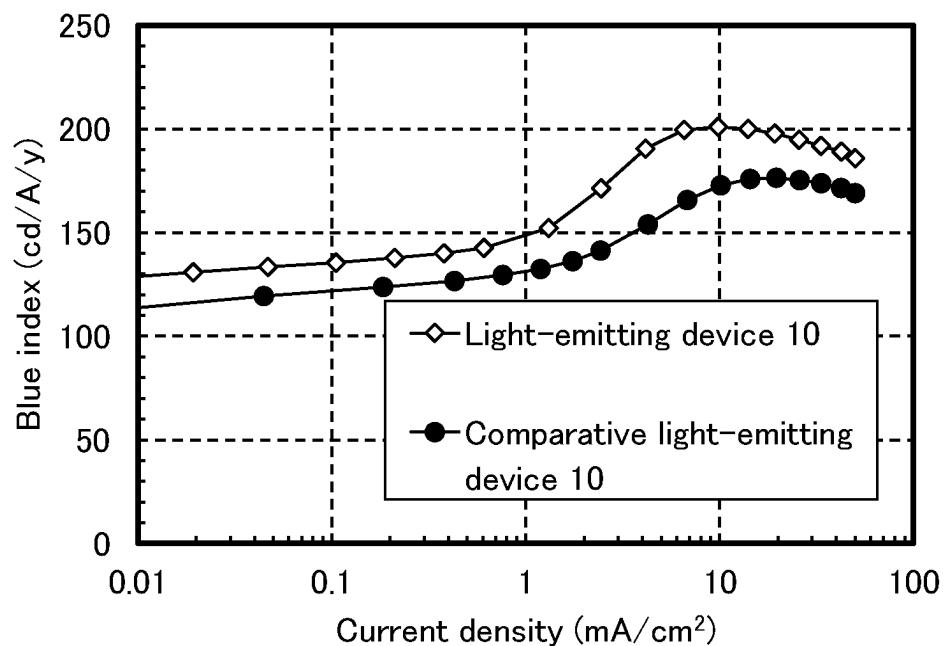


FIG. 30

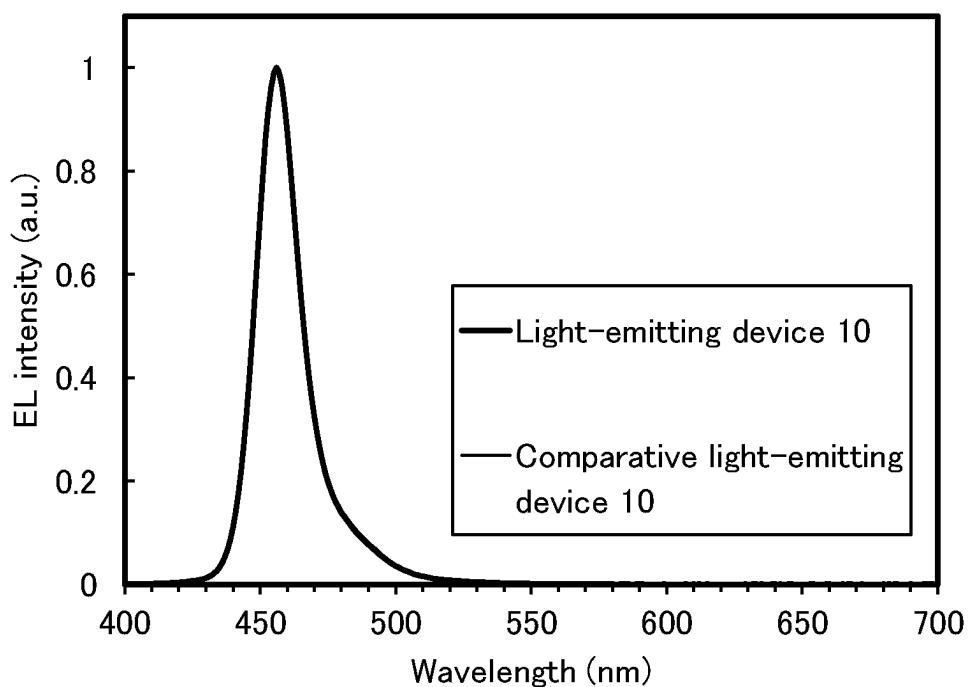


FIG. 31

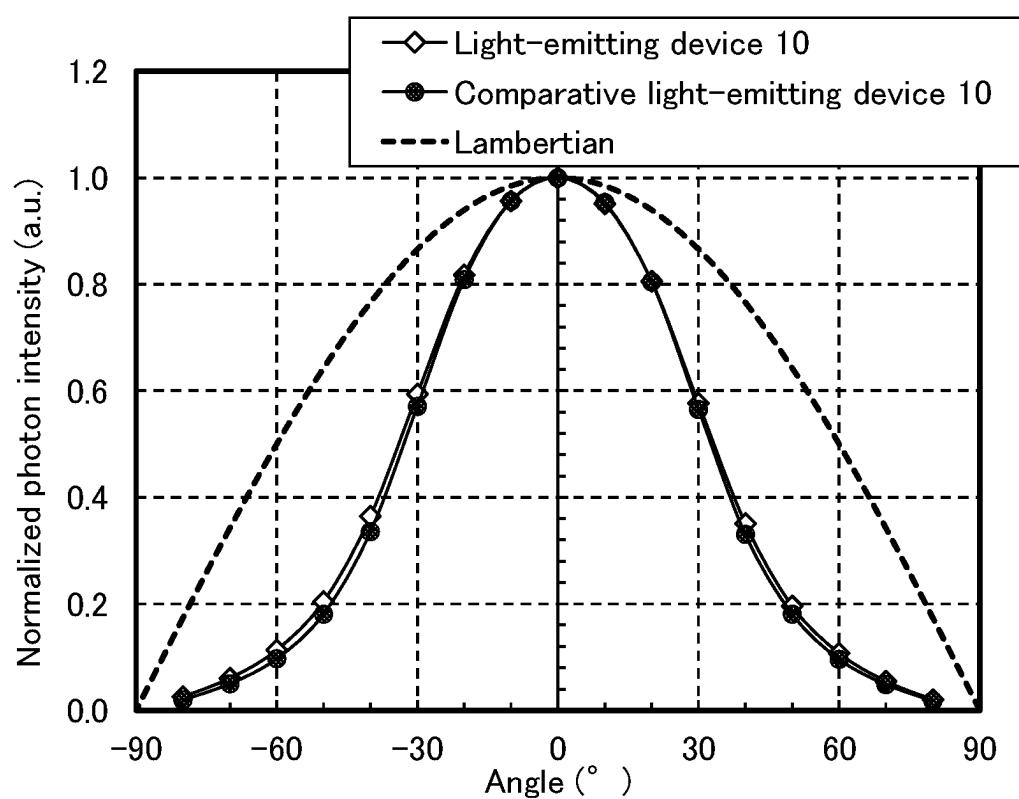


FIG. 32

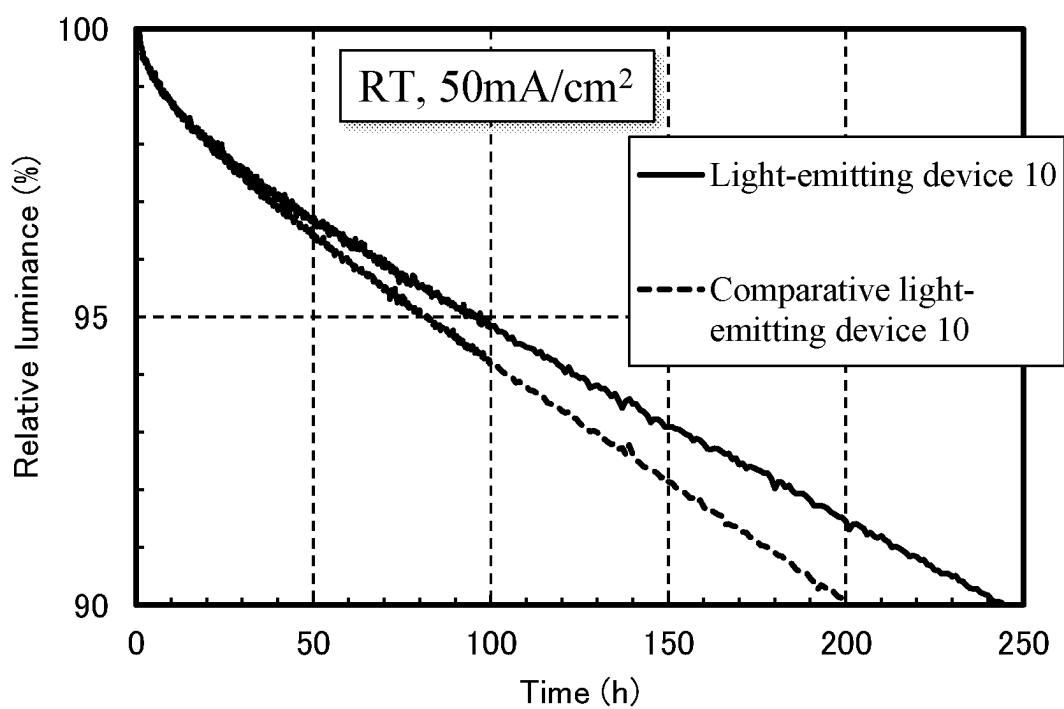


FIG. 33

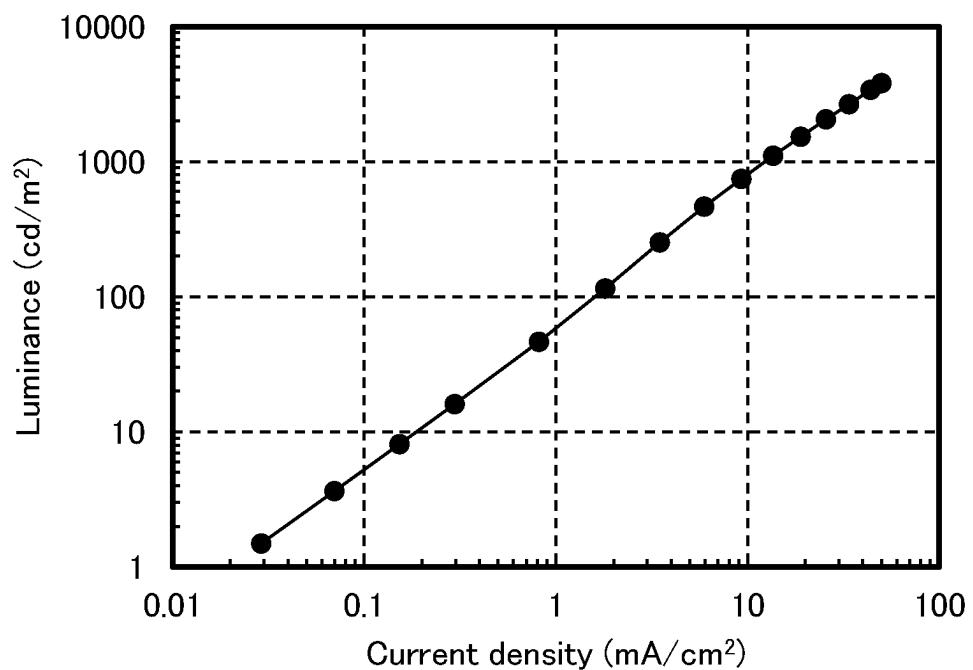


FIG. 34

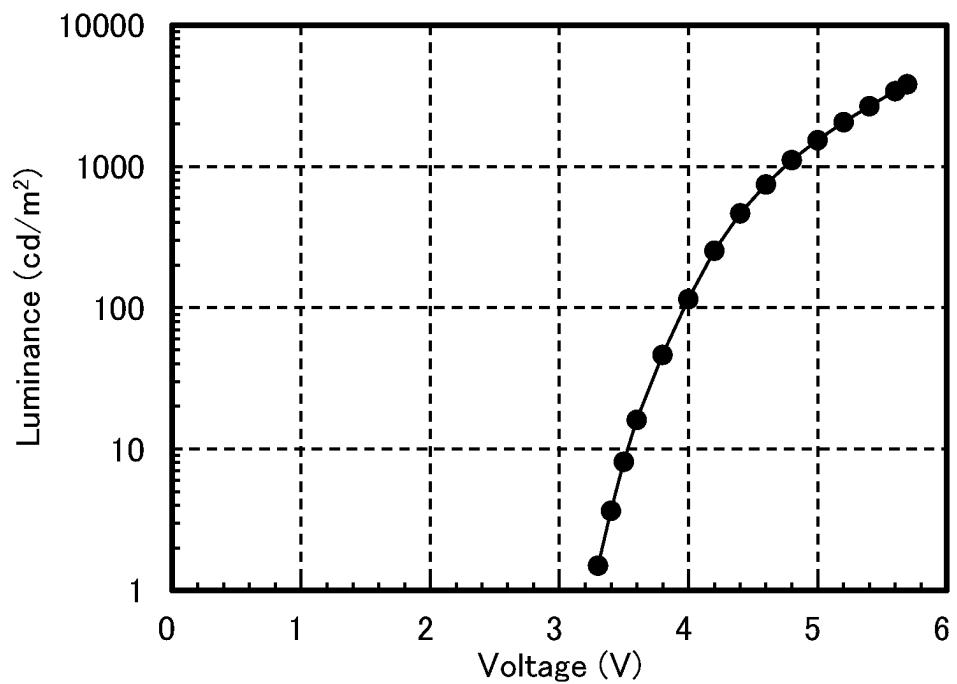


FIG. 35

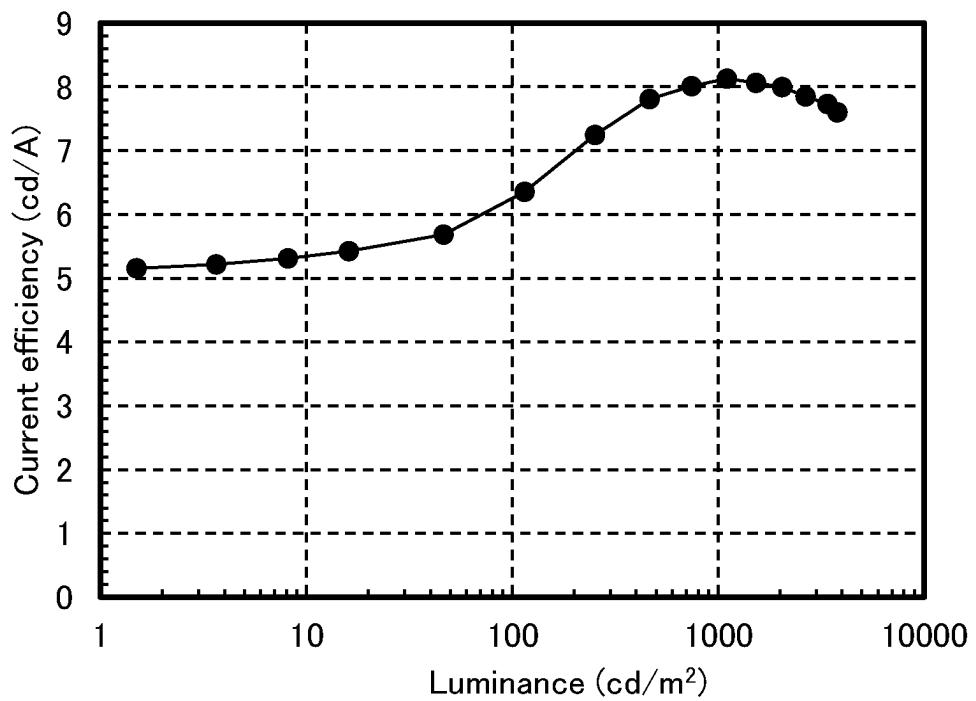


FIG. 36

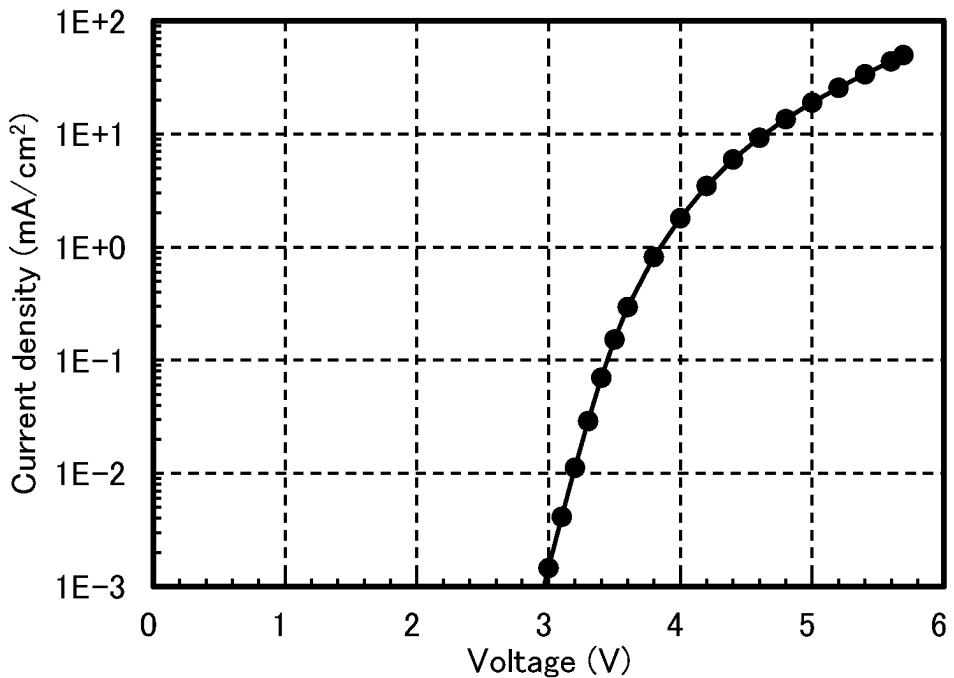


FIG. 37

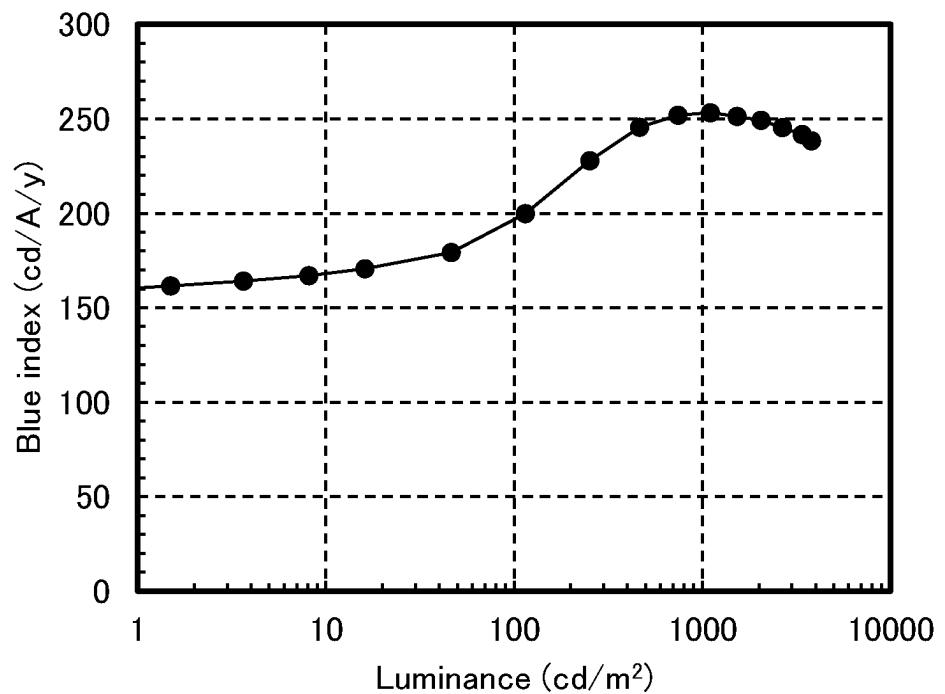
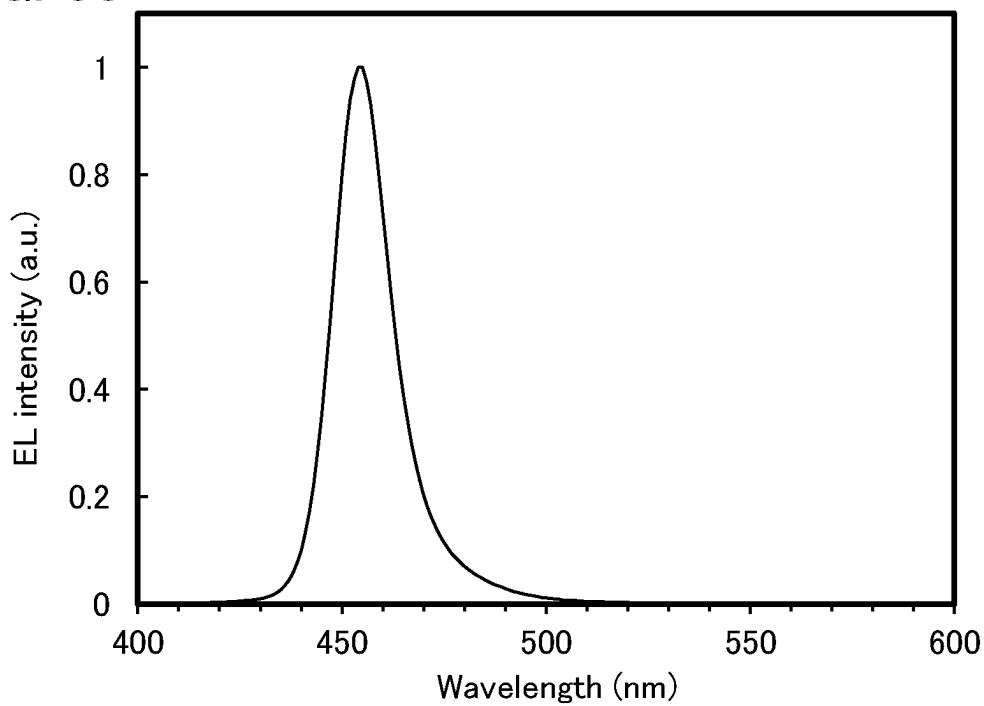


FIG. 38



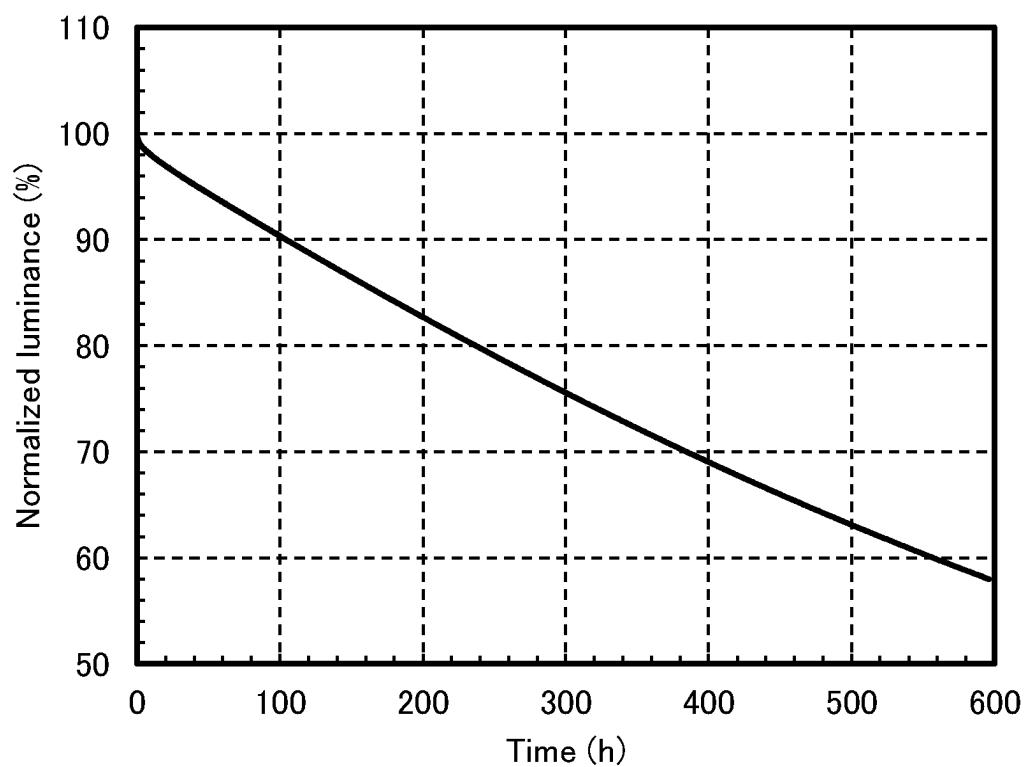
**FIG. 39**

FIG. 40

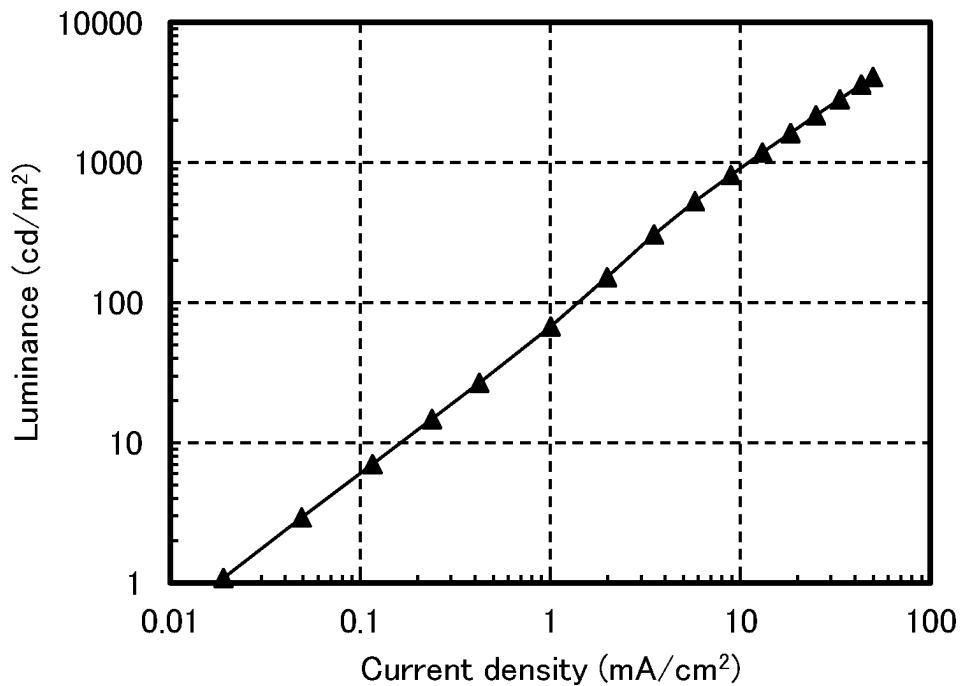


FIG. 41

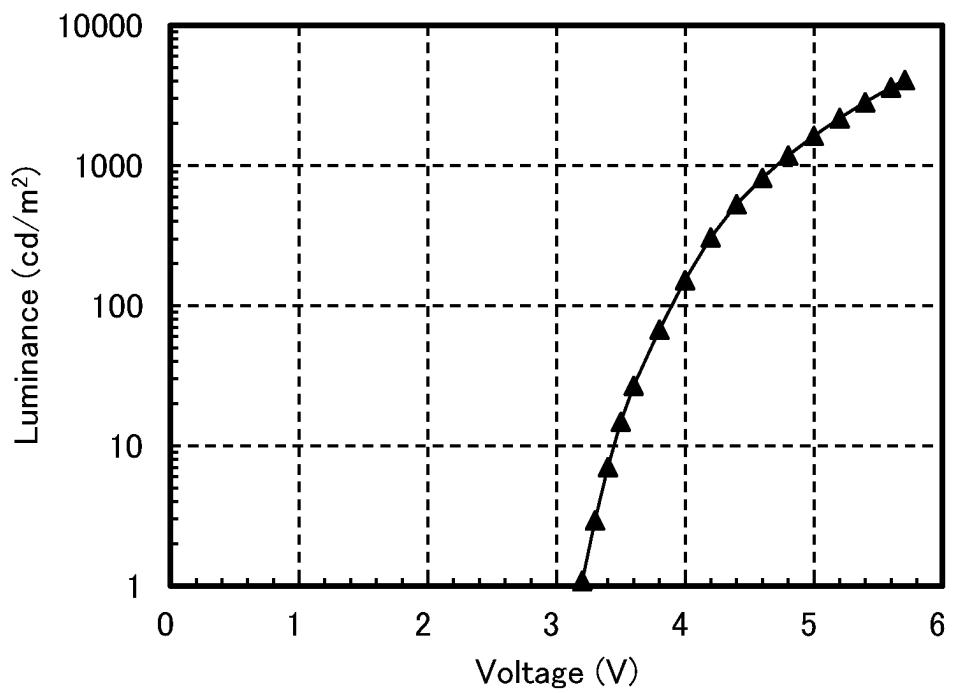


FIG. 42

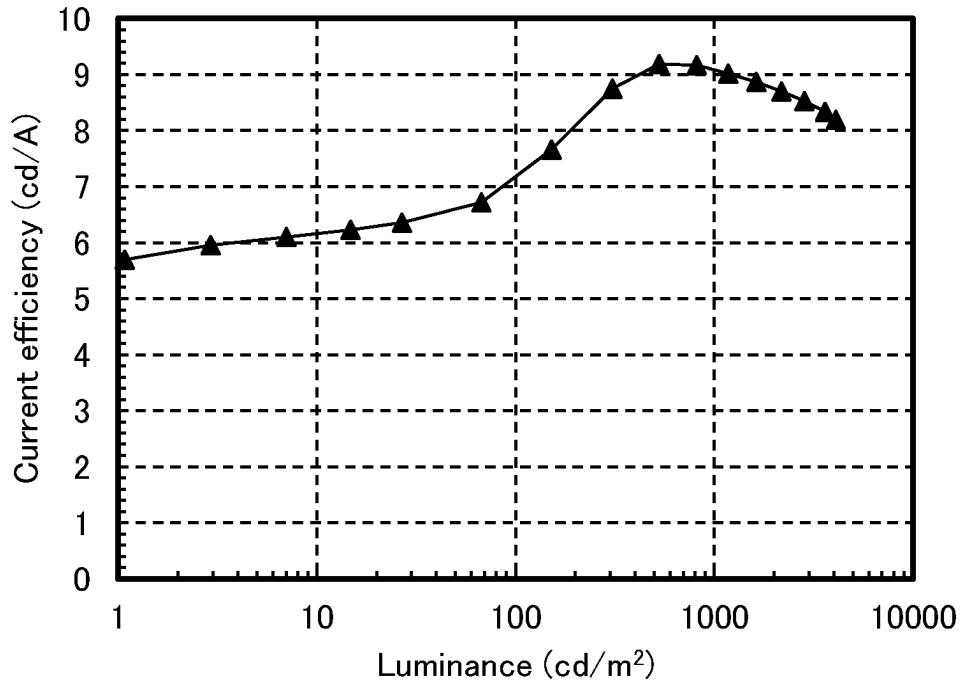


FIG. 43

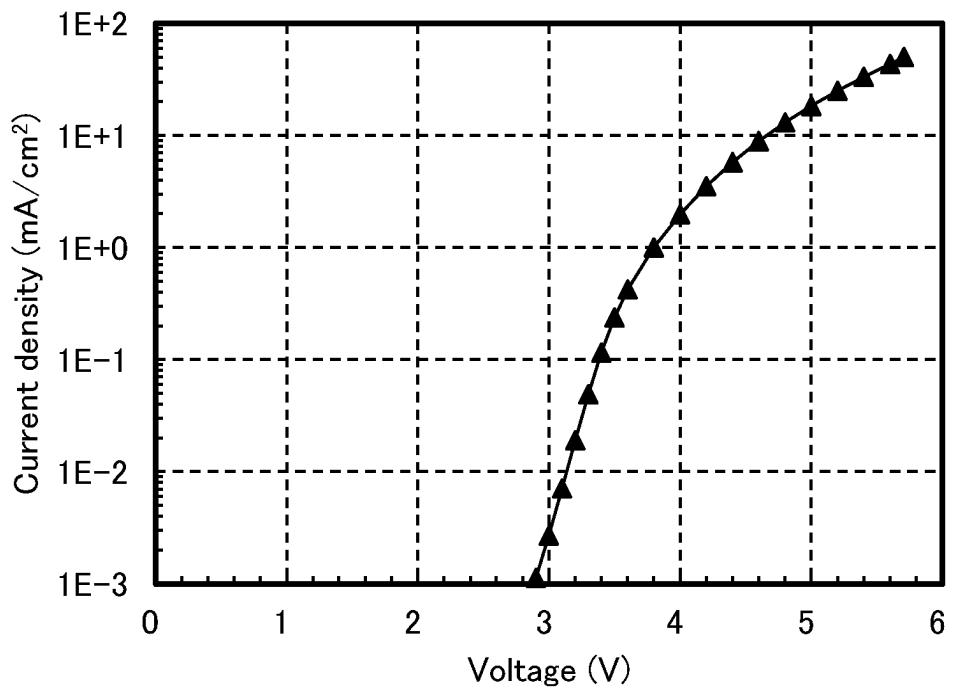


FIG. 44

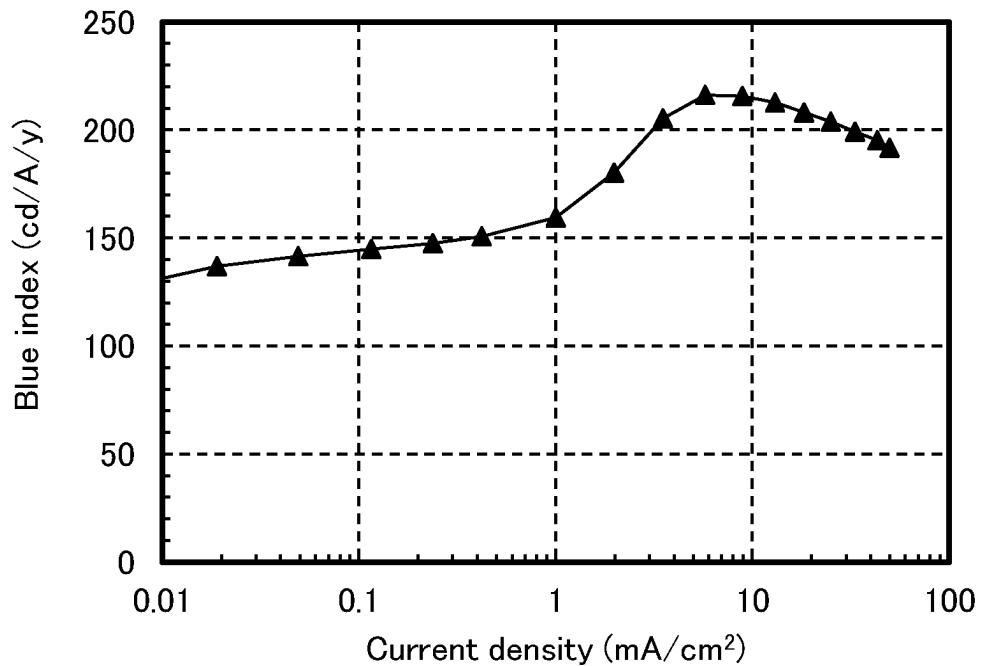


FIG. 45

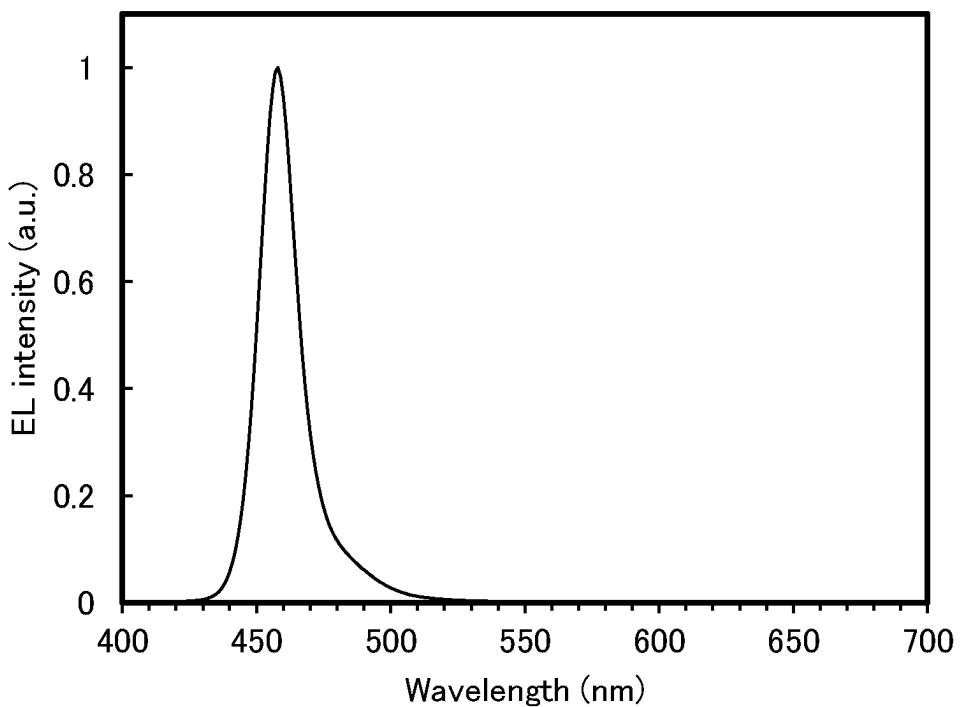


FIG. 46

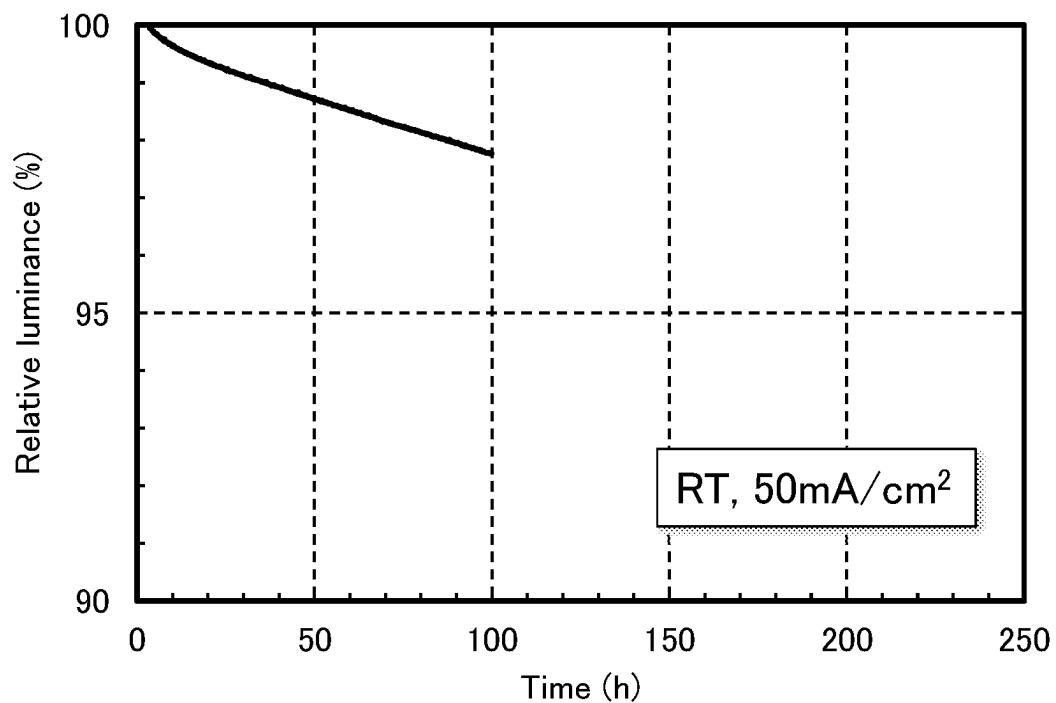


FIG. 47A

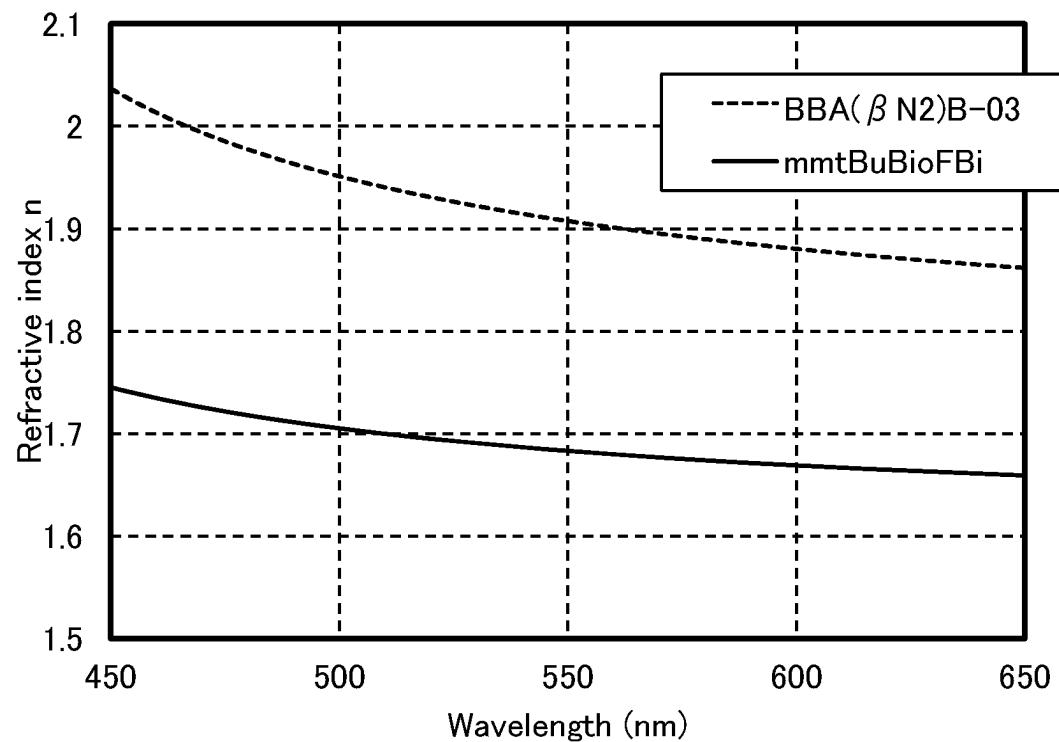


FIG. 47B

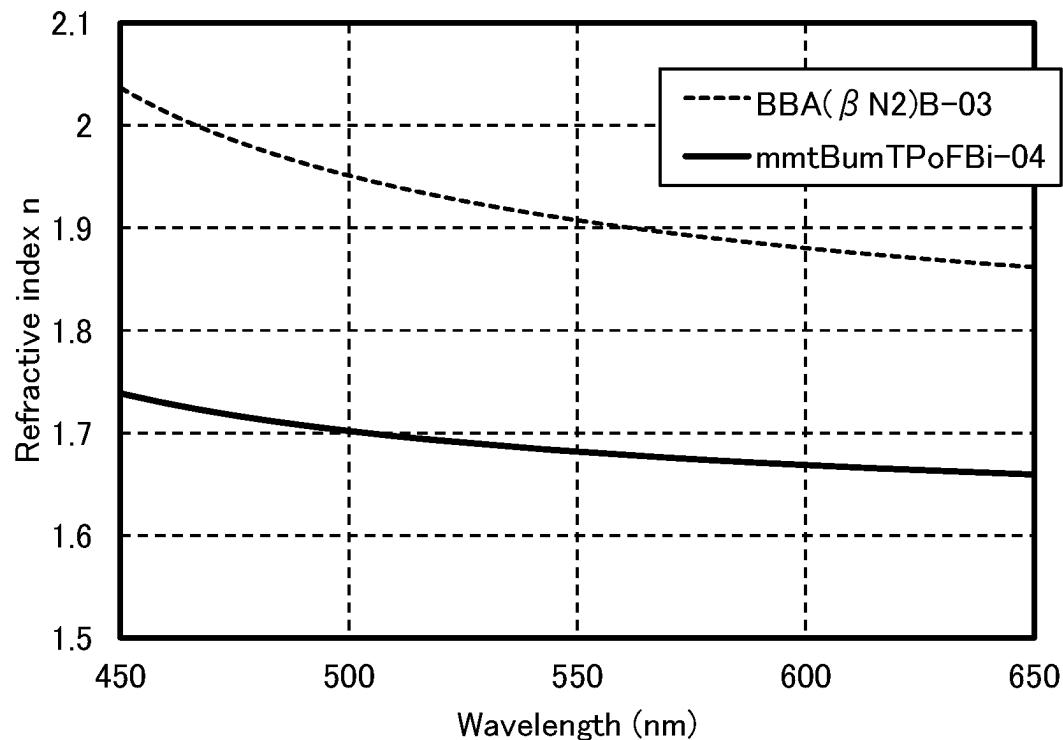


FIG. 48

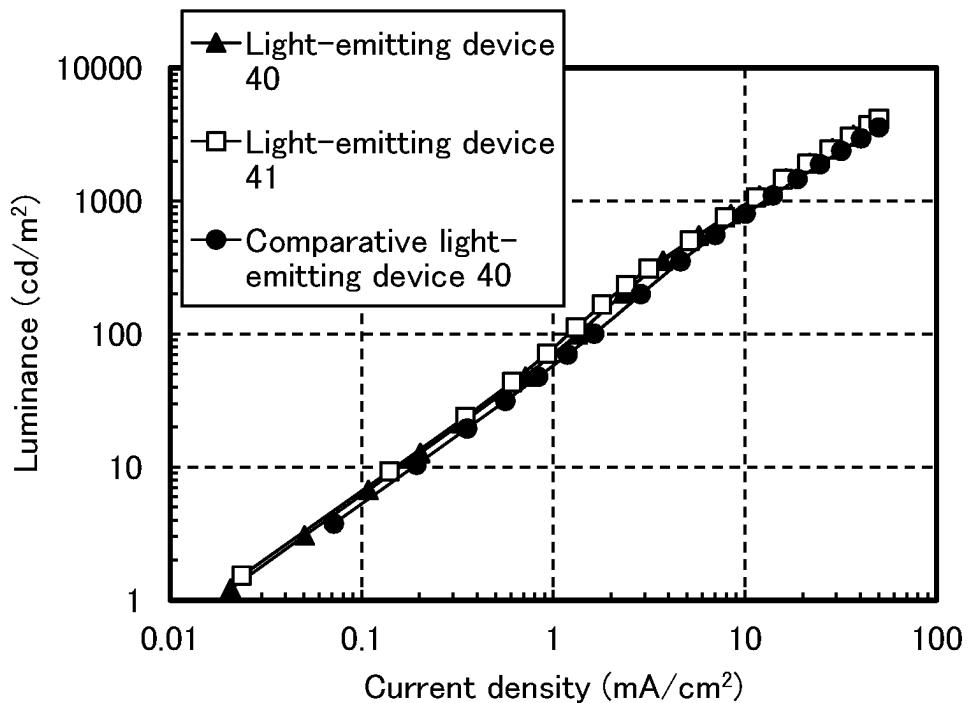


FIG. 49

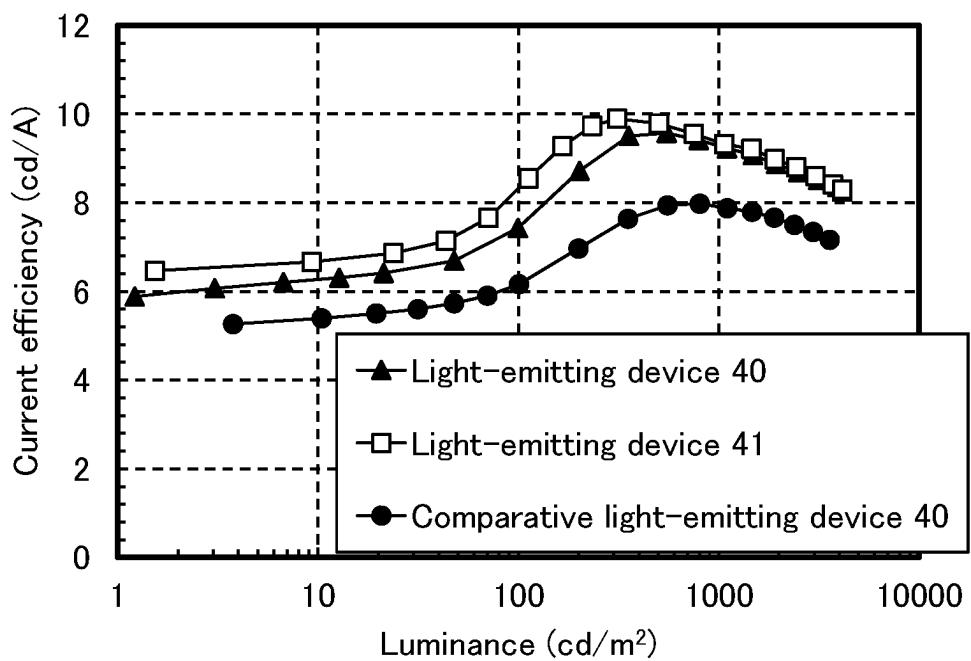


FIG. 50

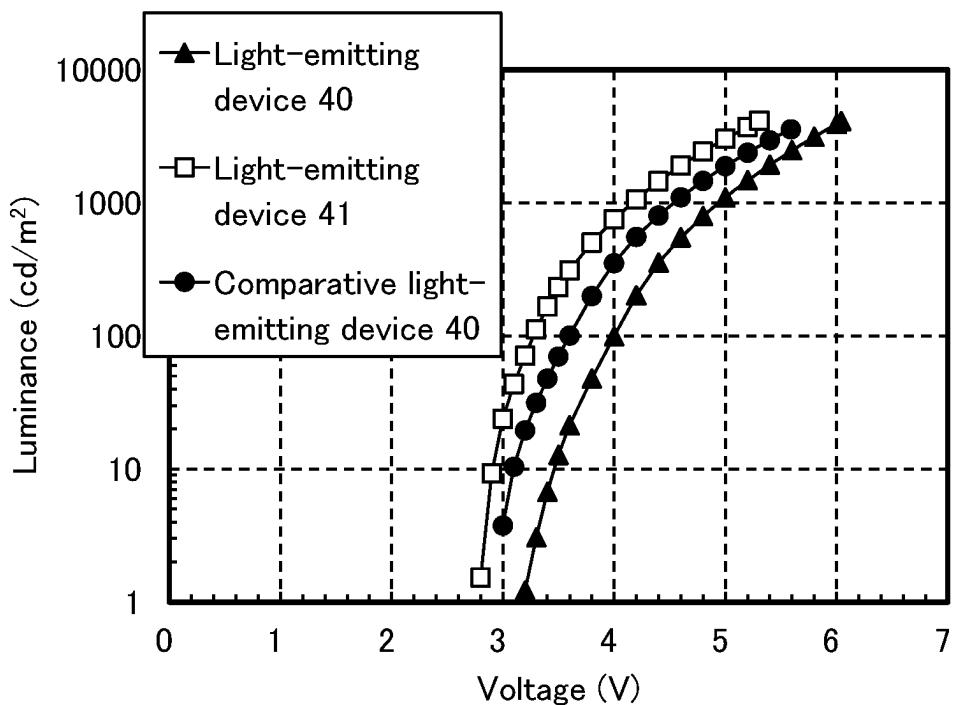


FIG. 51

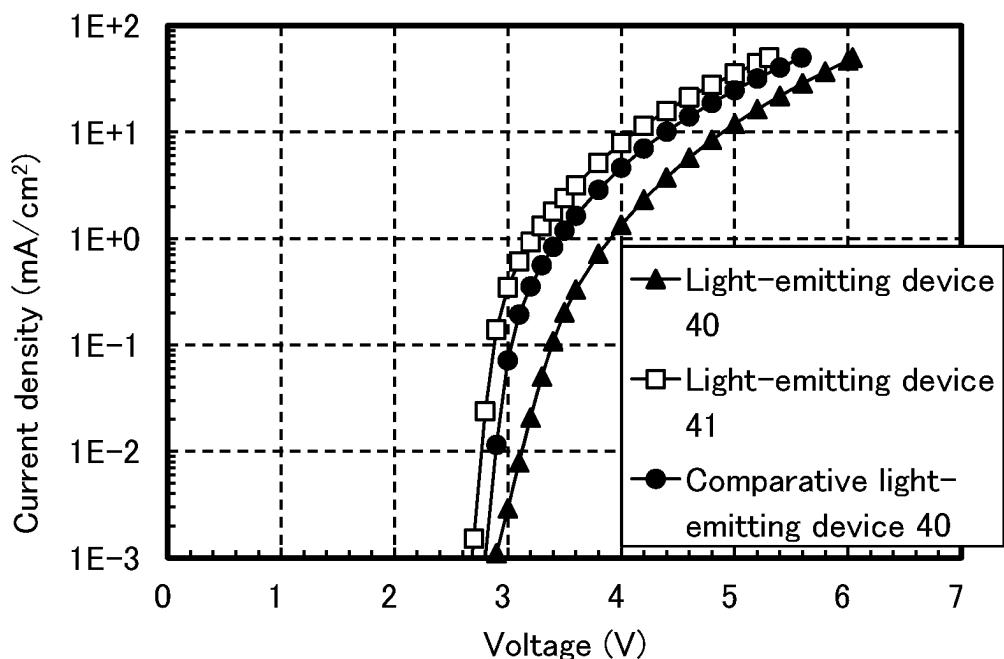


FIG. 52

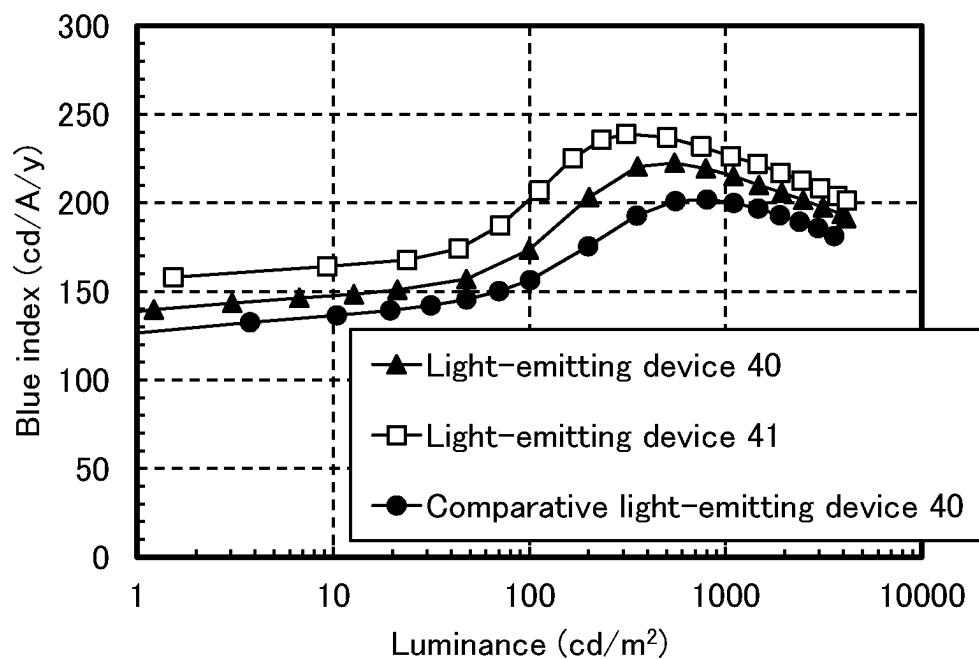


FIG. 53

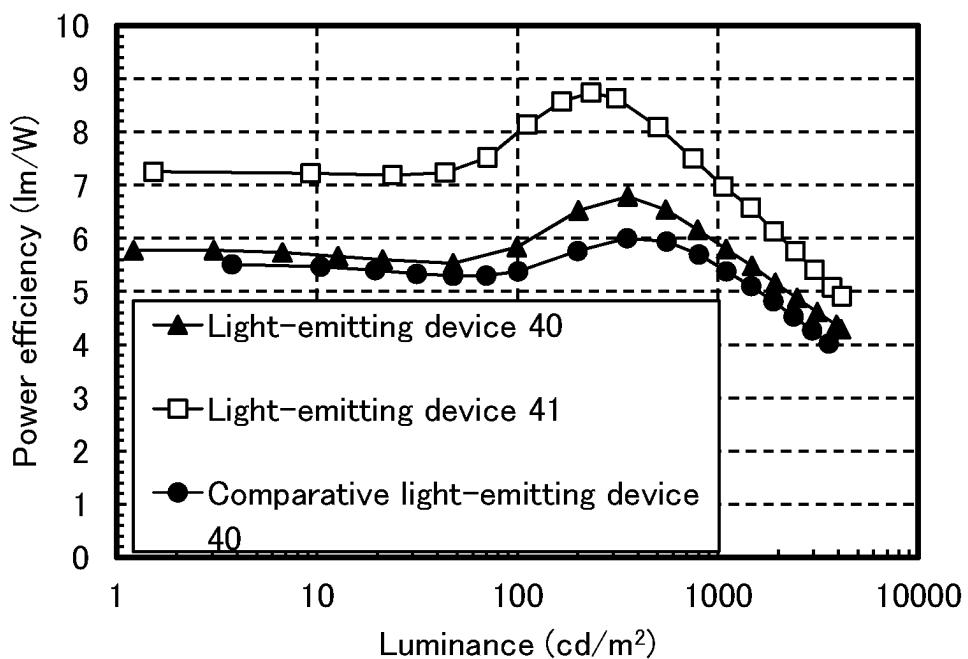


FIG. 54

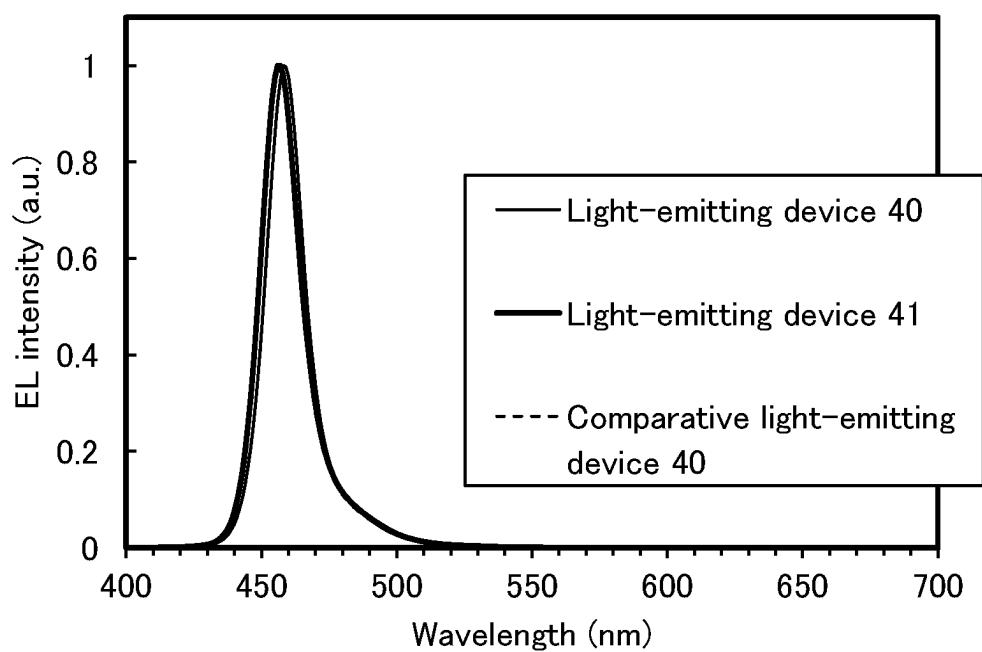


FIG. 55

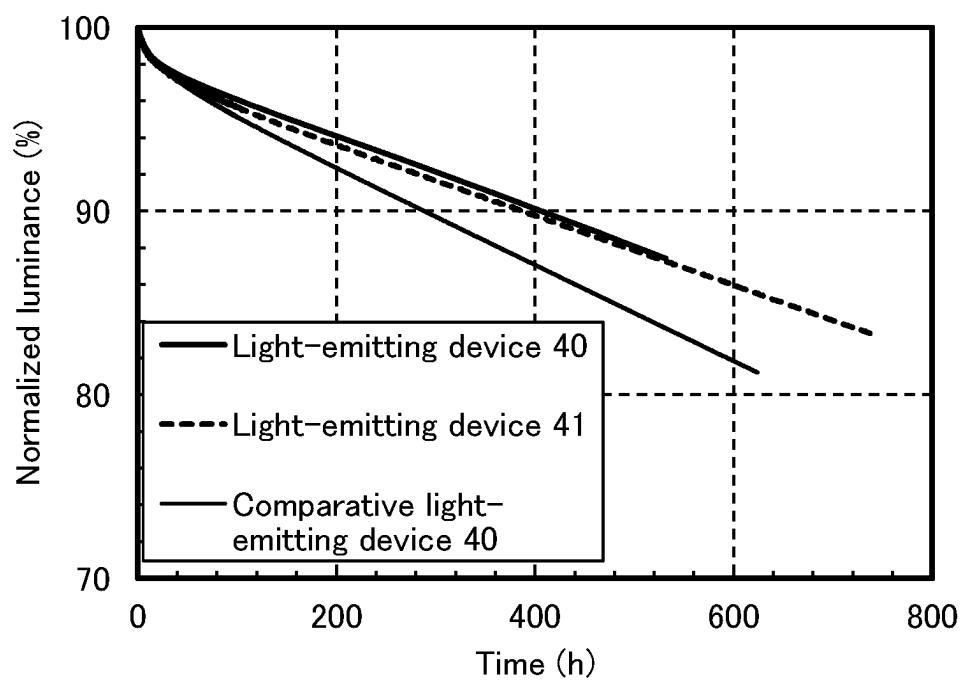


FIG. 56

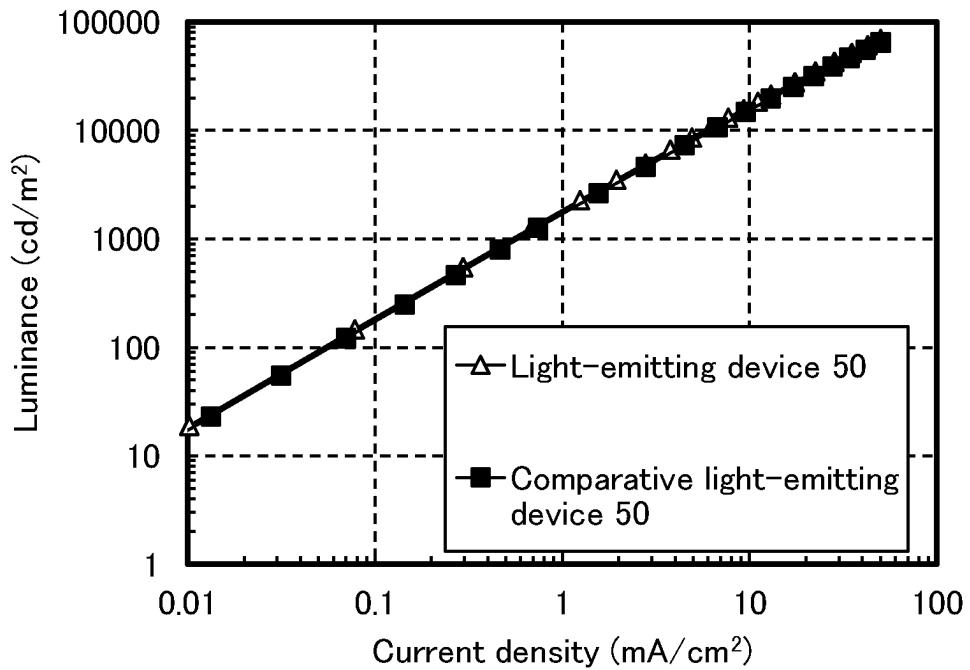


FIG. 57

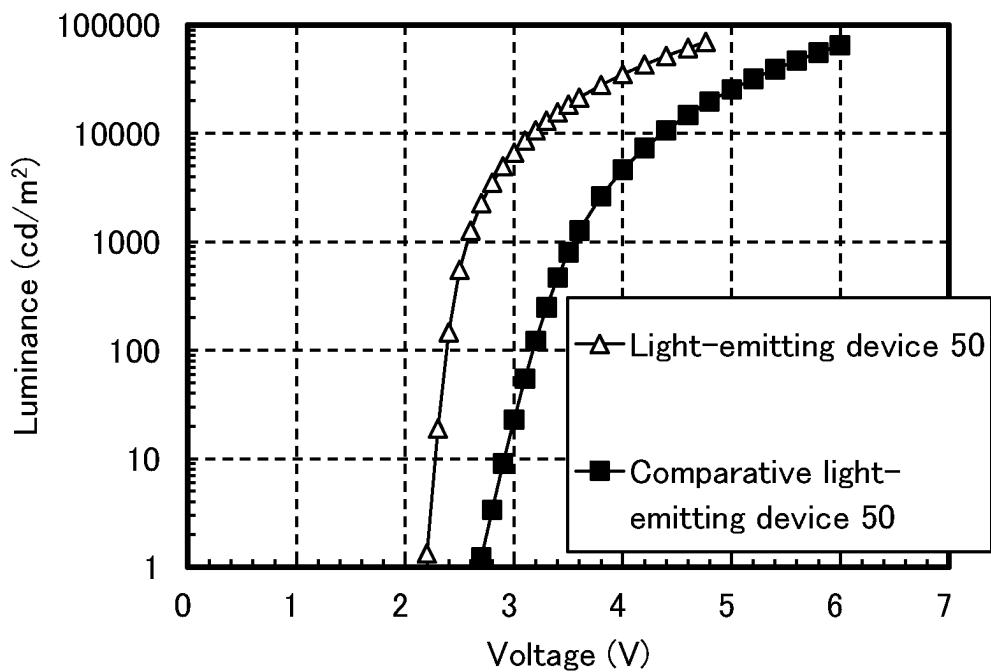


FIG. 58

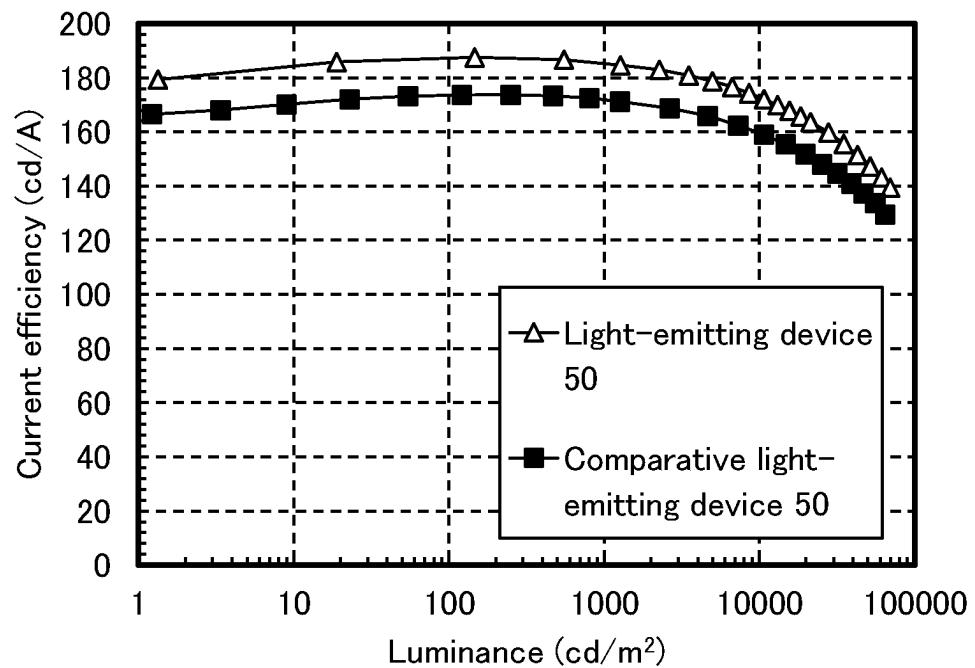


FIG. 59

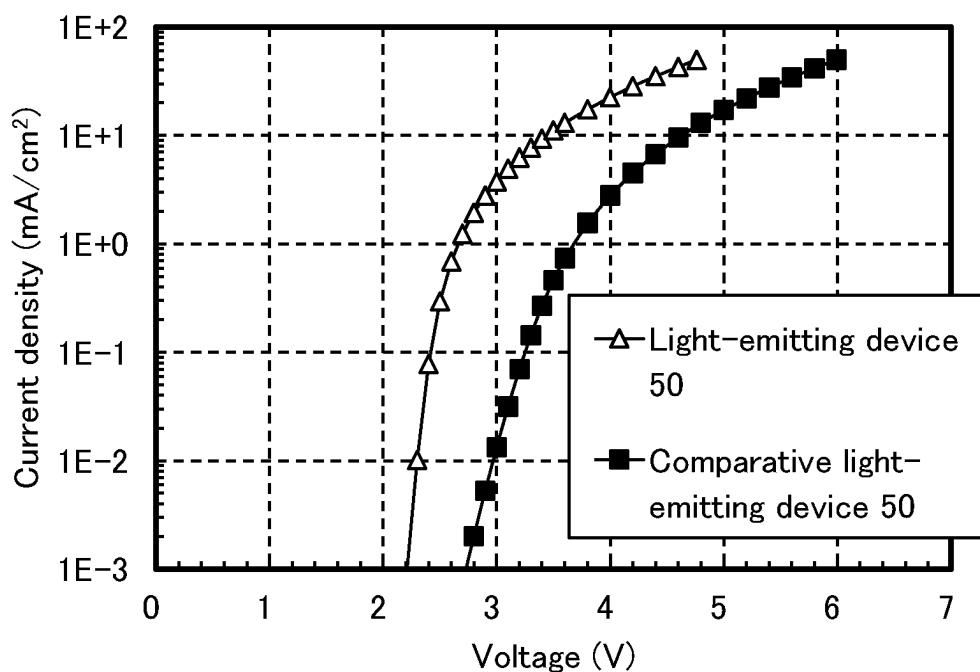


FIG. 60

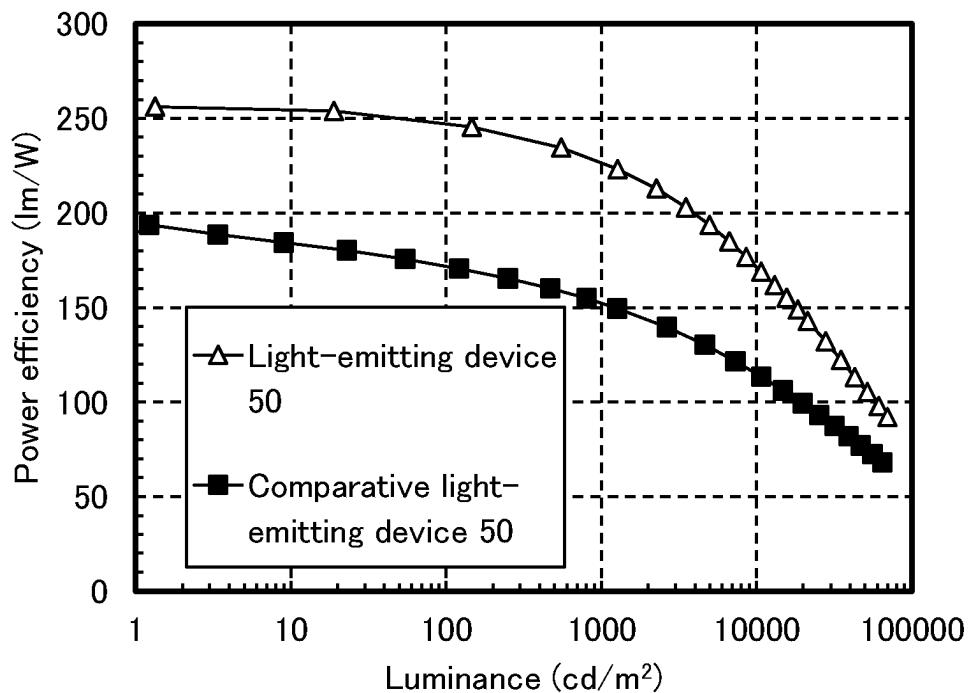
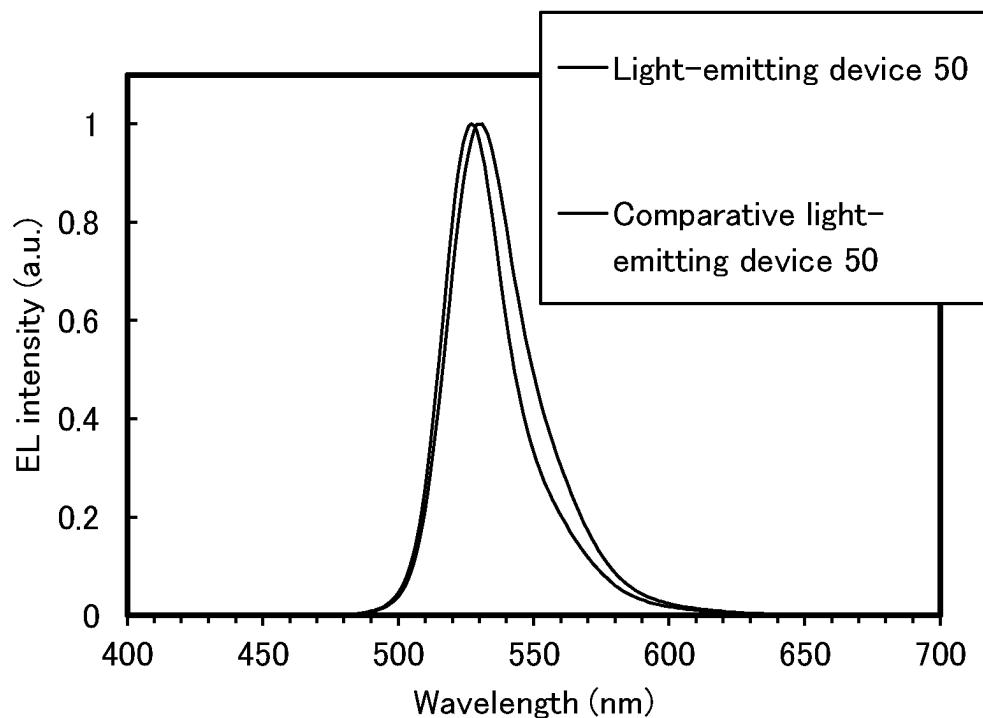


FIG. 61



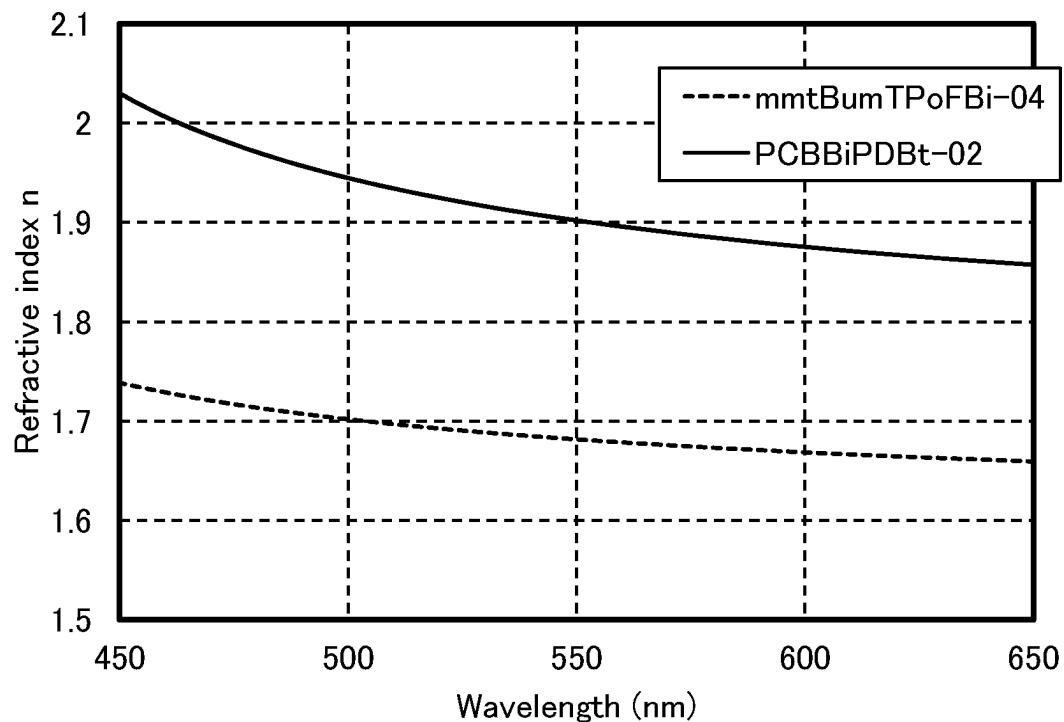
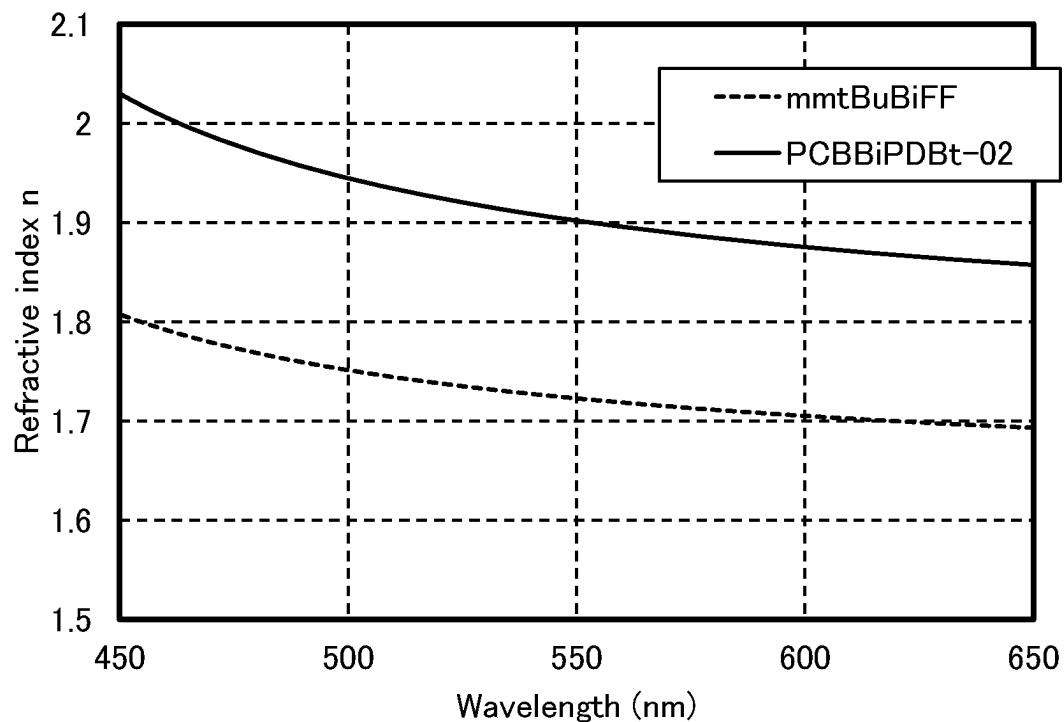
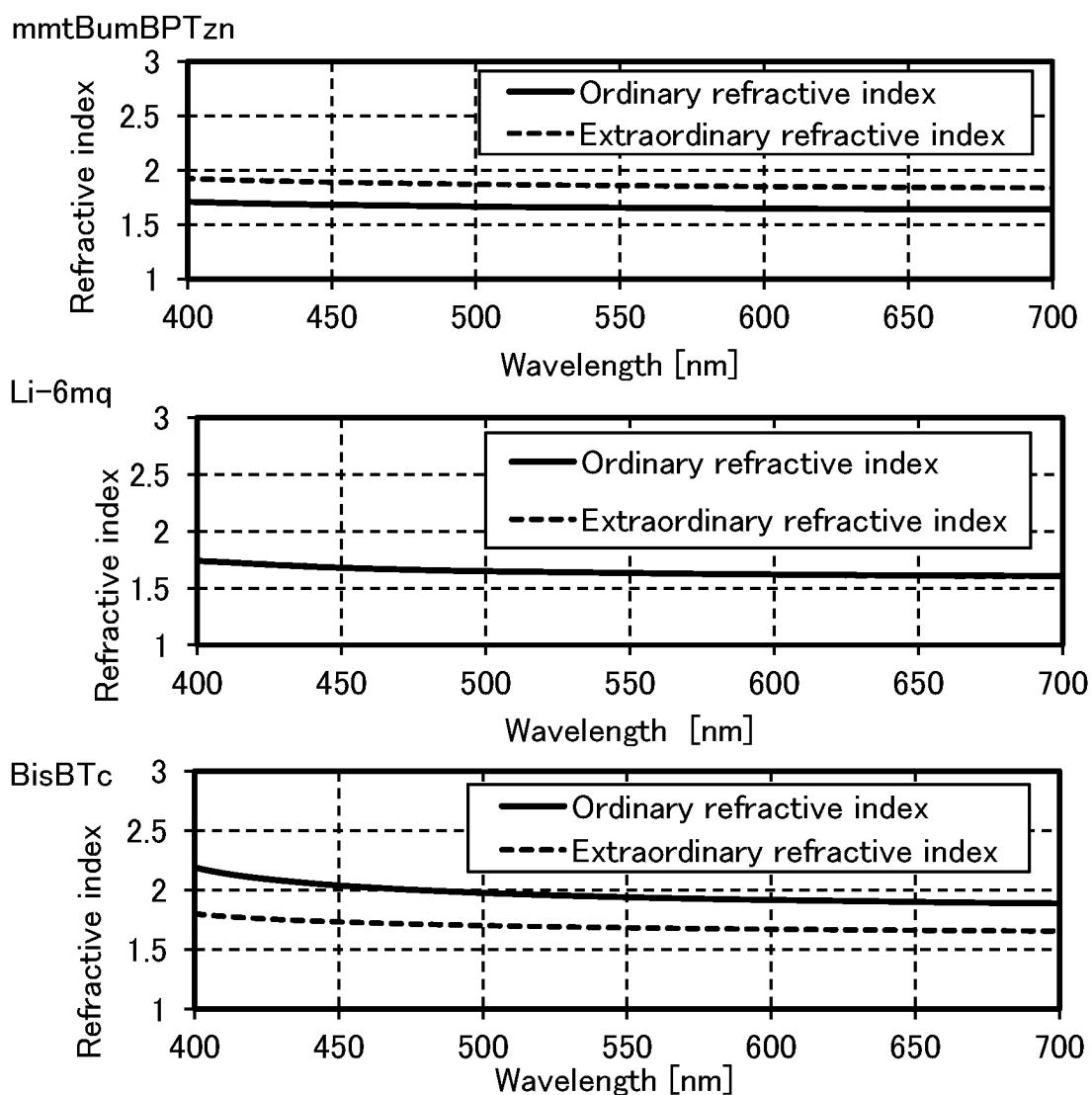
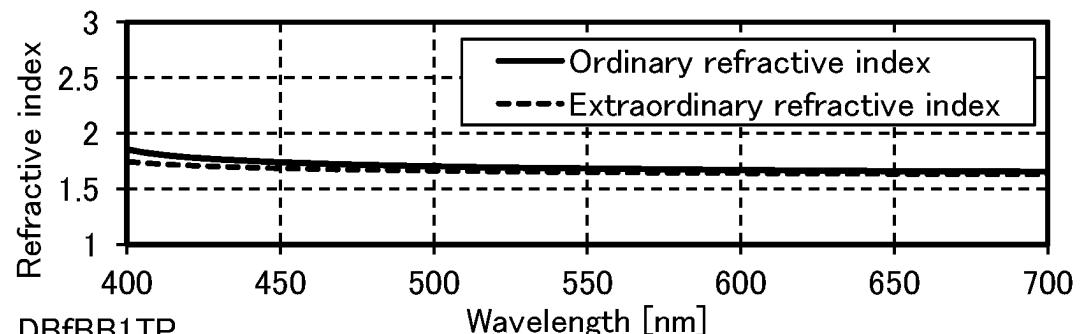
**FIG. 62A****FIG. 62B**

FIG. 63

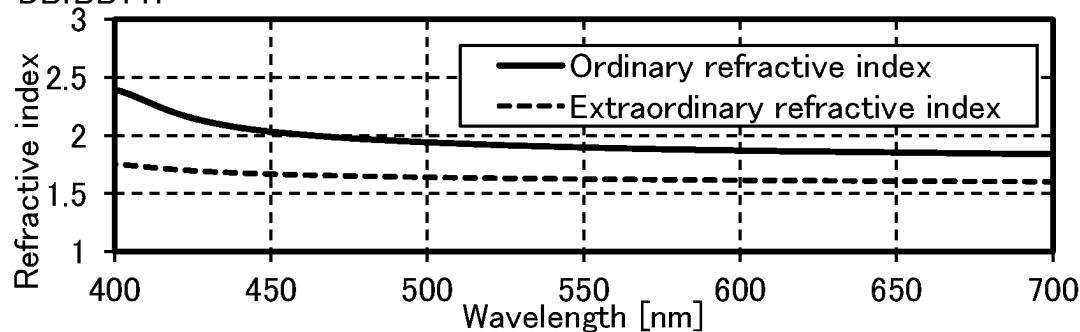


**FIG. 64**

mmtBumTPoFBi-04



DBfBB1TP



mmtBuBioFBi

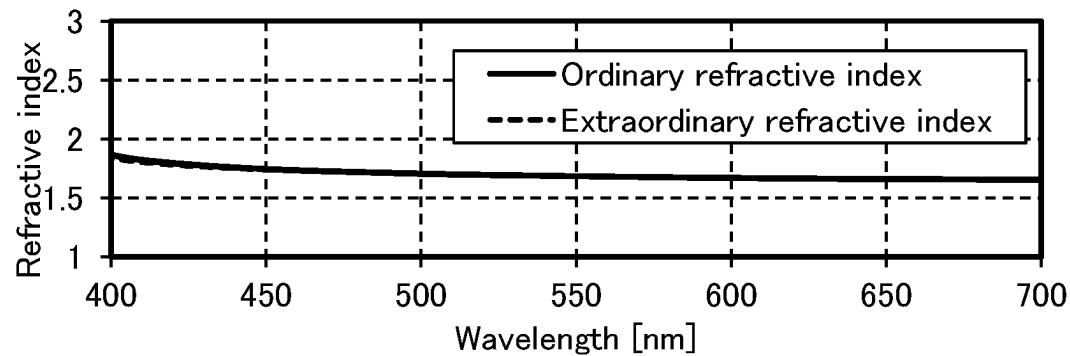


FIG. 65

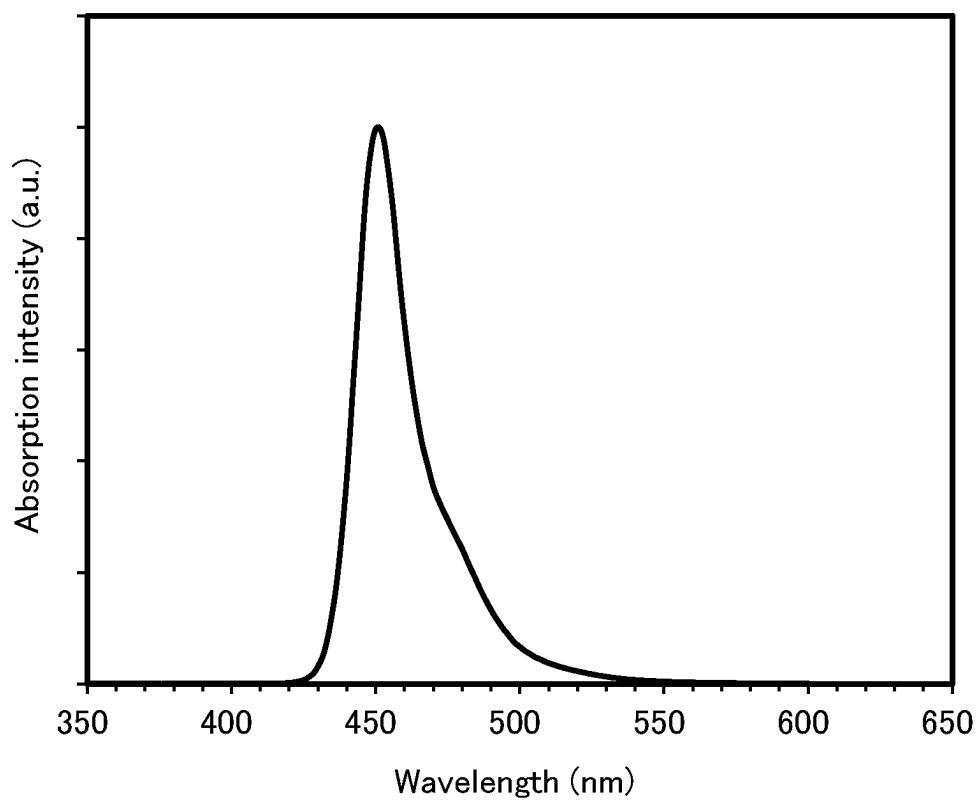


FIG. 66

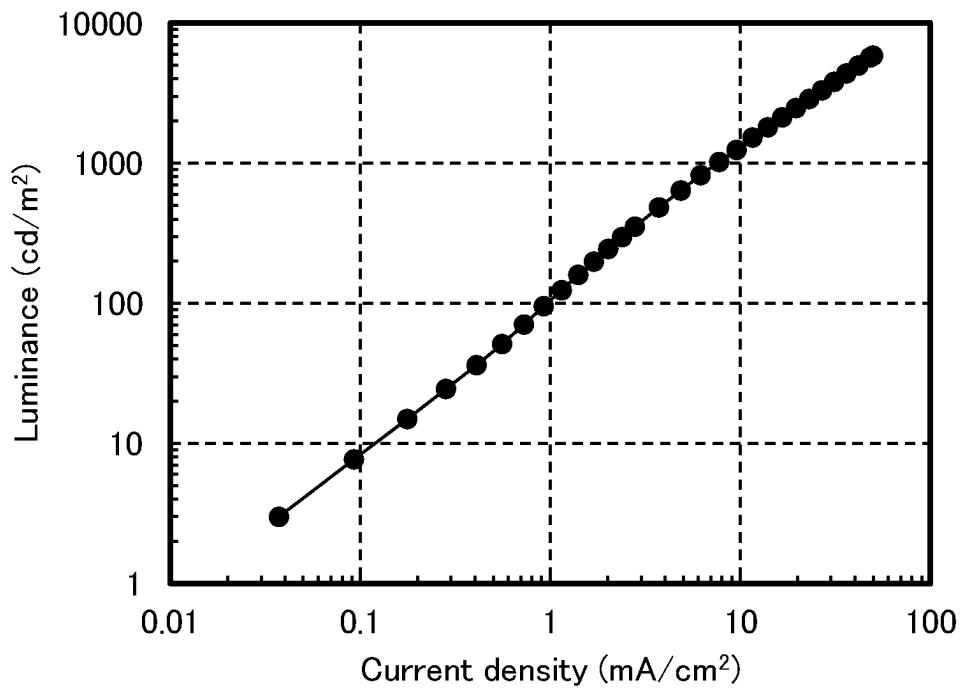


FIG. 67

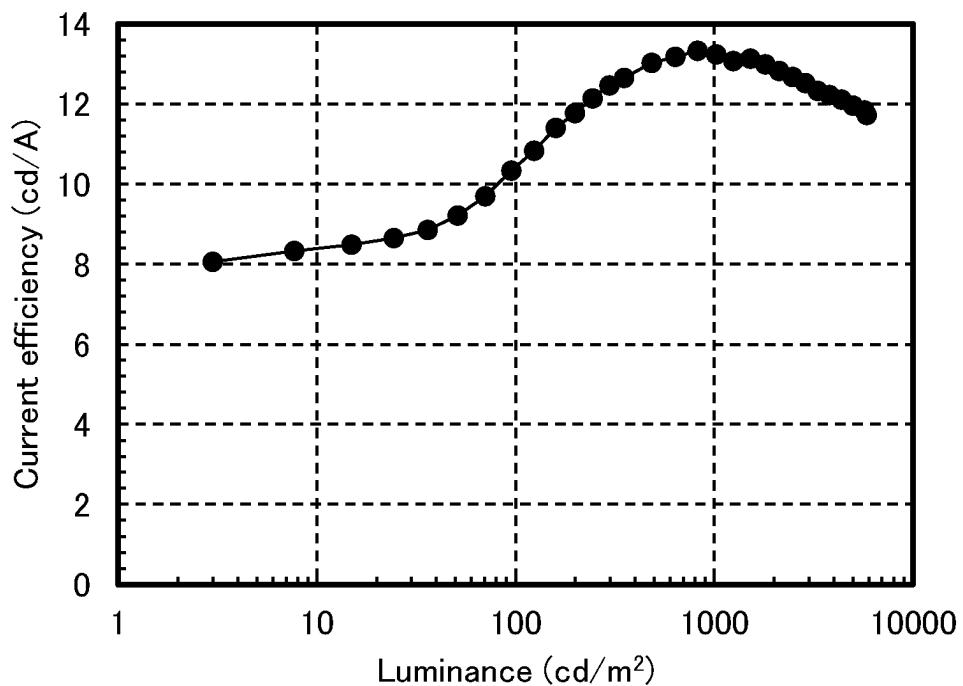


FIG. 68

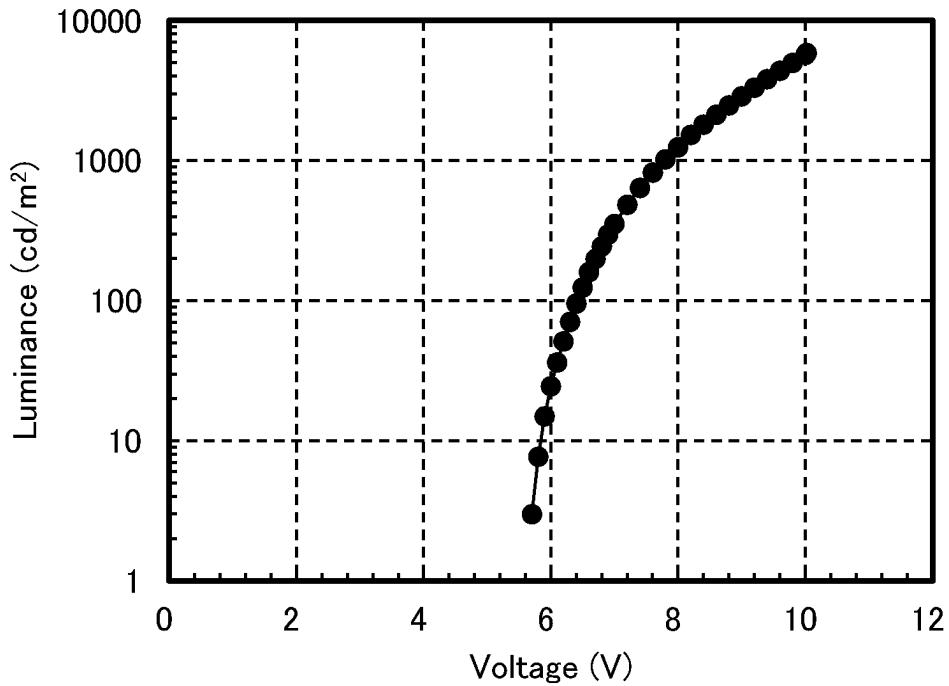


FIG. 69

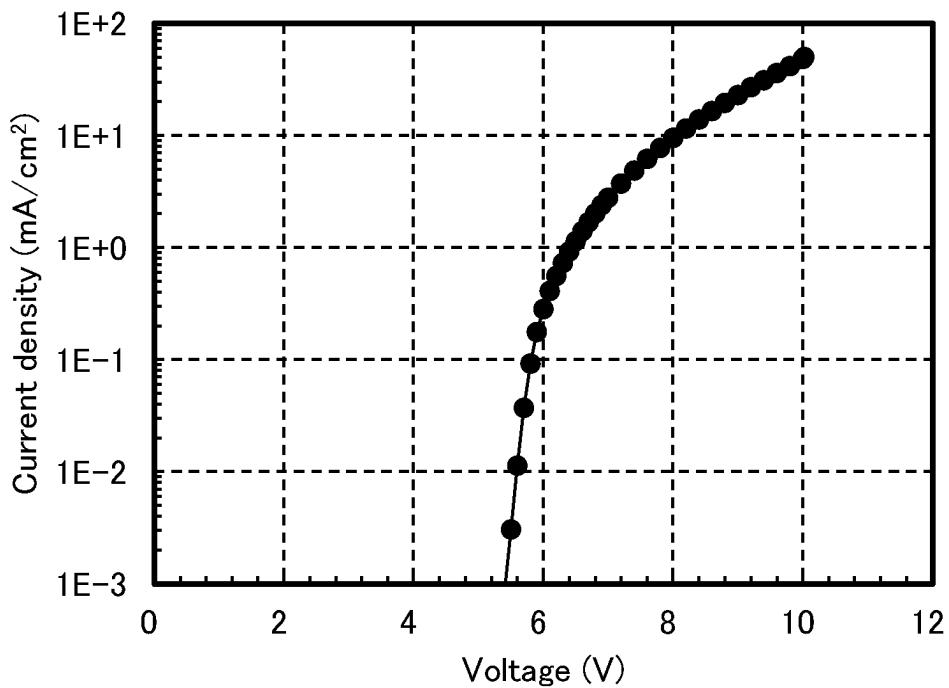


FIG. 70

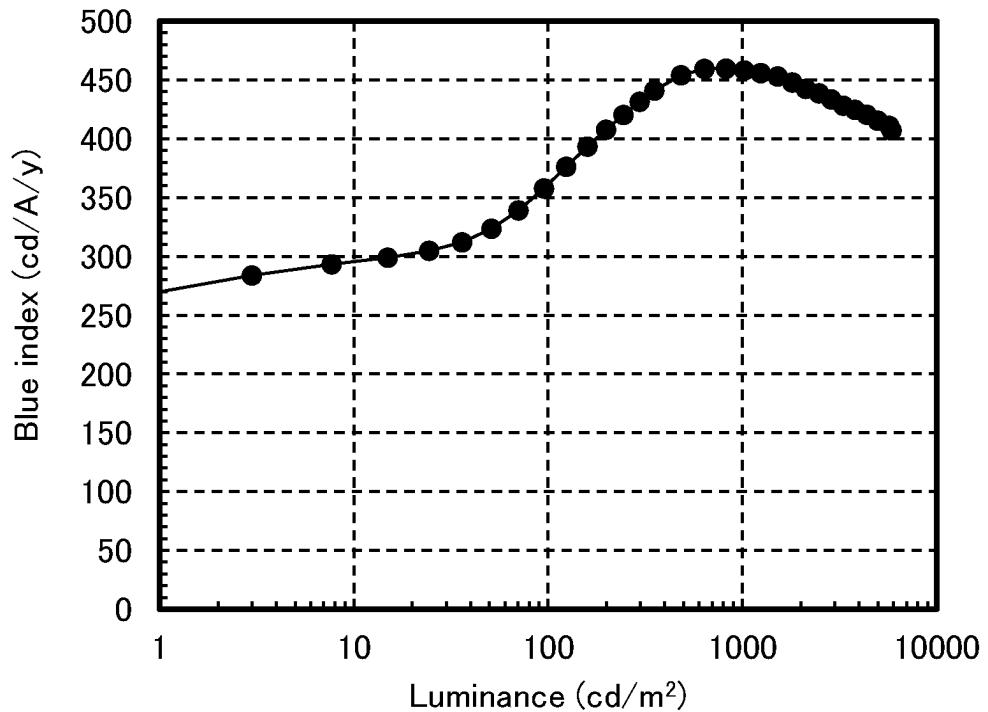
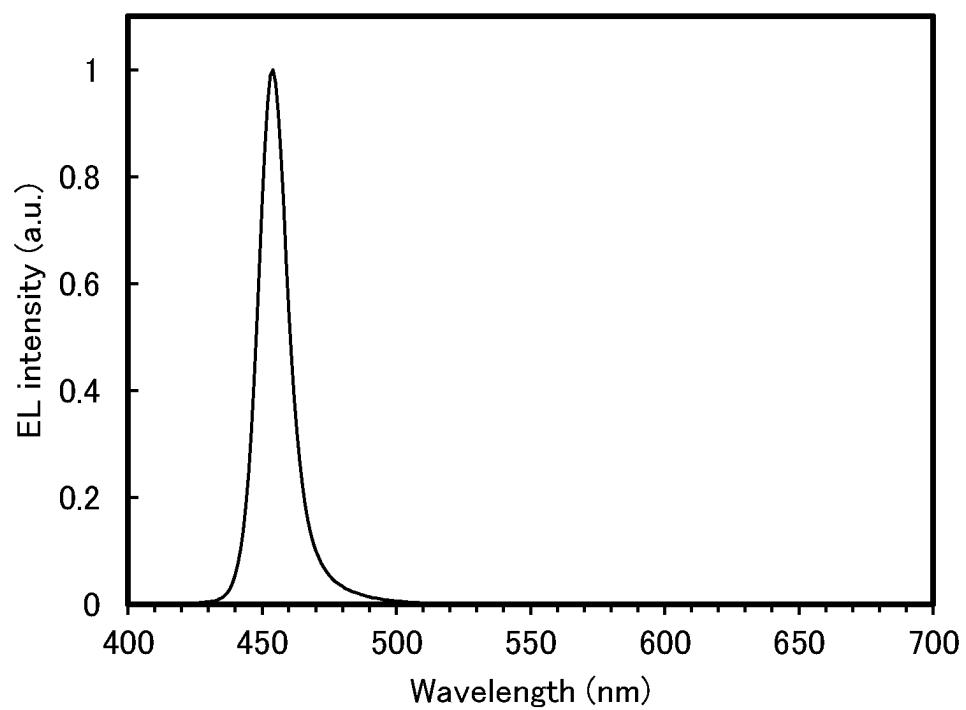


FIG. 71



**LIGHT-EMITTING DEVICE, DISPLAY DEVICE, LIGHT-EMITTING APPARATUS, ELECTRONIC DEVICE, AND LIGHTING DEVICE**

**BACKGROUND OF THE INVENTION**

**1. Field of the Invention**

One embodiment of the present invention relates to an organic compound, a light-emitting element, a light-emitting device, a display module, a lighting module, a display device, a light-emitting apparatus, an electronic device, a lighting device, and an electronic apparatus. Note that one embodiment of the present invention is not limited to the above technical field. The technical field of one embodiment of the invention disclosed in this specification and the like relates to an object, a method, or a manufacturing method. One embodiment of the present invention relates to a process, a machine, manufacture, or a composition of matter. Specifically, examples of the technical field of one embodiment of the present invention disclosed in this specification include a semiconductor device, a display device, a liquid crystal display device, a light-emitting apparatus, a lighting device, a power storage device, a memory device, an imaging device, a driving method thereof, and a manufacturing method thereof.

**2. Description of the Related Art**

Light-emitting devices (organic EL devices) utilizing electroluminescence (EL) of organic compounds have been put to more practical use. In the basic structure of such light-emitting devices, an organic compound layer containing a light-emitting material (an EL layer) is interposed between a pair of electrodes. Carriers are injected by application of voltage to the device, and recombination energy of the carriers is used, whereby light emission can be obtained from the light-emitting material.

Such light-emitting devices are of self-luminous type and thus have advantages over liquid crystal displays, such as high visibility and no need for backlight when used as pixels of a display, and are particularly suitable for flat panel displays. Displays including such light-emitting devices are also highly advantageous in that they can be thin and lightweight. Moreover, such light-emitting devices also have a feature of extremely fast response speed.

Since light-emitting layers of such light-emitting devices can be successively formed two-dimensionally, planar light emission can be achieved. This feature is difficult to realize with point light sources typified by incandescent lamps and LEDs or linear light sources typified by fluorescent lamps; thus, the light-emitting devices also have great potential as planar light sources, which can be applied to lighting devices and the like.

Displays or lighting devices including light-emitting devices are suitable for a variety of electronic devices as described above, and research and development of light-emitting devices have progressed for more favorable characteristics.

Low outcoupling efficiency is often a problem in an organic EL device. In order to improve the outcoupling efficiency, a structure including a layer formed using a low refractive index material in an EL layer has been proposed (see Patent Document 1, for example).

**REFERENCE**

**Patent Document**

- 5 [Patent Document 1] United States Patent Application Publication No. 2020/0176692

**Non-Patent Document**

- 10 [Non-Patent Document 1] Y. Noguchi et al., "Spontaneous Orientation Polarization of Polar Molecules and Interface Properties of Organic Electronic Devices", *Journal of the Vacuum Society of Japan*, 2015, Vol. 58, No. 3.

**SUMMARY OF THE INVENTION**

An object of one embodiment of the present invention is to provide a light-emitting device with high emission efficiency. Another object of one embodiment of the present invention is to provide any of a light-emitting device, a light-emitting apparatus, an electronic device, a display device, and an electronic apparatus each having low power consumption.

It is only necessary that at least one of the above-described objects be achieved in the present invention.

One embodiment of the present invention is a light-emitting device including a first electrode, a second electrode, and an EL layer positioned between the first electrode and the second electrode. The EL layer includes at least a light-emitting layer, a first layer, a second layer, and a third layer. The first layer is positioned between the first electrode and the light-emitting layer. The third layer is positioned between the first layer and the light-emitting layer. The second layer is positioned between the first layer and the third layer. The first layer includes a first organic compound. The second layer includes a second organic compound. The third layer includes a third organic compound. The ordinary refractive index of the second organic compound with respect to light with any of the wavelengths greater than or equal to 450 nm and less than or equal to 650 nm is higher than the ordinary refractive index of the first organic compound with respect to light with any of the wavelengths and the ordinary refractive index of the third organic compound with respect to light with any of the wavelengths.

Another embodiment of the present invention is a light-emitting device including a first electrode, a second electrode, and an EL layer positioned between the first electrode and the second electrode. The EL layer includes at least a light-emitting layer, a first layer, a second layer, and a third layer. The first layer is positioned between the first electrode and the light-emitting layer. The third layer is positioned between the first layer and the light-emitting layer. The second layer is positioned between the first layer and the third layer. The first layer includes a first organic compound.

The second layer includes a second organic compound. The third layer includes a third organic compound. The ordinary refractive index of the second organic compound with respect to light with any of the wavelengths greater than or equal to 455 nm and less than or equal to 465 nm is higher than the ordinary refractive index of the first organic compound with respect to light with any of the wavelengths and the ordinary refractive index of the third organic compound with respect to light with any of the wavelengths.

Another embodiment of the present invention is a light-emitting device including a first electrode, a second electrode, and an EL layer positioned between the first electrode and the second electrode. The EL layer includes at least a

light-emitting layer, a first layer, a second layer, and a third layer. The first layer is positioned between the first electrode and the light-emitting layer. The third layer is positioned between the first layer and the light-emitting layer. The second layer is positioned between the first layer and the third layer. The first layer includes a first organic compound. The second layer includes a second organic compound. The third layer includes a third organic compound. The ordinary refractive index of the second organic compound with respect to light with a wavelength of  $\lambda$  nm is higher than the ordinary refractive index of the first organic compound with respect to light with a wavelength of  $\lambda$  nm and the ordinary refractive index of the third organic compound with respect to light with a wavelength of  $\lambda$  nm.

Another embodiment of the present invention is a light-emitting device including a first electrode, a second electrode, and an EL layer positioned between the first electrode and the second electrode. The EL layer includes at least a light-emitting layer, a first layer, a second layer, and a third layer. The first layer is positioned between the first electrode and the light-emitting layer. The third layer is positioned between the first layer and the light-emitting layer. The second layer is positioned between the first layer and the third layer. The light-emitting layer includes a light-emitting material that emits light having a peak wavelength of  $\lambda$  nm. The first layer includes a first organic compound. The second layer includes a second organic compound. The third layer includes a third organic compound. The ordinary refractive index of the second organic compound with respect to light with the wavelength of  $\lambda$  nm is higher than the ordinary refractive index of the first organic compound with respect to light with the wavelength of  $\lambda$  nm and the ordinary refractive index of the third organic compound with respect to light with the wavelength of  $\lambda$  nm.

Another embodiment of the present invention is the light-emitting device having the above structure, in which a difference between the HOMO levels of the first organic compound and the third organic compound is less than or equal to 0.2 eV.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the first organic compound and the third organic compound are the same organic compound.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the third layer does not include fluorine.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the first layer to the third layer each have a thickness larger than or equal to 25 nm and smaller than or equal to 70 nm.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the thickness of the second layer is larger than the thickness of the first layer and the thickness of the third layer.

Another embodiment of the present invention is the light-emitting device having the above structure, in which a difference between the ordinary refractive indices of the second organic compound and the first organic compound with respect to light with the wavelength of  $\lambda$  nm and a difference between the ordinary refractive indices of the second organic compound and the third organic compound with respect to light with the wavelength of  $\lambda$  nm are each greater than or equal to 0.2 and less than 0.5.

Another embodiment of the present invention is the light-emitting device having the above structure, in which a difference between the ordinary refractive indices of the first

organic compound and the third organic compound with respect to light with the wavelength of  $\lambda$  nm is less than or equal to 0.1.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the product of the thickness of the second layer and the ordinary refractive index of the second organic compound with respect to light with the wavelength of  $\lambda$  nm is greater than or equal to 70% and less than or equal to 130% of  $\lambda$ .

Another embodiment of the present invention is the light-emitting device having the above structure, in which the  $\lambda$  is a peak wavelength of light emitted from the light-emitting material.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the  $\lambda$  is any of the values greater than or equal to 450 and less than or equal to 650.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the  $\lambda$  is any of the values greater than or equal to 455 and less than or equal to 465.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the  $\lambda$  is 633.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the first electrode includes a reflective electrode.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the value obtained by subtracting GSP (mV/nm) of the second layer from GSP (mV/nm) of the third layer is less than or equal to 10 (mV/nm).

Another embodiment of the present invention is the light-emitting device having the above structure, in which the value obtained by subtracting GSP (mV/nm) of the first layer from GSP (mV/nm) of the second layer is less than or equal to 10 (mV/nm).

Another embodiment of the present invention is the light-emitting device having the above structure, in which GSP (mV/nm) of the third layer is larger than GSP (mV/nm) of the second layer.

Another embodiment of the present invention is the light-emitting device having the above structure, in which GSP (mV/nm) of the third layer is larger than GSP (mV/nm) of the first layer.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the GSP (mV/nm) of the second layer is larger than the GSP (mV/nm) of the first layer.

Another embodiment of the present invention is the light-emitting device having the above structure, in which the first organic compound and the third organic compound are different organic compounds.

Another embodiment of the present invention is a display device including a first light-emitting device and a second light-emitting device. The first light-emitting device has any of the above structures. The second light-emitting device includes an EL layer between a third electrode and the second electrode. The EL layer of the second light-emitting device includes at least a light-emitting layer, a fourth layer, a fifth layer, and a sixth layer. The fourth layer, the fifth layer, and the sixth layer respectively have the same structure as the first layer, the second layer, and the third layer. A peak wavelength of light emitted from the light-emitting material included in the light-emitting layer of the first light-emitting device is different from a peak wavelength of light emitted from a light-emitting material included in the

light-emitting layer of the second light-emitting device by more than or equal to 30 nm.

Another embodiment of the present invention is a display device including any of the above light-emitting devices, and a sensor, an operation button, a speaker, or a microphone.

Another embodiment of the present invention is an electronic device including the above display device, and a sensor, an operation button, a speaker, or a microphone.

Another embodiment of the present invention is a light-emitting apparatus including any of the above light-emitting devices, and a transistor or a substrate.

Another embodiment of the present invention is a lighting device including any of the above light-emitting devices and a housing.

Note that the light-emitting apparatus in this specification includes, in its category, an image display device that uses a light-emitting device. The light-emitting apparatus may also include a module in which a light-emitting device is provided with a connector such as an anisotropic conductive film or a tape carrier package (TCP), a module in which a printed wiring board is provided at the end of a TCP, and a module in which an integrated circuit (IC) is directly mounted on a light-emitting device by a chip on glass (COG) method. Furthermore, a lighting device or the like may include the light-emitting apparatus.

According to one embodiment of the present invention, a light-emitting device with high emission efficiency can be provided. According to another embodiment of the present invention, any of a light-emitting device, a light-emitting apparatus, an electronic apparatus, a display device, and an electronic apparatus each having low power consumption can be provided.

Note that the description of these effects does not preclude the existence of other effects. One embodiment of the present invention does not necessarily have all these effects. Other effects will be apparent from and can be derived from the description of the specification, the drawings, the claims, and the like.

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIGS. 1A to 1D are schematic views of light-emitting devices.

FIGS. 2A and 2B illustrate an active matrix light-emitting apparatus.

FIGS. 3A and 3B illustrate active matrix light-emitting apparatuses.

FIG. 4 illustrates an active matrix light-emitting apparatus.

FIGS. 5A and 5B illustrate a passive matrix light-emitting apparatus.

FIGS. 6A and 6B illustrate a lighting device.

FIGS. 7A, 7B1, 7B2, and 7C illustrate electronic devices.

FIGS. 8A to 8C illustrate electronic devices.

FIG. 9 illustrates a lighting device.

FIG. 10 illustrates a lighting device.

FIG. 11 illustrates in-vehicle display devices and lighting devices.

FIGS. 12A and 12B illustrate an electronic device.

FIGS. 13A to 13C illustrate an electronic device.

FIGS. 14A to 14D show a structure example of a display device.

FIGS. 15A to 15F show a manufacturing method example of a display device.

FIGS. 16A to 16F show a manufacturing method example of a display device.

FIG. 17 shows the refractive indices of PCBDBtBB-02 and mmTBumTPoFBi-02.

FIG. 18 shows the luminance-current density characteristics of a light-emitting device 1, a comparative light-emitting device 1, and a comparative light-emitting device 2.

FIG. 19 shows the luminance-voltage characteristics of the light-emitting device 1, the comparative light-emitting device 1, and the comparative light-emitting device 2.

FIG. 20 shows the current efficiency-luminance characteristics of the light-emitting device 1, the comparative light-emitting device 1, and the comparative light-emitting device 2.

FIG. 21 shows the current density-voltage characteristics of the light-emitting device 1, the comparative light-emitting device 1, and the comparative light-emitting device 2.

FIG. 22 shows the blue index-luminance characteristics of the light-emitting device 1, the comparative light-emitting device 1, and the comparative light-emitting device 2.

FIG. 23 shows the emission spectra of the light-emitting device 1, the comparative light-emitting device 1, and the comparative light-emitting device 2.

FIG. 24 shows the refractive indices of BBA( $\beta$ N2)B-03 and mmTBumTPoFBi.

FIG. 25 shows the luminance-current density characteristics of a light-emitting device 10 and a comparative light-emitting device 10.

FIG. 26 shows the luminance-voltage characteristics of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 27 shows the current efficiency-luminance characteristics of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 28 shows the current density-voltage characteristics of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 29 shows the blue index-current density characteristics of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 30 shows the emission spectra of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 31 shows the viewing angle characteristics of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 32 shows the time dependence of relative luminance of the light-emitting device 10 and the comparative light-emitting device 10.

FIG. 33 shows the luminance-current density characteristics of a light-emitting device 20.

FIG. 34 shows the luminance-voltage characteristics of the light-emitting device 20.

FIG. 35 shows the current efficiency-luminance characteristics of the light-emitting device 20.

FIG. 36 shows the current density-voltage characteristics of the light-emitting device 20.

FIG. 37 shows the blue index-luminance characteristics of the light-emitting device 20.

FIG. 38 shows the emission spectrum of the light-emitting device 20.

FIG. 39 shows the time dependence of normalized luminance of the light-emitting device 20.

FIG. 40 shows the luminance-current density characteristics of a light-emitting device 30.

FIG. 41 shows the luminance-voltage characteristics of the light-emitting device 30.

FIG. 42 shows the current efficiency-luminance characteristics of the light-emitting device 30.

FIG. 43 shows the current density-voltage characteristics of the light-emitting device 30.

FIG. 44 shows the blue index-current density characteristics of the light-emitting device 30.

FIG. 45 shows the emission spectrum of the light-emitting device 30.

FIG. 46 shows the time dependence of normalized luminescence of the light-emitting device 30.

FIGS. 47A and 47B show the refractive indices of BBA (BN2)B-03, mmtBuBioFBi, and mmtBumTPoFBi-04.

FIG. 48 shows the luminance-current density characteristics of a light-emitting device 40, a light-emitting device 41, and a comparative light-emitting device 40.

FIG. 49 shows the current efficiency-luminance characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 50 shows the luminance-voltage characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 51 shows the current density-voltage characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 52 shows the blue index-luminance characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 53 shows the power efficiency-luminance characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 54 shows the emission spectra of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 55 shows the time dependence of normalized luminescence of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40.

FIG. 56 shows the luminance-current density characteristics of a comparative light-emitting device 50 and a light-emitting device 50.

FIG. 57 shows the luminance-voltage characteristics of the comparative light-emitting device 50 and the light-emitting device 50.

FIG. 58 shows the current efficiency-luminance characteristics of the comparative light-emitting device 50 and the light-emitting device 50.

FIG. 59 shows the current density-voltage characteristics of the comparative light-emitting device 50 and the light-emitting device 50.

FIG. 60 shows the power efficiency-luminance characteristics of the comparative light-emitting device 50 and the light-emitting device 50.

FIG. 61 shows the emission spectra of the comparative light-emitting device 50 and the light-emitting device 50.

FIGS. 62A and 62B show the refractive indices of mmtBumTPoFBi-04, PCBBiPDBt-02, and mmtBuBiFF.

FIG. 63 shows the refractive indices of mmtBumBPTzn, Li-6mq, and BisBTc.

FIG. 64 shows the refractive indices of mmtBumTPoFBi-04, DBfBB1TP, and mmtBuBioFBi.

FIG. 65 shows the emission spectrum of DPhA-tBu4DABNA in a toluene solution.

FIG. 66 shows the luminance-current density characteristics of a light-emitting device 60.

FIG. 67 shows the current efficiency-luminance characteristics of the light-emitting device 60.

FIG. 68 shows the luminance-voltage characteristics of the light-emitting device 60.

FIG. 69 shows the current density-voltage characteristics of the light-emitting device 60.

FIG. 70 shows the blue index-luminance characteristics of the light-emitting device 60.

FIG. 71 shows the emission spectra of the light-emitting device 60.

## DETAILED DESCRIPTION OF THE INVENTION

Embodiments of the present invention will be described in detail below with reference to the drawings. Note that the present invention is not limited to the following description, and it will be readily appreciated by those skilled in the art that modes and details of the present invention can be modified in various ways without departing from the spirit and scope of the present invention. Therefore, the present invention should not be construed as being limited to the description in the following embodiments.

Note that in the case where light is incident on a material having optical anisotropy, light with a plane of vibration parallel to the light axis is referred to as extraordinary light (rays) and light with a plane of vibration perpendicular to the light axis is referred to as ordinary light (rays); the refractive index of the material with respect to ordinary light might differ from that with respect to extraordinary light. In such a case, the ordinary refractive index and the extraordinary refractive index can be separately calculated by anisotropy analysis. Note that in the case where the measured material has both the ordinary refractive index and the extraordinary refractive index, the ordinary refractive index is used as an index in this specification.

### Embodiment 1

FIG. 1A illustrates a light-emitting device of one embodiment of the present invention. The light-emitting device illustrated in FIG. 1A includes a first electrode 101, a second electrode 102, and an EL layer 103. The EL layer 103 includes at least a hole-transport layer 112 (a first hole-transport layer 112-1, a second hole-transport layer 112-2, and a third hole-transport layer 112-3) and a light-emitting layer 113. The light-emitting layer 113 includes at least a light-emitting material. The hole-transport layer 112 is positioned between the light-emitting layer 113 and the first electrode 101. The EL layer 103 may include a hole-injection layer 111, an electron-transport layer 114, an electron-injection layer 115, and the like as illustrated in the drawings and may additionally include other functional layers such as a carrier-blocking layer, an exciton-blocking layer, and an intermediate layer.

In one embodiment of the present invention, the first hole-transport layer 112-1, the second hole-transport layer 112-2, and the third hole-transport layer 112-3 include a first organic compound, a second organic compound, and a third organic compound, respectively. The first organic compound 55 to the third organic compound respectively account for higher than or equal to 80% of the first hole-transport layer 112-1 to the third hole-transport layer 112-3.

In the light-emitting device of one embodiment of the present invention, the second organic compound has a higher refractive index than the first organic compound and the third organic compound. That is, the second hole-transport layer 112-2 has a higher refractive index than the first hole-transport layer 112-1 and the third hole-transport layer 112-3.

60 In the light-emitting device of one embodiment of the present invention with such a structure, light emitted from the light-emitting material is reflected at the interface

between the hole-transport layers, which allows a larger amount of light to be reflected than in the case where light is reflected only with a reflective electrode, and improves external quantum efficiency. At the same time, the influence of surface plasmon generated in the reflective electrode can be decreased, which reduces energy loss to extract light efficiently.

In one embodiment of the present invention, the difference between the ordinary refractive indices of the second organic compound and the first organic compound with respect to light with a wavelength of  $\lambda$  nm and the difference between the ordinary refractive indices of the second organic compound and the third organic compound with respect to light with the wavelength of  $\lambda$  nm are each preferably greater than or equal to 0.1 and less than 0.5, further preferably greater than or equal to 0.2 and less than 0.5, and still further preferably greater than or equal to 0.3 and less than 0.5. When the difference in ordinary refractive index is in the above ranges, the hole-transport layer 112 having the same structure as that in this light-emitting device can be applied to another light-emitting device including light-emitting materials that emit light with different wavelengths with little problem; thus, the hole-transport layer 112 does not need to be separately formed for each emission color, resulting in advantages in cost.

The difference between the ordinary refractive indices of the second hole-transport layer 112-2 and the first hole-transport layer 112-1 with respect to light with the wavelength of  $\lambda$  nm and the difference between the ordinary refractive indices of the second hole-transport layer 112-2 and the third hole-transport layer 112-3 with respect to light with the wavelength of  $\lambda$  nm are each preferably greater than or equal to 0.2 and less than 0.5. The wavelength of  $\lambda$  nm is any or all of the wavelengths greater than or equal to 450 nm and less than or equal to 650 nm. In the case where the light-emitting device exhibits light in the blue region, the wavelength of  $\lambda$  nm is any or all of the wavelengths greater than or equal to 455 nm and less than or equal to 465 nm. The wavelength of  $\lambda$  nm is preferably an emission peak wavelength of the light-emitting material. Note that the wavelength of  $\lambda$  nm typically used as an index of refractive index is 633 nm. Any of these values can be used as an index of the wavelength of  $\lambda$  nm.

The ordinary refractive indices of the first organic compound and the third organic compound with respect to light with the wavelength of  $\lambda$  nm are each preferably greater than or equal to 1.40 and less than or equal to 1.75; specifically, the ordinary refractive index in the blue light emission range (455 nm to 465 nm) is preferably greater than or equal to 1.40 and less than or equal to 1.75 or the ordinary refractive index with respect to light with a wavelength of 633 nm, which is typically used in measurement of refractive index, is preferably greater than or equal to 1.40 and less than or equal to 1.70. It is further preferable that the ordinary refractive indices of the first hole-transport layer 112-1 and the third hole-transport layer 112-3 be within the above range. The difference between the ordinary refractive indices of the first organic compound and the third organic compound with respect to light with the wavelength of  $\lambda$  nm is preferably less than or equal to 0.1.

The ordinary refractive index of the second organic compound with respect to light with the wavelength of  $\lambda$  nm is preferably greater than or equal to 1.75; specifically, the ordinary refractive index in the blue light emission range (455 nm to 465 nm) is preferably greater than or equal to 1.75 and less than or equal to 2.4 or the ordinary refractive index with respect to light with a wavelength of 633 nm,

which is typically used in measurement of refractive index, is preferably greater than or equal to 1.75 and less than or equal to 2.30. It is further preferable that the ordinary refractive index of the second hole-transport layer 112-2 be within the above range.

The difference between the HOMO levels of the first organic compound and the third organic compound is preferably less than or equal to 0.2 eV, further preferably less than or equal to 0.1 eV, in which case the hole-transport property is increased. The difference between the HOMO levels of the second organic compound and each of the first organic compound and the third organic compound is preferably less than or equal to 0.2 eV, further preferably less than or equal to 0.1 eV, in which case the hole-transport property is increased.

In the light-emitting device of one embodiment of the present invention, the thickness of each of the first hole-transport layer 112-1 to the third hole-transport layer 112-3 is preferably larger than or equal to 25 nm and smaller than 10 or equal to 70 nm. The properties of the light-emitting device of one embodiment of the present invention are improved when a buffer layer such as a hole-injection layer or an electron-blocking layer is provided on the side of each of the first hole-transport layer 112-1 and the third hole-transport layer 112-3 that is not in contact with the second hole-transport layer 112-2; thus, the thickness of the second hole-transport layer 112-2 is preferably larger than that of each of the first hole-transport layer 112-1 and the third hole-transport layer 112-3. In addition, the product of the thickness and the ordinary refractive index of the second hole-transport layer 112-2 with respect to light with the wavelength of  $\lambda$  nm is preferably greater than or equal to 70% and less than or equal to 130% of  $\lambda/4$ , in which case optical interference increases. With such a structure, light reflected at the interface between the third hole-transport layer 112-3 and the second hole-transport layer 112-2 and light reflected at the interface between the second hole-transport layer 112-2 and the first hole-transport layer 112-1 can be amplified by interference, so that a light-emitting device with higher efficiency can be provided.

The first organic compound and the third organic compound are preferably the same organic compound in order to ensure a high hole-transport property and reduce the number of materials used in fabrication of the light-emitting device. 45 In order to ensure the reliability, it is preferable that the third hole-transport layer 112-3 not contain fluorine. Furthermore, when a common hole-transport layer is used in a display device exhibiting light of the three primary colors of red, green, and blue, too much optical interference that intensifies a specific light is not preferable; hence, it is preferable that the first organic compound and the third organic compound not contain fluorine with low atomic refraction.

The first hole-transport layer 112-1 and the third hole-transport layer 112-3 are formed using a material with a 55 relatively low refractive index. In general, a high carrier-transport property and a low refractive index have a trade-off relationship. This is because the carrier-transport property of an organic compound largely depends on an unsaturated bond, and an organic compound having many unsaturated bonds tends to have a high refractive index. Even having a low refractive index, a material with a low carrier-transport property causes a problem such as a decrease in emission efficiency or reliability due to an increase in driving voltage or poor carrier balance, so that a light-emitting device with favorable characteristics cannot be obtained. Furthermore, even when a material has a sufficient carrier-transport property and a low refractive index, a highly reliable light-

emitting device cannot be obtained if the material has a low glass transition temperature ( $T_g$ ) or a heat resistance problem due to an unstable structure.

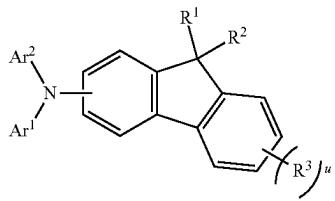
Thus, an organic compound that can be used in the first hole-transport layer **112-1** and the third hole-transport layer **112-3** is preferably a monoamine compound including a first aromatic group, a second aromatic group, and a third aromatic group, in which the first aromatic group, the second aromatic group, and the third aromatic group are bonded to the same nitrogen atom.

In the monoamine compound, the proportion of carbon atoms each forming a bond by the  $sp^3$  hybrid orbitals to the total number of carbon atoms in the molecule is preferably higher than or equal to 23% and lower than or equal to 55%. In addition, it is preferable that the integral value of signals at lower than 4 ppm exceed the integral value of signals at 4 ppm or higher in the results of  $^1H$ -NMR measurement conducted on the monoamine compound.

The monoamine compound preferably has at least one fluorene skeleton. One or more of the first aromatic group, the second aromatic group, and the third aromatic group are a fluorene skeleton. Since fluorenylamine has an effect of increasing the HOMO level, bonding of three fluorenes to nitrogen of the monoamine compound possibly increases the HOMO level significantly. In that case, a difference in HOMO level between the monoamine compound and peripheral materials (e.g., the second organic compound) becomes large, which might affect driving voltage, reliability, and the like. Thus, any one or two of the first to third aromatic groups are further preferably fluorene skeletons.

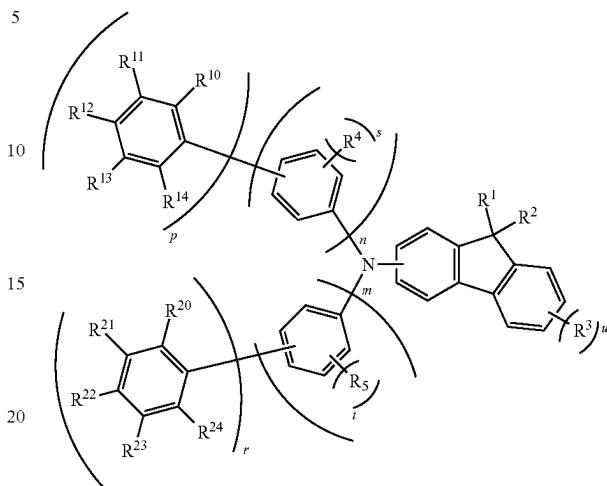
Examples of the above-described organic compound having a hole-transport property include organic compounds having structures represented by General formulae  $(G_{h1}1)$  to  $(G_{h1}4)$  shown below.

[Chemical Formula 1]



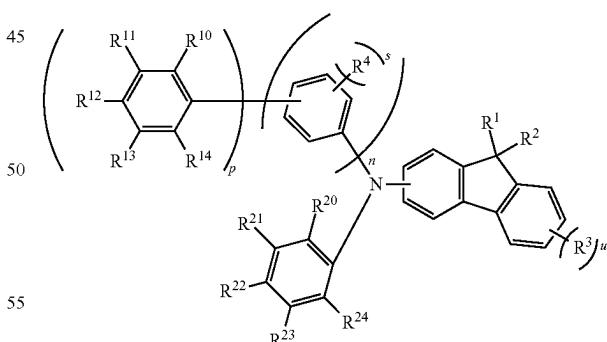
In General formula (G<sub>h1</sub>), Ar<sup>1</sup> and Ar<sup>2</sup> each independently represent a substituent with a benzene ring or a substituent in which two or three benzene rings are bonded to each other. Note that one or both of Ar<sup>1</sup> and Ar<sup>2</sup> have one or more hydrocarbon groups each having 1 to 12 carbon atoms each forming a bond only by the  $sp^3$  hybrid orbitals. The total number of carbon atoms contained in all of the hydrocarbon groups bonded to Ar<sup>1</sup> and Ar<sup>2</sup> is 8 or more and the total number of carbon atoms contained in all of the hydrocarbon groups bonded to Ar<sup>1</sup> or Ar<sup>2</sup> is 6 or more. Note that in the case where a plurality of straight-chain alkyl groups each having one or two carbon atoms are bonded to Ar<sup>1</sup> or Ar<sup>2</sup> as the hydrocarbon groups, the straight-chain alkyl groups may be bonded to each other to form a ring.

[Chemical Formula 2]

(G<sub>h1</sub>2)

In General formula (G<sub>h1</sub>2), m and r each independently represent 1 or 2 and m+r is 2 or 3. Furthermore, t represents an integer of 0 to 4 and is preferably 0. R<sup>s</sup> represents hydrogen or any of hydrocarbon groups having 1 to 3 carbon atoms. When m is 2, the kind and number of substituents and the position of bonds included in one phenylene group may be the same as or different from those of the other phenylene group; and when r is 2, the kind and number of substituents and the position of bonds included in one phenyl group may be the same as or different from those of the other phenyl group. In the case where t is an integer of 2 to 4, R<sup>s</sup>'s may be the same as or different from each other; and adjacent groups (adjacent R<sup>s</sup>'s) may be bonded to each other to form a ring.

[Chemical Formula 3]

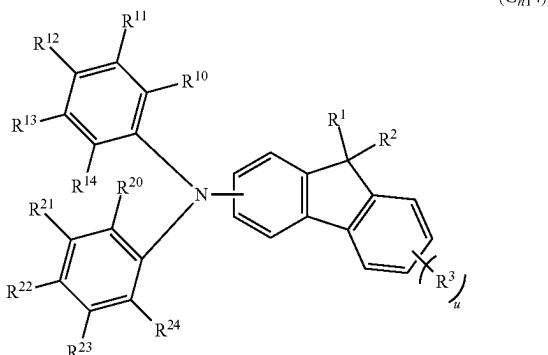
(G<sub>h1</sub>3)

In General formulae (G<sub>h1</sub>2) and (G<sub>h1</sub>3), n and p each independently represent 1 or 2 and n+p is 2 or 3. In addition, s represents an integer of 0 to 4 and is preferably 0. R<sup>4</sup> represents hydrogen or any of hydrocarbon groups having 1 to 3 carbon atoms. When n is 2, the kind and number of substituents and the position of bonds in one phenylene group may be the same as or different from those of the other phenylene group; and when p is 2, the kind and number of substituents and the position of bonds in one phenyl group

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may be the same as or different from those of the other phenyl group. In the case where s is an integer of 2 to 4, R<sup>s</sup> may be the same as or different from each other.

[Chemical Formula 4]



In General formulae (G<sub>h1</sub>2) to (G<sub>h1</sub>4), R<sup>10</sup> to R<sup>14</sup> and R<sup>20</sup> to R<sup>24</sup> each independently represent hydrogen or a hydrocarbon group having 1 to 12 carbon atoms each forming a bond only by the sp<sup>3</sup> hybrid orbitals. Note that at least three of R<sup>10</sup> to R<sup>14</sup> and at least three of R<sup>20</sup> to R<sup>24</sup> are preferably hydrogen. As the hydrocarbon group having 1 to 12 carbon atoms each forming a bond only by the sp<sup>3</sup> hybrid orbitals, a tert-butyl group and a cyclohexyl group are preferable. The total number of carbon atoms contained in R<sup>10</sup> to R<sup>14</sup> and R<sup>20</sup> to R<sup>24</sup> is 8 or more and the total number of carbon atoms contained in either R<sup>10</sup> to R<sup>14</sup> or R<sup>20</sup> to R<sup>24</sup> is 6 or more. Note that adjacent groups of R<sup>4</sup>, R<sup>10</sup> to R<sup>14</sup> and R<sup>2</sup> to R<sup>24</sup> may be bonded to each other to form a ring.

In General formulae (G<sub>h1</sub>1) to (G<sub>h1</sub>4), each u independently represents an integer of 0 to 4 and is preferably 0. Note that in the case where u is an integer of 2 to 4, R<sup>3</sup> may be the same as or different from each other. In addition, R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> each independently represent an alkyl group having 1 to 4 carbon atoms and R<sup>1</sup> and R<sup>2</sup> may be bonded to each other to form a ring.

An arylamine compound that has at least one aromatic group having first to third benzene rings and at least three alkyl groups is also preferable as one of the materials having a hole-transport property that can be used for the first hole-transport layer 112-1 and the third hole-transport layer 112-3. Note that the first to third benzene rings are bonded in this order and the first benzene ring is directly bonded to nitrogen of amine. The first benzene ring may further include a substituted or unsubstituted phenyl group and preferably includes an unsubstituted phenyl group. Furthermore, the second benzene ring or the third benzene ring may include a phenyl group substituted by an alkyl group.

Note that hydrogen is not directly bonded to carbon atoms at 1- and 3-positions in two or more of, preferably all of the first to third benzene rings, and the carbon atoms are bonded to any of the first to third benzene rings, the phenyl group substituted by the alkyl group, the at least three alkyl groups, and the nitrogen of the amine.

It is preferable that the arylamine compound further include a second aromatic group. It is preferable that the second aromatic group have an unsubstituted monocyclic ring or a substituted or unsubstituted bicyclic or tricyclic condensed ring; in particular, it is further preferable that the second aromatic group be a group having a substituted or

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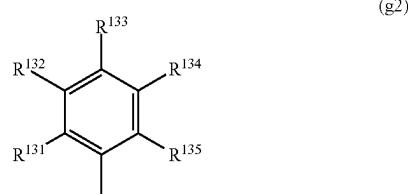
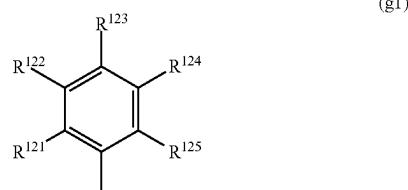
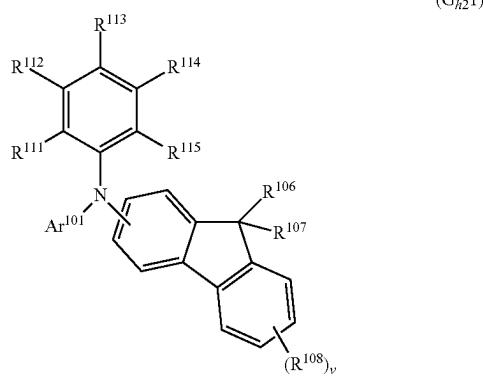
unsubstituted bicyclic, or tricyclic condensed ring where the number of carbon atoms forming the ring is 6 to 13. It is still further preferable that the second aromatic group be a group including a fluorene ring. Note that a dimethylfluorenyl group is preferable as the second aromatic group.

It is preferable that the arylamine compound further include a third aromatic group. The third aromatic group is a group having 1 to 3 substituted or unsubstituted benzene rings.

It is preferable that the at least three alkyl groups and the alkyl group substituted for the phenyl group be each a chain alkyl group having 2 to 5 carbon atoms. In particular, as the alkyl group, a chain alkyl group having a branch formed of 3 to 5 carbon atoms is preferable, and a t-butyl group is further preferable.

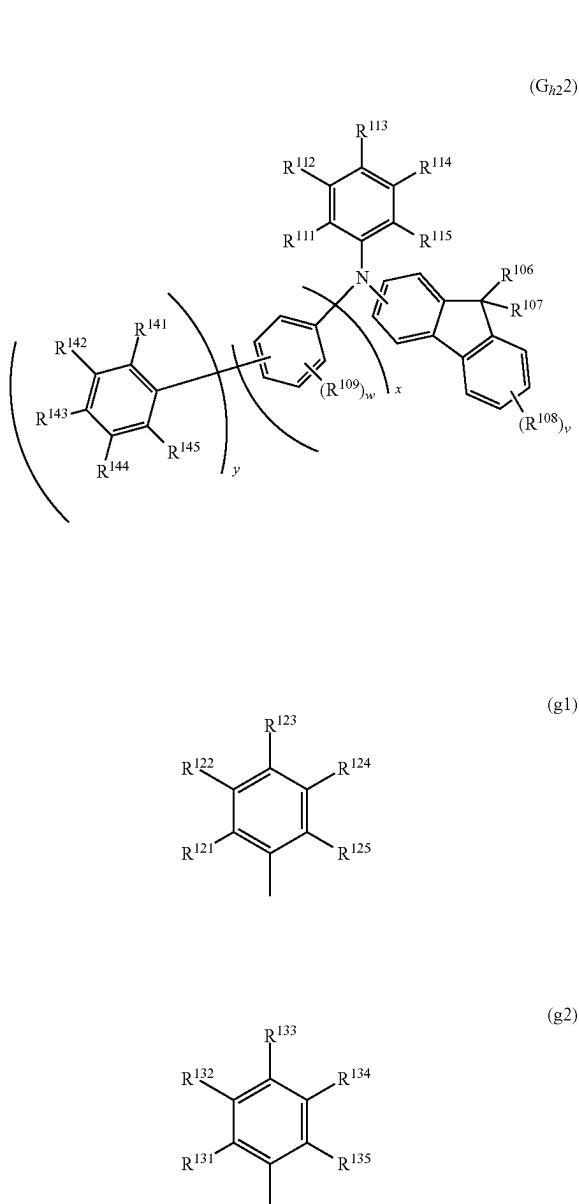
Examples of the above-described material having a hole-transport property include organic compounds having structures represented by General formulae (G<sub>h2</sub>1) to (G<sub>h2</sub>3) shown below.

[Chemical Formula 5]



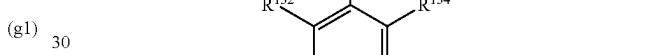
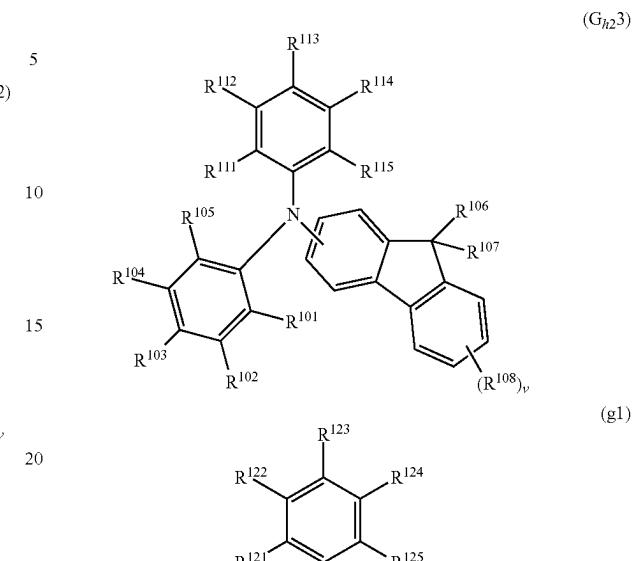
Note that in General formula (G<sub>h2</sub>1), Ar<sup>101</sup> represents a substituted or unsubstituted benzene ring or a substituent in which two or three substituted or unsubstituted benzene rings are bonded to one another.

[Chemical Formula 6]



Note that in General formula (G<sub>h2</sub>2), x and y each independently represent 1 or 2 and x+y is 2 or 3. Furthermore, R<sup>109</sup> represents an alkyl group having 1 to 4 carbon atoms, and w represents an integer of 0 to 4. R<sup>141</sup> to R<sup>145</sup> each independently represent any one of hydrogen, an alkyl group having 1 to 6 carbon atoms, and a cycloalkyl group having 5 to 12 carbon atoms. When w is an integer of 2 or more, R<sup>109</sup>'s may be the same as or different from each other. When x is 2, the kind and number of substituents and the position of bonds included in one phenylene group may be the same as or different from those of the other phenylene group. When y is 2, the kind and number of substituents and the position of bonds included in one phenyl group including R<sup>141</sup> to R<sup>145</sup> may be the same as or different from those of the other phenyl group including R<sup>141</sup> to R<sup>145</sup>.

[Chemical Formula 7]



35 In General formula (G<sub>h2</sub>3), R<sup>101</sup> to R<sup>105</sup> each independently represent any one of hydrogen, an alkyl group having 1 to 6 carbon atoms, a cycloalkyl group having 6 to 12 carbon atoms, and a substituted or unsubstituted phenyl group.

40 In General formulae (G<sub>h2</sub>1) to (G<sub>h2</sub>3), R<sup>106</sup>, R<sup>107</sup>, and R<sup>108</sup> each independently represent an alkyl group having 1 to 4 carbon atoms, and v represents an integer of 0 to 4. Note that when v is 2 or more, R<sup>108</sup>'s may be the same as or

45 different from each other. One of R<sup>111</sup> to R<sup>125</sup> represents a substituent represented by General formula (g1), and the others each independently represent any one of hydrogen, an alkyl group having 1 to 6 carbon atoms, and a substituted or unsubstituted phenyl group. In General formula (g1), one of

50 R<sup>121</sup> to R<sup>125</sup> represents a substituent represented by General formula (g2), and the others each independently represent any one of hydrogen, an alkyl group having 1 to 6 carbon atoms, and a phenyl group substituted by an alkyl group having 1 to 6 carbon atoms. In General formula (g2), R<sup>131</sup> to R<sup>135</sup> each independently represent any one of hydrogen,

55 an alkyl group having 1 to 6 carbon atoms, and a phenyl group substituted by an alkyl group having 1 to 6 carbon atoms. Note that at least three of R<sup>111</sup> to R<sup>115</sup>, R<sup>121</sup> to R<sup>125</sup>, and R<sup>131</sup> to R<sup>135</sup> are each an alkyl group having 1 to 6 carbon atoms; the number of substituted or unsubstituted phenyl groups in R<sup>111</sup> to R<sup>115</sup> is one or less; and the number of phenyl groups substituted by an alkyl group having 1 to 6 carbon atoms in R<sup>121</sup> to R<sup>125</sup> and R<sup>131</sup> to R<sup>135</sup> is one or less.

60 In at least two combinations of the three combinations R<sup>112</sup> and R<sup>114</sup>, R<sup>122</sup> and R<sup>124</sup>, and R<sup>132</sup> and R<sup>134</sup>, at least one R represents any of the substituents other than hydrogen.

The above-described organic compounds having a hole-transport property each have an ordinary refractive index greater than or equal to 1.40 and less than or equal to 1.75 in the blue light emission range (455 nm to 465 nm) or an ordinary refractive index greater than or equal to 1.40 and less than or equal to 1.70 with respect to light with a wavelength of 633 nm, which is typically used in measurement of refractive index. The organic compounds have both a high hole-transport property and a high T<sub>g</sub> to achieve favorable reliability. These organic compounds having a sufficient hole-transport property can be favorably used as the materials for the first hole-transport layer 112-1 and the third hole-transport layer 112-3.

Alternatively, 1,1-bis[4-[N,N-di(p-tolyl)amino]phenyl]cyclohexane (abbreviation: TAPC) or the like can be used.

The second hole-transport layer 112-2 is formed using an organic compound having a relatively high refractive index; the organic compound preferably has a condensed aromatic hydrocarbon ring or a condensed heteroaromatic ring. The condensed aromatic hydrocarbon ring preferably has a naphthalene structure such as a naphthalene ring, an anthracene ring, a phenanthrene ring, or a triphenylene ring, and the condensed heteroaromatic ring preferably has a structure such as a carbazole ring, a dibenzofuran ring, or a dibenzothiophene ring. For example, benzo[b]naphtho[1,2-d]furan is preferable because having a structure of a dibenzofuran ring.

It is preferable to use an organic compound having one or more elements of the third and later periods, an organic compound having a terphenyl skeleton, an organic compound having both of them, or the like. For example, a biphenyl group substituted by a naphthyl group, or a phenyl group substituted by a dibenzofuranyl group can be said to have a terphenyl skeleton. Specifically, N,N-bis[4-(6-phenylbenzo[b]naphtho[1,2-d]furan-8-yl)phenyl]-4-amino-p-terphenyl (abbreviation: BnfBB1TP), 4,4'-bis[4-(2-naphthyl)phenyl]-4"-phenyltriphenylamine (abbreviation: PNBiB1BP), N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBfBB1TP), 4-[4'-(carbazol-9-yl)biphenyl-4-yl]-4'-(2-naphthyl)-4"-phenyltriphenylamine (abbreviation: YGTBiβNB), 5,5'-diphenyl-2,2'-di-5H-[1]benzothieno[3,2-c]carbazole (abbreviation: BisBTc), or the like can be favorably used.

The first electrode 101 preferably includes a reflective electrode having a visible light reflectivity of 40% or more, preferably 70% or more. The second electrode 102 is preferably a transreflective electrode having a visible light reflectivity of 20% to 80%, preferably 40% to 70%. With such a structure, the light-emitting device of one embodiment of the present invention is a top-emission light-emitting device that emits light through the second electrode 102, and can have a microcavity structure by adjusting the thickness of the EL layer.

In that case, the optical path length between the interface of the reflective electrode on the EL layer 103 side and the interface of the first hole-transport layer 112-1 on the reflective electrode side is preferably  $\lambda/8$  to  $3\lambda/8$ . The optical path length between the main emission region of the light-emitting layer 113 and the interface of the first hole-transport layer 112-1 on the reflective electrode side is preferably  $3\lambda/8$  to  $5\lambda/8$ . The optical path length between the interface of the reflective electrode on the EL layer 103 side and the interface of the third hole-transport layer 112-3 on the reflective electrode side is preferably  $3D8$  to  $58$ . The optical path length between the main emission region of the light-emitting layer 113 and the interface of the third hole-transport layer 112-3 on the light-emitting layer side is

preferably  $\lambda/8$  to  $3\lambda/8$ . Such a structure enables light reflected on the interface of each layer and the reflective electrode to be amplified and the light-emitting device to have favorable efficiency and color purity. Note that if the main emission region of the light-emitting layer is difficult to determine accurately, it can be determined on the basis of the position estimated in consideration of the transport property of the light-emitting layer.

In one embodiment of the present invention, the light extraction efficiency is improved with a stack of a plurality of hole-transport layers having different refractive indices. However, the light-emitting device includes more layers than typical light-emitting devices, and thus includes more interfaces of layers, which might easily generate a resistance due to interfaces and increase driving voltage.

In general, holes need to be sequentially injected into layers formed of organic compounds with different HOMO levels between a hole-transport region of an organic semiconductor device and an active layer or a light-emitting layer, in consideration of donation and acceptance of holes with electrodes. Since an excessively large difference in HOMO level between layers naturally increases driving voltage, a difference in HOMO level is reduced by provision of a layer, between an electrode and an active layer (a light-emitting layer), that is formed of an organic compound having a HOMO level positioned between those of the electrode and the active layer (the light-emitting layer). However, layers whose difference between HOMO levels is not so large may lead to a significant increase in driving voltage depending on a combination of organic compounds to be used. There is no guideline for avoiding the above problem so far, and it has been considered that the cause of the problem is the incompatibility of materials.

In contrast, a polar molecule and a non-polar molecule exist in an organic compound. The polar molecule has a permanent dipole moment. When the polar molecule is evaporated and the evaporated film has random orientation, unbalanced polarity is canceled out and polarization derived from the polarity of the molecule does not occur in the film. However, when the evaporated film has molecular orientation, the giant surface potential derived from unbalanced polarization is sometimes observed.

The giant surface potential refers to a phenomenon in which a surface potential of an evaporated film increases in proportion to a film thickness, and can be described as spontaneous orientation polarization due to slight deviation of a permanent dipole moment of an organic compound to the thickness direction. In order to treat the surface potential as a value independent of a film thickness, a value obtained by dividing the surface potential of an evaporated film by the film thickness, that is, the potential gradient (slope) of a surface potential of an evaporated film, is used. In this specification, the potential gradient of a surface potential of an evaporated film is denoted by GSP (mV/nm).

The consideration of the value of GSP can eliminate the mismatch that has been thought to be caused by the incompatibility of materials and enables an organic semiconductor device with excellent properties to be easily obtained.

In one embodiment of the present invention, the value (ΔGSP<sub>1,3</sub>) obtained by subtracting GSP of the third hole-transport layer 112-3 from GSP of the first hole-transport layer 112-1 is preferably less than or equal to 10 (mV/nm), further preferably less than or equal to 0 (mV/nm). The value (ΔGSP<sub>2,3</sub>) obtained by subtracting GSP of the third hole-transport layer 112-3 from GSP of the second hole-transport layer 112-2 is preferably less than or equal to 10 (mV/nm), further preferably less than or equal to 0 (mV/nm). Further

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preferably, both  $\Delta GSP_{1-3}$  and  $\Delta GSP_{2-3}$  are less than or equal to 10 (mV/nm), still further preferably less than or equal to 0 (mV/nm).

The value ( $\Delta GSP_{1-2}$ ) obtained by subtracting GSP of the second hole-transport layer **112-2** from GSP of the first hole-transport layer **112-1** is preferably less than or equal to 10 (mV/nm), further preferably less than or equal to 0 (mV/nm).

With such a structure, a light-emitting device having excellent properties such as a low driving voltage, low power consumption, or a high power efficiency can be easily obtained.

In addition, GSP of the third hole-transport layer **112-3** is preferably larger than GSP of the first hole-transport layer **112-1**, and GSP of the third hole-transport layer **112-3** is preferably larger than GSP of the second hole-transport layer **112-2**. Note that GSP of the third hole-transport layer **112-3** is preferably larger than GSP of the first hole-transport layer **112-1** and GSP of the second hole-transport layer **112-2**. With such a structure, a light-emitting device having excellent properties such as a low driving voltage, low power consumption, or a high power efficiency can be easily obtained.

Furthermore, GSP of the second hole-transport layer **112-2** is preferably larger than GSP of the first hole-transport layer **112-1**. With such a structure, a light-emitting device having excellent properties such as a low driving voltage, low power consumption, or a high power efficiency can be easily obtained.

Note that GSP of each layer can be obtained by measurement of GSP of an evaporated film of a material (an organic compound) in the layer.

A method for obtaining GSP of an organic compound will be described.

In general, a slope of a plot of a surface potential of an evaporated film in the thickness direction by Kelvin probe measurement is assumed as the level of the giant surface potential, that is, GSP (mV/nm); in the case where two different layers are stacked, a change in the density of polarization charges ( $mC/m^2$ ) accumulated at the interface, which is in association with GSP, can be utilized to estimate GSP.

Non-Patent Document 1 discloses that the following formulae hold when current is made to flow through a stack of organic thin films (a thin film 1 positioned on the anode side and a thin film 2 positioned on the cathode side) with different kinds of spontaneous polarization.

[Formula 1]

$$\sigma_{if} = \frac{Q_{if}}{S} = (V_i - V_{bi}) \frac{\epsilon_2}{d_2} \quad (1)$$

[Formula 2]

$$\sigma_{if} = P_1 - P_2 = \frac{\epsilon_1 V_1}{d_1} - \frac{\epsilon_2 V_2}{d_2} \quad (2)$$

In Formula (1),  $\sigma_{if}$  is a polarization charge density,  $V_i$  is a hole-injection voltage,  $V_{bi}$  is a threshold voltage,  $d_2$  is a thickness of the thin film 2, and  $\epsilon_2$  is a dielectric constant of the thin film 2. Note that  $V_i$  and  $V_{bi}$  can be estimated from the capacity-voltage characteristics of a device. The square of an ordinary refractive index  $n_o$  (633 nm) can be used as the dielectric constant. As described above, according to Formula (1), the polarization charge density  $\sigma_{if}$  can be calculated using  $V_i$  and  $V_{bi}$  estimated from the capacity-voltage

## 20

characteristics, the dielectric constant  $\epsilon_2$  of the thin film 2 calculated from the refractive index, and the thickness  $d_2$  of the thin film 2.

Next, in Formula (2), as is a polarization charge density,  $P_n$  is GSP of a thin film n, and  $\epsilon_n$  is a dielectric constant of the thin film n. Since the polarization charge density  $\sigma_{if}$  can be obtained from Formula (1), the use of a substance with known GSP for the thin film 2 enables GSP of the thin film 1 to be estimated.

10 In this manner, an evaporated film of an organic compound with GSP to be obtained is formed as the thin film 1, and GSP of the organic compound can be obtained by the above method.

Note that in this specification,  $Alq_3$  whose GSP is known 15 to be 48 (mV/nm) is used for the thin film 2, and GSP of each thin film is obtained.

The orientation of an evaporated film is known to depend 20 on the substrate temperature in evaporation, and the value of GSP also possibly depends on the substrate temperature in evaporation. The measured values in this specification are values of films evaporated with the substrate temperature set to room temperature.

Next, examples of other structures and materials of the 25 light-emitting device of one embodiment of the present invention will be described. The light-emitting device of one embodiment of the present invention includes, as described above, the EL layer **103** including a plurality of layers between the first electrode **101** and the second electrode **102**, and the EL layer **103** includes at least the light-emitting layer **113** containing a light-emitting material and the hole-transport layer **112** (the first hole-transport layer **112-1**, the second hole-transport layer **112-2**, and the third hole-transport layer **112-3**).

The first electrode **101** is preferably formed using any of 30 metals, alloys, and conductive compounds with a high work function (specifically, higher than or equal to 4.0 eV), mixtures thereof, and the like. Specific examples include indium oxide-tin oxide (ITO: indium tin oxide), indium oxide-tin oxide containing silicon or silicon oxide, indium 35 oxide-zinc oxide, and indium oxide containing tungsten oxide and zinc oxide (IWZO). Such conductive metal oxide films are usually formed by a sputtering method, but may be formed by application of a sol-gel method or the like. In an example of the formation method, indium oxide-zinc oxide is deposited by a sputtering method using a target obtained by adding 1 wt % to 20 wt % of zinc oxide to indium oxide. Furthermore, a film of indium oxide containing tungsten 40 oxide and zinc oxide (IWZO) can be formed by a sputtering method using a target in which tungsten oxide and zinc oxide are added to indium oxide at 0.5 wt % to 5 wt % and 45 0.1 wt % to 1 wt %, respectively. Alternatively, gold (Au), platinum (Pt), nickel (Ni), tungsten (W), chromium (Cr), molybdenum (Mo), iron (Fe), cobalt (Co), copper (Cu), palladium (Pd), nitride of a metal material (e.g., titanium 50 nitride), or the like can be used for the first electrode **101**. Graphene can also be used for the first electrode **101**. Note that when a composite material described later is used for a 55 layer (typically, the hole-injection layer **111**) that is in contact with the first electrode **101** in the EL layer **103**, an electrode material can be selected regardless of its work function.

When the first electrode **101** is formed using a material 60 that transmits light emitted from the light-emitting device, the light-emitting device can emit light through the first electrode **101** as illustrated in FIG. 1C. When the first electrode **101** includes an electrode that reflects light emitted from the light-emitting device and the second electrode **102**

is formed using a material that transmits light emitted from the light-emitting device, the light-emitting device can emit light through the second electrode 102 as illustrated in FIG. 1D. Since the hole-transport layer 112 is provided between the light-emitting layer 113 and the first electrode 101, the first electrode 101 preferably reflects light emitted from the light-emitting device.

Although the EL layer 103 preferably has a stacked-layer structure, there is no particular limitation on the stacked-layer structure except for the light-emitting layer 113 and the hole-transport layer 112, and a variety of functional layers such as a hole-injection layer, an electron-transport layer, an electron-injection layer, a carrier-blocking layer (a hole-blocking layer and an electron-blocking layer), an exciton-blocking layer, an intermediate layer, and a charge-generation layer can be employed. Note that one or more of the above layers are not necessarily provided. Two kinds of structures are described in this embodiment: the structure illustrated in FIG. 1A, which includes the hole-injection layer 111, the electron-transport layer 114, and the electron-injection layer 115 in addition to the light-emitting layer 113 and the hole-transport layer 112; and the structure illustrated in FIG. 1B, which includes a charge-generation layer 116 instead of the electron-injection layer 115.

The hole-injection layer 111 is provided in contact with the first electrode 101 and has a function of facilitating injection of holes into the EL layer 103. The hole-injection layer 111 can be formed using a phthalocyanine-based complex compound such as phthalocyanine (abbreviation: H<sub>2</sub>Pc) and copper phthalocyanine (abbreviation: CuPc), an aromatic amine compound such as 4,4'-bis[N-(4-diphenylaminophenyl)-N-phenylamino]biphenyl (abbreviation: DPAB) or 4,4'-bis(N-[4-[N'-(3-methylphenyl)-N'-phenylamino]phenyl]-N-phenylamino)biphenyl (abbreviation: DNTPD), or a high molecular compound such as poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonic acid) (PEDOT/PSS).

The hole-injection layer 111 may be formed using a substance having an acceptor property. Examples of the substance having an acceptor property include an organic compound having an electron-withdrawing group (a halogen group or a cyano group), such as 7,7,8,8-tetracyano-2,3,5,6-tetrafluoroquinodimethane (abbreviation: F4-TCNQ), chloranil, 2,3,6,7,10,11-hexacyano-1,4,5,8,9,12-hexaaazatriphenylene (abbreviation: HAT-CN), 1,3,4,5,7,8-hexafluorotetracyano-naphthoquinodimethane (abbreviation: F6-TCNNQ), or 2-(7-dicyanomethylene-1,3,4,5,6,8,9,10-octafluoro-7H-pyren-2-ylidene)malononitrile. A compound in which electron-withdrawing groups are bonded to a condensed aromatic ring having a plurality of heteroatoms, such as HAT-CN, is particularly preferable because it is thermally stable. A [3]radialene derivative having an electron-withdrawing group (in particular, a cyano group or a halogen group such as a fluoro group) has a very high electron-accepting property and thus is preferable. Specific examples include  $\alpha,\alpha',\alpha''$ -1,2,3-cyclopropanetriylidenetris [4-cyano-2,3,5,6-tetrafluorobenzeneacetonitrile],  $\alpha,\alpha',\alpha''$ -1,2,3-cyclopropanetriylidenetris[2,6-dichloro-3,5-difluoro-4-(trifluoromethyl)benzenecacetonitrile], and  $\alpha,\alpha',\alpha''$ -1,2,3-cyclopropanetriylidenetris[2,3,4,5,6-pentafluorobenzeneacetonitrile]. As the substance having an acceptor property, molybdenum oxide, vanadium oxide, ruthenium oxide, tungsten oxide, manganese oxide, or the like can also be used, other than the above-described organic compounds. The substance having an acceptor property can

extract electrons from an adjacent hole-transport layer (or hole-transport material) by the application of voltage between the electrodes.

The hole-injection layer 111 may be formed using a composite material containing any of the aforementioned materials having an acceptor property and a material having a hole-transport property. As the material having a hole-transport property that is used in the composite material, any of a variety of organic compounds such as aromatic amine compounds, heteroaromatic compounds, aromatic hydrocarbons, and high molecular compounds (e.g., oligomers, dendrimers, or polymers) can be used. Note that the material having a hole-transport property that is used in the composite material preferably has a hole mobility of  $1 \times 10^{-6} \text{ cm}^2/\text{Vs}$  or higher. The material having a hole-transport property that is used in the composite material is preferably a compound having a condensed aromatic hydrocarbon ring or a  $\pi$ -electron rich heteroaromatic ring. As the condensed aromatic hydrocarbon ring, an anthracene ring, a naphthalene ring, or the like is preferable. As the  $\pi$ -electron rich heteroaromatic ring, a condensed aromatic ring having at least one of a pyrrole skeleton, a furan skeleton, and a thiophene skeleton is preferable; specifically, a carbazole ring, a dibenzothiophene ring, or a ring in which an aromatic ring or a heteroaromatic ring is further condensed to the carbazole ring or the dibenzothiophene ring is preferable.

The material having a hole-transport property further preferably has any of a carbazole skeleton, a dibenzofuran skeleton, a dibenzothiophene skeleton, and an anthracene skeleton. In particular, an aromatic amine having a substituent that includes a dibenzofuran ring or a dibenzothiophene ring, an aromatic monoamine that includes a naphthalene ring, or an aromatic monoamine in which a 9-fluorenyl group is bonded to nitrogen of amine through an arylene group may be used. Note that the hole-transport material having an N,N-bis(4-biphenyl)amino group is preferable because a light-emitting device having a long lifetime can be fabricated. Specific examples of the hole-transport material include N-(4-biphenyl)-6,N-diphenylbenzo[b]naphtho[1,2-d]furan-8-amine (abbreviation: BnfABP), N,N-bis(4-biphenyl)-6-phenylbenzo[b]naphtho[1,2-d]furan-8-amine (abbreviation: BBABnf), 4,4'-bis(6-phenylbenzo[b]naphtho[1,2-d]furan-8-yl)-4"-phenyltriphenylamine (abbreviation: BnfBB1BP), N,N-bis(4-biphenyl)benzo[b]naphtho[1,2-d]furan-6-amine (abbreviation: BBABnf(6)), N,N-bis(4-biphenyl)benzo[b]naphtho[1,2-d]furan-8-amine (abbreviation: BBABnf(8)), N,N-bis(4-biphenyl)benzo[b]naphtho[2,3-d]furan-4-amine (abbreviation: BBABnf(II) (4)), N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBIBB1TP), N-[4-(dibenzothiophen-4-yl)phenyl]-N-phenyl-4-biphenylamine (abbreviation: ThBA1BP), 4-(2-naphthyl)-4',4"-diphenyltriphenylamine (abbreviation: BBA $\beta$ NB), 4-[4-(2-naphthyl)phenyl]-4',4"-diphenyltriphenylamine (abbreviation: BBA $\beta$ NBi), 4,4'-diphenyl-4"-6,1'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA $\alpha$ N $\beta$ NB), 4,4'-diphenyl-4"-7,1'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA $\alpha$ N $\beta$ NB-03), 4,4'-diphenyl-4"-7-phenyl)naphthyl-2-yl)triphenylamine (abbreviation: BBAP $\beta$ NB-03), 4,4'-diphenyl-4"-6,2'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA( $\beta$ N2)B), 4,4'-diphenyl-4"-7,2'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA( $\beta$ N2>B-03), 4,4'-diphenyl-4"-4,2'-binaphthyl-1-yl)triphenylamine (abbreviation: BBA $\beta$ N $\alpha$ NB), 4,4'-diphenyl-4"-5,2'-binaphthyl-1-yl)triphenylamine (abbreviation: BBA $\beta$ N $\alpha$ NB-02), 4-(4-biphenyl)-4'(2-naphthyl)-4"-phenyltriphenylamine (abbreviation: TPBi $\beta$ NB), 4-(3-biphenyl)-4'-[4-(2-naphthyl)phenyl]-

4"-phenyltriphenylamine (abbreviation: mTPBiA $\beta$ NBi), 4-(4-biphenyl)-4'-[4-(2-naphthyl)phenyl]-4"-phenyltriphenylamine (abbreviation: TPBiA $\beta$ NBi), 4-phenyl-4'-(1-naphthyl)triphenylamine (abbreviation:  $\alpha$ NBA1BP), 4,4'-bis(1-naphthyl)triphenylamine (abbreviation:  $\alpha$ NBB1BP), 4,4'-diphenyl-4"-[(4-(carbazol-9-yl)biphenyl-4-yl)triphenylamine (abbreviation: YGTBi1BP), 4'-[4-(3-phenyl-9H-carbazol-9-yl)phenyl]tris(1,1'-biphenyl-4-yl)amine (abbreviation: YGTBi1BP-02), 4-[4'-(carbazole-9-yl)biphenyl-4-yl]-4'-(2-naphthyl)-4"-phenyltriphenylamine (abbreviation: YGTBi $\beta$ NB), N-[4-(9-phenyl-9H-carbazol-3-yl)phenyl]-N-[4-(1-naphthyl)phenyl]-9,9'-spirobi[9H-fluoren]-2-amine (abbreviation: PCBNBSF), N,N-bis([1,1'-biphenyl]-4-yl)-9,9'-spirobi[9H-fluoren]-2-amine (abbreviation: BBASF), N,N-bis([1,1'-biphenyl]-4-yl)-9,9'-spirobi[9H-fluoren]-4-amine (abbreviation: BBASF(4)), N-(1,1'-biphenyl-2-yl)-N-(9,9-dimethyl-9H-fluoren-2-yl)-9,9'-spirobi[9H-fluoren]-4-amine (abbreviation: oFBiSF), N-(4-biphenyl)-N-(9,9-dimethyl-9H-fluoren-2-yl)dibenzofuran-4-amine (abbreviation: FrBiF), N-[4-(1-naphthyl)phenyl]-N-[3-(6-phenyldibenzofuran-4-yl)phenyl]-1-naphthylamine (abbreviation: mPDBfBNBN), 4-phenyl-4'-(9-phenylfluoren-9-yl)triphenylamine (abbreviation: BPAFLP), 4-phenyl-3'-(9-phenylfluoren-9-yl)triphenylamine (abbreviation: mBPAFLP), 4-phenyl-4'[4-(9-phenylfluoren-9-yl)phenyl]triphenylamine (abbreviation: BPAFLBi), 4-phenyl-4'-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBA1BP), 4,4'-diphenyl-4"--(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBBI1BP), 4-(1-naphthyl)-4'-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBANB), 4,4'-di(1-naphthyl)-4"--(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBNBB), N-phenyl-N-[4-(9-phenyl-9H-carbazol-3-yl)phenyl]-9,9'-spirobi[9H-fluoren]-2-amine (abbreviation: PCBASF), N-(1,1'-biphenyl-4-yl)-N-[4-(9-phenyl-9H-carbazol-3-yl)phenyl]-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: PCBBI1F), N,N-bis(9,9-dimethyl-9H-fluoren-2-yl)-9,9'-spirobi[9H-fluoren-4-amine], N,N-bis(9,9-dimethyl-9H-fluoren-2-yl)-9,9'-spirobi[9H-fluoren-3-amine], N,N-bis(9,9-dimethyl-9H-fluoren-2-yl)-9,9'-spirobi[9H-fluoren-2-amine], and N,N-bis(9,9-dimethyl-9H-fluoren-2-yl)-9,9'-spirobi[9H-fluoren-1-amine].

As the material having a hole-transport property, the following aromatic amine compounds can also be used: N,N-di(p-tolyl)-N,N-diphenyl-p-phenylenediamine (abbreviation: DTDPAA), 4,4'-bis[N-(4-diphenylaminophenyl)-N-phenylamino]biphenyl (abbreviation: DPAB), 4,4'-bis(N-[4-[N-(3-methylphenyl)-N-phenylamino]phenyl]-N-phenylamino)biphenyl (abbreviation: DNTPD), and 1,3,5-tris[N-(4-diphenylaminophenyl)-N-phenylamino]benzene (abbreviation: DPA3B). In addition, any of the organic compounds used for the first hole-transport layer 112-1 and the third hole-transport layer 112-3 can be favorably used.

Further preferably, the material having a hole-transport property that is used in the composite material has a relatively deep HOMO level higher than or equal to -5.7 eV and lower than or equal to -5.4 eV. When the material having a hole-transport property that is used in the composite material has a relatively deep HOMO level, holes can be easily injected into the hole-transport layer 112 to easily provide a light-emitting device having a long lifetime. In addition, when the material having a hole-transport property that is used in the composite material has a relatively deep HOMO level, induction of holes can be inhibited properly so that a light-emitting device having a longer lifetime can be easily obtained.

Note that mixing the above composite material with a fluoride of an alkali metal or an alkaline earth metal (the proportion of fluorine atoms in a layer using the mixed material is preferably greater than or equal to 20%) can lower the refractive index of the layer. This also enables a layer with a low refractive index to be formed in the EL layer 103, improving the external quantum efficiency of the light-emitting device.

The formation of the hole-injection layer 111 can improve 10 the hole-injection property, offering the light-emitting device with a low driving voltage.

Among substances having an acceptor property, the organic compound having an acceptor property is easy to use because it is easily deposited by vapor deposition.

15 The hole-transport layer 112 is formed using a material having a hole-transport property. The material having a hole-transport property preferably has a hole mobility higher than or equal to  $1 \times 10^{-6}$  cm<sup>2</sup>/Vs. With the hole-transport layer 112 having the aforementioned structure, the light-emitting device of one embodiment of the present invention can have excellent properties. For example, one or more of the external quantum efficiency, the current efficiency, and the blue index of the light-emitting device can be improved. In the case where the aforementioned structure is not employed, the hole-transport layer 112 can be formed with a single layer or a plurality of layers using any of the aforementioned materials having a hole-transport property that can be used in the composite material of the hole-injection layer.

20 An electron-blocking layer may be provided between the hole-transport layer 112 and the light-emitting layer 113. The electron-blocking layer is preferably formed using a substance that has a hole-transport property and a higher LUMO level than the light-emitting layer 113 by 0.25 eV or 25 higher.

The light-emitting layer 113 preferably includes a light-emitting substance and a host material. The light-emitting layer 113 may additionally include other materials. Alternatively, the light-emitting layer 113 may be a stack of two layers with different compositions.

30 As the light-emitting substance, fluorescent substances, phosphorescent substances, substances exhibiting thermally activated delayed fluorescence (TADF), or other light-emitting substances may be used. Note that one embodiment of the present invention is more suitably used in the case where the light-emitting layer 113 exhibits fluorescence, specifically, blue fluorescence.

Examples of the material that can be used as a fluorescent substance in the light-emitting layer 113 are as follows. Other fluorescent substances can also be used.

35 The examples include 5,6-bis[4-(10-phenyl-9-anthryl)phenyl]-2,2'-bipyridine (abbreviation: PAP2BPy), 5,6-bis[4'-(10-phenyl-9-anthryl)biphenyl-4-yl]-2,2'-bipyridine (abbreviation: PAPP2BPy), N,N'-diphenyl-N,N'-bis[4-(9-phenyl-9H-fluoren-9-yl)phenyl]pyrene-1,6-diamine (abbreviation: 1,6FLPAPM), N,N'-bis(3-methylphenyl)-N,N'-bis[3-(9-phenyl-9H-fluoren-9-yl)phenyl]pyrene-1,6-diamine (abbreviation: 1,6mMemFLPAPM), N,N'-bis[4-(9H-carbazol-9-yl)phenyl]-N,N'-diphenylstilbene-4,4'-diamine (abbreviation: YGA2S), 4-(9H-carbazol-9-yl)-4'-(10-phenyl-9-anthryl)triphenylamine (abbreviation: YGAPA), 4-(9H-carbazol-9-yl)-4'-(9,10-diphenyl-2-anthryl)triphenylamine (abbreviation: 2YGAPPA), N,9-diphenyl-N-[4-(10-phenyl-9-anthryl)phenyl]-9H-carbazol-3-amine (abbreviation: PCAPPA), perylene, 2,5,8,11-tetra-tert-butylperylene (abbreviation: TBP), 4-(10-phenyl-9-anthryl)-4'-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBAPA),

N,N'-(2-tert-butylanthracene-9,10-diyldi-4,1-phenylene)bis[N,N',N'-triphenyl-1,4-phenylenediamine] (abbreviation: DPABPA), N,9-diphenyl-N-[4-(9,10-diphenyl-2-anthryl)phenyl]-9H-carbazol-3-amine (abbreviation: 2PCAPPA), N-[4-(9,10-diphenyl-2-anthryl)phenyl]-N,N',N'-triphenyl-1,4-phenylenediamine (abbreviation: 2DPAPPA), N,N',N'',N''-octaphenylbenzo[g,p]chrysene-2,7,10,15-tetraamine (abbreviation: DBC1), coumarin 30, N-(9,10-diphenyl-2-anthryl)-N,9-diphenyl-9H-carbazol-3-amine (abbreviation: 2PCAPA), N-[9,10-bis(1,1'-biphenyl-2-yl)-2-anthryl]-N,9-diphenyl-9H-carbazol-3-amine (abbreviation: 2PCABPhA), N-(9,10-diphenyl-2-anthryl)-N,N',N'-triphenyl-1,4-phenylenediamine (abbreviation: 2DPAPA), N-[9,10-bis(1,1'-biphenyl-2-yl)-2-anthryl]-N,N',N'-triphenyl-1,4-phenylenediamine (abbreviation: 2DPABPhA), 9,10-bis(1,1'-biphenyl-2-yl)-N-[4-(9H-carbazol-9-yl)phenyl]-N-phenylanthracen-2-amine (abbreviation: 2YGABPhA), N,N,9-triphenylanthracen-9-amine (abbreviation: DPhAPhA), coumarin 545T, N,N'-diphenylquinacridone (abbreviation: DPQd), rubrene, 5,12-bis(1,1'-biphenyl-4-yl)-6,11-diphenyltetracene (abbreviation: BPT), 2-(2-{2-[4-(dimethylamino)phenyl]ethenyl}-6-methyl-4H-pyran-4-ylidene)propanedinitrile (abbreviation: DCM1), 2-{2-methyl-6-[2-(2,3,6,7-tetrahydro-1H,5H-benzo[ij]quinolizin-9-yl)ethenyl]-4H-pyran-4-ylidene}propanedinitrile (abbreviation: DCM2), N,N,N',N'-tetrakis(4-methylphenyl)tetracene-5,11-diamine (abbreviation: p-mPhTD), 7,14-diphenyl-N,N,N',N'-tetrakis(4-methylphenyl)acenaphtho[1,2-a]fluoranthene-3,10-diamine (abbreviation: p-mPhAFD), 2-{2-isopropyl-6-[2-(1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1H,5H-benzo[ij]quinolizin-9-yl)ethenyl]-4H-pyran-4-ylidene}propanedinitrile (abbreviation: DCJTI), 2-{2-tert-butyl-6-[2-(1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1H,5H-benzo[ij]quinolizin-9-yl)ethenyl]-4H-pyran-4-ylidene}propanedinitrile (abbreviation: DCJTB), 2-(2,6-bis{2-[4-(dimethylamino)phenyl]ethenyl}-4H-pyran-4-ylidene)propanedinitrile (abbreviation: BisDCM), 2-{2,6-bis[2-(8-methoxy-1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1H,5H-benzo[ij]quinolizin-9-yl)ethenyl]-4H-pyran-4-ylidene}propanedinitrile (abbreviation: BisDCJTM), N,N'-diphenyl-N,N'-(1,6-pyrene-diyl)bis[(6-phenylbenzo[b]naphtho[1,2-d]furan)-8-amine] (abbreviation: 1,6BnfAPrn-03), 3,10-bis[N-(9-phenyl-9H-carbazol-2-yl)-N-phenylamino]naphtho[2,3-b;6,7-b']bisbenzofuran (abbreviation: 3,10PCA2Nbf(IV)-02), and 3,10-bis[N-(dibenzo[furan-3-yl)-N-phenylamino]naphtho[2,3-b;6,7-b']bisbenzofuran (abbreviation: 3,10FrA2Nbf(IV)-02). Condensed aromatic diamine compounds typified by pyrenediamine compounds such as 1,6FLPAPm, 1,6mMemFLPAPm, and 1,6BnfAPm-03 are particularly preferable because of their high hole-trapping properties, high emission efficiency, or high reliability.

Examples of the material that can be used when a phosphorescent substance is used as the light-emitting substance in the light-emitting layer 113 are as follows.

The examples include an organometallic iridium complex having a 4H-triazole skeleton, such as tris{2-[5-(2-methylphenyl)-4-(2,6-dimethylphenyl)-4H-1,2,4-triazol-3-yl- $\kappa$ N2]phenyl- $\kappa$ C}iridium(III) (abbreviation: [Ir(mpptz-dnp)<sub>3</sub>]), tris(5-methyl-3,4-diphenyl-4H-1,2,4-triazolato)iridium(III) (abbreviation: [Ir(Mptz)<sub>3</sub>]), or tris[4-(3-biphenyl)-5-isopropyl-3-phenyl-4H-1,2,4-triazolato]iridium(III) (abbreviation: [Ir(iPrtz-3b)<sub>3</sub>]); an organometallic iridium complex having a 1H-triazole skeleton, such as tris[3-methyl-1-(2-methylphenyl)-5-phenyl-1H-1,2,4-triazolato]iridium(III) (abbreviation: [Ir(Mptz1-mp)<sub>3</sub>]) or tris(1-methyl-5-phenyl-3-propyl-1H-1,2,4-triazolato)iridium(III)

(abbreviation: [Ir(Prptz1-Me)<sub>3</sub>]); an organometallic iridium complex having an imidazole skeleton, such as fac-tris[1-(2,6-diisopropylphenyl)-2-phenyl-1H-imidazole]iridium(III) (abbreviation: [Ir(iPrpmi)<sub>3</sub>]) or tris[3-(2,6-dimethylphenyl)-7-methylimidazo[1,2-f]phenanthridinato]iridium(III) (abbreviation: [Ir(dmpimpt-Me)<sub>3</sub>]); and an organometallic iridium complex in which a phenylpyridine derivative having an electron-withdrawing group is a ligand, such as bis[2-(4',6'-difluorophenyl)pyridinato-N,C<sup>2</sup>]iridium(III) tetrakis(1-pyrazolyl)borate (abbreviation: FIr6), bis[2-(4',6'-difluorophenyl)pyridinato-N,C<sup>2</sup>]iridium(III) picolinate (abbreviation: FIrpic), bis{2-[3',5'-bis(trifluoromethyl)phenyl]pyridinato-N,C<sup>2</sup>}iridium(III) picolinate (abbreviation: [Ir(CF<sub>3</sub>ppy)<sub>2</sub>(pic)]), or bis[2-(4',6'-difluorophenyl)pyridinato-N,C<sup>2</sup>]iridium(III) acetylacetone (abbreviation: FIr(acac)). These compounds exhibit blue phosphorescence and have an emission peak in the wavelength range of 440 nm to 520 nm.

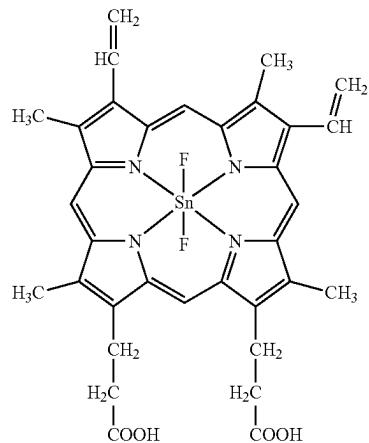
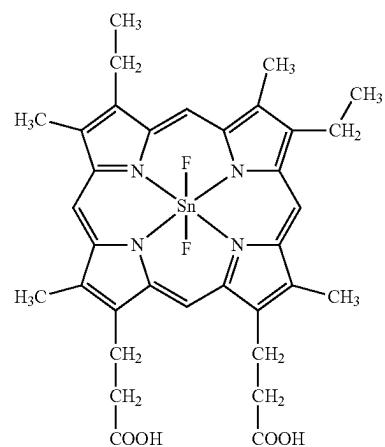
Other examples include an organometallic iridium complex having a pyrimidine skeleton, such as tris(4-methyl-6-phenylpyrimidinato)iridium(III) (abbreviation: [Ir(mppm)<sub>3</sub>]), tris(4-t-butyl-6-phenylpyrimidinato)iridium(III) (abbreviation: [Ir(tBuppm)<sub>3</sub>]), (acetylacetato)bis(6-methyl-4-phenylpyrimidinato)iridium(III) (abbreviation: [Ir(mppm)<sub>2</sub>(acac)]), (acetylacetato)bis(6-tert-butyl-4-phenylpyrimidinato)iridium(III) (abbreviation: [Ir(tBuppm)<sub>2</sub>(acac)]), (acetylacetato)bis[6-(2-norbornyl)-4-phenylpyrimidinato]iridium(III) (abbreviation: [Ir(nbppm)<sub>2</sub>(acac)]), (acetylacetato)bis[5-methyl-6-(2-methylphenyl)-4-phenylpyrimidinato]iridium(III) (abbreviation: [Ir(mpmpm)<sub>2</sub>(acac)]), or (acetylacetato)bis(4,6-diphenylpyrimidinato)iridium(III) (abbreviation: [Ir(dppm)<sub>2</sub>(acac)]); an organometallic iridium complex having a pyrazine skeleton, such as (acetylacetato)bis(3,5-dimethyl-2-phenylpyrazinato)iridium(III) (abbreviation: [Ir(mprr-Me)<sub>2</sub>(acac)])) or (acetylacetato)bis(5-isopropyl-3-methyl-2-phenylpyrazinato)iridium(III) (abbreviation: [Ir(mprr-iPr)<sub>2</sub>(acac)]); an organometallic iridium complex having a pyridine skeleton, such as tris(2-phenylpyridinato-N,C<sup>2</sup>)iridium(III) (abbreviation: [Ir(ppy)<sub>3</sub>]), bis(2-phenylpyridinato-N,C<sup>2</sup>)iridium(III) acetylacetone (abbreviation: [Ir(ppy)<sub>2</sub>(acac)]), bis(benzo[h]quinolinato)iridium(III) acetylacetone (abbreviation: [Ir(bzq)<sub>2</sub>(acac)]), tris(benzo[h]quinolinato)iridium(III) (abbreviation: [Ir(bzq)<sub>3</sub>]), tris(2-phenylquinolinato-N,C<sup>2</sup>)iridium(III) (abbreviation: [Ir(pq)<sub>3</sub>]), bis(2-phenylquinolinato-N,C<sup>2</sup>)iridium(III) acetylacetone (abbreviation: [Ir(pq)<sub>2</sub>(acac)]), [2-d<sub>3</sub>-methyl-8-(2-pyridinyl- $\kappa$ N)benzofuro[2,3-b]pyridine- $\kappa$ C]bis[2-(5-d<sub>3</sub>-methyl-2-pyridinyl-N2)phenyl- $\kappa$ C]iridium(III) (abbreviation: [Ir(5mppy-d3)<sub>2</sub>(mbfpypy-d3)]), [2-(methyl-d3)-8-[4-(1-methylethyl-1-d)-2-pyridinyl- $\kappa$ N]benzofuro[2,3-b]pyridin-7-yl- $\kappa$ C]bis[5-(methyl-d3)-2-[5-(methyl-d3)-2-pyridinyl-N]phenyl- $\kappa$ C]iridium(III) (abbreviation: Ir(5mtpy-d6)<sub>2</sub>(mbfpypy-iPr-d4)), [2-d<sub>3</sub>-methyl-(2-pyridinyl- $\kappa$ N)benzofuro[2,3-b]pyridine- $\kappa$ C]bis[2-(2-pyridinyl- $\kappa$ N)phenyl- $\kappa$ C]iridium(III) (abbreviation: [Ir(5mtpy-d6)<sub>2</sub>(mbfpypy-iPr-d4)]), [2-d<sub>3</sub>-methyl-(2-pyridinyl- $\kappa$ N)phenyl- $\kappa$ C]iridium(III) (abbreviation: [Ir(ppy)<sub>2</sub>(mbfpypy-d3)]), or [2-(4-methyl-5-phenyl-2-pyridinyl- $\kappa$ N)phenyl- $\kappa$ C]bis[2-(2-pyridinyl- $\kappa$ N)phenyl- $\kappa$ C]iridium(III) (abbreviation: [Ir(ppy)<sub>2</sub>(mdppy)]); and a rare earth metal complex such as tris(acetylacetato)(monophenanthroline)terbium(III) (abbreviation: [Tb(acac)<sub>3</sub>(Phen)]). These are mainly compounds that exhibit green phosphorescence and have an emission peak in the wavelength range of 500 nm to 600 nm. Note that organometallic iridium complexes having a pyrimidine skeleton have distinctively high reliability or emission efficiency and thus are particularly preferable.

Other examples include an organometallic iridium complex having a pyrimidine skeleton, such as (diisobutyryl-methanato)bis[4,6-bis(3-methylphenyl)pyrimidinato]iridium(III) (abbreviation: [Ir(5mdppm)<sub>2</sub>(dibm)]), bis[4,6-bis(3-methylphenyl)pyrimidinato](dipivaloylmethanato)iridium(III) (abbreviation: [Ir(5mdppm)<sub>2</sub>(dpm)]), or bis[4,6-di(naphthalen-1-yl)pyrimidinato](dipivaloylmethanato)iridium(III) (abbreviation: [Ir(d1npm)<sub>2</sub>(dpm)]); an organometallic iridium complex having a pyrazine skeleton, such as (acetylacetone)bis(2,3,5-triphenylpyrazinato)iridium(III) (abbreviation: [Ir(tppr)<sub>2</sub>(acac)]), bis(2,3,5-triphenylpyrazinato)(dipivaloylmethanato)iridium(III) (abbreviation: [Ir(tppr)<sub>2</sub>(dpm)]), or (acetylacetone)bis[2,3-bis(4-fluorophenyl)quinoxalinato]iridium(III) (abbreviation: [Ir(Fdpq)<sub>2</sub>(acac)]); an organometallic iridium complex having a pyridine skeleton, such as tris(1-phenylisoquinolinato-N, C<sup>2</sup>)iridium(III) (abbreviation: [Ir(piq)<sub>3</sub>]) or bis(1-phenylisoquinolinato-N,C<sup>2</sup>)iridium(III) acetylacetone (abbreviation: [Ir(piq)<sub>2</sub>(acac)]); a platinum complex such as 2,3,7,8,12,13,17,18-octaethyl-21H,23H-porphyrinplatinum(II) (abbreviation: PtOEP); and a rare earth metal complex such as tris(1,3-diphenyl-1,3-propanedionato)(monophenanthroline)europium(III) (abbreviation: [Eu(DBM)<sub>3</sub>(Phen)]) or tris[1-(2-thenoyl)-3,3,3-trifluoroacetonato](monophenanthroline)europium(III) (abbreviation: [Eu(TTA)<sub>3</sub>(Phen)]). These compounds exhibit red phosphorescence and have an emission peak in the wavelength range of 600 nm to 700 nm. Organometallic iridium complexes having a pyrazine skeleton can provide red light emission with favorable chromaticity.

Besides the above phosphorescent compounds, known phosphorescent compounds may be selected and used.

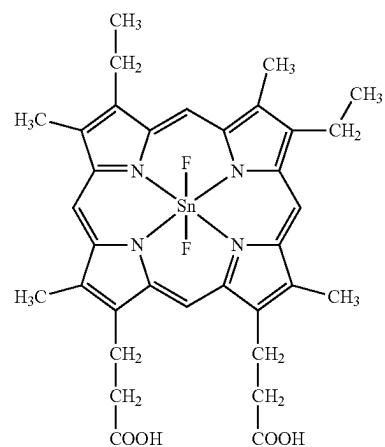
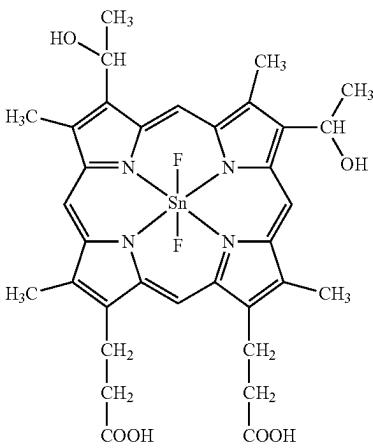
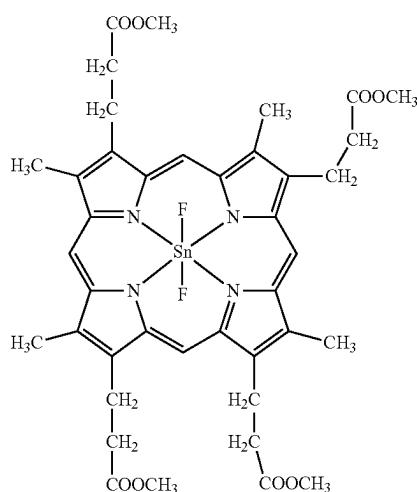
Examples of the TADF material include a fullerene, a derivative thereof, an acridine, a derivative thereof, and an eosin derivative. Furthermore, a metal-containing porphyrin, such as a porphyrin containing magnesium (Mg), zinc (Zn), cadmium (Cd), tin (Sn), platinum (Pt), indium (In), or palladium (Pd), can be given. Examples of the metal-containing porphyrin include a protoporphyrin-tin fluoride complex (SnF<sub>2</sub>(Proto IX)), a mesoporphyrin-tin fluoride complex (SnF<sub>2</sub>(Meso IX)), a hematoporphyrin-tin fluoride complex (SnF<sub>2</sub>(Hemato IX)), a coproporphyrin tetramethyl ester-tin fluoride complex (SnF<sub>2</sub>(Copro III-4Me)), an octaethylporphyrin-tin fluoride complex (SnF<sub>2</sub>(OEP)), an etioporphyrin-tin fluoride complex (SnF<sub>2</sub>(Etio I)), and an octaethylporphyrin-platinum chloride complex (PtCl<sub>2</sub>OEP), which are represented by the following structural formulae.

[Chemical Formula 8]

SnF<sub>2</sub>(Proto IX)SnF<sub>2</sub>(Meso IX)

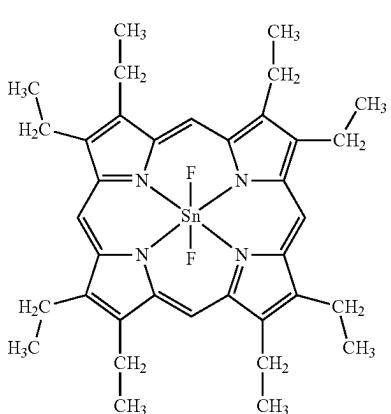
## 28

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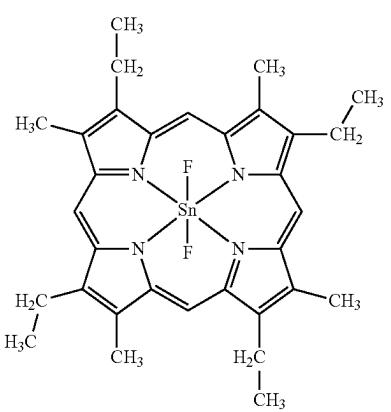
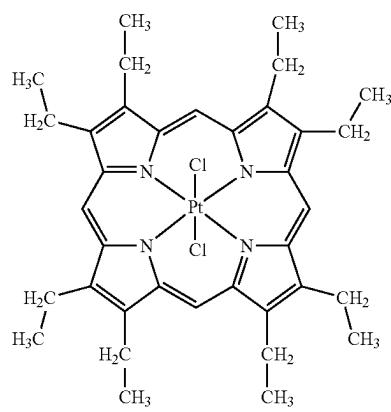
SnF<sub>2</sub>(Hemato IX)SnF<sub>2</sub>(Copro III-4Me)

**29**

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SnF<sub>2</sub>(OEP)**30**

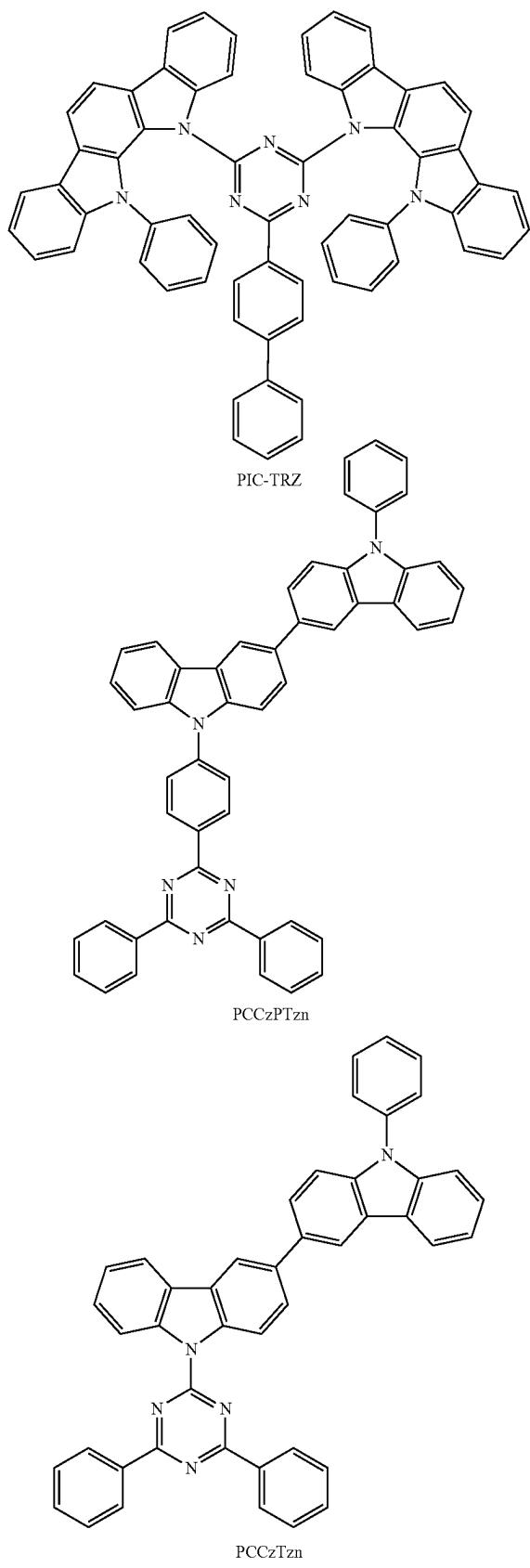
6-bis(12-phenylindolo[2,3-a]carbazol-11-yl)-1,3,5-triazine (abbreviation: PIC-TRZ), 9-(4,6-diphenyl-1,3,5-triazin-2-yl)-9'-phenyl-9H,9'H-3,3'-bicarbazole (abbreviation: PCCzTzn), 2-[4-[3-(N-phenyl-9H-carbazol-3-yl)-9H-carbazol-9-yl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: PCCzPTzn), 2-[4-(10H-phenoxyazin-10-yl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: PXZ-TRZ), 3-[4-(5-phenyl-5,10-dihydrophenazin-10-yl)phenyl]-4,5-diphenyl-1,2,4-triazole (abbreviation: PPZ-3TPT), 3-(9,9-dimethyl-9H-acridin-10-yl)-9H-xanthan-9-one (abbreviation: ACRXTN), bis[4-(9,9-dimethyl-9,10-dihydroacridine)phenyl]sulfone (abbreviation: DMAC-DPS), or 10-phenyl-10H, 10'H-spiro[acridin-9,9'-anthracen]-10'-one (abbreviation: ACRSA) can be used. Such a heterocyclic compound is preferable because of having excellent electron-transport and hole-transport properties owing to a π-electron rich heteroaromatic ring and a π-electron deficient heteroaromatic ring. Among skeletons having the π-electron deficient heteroaromatic ring, a pyridine skeleton, a diazine skeleton (a pyrimidine skeleton, a pyrazine skeleton, and a pyridazine skeleton), and a triazine skeleton are preferred because of their high stability and reliability. In particular, a benzofuropyrimidine skeleton, a benzothienopyrimidine skeleton, a benzofuropyrazine skeleton, and a benzothienopyrazine skeleton are preferred because of their high acceptor properties and high reliability. Among skeletons having the π-electron rich heteroaromatic ring, an acridine skeleton, a phenoxazine skeleton, a phenothiazine skeleton, a furan skeleton, a thiophene skeleton, and a pyrrole skeleton have high stability and reliability; thus, at least one of these skeletons is preferably included. A dibenzofuran skeleton is preferable as a furan skeleton, and a dibenzothiophene skeleton is preferable as a thiophene skeleton. As a pyrrole skeleton, an indole skeleton, a carbazole skeleton, an indocarbazole skeleton, a bicarbazole skeleton, and a 3-(9-phenyl-9H-carbazol-3-yl)-9H-carbazole skeleton are particularly preferable. Note that a substance in which the π-electron rich heteroaromatic ring is directly bonded to the π-electron deficient heteroaromatic ring is particularly preferred because the electron-donating property of the π-electron rich heteroaromatic ring and the electron-accepting property of the π-electron deficient heteroaromatic ring are both improved, the energy difference between the S1 level and the T1 level becomes small, and thus thermally activated delayed fluorescence can be obtained with high efficiency. Note that an aromatic ring to which an electron-withdrawing group such as a cyano group is bonded may be used instead of the π-electron deficient heteroaromatic ring. As a π-electron rich skeleton, an aromatic amine skeleton, a phenazine skeleton, or the like can be used. As a π-electron deficient skeleton, a xanthene skeleton, a thioxanthene dioxide skeleton, an oxadiazole skeleton, a triazole skeleton, an imidazole skeleton, an anthraquinone skeleton, a skeleton containing boron such as phenylborane or boranthrene, an aromatic ring or a heteroaromatic ring having a cyano group or a nitrile group such as benzonitrile or cyanobenzene, a carbonyl skeleton such as benzophenone, a phosphine oxide skeleton, a sulfone skeleton, or the like can be used. As described above, a π-electron deficient skeleton and a π-electron rich skeleton can be used instead of at least one of the π-electron deficient heteroaromatic ring and the π-electron rich heteroaromatic ring.

SnF<sub>2</sub>(Etio I)PtCl<sub>2</sub>OEP

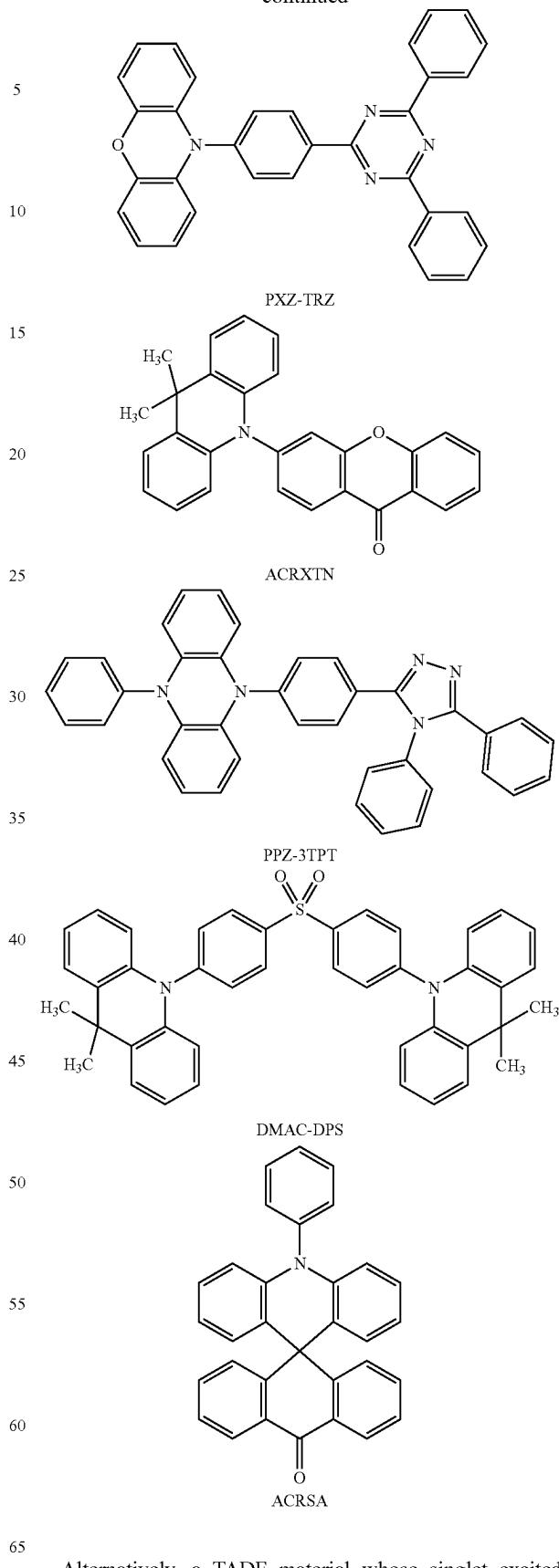
Alternatively, a heterocyclic compound having one or both of a π-electron rich heteroaromatic ring and a π-electron deficient heteroaromatic ring that is represented by the following structural formulae, such as 2-(biphenyl-4-yl)-4,

**31**

[Chemical Formula 9]

**32**

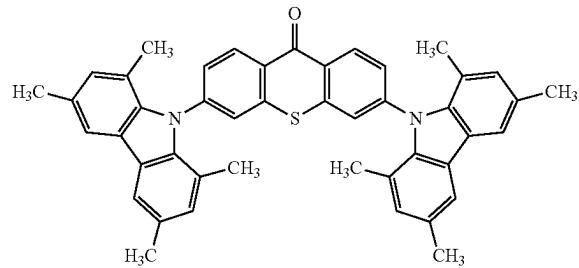
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Alternatively, a TADF material whose singlet excited state and triplet excited state are in a thermal equilibrium

state may be used. Such a TADF material has a short emission lifetime (excitation lifetime), which allows inhibiting a decrease in efficiency in a high-luminance region of a light-emitting element. Specifically, a material having the following molecular structure can be used.

[Chemical Formula 10]



Note that a TADF material is a material having a small difference between the S1 level and the T1 level and a function of converting triplet excitation energy into singlet excitation energy by reverse intersystem crossing. Thus, a TADF material can upconvert triplet excitation energy into singlet excitation energy (i.e., reverse intersystem crossing) using a small amount of thermal energy and efficiently generate a singlet excited state. In addition, the triplet excitation energy can be converted into luminescence.

An exciplex whose excited state is formed of two kinds of substances has an extremely small difference between the S1 level and the T1 level and functions as a TADF material capable of converting triplet excitation energy into singlet excitation energy.

A phosphorescent spectrum observed at a low temperature (e.g., 77 K to 10 K) is used for an index of the T1 level. When the level of energy with a wavelength of the line obtained by extrapolating a tangent to the fluorescent spectrum at a tail on the short wavelength side is the S1 level and the level of energy with a wavelength of the line obtained by extrapolating a tangent to the phosphorescent spectrum at a tail on the short wavelength side is the T1 level, the difference between the S1 level and the T1 level of the TADF material is preferably smaller than or equal to 0.3 eV, further preferably smaller than or equal to 0.2 eV.

When a TADF material is used as the light-emitting substance, the S1 level of the host material is preferably higher than that of the TADF material. In addition, the T1 level of the host material is preferably higher than that of the TADF material.

As the host material in the light-emitting layer, various carrier-transport materials such as materials having an electron-transport property, materials having a hole-transport property, and the TADF materials can be used.

The material having a hole-transport property is preferably an organic compound having an amine skeleton or a  $\pi$ -electron rich heteroaromatic ring skeleton, for example. Examples of the material include a compound having an aromatic amine skeleton, such as 4,4'-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (abbreviation: NPB), N,N'-bis(3-methylphenyl)-N,N'-diphenyl-[1,1'-biphenyl]-4,4'-diamine (abbreviation: TPD), 4,4'-bis[N-(spiro-9,9'-bifluoren-2-yl)-N-phenylamino]biphenyl (abbreviation: BSPB), 4-phenyl-4'-(9-phenylfluoren-9-yl)triphenylamine (abbreviation: BPAFLP), 4-phenyl-3'-(9-phenylfluoren-9-yl)triphenylamine (abbreviation: mBPAFLP), 4-phenyl-4'-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBA1BP),

4,4'-diphenyl-4''-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBBi1BP), 4-(1-naphthyl)-4'-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBANB), 4,4'-di(1-naphthyl)-4''-(9-phenyl-9H-carbazol-3-yl)triphenylamine (abbreviation: PCBNB), 9,9-dimethyl-N-phenyl-N-[4-(9-phenyl-9H-carbazol-3-yl)phenyl]fluoren-2-amine (abbreviation: PCBAF), or N-phenyl-N-[4-(9-phenyl-9H-carbazol-3-yl)phenyl]-9,9'-spirobi[9H-fluoren]-2-amine (abbreviation: PCBASF); a compound having a carbazole skeleton, such as 1,3-bis(N-carbazolyl)benzene (abbreviation: mCP), 4,4'-di(N-carbazolyl)biphenyl (abbreviation: CBP), 3,6-bis(3,5-diphenylphenyl)-9-phenylcarbazole (abbreviation: CzTP), or 3,3'-bis(9-phenyl-9H-carbazole) (abbreviation: PCCP); a compound having a thiophene skeleton, such as 4,4',4''-(benzene-1,3,5-triyl)tri(dibenzothiophene) (abbreviation: DBT3P-II), 2,8-diphenyl-4-[4-(9-phenyl-9H-fluoren-9-yl)phenyl]dibenzothiophene (abbreviation: DBTFLP-III), or 4-[4-(9-phenyl-9H-fluoren-9-yl)phenyl]-6-phenyldibenzothiophene (abbreviation: DBTFLP-IV); and a compound having a furan skeleton, such as 4,4',4''-(benzene-1,3,5-triyl)tri(dibenzofuran) (abbreviation: DBF3P-II) or 4-[3-[3-(9-phenyl-9H-fluoren-9-yl)phenyl]phenyl]dibenzofuran (abbreviation: mmDBFFLBi-II). Among the above materials, the compound having an aromatic amine skeleton and the compound having a carbazole skeleton are preferable because these compounds are highly reliable and have high hole-transport properties to contribute to a reduction in driving voltage. In addition, the organic compounds given as examples of the material having a hole-transport property that can be used for the hole-transport layer 112 can also be used.

As the material having an electron-transport property, for example, a metal complex such as bis(10-hydroxybenzo[h]quinolinato)beryllium(II) (abbreviation: BeBq<sub>2</sub>), bis(2-methyl-8-quinolinolato)(4-phenylphenolato)aluminum(III) (abbreviation: BAlq), bis(8-quinolinolato)zinc(II) (abbreviation: Znq), bis[2-(2-benzoxazolyl)phenolato]zinc(II) (abbreviation: ZnPBO), or bis[2-(2-benzothiazolyl)phenolato]zinc(II) (abbreviation: ZnBTZ); or an organic compound having a  $\pi$ -electron deficient heteroaromatic ring is preferable. Examples of the organic compound having a  $\pi$ -electron deficient heteroaromatic ring include an organic compound having an azole skeleton, such as 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (abbreviation: PBD), 3-(4-biphenyl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole (abbreviation: TAZ), 1,3-bis[5-(p-tert-butylphenyl)-1,3,4-oxadiazol-2-yl]benzene (abbreviation: OXD-7), 9-[4-(5-phenyl-1,3,4-oxadiazol-2-yl)phenyl]-9H-carbazole (abbreviation: CO11), 2,2',2''-(1,3,5-benzenetriyl)tris(1-phenyl-1H-benzimidazole) (abbreviation: TPBI), 2-[3-(dibenzothiophen-4-yl)phenyl]-1-phenyl-1H-benzimidazole (abbreviation: mDBTBIm-II), or 4,4'-bis(5-methylbenzoxazol-2-yl)stilbene (abbreviation: BzOs); an organic compound having a heteroaromatic ring having a pyridine skeleton, such as 3,5-bis[3-(9H-carbazol-9-yl)phenyl]pyridine (abbreviation: 35DCzPPy), 1,3,5-tri[3-(3-pyridyl)phenyl]benzene (abbreviation: TmPyPB), bathophenanthroline (abbreviation: BPhen), bathocuproine (abbreviation: BCP), 2,9-di(2-naphthyl)-4,7-diphenyl-1,10-phenanthroline (abbreviation: NBPhen), or 2,2-(1,3-phenylene)bis[9-phenyl-1,10-phenanthroline] (abbreviation: mPPhen2P), an organic compound having a diazine skeleton, such as 2-[3-(dibenzothiophen-4-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 2mDBTPDBq-II), 2-[3'-(dibenzothiophen-4-yl)biphenyl-3-yl]dibenzo[f,h]quinoxaline (abbreviation: 2mDBTPDBq-II), 2-[3'-(9H-carbazol-9-yl)biphenyl-3-yl]

dibenzo[f,h]quinoxaline (abbreviation: 2mCzBPDBq), 2-[4'-(9-phenyl-9H-carbazol-3-yl)-3,1'-biphenyl-1-yl]dibenzo[f,h]quinoxaline (abbreviation: 2mpPCBPDDBq), 2-[4-(3,6-diphenyl-9H-carbazol-9-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 2CzPDBq-III), 7-[3-(dibenzothiophen-4-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 7mDBTPDBq-II), 6-[3-(dibenzothiophen-4-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 6mDBTPDBq-II), 9-[3'-dibenzothiophen-4-yl]biphenyl-3-yl]naphtho[1',2':4,5]furo[2,3-b]pyrazine (abbreviation: 9mDBtBPNfpr), 9-[3'-dibenzothiophen-4-yl]biphenyl-4-yl]naphtho[1',2':4,5]furo[2,3-b]pyrazine (abbreviation: 9pmDBtBPNfpr), 4,6-bis[3-(phenanthren-9-yl)phenyl]pyrimidine (abbreviation: 4,6mPnP2Pm), 4,6-bis[3-(4-dibenzothienyl)phenyl]pyrimidine (abbreviation: 4,6mDBTP2Pm-II), 4,6-bis[3-(9H-carbazol-9-yl)phenyl]pyrimidine (abbreviation: 4,6mCzP2Pm), 9,9'-(pyrimidine-4,6-diyl)bis(biphenyl-3,3'-diyl)]bis(9H-carbazole) (abbreviation: 4,6mCzBP2Pm), 8-(1,1'-biphenyl-4-yl)-4-[3-(dibenzothiophen-4-yl)phenyl]-[1]benzofuro[3,2-d]pyrimidine (abbreviation: 8BP-4mDBtPBfpm), 3,8-bis[3-(dibenzothiophen-4-yl)phenyl]benzofuro[2,3-b]pyrazine (abbreviation: 3,8mDBtP2Bfpr), 4,8-bis[3-(dibenzothiophen-4-yl)phenyl]-[1]benzofuro[3,2-d]pyrimidine (abbreviation: 4,8mDBtP2Bfpm), 8-[3-(dibenzothiophen-4-yl)(1,1'-biphenyl-3-yl)]naphtho[1',2':4,5]furo[3,2-d]pyrimidine (abbreviation: 8mDBtBPNfpm), 8-[2,(2'-binaphthalen-6-yl)-4-[3-(dibenzothiophen-4-yl)phenyl]-[1]benzofuro[3,2-d]pyrimidine (abbreviation: 8( $\beta$ N2)-4mDBtPBfpm), 2,2'-(pyridine-2,6-diyl)bis(4-phenylbenzo[h]quinazoline) (abbreviation: 2,6(P-Bqn)2Py), 2,2'-(pyridine-2,6-diyl)bis(4-[4-(2-naphthyl)phenyl]-6-phenylpyrimidine) (abbreviation: 2,6(NP-PPm)2Py), 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm), 2,6-bis(4-naphthalen-1-ylphenyl)-4-[4-(3-pyridyl)phenyl]pyrimidine (abbreviation: 2,4NP-6PyPPm), 4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenyl-6-(1,1'-biphenyl-4-yl)pyrimidine (abbreviation: 6BP-4Cz2PPm), or 7-[4-(9-phenyl-9H-carbazol-2-yl)quinazolin-2-yl]-7H-dibenzo[c,g]carbazol (abbreviation: PC-cgDBCzQz); and an organic compound having a heteroaromatic ring having a triazine skeleton, such as 2-[(1,1'-biphenyl)-4-yl]-4-phenyl-6-[9,9'-spirobi(9H-fluoren-2-yl)]-1,3,5-triazine (abbreviation: BP-SFTzn), 2-[3-[3-(benzo[b]naphtho[1,2-d]furan-8-yl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mBnfBPTzn), 2-[3-[3-(benzo[b]naphtho[1,2-d]furan-6-yl)phenyl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mBnfBPTzn-02), 2-[4-[3-(N-phenyl-9H-carbazol-3-yl)-9H-carbazol-9-yl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: PCCzPTzn), 9-[3-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-9'-phenyl-2,3'-bi-9H-carbazole (abbreviation: mPCCzPTzn-02), 2-[3'-(9,9-dimethyl-9H-fluoren-2-yl)-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mFBPTzn), 5-[3-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-7,7-dimethyl-5H,7H-indeno[2,1-b]carbazole (abbreviation: mINc(II)PTzn), 2-[3-[3-(dibenzothiophen-4-yl)phenyl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mDBtBPTzn), 2,4,6-tris[3'-(pyridin-3-yl)biphenyl-3-yl]-1,3,5-triazine (abbreviation: TmPPPbTz), 2-[3-(2,6-dimethyl-3-pyridinyl)-5-(9-phenanthrenyl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mPn-mDMePyPTzn), 11-(4-[1,1'-diphenyl]-4-yl-6-phenyl-1,3,5-triazin-2-yl)-11,12-dihydro-12-phenyl-indolo[2,3-a]carbazole (abbreviation: BP-Icz(II)Tzn), 2-[3'-(triphenylene-2-yl)-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mTpBPTzn), 9-[4-(4,6-diphenyl-1,3,5-triazin-2-yl)-2-dibenzothiophenyl]-2-phenyl-9H-carbazole (abbreviation: PCDBfTzn), or 2-[1,1'-

biphenyl]-3-yl-4-phenyl-6-(8-[1,1':4',1"-terphenyl]-4-yl-1-dibenzofuranyl)-1,3,5-triazine (abbreviation: mBP-TPDBfTzn). Among the above materials, the organic compound having a heteroaromatic ring having a diazine skeleton, the organic compound having a heteroaromatic ring having a pyridine skeleton, and the organic compound having a heteroaromatic ring having a triazine skeleton have high reliability and thus are preferable. In particular, the organic compound having a heteroaromatic ring having a diazine (pyrimidine or pyrazine) skeleton and the organic compound having a heteroaromatic ring having a triazine skeleton have a high electron-transport property to contribute to a reduction in driving voltage.

As the TADF material that can be used as the host material, the above materials mentioned as the TADF material can also be used. When the TADF material is used as the host material, triplet excitation energy generated in the TADF material is converted into singlet excitation energy by reverse intersystem crossing and transferred to the light-emitting substance, whereby the emission efficiency of the light-emitting device can be increased. Here, the TADF material functions as an energy donor, and the light-emitting substance functions as an energy acceptor.

This is very effective in the case where the light-emitting substance is a fluorescent substance. In that case, the S1 level of the TADF material is preferably higher than that of the fluorescent substance in order that high emission efficiency can be achieved. Furthermore, the T1 level of the TADF material is preferably higher than the S1 level of the fluorescent substance. Therefore, the T1 level of the TADF material is preferably higher than that of the fluorescent substance.

It is also preferable to use a TADF material that emits light whose wavelength overlaps with the wavelength on a lowest-energy-side absorption band of the fluorescent substance, in which case excitation energy is transferred smoothly from the TADF material to the fluorescent substance and light emission can be obtained efficiently.

In addition, in order to efficiently generate singlet excitation energy from the triplet excitation energy by reverse intersystem crossing, carrier recombination preferably occurs in the TADF material. It is also preferable that the triplet excitation energy generated in the TADF material not be transferred to the triplet excitation energy of the fluorescent substance. For that reason, the fluorescent substance preferably has a protective group around a luminophore (a skeleton which causes light emission) of the fluorescent substance. As the protective group, a substituent having no a bond and a saturated hydrocarbon are preferably used. Specific examples include an alkyl group having 3 to 10 carbon atoms, a substituted or unsubstituted cycloalkyl group having 3 to 10 carbon atoms, and a trialkylsilyl group having 3 to 10 carbon atoms. It is further preferable that the fluorescent substance have a plurality of protective groups. The substituents having no a bond are poor in carrier transport performance, whereby the TADF material and the luminophore of the fluorescent substance can be made away from each other with little influence on carrier transportation or carrier recombination. Here, the luminophore refers to an atomic group (skeleton) that causes light emission in a fluorescent substance. The luminophore is preferably a skeleton having a  $\pi$  bond, further preferably includes an aromatic ring, and still further preferably includes a condensed aromatic ring or a condensed heteroaromatic ring. Examples of the condensed aromatic ring or the condensed heteroaromatic ring include a phenanthrene skeleton, a stilbene skeleton, an acridone skeleton, a phenoxazine skeleton, and a

phenothiazine skeleton. Specifically, a fluorescent substance having any of a naphthalene skeleton, an anthracene skeleton, a fluorene skeleton, a chrysene skeleton, a triphenylene skeleton, a tetracene skeleton, a pyrene skeleton, a perylene skeleton, a coumarin skeleton, a quinacridone skeleton, and a naphthobisbenzofuran skeleton is preferred because of its high fluorescence quantum yield.

In the case where a fluorescent substance is used as the light-emitting substance, a material having an anthracene skeleton is suitably used as the host material. The use of a substance having an anthracene skeleton as the host material for the fluorescent substance makes it possible to obtain a light-emitting layer with high emission efficiency and high durability. Among the substances having an anthracene skeleton, a substance having a diphenylanthracene skeleton, in particular, a substance having a 9,10-diphenylanthracene skeleton, is chemically stable and thus is preferably used as the host material. The host material preferably has a carbazole skeleton because the hole-injection and hole-transport properties are improved; further preferably, the host material has a benzocarbazole skeleton in which a benzene ring is further condensed to carbazole because the HOMO level thereof is shallower than that of carbazole by approximately 0.1 eV and thus holes enter the host material easily. In particular, the host material preferably has a dibenzocarbazole skeleton because the HOMO level thereof is shallower than that of carbazole by approximately 0.1 eV so that holes enter the host material easily, the hole-transport property is improved, and the heat resistance is increased. Accordingly, a substance that has both a 9,10-diphenylanthracene skeleton and a carbazole skeleton (or a benzocarbazole or dibenzocarbazole skeleton) is further preferable as the host material. Note that in terms of the hole-injection and hole-transport properties described above, instead of a carbazole skeleton, a benzofluorene skeleton or a dibenzofluorene skeleton may be used. Examples of such a substance include 9-phenyl-3-[4-(10-phenyl-9-anthryl)phenyl]-9H-carbazole (abbreviation: PCzPA), 3-[4-(1-naphthyl)-phenyl]-9-phenyl-9H-carbazole (abbreviation: PCPN), 9-[4-(10-phenyl-9-anthracenyl)phenyl]-9H-carbazole (abbreviation: CzPA), 7-[4-(10-phenyl-9-anthryl)phenyl]-7H-dibenzo[c,g]carbazole (abbreviation: cgDBCzPA), 6-[3-(9,10-diphenyl-2-anthryl)phenyl]-benzo[b]naphtho[1,2-d]furan (abbreviation: 2mBnfPPA), 9-phenyl-10-[4-(9-phenyl-9H-fluoren-9-yl)biphenyl-4'-yl]anthracene (abbreviation: FLPPA), 9-(1-naphthyl)-10-[4-(2-naphthyl)phenyl]anthracene (abbreviation:  $\alpha$ N- $\beta$ NPAnt), 9-(1-naphthyl)-10-(2-naphthyl)anthracene (abbreviation:  $\alpha$ , $\beta$ ADN), 2-(10-phenylanthracen-9-yl)dibenzofuran, 2-(10-phenyl-9-anthracenyl)-benzo[b]naphtho[2,3-d]furan (abbreviation: Bnf(II)PhA), 9-(2-naphthyl)-10-[3-(2-naphthyl)phenyl]anthracene (abbreviation:  $\beta$ N-m $\beta$ NPAnt), and 1-[4-(10-[1,1'-biphenyl]-4-yl-9-anthracenyl)phenyl]-2-ethyl-1H-benzimidazole (abbreviation: EtBImPBPhA). In particular, CzPA, cgDBCzPA, 2mBnfPPA, and PCzPA exhibit excellent properties and thus are preferably selected.

Note that the host material may be a mixture of a plurality of kinds of substances; in the case of using a mixed host material, it is preferable to mix a material having an electron-transport property with a material having a hole-transport property. By mixing the material having an electron-transport property with the material having a hole-transport property, the transport property of the light-emitting layer 113 can be easily adjusted and a recombination region can be easily controlled. The weight ratio of the content of the

material having a hole-transport property to the content of the material having an electron-transport property may be 1:19 to 19:1.

Note that a phosphorescent substance can be used as part of the mixed material. When a fluorescent substance is used as the light-emitting substance, a phosphorescent substance can be used as an energy donor for supplying excitation energy to the fluorescent substance.

An exciplex may be formed of these mixed materials. 10 These mixed materials are preferably selected so as to form an exciplex that exhibits light emission whose wavelength overlaps with the wavelength on a lowest-energy-side absorption band of the light-emitting substance, in which case energy can be transferred smoothly and light emission 15 can be obtained efficiently. The use of such a structure is preferable because the driving voltage can also be reduced.

Note that at least one of the materials forming an exciplex 20 may be a phosphorescent substance. In this case, triplet excitation energy can be efficiently converted into singlet excitation energy by reverse intersystem crossing.

Combination of a material having an electron-transport 25 property and a material having a hole-transport property whose HOMO level is higher than or equal to that of the material having an electron-transport property is preferable for forming an exciplex efficiently. In addition, the LUMO level of the material having a hole-transport property is 30 preferably higher than or equal to that of the material having an electron-transport property. Note that the LUMO levels and the HOMO levels of the materials can be derived from the electrochemical characteristics (the reduction potentials and the oxidation potentials) of the materials that are measured by cyclic voltammetry (CV).

The formation of an exciplex can be confirmed by a 35 phenomenon in which the emission spectrum of the mixed film in which the material having a hole-transport property and the material having an electron-transport property are mixed is shifted to the longer wavelength side than the emission spectrum of each of the materials (or has another peak on the longer wavelength side) observed by comparison of the emission spectra of the material having a hole-transport property, the material having an electron-transport property, and the mixed film of these materials, for example. Alternatively, the formation of an exciplex can be confirmed 45 by a difference in transient response, such as a phenomenon in which the transient PL lifetime of the mixed film has longer lifetime components or has a larger proportion of delayed components than that of each of the materials, observed by comparison of transient photoluminescence (PL) of the material having a hole-transport property, the material having an electron-transport property, and the mixed film of these materials. The transient PL can be rephrased as transient electroluminescence (EL). That is, the formation of an exciplex can also be confirmed by a difference in transient response observed by comparison of the 55 transient EL of the material having a hole-transport property, the material having an electron-transport property, and the mixed film of these materials.

The electron-transport layer 114 contains a substance 60 having an electron-transport property. The substance having an electron-transport property preferably has an electron mobility higher than or equal to  $1 \times 10^{-6}$  cm<sup>2</sup>/Vs in the case where the square root of the electric field strength [V/cm] is 600. Note that any other substance can also be used as long as the substance has an electron-transport property higher than a hole-transport property. An organic compound having a  $\pi$ -electron deficient heteroaromatic ring is preferable as the above organic compound. The organic compound having a

$\pi$ -electron deficient heteroaromatic ring is preferably one or more of an organic compound having a heteroaromatic ring having a polyazole skeleton, an organic compound having a heteroaromatic ring having a pyridine skeleton, an organic compound having a heteroaromatic ring having a diazine skeleton, and an organic compound having a heteroaromatic ring having a triazine skeleton.

Specific examples of the material having an electron-transport property that can be used for the above electron-transport layer include an organic compound having an azole skeleton, such as 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (abbreviation: PBD), 3-(4-biphenyl)-4-phenyl-5-(4-tert-butylphenyl)-1,2,4-triazole (abbreviation: TAZ), 1,3-bis[5-(p-tert-butylphenyl)-1,3,4-oxadiazol-2-yl]benzene (abbreviation: OXD-7), 9-[4-(5-phenyl-1,3,4-oxadiazol-2-yl)phenyl]-9H-carbazole (abbreviation: CO11), 2,2',2"-(1,3,5-benzenetriyl)tris(1-phenyl-1H-benzimidazole) (abbreviation: TPBI), 2-[3-(dibenzothiophen-4-yl)phenyl]-1-phenyl-1H-benzimidazole (abbreviation: mDBTBIm-II), or 4,4'-bis(5-methylbenzoxazol-2-yl)stilbene (abbreviation: BzOs); an organic compound having a heteroaromatic ring having a pyridine skeleton, such as 3,5-bis[3-(9H-carbazol-9-yl)phenyl]pyridine (abbreviation: 35DCzPPy), 1,3,5-tri[3-(3-pyridyl)phenyl]benzene (abbreviation: TmPyPB), bathophenanthroline (abbreviation: BPhen), bathocuproine (abbreviation: BCP), 2,9-di(2-naphthyl)-4,7-diphenyl-1,10-phenanthroline (abbreviation: NBPhen), or 2,2-(1,3-phenylene)bis[9-phenyl-1,10-phenanthroline] (abbreviation: mPPhen2P), an organic compound having a diazine skeleton, such as 2-[3-(dibenzothiophen-4-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 2mDBTPDBq-II), 2-[3'-(dibenzothiophen-4-yl)biphenyl-3-yl]dibenzo[f,h]quinoxaline (abbreviation: 2mDBBPDBq-II), 2-[3'-(9H-carbazol-9-yl)biphenyl-3-yl]dibenzo[f,h]quinoxaline (abbreviation: 2mCzBPDBq), 2-[4'-(9-phenyl-9H-carbazol-3-yl)-3,1'-biphenyl-1-yl]dibenzo[f,h]quinoxaline (abbreviation: 2mpPCBPDQ), 2-[4-(3,6-diphenyl-9H-carbazol-9-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 2CzPDBq-III), 7-[3-(dibenzothiophen-4-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 7mDBTPDBq-II), 6-[3-(dibenzothiophen-4-yl)phenyl]dibenzo[f,h]quinoxaline (abbreviation: 6mDBTPDBq-II), 9-[3'(dibenzothiophen-4-yl)biphenyl-3-yl]naphtho[1',2':4,5]furo[2,3-b]pyrazine (abbreviation: 9mDBtBPNfpr), 9-[3'-dibenzothiophen-4-yl)biphenyl-4-yl]naphtho[1',2':4,5]furo[2,3-b]pyrazine (abbreviation: 9pmDBtBPNfpr), 4,6-bis[3-(phenanthren-9-yl)phenyl]pyrimidine (abbreviation: 4,6mPnP2Pm), 4,6-bis[3-(4-dibenzothienyl)phenyl]pyrimidine (abbreviation: 4,6mDBTP2Pm-II), 4,6-bis[3-(9H-carbazol-9-yl)phenyl]pyrimidine (abbreviation: 4,6mCzP2Pm), 9,9'-[pyrimidine-4,6-diyl]bis(biphenyl-3,3'-diyl)]bis(9H-carbazole) (abbreviation: 4,6mCzBP2Pm), 8-(1,1'-biphenyl-4-yl)-4-[3-(dibenzothiophen-4-yl)phenyl]-[1]benzofuro[3,2-d]pyrimidine (abbreviation: 8BP-4mDBtPBfpm), 3,8-bis[3-(dibenzothiophen-4-yl)phenyl]benzofuro[2,3-b]pyrazine (abbreviation: 3,8mDBtP2Bfpm), 4,8-bis[3-(dibenzothiophen-4-yl)phenyl]-[1]benzofuro[3,2-d]pyrimidine (abbreviation: 4,8mDBtP2Bfpm), 8-[3'(dibenzothiophen-4-yl)(1,1'-biphenyl-3-yl)]naphtho[1',2':4,5]furo[3,2-d]pyrimidine (abbreviation: 8mDBtBPNfpm), 8-[2,(2'-binaphthalen)-6-yl]-4-[3-(dibenzothiophen-4-yl)phenyl]-[1]benzofuro[3,2-d]pyrimidine (abbreviation: 8(BN2)-4mDBtPBfpm), 2,2'-(pyridine-2,6-diyl)bis(4-phenylbenzo[h]quinazoline) (abbreviation: 2,6(P-Bqn)2Py), 2,2'-(pyridine-2,6-diyl)bis{4-[4-(2-naphthyl)phenyl]-6-phenylpyrimidine} (abbreviation: 2,6(NP-PPm)2Py), 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm), 2,6-bis(4-naphthalen-1-ylphenyl)-4-[4-(3-pyridyl)phenyl]pyrimidine (abbreviation: 2,4NP-6PyPPm), 4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenyl-6-(1,1'-biphenyl-4-yl)pyrimidine (abbreviation: 6BP-4Cz2PPm), or 7-[4-(9-phenyl-9H-carbazol-2-yl)quinazolin-2-yl]-7H-dibenzo[c,g]carbazol (abbreviation: PC-cgDBCzQz); and an organic compound having a triazine skeleton, such as 2-[(1,1'-biphenyl)-4-yl]-4-phenyl-6-[9,9'-spirobi(9H-fluoren)-2-yl]-1,3,5-triazine (abbreviation: BP-SFTzn), 2-[3-[3-(benzo[b]naphtho[1,2-d]furan-8-yl)phenyl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mBnfBPTzn), 2-[3-[3-(benzo[b]naphtho[1,2-d]furan-6-yl)phenyl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mBnfBPTzn-02), 2-[4-[3-(N-phenyl-9H-carbazol-3-yl)-9H-carbazol-9-yl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: PCCzPTzn), 9-[3-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-9'-phenyl-2,3'-bi-9H-carbazole (abbreviation: mPCCzPTzn-02), 2-[3'-(9,9-dimethyl-9H-fluoren-2-yl)-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mFBPTzn), 5-[3-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-7,7-dimethyl-5H,7H-indeno[2,1-b]carbazole (abbreviation: mINc(II)PTzn), 2-[3-[3-(dibenzothiophen-4-yl)phenyl]phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mDBtBPTzn), 2,4,6-tris[3'-(pyridin-3-yl)biphenyl-3-yl]-1,3,5-triazine (abbreviation: TmPPPyTz), 2-[3-(2,6-dimethyl-3-pyridinyl)-5-(9-phenanthrenyl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mPn-mDMePyPTzn), 11-(4-[1,1'-diphenyl]-4-yl-6-phenyl-1,3,5-triazin-2-yl)-11,12-dihydro-12-phenyl-indolo[2,3-a]carbazole (abbreviation: BP-Icz(II)Tm), 2-[3'-(triphenyl-2-yl)-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mTpBPTzn), 9-[4-(4,6-diphenyl-1,3,5-triazin-2-yl)-2-dibenzothiophenyl]-2-phenyl-9H-carbazole (abbreviation: PCDBfTzn), or 2-[1,1'-biphenyl]-3-yl-4-phenyl-6-(8-[1,1':4',1"-terphenyl]-4-yl-1-dibenzofuranyl)-1,3,5-triazine (abbreviation: mBP-TPDBfTzn). Among the above materials, the organic compound having a heteroaromatic ring having a diazine skeleton, the organic compound having a heteroaromatic ring having a pyridine skeleton, and the organic compound having a heteroaromatic ring having a triazine skeleton have high reliability and thus are preferable. In particular, the organic compound having a heteroaromatic ring having a diazine (pyrimidine or pyrazine) skeleton and the organic compound having a heteroaromatic ring having a triazine skeleton have a high electron-transport property to contribute to a reduction in driving voltage.

The use of a material having an electron-transport property and a low refractive index for the electron-transport layer **114** can offer a light-emitting device with better properties.

The organic compound having an electron-transport property and a low refractive index that can be used for the electron-transport layer preferably includes at least one six-membered heteroaromatic ring having 1 to 3 nitrogen atoms, a plurality of aromatic hydrocarbon rings each of which has 6 to 14 carbon atoms forming a ring and at least two of which are benzene rings, and a plurality of hydrocarbon groups forming a bond by  $sp^3$  hybrid orbitals.

In the above organic compound, carbon atoms forming a bond by  $sp^3$  hybrid orbitals preferably account for higher than or equal to 10% and lower than or equal to 60%, further preferably higher than or equal to 10% and lower than or equal to 50% of total carbon atoms in molecules of the organic compound. Alternatively, when the above organic compound is subjected to  $^1\text{H-NMR}$  measurement, the inte-

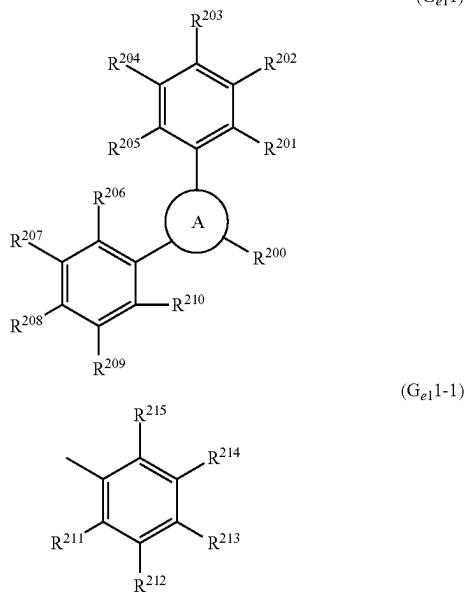
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gral value of signals at lower than 4 ppm is preferably 1/2 or more of the integral value of signals at 4 ppm or higher.

The molecular weight of the organic compound having an electron-transport property is preferably greater than or equal to 500 and less than or equal to 2000. It is preferable that all the hydrocarbon groups forming a bond by  $sp^3$  hybrid orbitals in the above organic compound be bonded to the aromatic hydrocarbon rings each having 6 to 14 carbon atoms forming a ring, and the LUMO of the organic compound not be distributed in the aromatic hydrocarbon rings.

The organic compound having an electron-transport property is preferably an organic compound represented by General Formula (G<sub>e1</sub>1) or (G<sub>e1</sub>1-1).

[Chemical Formula 11]



In the formula, A represents a six-membered heteroaromatic ring having 1 to 3 nitrogen atoms, and is preferably any one of a pyridine ring, a pyrimidine ring, a pyrazine ring, a pyridazine ring, and a triazine ring.

R<sup>200</sup> represents any one of hydrogen, an alkyl group having 1 to 6 carbon atoms, an alicyclic group having 3 to 10 carbon atoms, and a substituent represented by Formula (G-1-1).

At least one of R<sup>201</sup> to R<sup>215</sup> represents a phenyl group having a substituent and the others each independently represent any of hydrogen, an alkyl group having 1 to 6 carbon atoms, an alicyclic group having 3 to 10 carbon atoms, a substituted or unsubstituted aromatic hydrocarbon group having 6 to 14 carbon atoms in a ring, and a substituted or unsubstituted pyridyl group. Note that R<sup>201</sup>, R<sup>203</sup>, R<sup>205</sup>, R<sup>206</sup>, R<sup>208</sup>, R<sup>210</sup>, R<sup>211</sup>, R<sup>213</sup>, and R<sup>215</sup> are preferably hydrogen. The phenyl group having a substituent has one or two substituents, which each independently represent any of an alkyl group having 1 to 6 carbon atoms, an alicyclic group having 3 to 10 carbon atoms, and a substituted or unsubstituted aromatic hydrocarbon group having 6 to 14 carbon atoms in a ring.

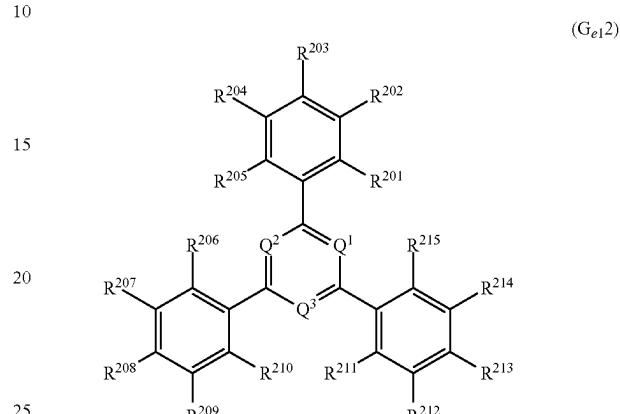
The organic compound represented by General Formula (G<sub>e1</sub>1) has a plurality of hydrocarbon groups selected from an alkyl group having 1 to 6 carbon atoms and an alicyclic group having 3 to 10 carbon atoms, and carbon atoms

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forming a bond by  $sp^3$  hybrid orbitals account for higher than or equal to 10% and lower than or equal to 60% of total carbon atoms in molecules of the organic compound.

The organic compound having an electron-transport property is preferably an organic compound represented by General Formula (G<sub>e1</sub>2).

[Chemical Formula 12]



In the formula, two or three of Q<sup>1</sup> to Q<sup>3</sup> represent N; in the case where two of Q<sup>1</sup> to Q<sup>3</sup> are N, the rest represents CH.

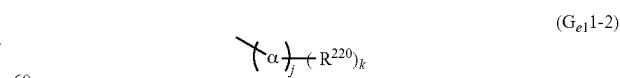
At least one of R<sup>201</sup> to R<sup>215</sup> represents a phenyl group having a substituent and the others each independently represent any of hydrogen, an alkyl group having 1 to 6 carbon atoms, an alicyclic group having 3 to 10 carbon atoms, a substituted or unsubstituted aromatic hydrocarbon group having 6 to 14 carbon atoms in a ring, and a substituted or unsubstituted pyridyl group. Note that R<sup>201</sup>, R<sup>203</sup>, R<sup>205</sup>, R<sup>206</sup>, R<sup>208</sup>, R<sup>210</sup>, R<sup>211</sup>, R<sup>213</sup>, and R<sup>215</sup> are preferably hydrogen. The phenyl group having a substituent has one or two substituents, which each independently represent any of an alkyl group having 1 to 6 carbon atoms, an alicyclic group having 3 to 10 carbon atoms, and a substituted or unsubstituted aromatic hydrocarbon group having 6 to 14 carbon atoms in a ring.

The organic compound represented by General Formula (G<sub>e1</sub>2) has a plurality of hydrocarbon groups selected from an alkyl group having 1 to 6 carbon atoms and an alicyclic group having 3 to 10 carbon atoms, and carbon atoms forming a bond by  $sp^3$  hybrid orbitals account for higher than or equal to 10% and lower than or equal to 60% of total carbon atoms in molecules of the organic compound.

In the organic compound represented by General formula (G<sub>e1</sub>1) or (G<sub>e1</sub>2), the phenyl group having a substituent is preferably a group represented by Formula (G<sub>e1</sub>1-2).

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[Chemical Formula 13]



In the formula, a represents a substituted or unsubstituted phenylene group and is preferably a meta-substituted phenylene group. In the case where the meta-substituted phenylene group has a substituent, the substituent is also preferably meta-substituted. The substituent is preferably an alkyl group having 1 to 6 carbon atoms or an alicyclic group

having 3 to 10 carbon atoms, further preferably an alkyl group having 1 to 6 carbon atoms, and still further preferably a t-butyl group.

$R^{220}$  represents an alkyl group having 1 to 6 carbon atoms, an alicyclic group having 3 to 10 carbon atoms, or a substituted or unsubstituted aromatic hydrocarbon group having 6 to 14 carbon atoms in a ring.

In addition, j and n each represent 1 or 2. In the case where j is 2, a plurality of a may be the same or different from each other. In the case where k is 2, a plurality of  $R^{220}$  may be the same or different from each other.  $R^{220}$  is preferably a phenyl group, further preferably a phenyl group that has an alkyl group having 1 to 6 carbon atoms or an alicyclic group having 3 to 10 carbon atoms at one or both of the two meta-positions. The substituent at one or both of the two meta-positions of the phenyl group is preferably an alkyl group having 1 to 6 carbon atoms, further preferably a t-butyl group.

The above-described organic compound having an electron-transport property has a high electron-transport property and has an ordinary refractive index greater than or equal to 1.50 and less than or equal to 1.75 in the blue light emission range (455 nm to 465 nm) or an ordinary refractive index greater than or equal to 1.45 and less than or equal to 1.70 with respect to light with a wavelength of 633 nm, which is typically used in measurement of refractive index.

In the case where the organic compound having an electron-transport property is used for the electron-transport layer 114, the electron-transport layer 114 preferably includes a metal complex of an alkali metal or an alkaline earth metal. A heterocyclic compound having a diazine skeleton, a heterocyclic compound having a triazine skeleton, and a heterocyclic compound having a pyridine skeleton are preferable in terms of driving lifetime because they are likely to form an exciplex with an organometallic complex of an alkali metal with stable energy (the emission wavelength of the exciplex easily becomes longer). In particular, the heterocyclic compound having a diazine skeleton or the heterocyclic compound having a triazine skeleton has a deep LUMO level and thus is preferred for stabilization of energy of an exciplex.

Note that the organometallic complex of an alkali metal is preferably a metal complex of sodium or lithium. Alternatively, the organometallic complex of an alkali metal preferably has a ligand having a quinolinol skeleton. Further preferably, the organometallic complex of an alkali metal is preferably a lithium complex having an 8-quinolinolato structure or a derivative thereof. The derivative of a lithium complex having an 8-quinolinolato structure is preferably a lithium complex having an 8-quinolinolato structure having an alkyl group, and further preferably has a methyl group.

It is possible that 8-quinolinolato-lithium having an alkyl group be a metal complex with a low refractive index. Specifically, the ordinary refractive index of the metal complex in a thin film state with respect to light with a wavelength in the range of 455 nm to 465 nm can be greater than or equal to 1.45 and less than or equal to 1.70, and the ordinary refractive index thereof with respect to light with a wavelength of 633 nm can be greater than or equal to 1.40 and less than or equal to 1.65.

Specific examples of the metal complex include 8-hydroxyquinolinolato-lithium (abbreviation: Liq) and 8-hydroxyquinolinolato-sodium (abbreviation: Naq). In particular, a complex of a monovalent metal ion, especially a complex of lithium is preferable, and Liq is further preferable. Note that in the case where the 8-hydroxyquinolinolato structure is included, a methyl-substituted product (e.g., a 2-methyl-

substituted product, a 5-methyl-substituted product, or 6-methyl-substituted product) thereof is also preferably used, for example. In particular, the use of an alkali metal complex having an 8-quinolinolato structure having an alkyl group at the 6 position results in lowering the driving voltage of a light-emitting device.

The electron mobility of the electron-transport layer 114 in the case where the square root of the electric field strength [V/cm] is 600 is preferably higher than or equal to  $1 \times 10^{-7}$  cm<sup>2</sup>/Vs and lower than or equal to  $5 \times 10^{-5}$  cm<sup>2</sup>/Vs. The amount of electrons injected into the light-emitting layer can be controlled by the reduction in the electron-transport property of the electron-transport layer 114, whereby the light-emitting layer can be prevented from having excess electrons. It is particularly preferable to employ this structure when the hole-injection layer is formed using a composite material that includes a material having a hole-transport property with a relatively deep HOMO level of -5.7 eV or higher and -5.4 eV or lower, in which case a long lifetime can be achieved. In this case, the material having an electron-transport property preferably has a HOMO level of -6.0 eV or higher.

A layer including an alkali metal, an alkaline earth metal, a compound thereof, or a complex thereof such as lithium fluoride (LiF), cesium fluoride (CsF), calcium fluoride (CaF<sub>2</sub>), or 8-hydroxyquinolinolato-lithium (Liq) may be provided as the electron-injection layer 115 between the electron-transport layer 114 and the second electrode 102. For example, an electride or a layer that is formed using a substance having an electron-transport property and that includes an alkali metal, an alkaline earth metal, or a compound thereof can be used as the electron-injection layer 115. Examples of the electride include a substance in which electrons are added at high concentration to calcium oxide-aluminum oxide.

Note that as the electron-injection layer 115, it is possible to use a layer including a substance that has an electron-transport property (preferably an organic compound having a bipyridine skeleton) and includes a fluoride of the alkali metal or the alkaline earth metal at a concentration higher than that at which the electron-injection layer 115 becomes in a microcrystalline state (50 wt % or higher). Since the layer has a low refractive index, a light-emitting device including the layer can have high external quantum efficiency.

Instead of the electron-injection layer 115 in FIG. 1A, the charge-generation layer 116 may be provided (FIG. 1B). The charge-generation layer 116 refers to a layer capable of injecting holes into a layer in contact with the cathode side 50 of the charge-generation layer and electrons into a layer in contact with the anode side thereof when a potential is applied. The charge-generation layer 116 includes at least a p-type layer 117. The p-type layer 117 is preferably formed using any of the composite materials given above as 55 examples of materials that can be used for the hole-injection layer 111. The p-type layer 117 may be formed by stacking a film including the above-described acceptor material as a material included in the composite material and a film including a hole-transport material. When a potential is 60 applied to the p-type layer 117, electrons are injected into the electron-transport layer 114 and holes are injected into the second electrode 102 which is the cathode; thus, the light-emitting device operates.

Note that the charge-generation layer 116 preferably includes one or both of an electron-relay layer 118 and an electron-injection buffer layer 119 in addition to the p-type layer 117.

The electron-relay layer 118 includes at least the substance having an electron-transport property and has a function of preventing an interaction between the electron-injection buffer layer 119 and the p-type layer 117 and smoothly transferring electrons. The LUMO level of the substance having an electron-transport property included in the electron-relay layer 118 is preferably between the LUMO level of the acceptor substance in the p-type layer 117 and the LUMO level of a substance included in a layer of the electron-transport layer 114 that is in contact with the charge-generation layer 116. As a specific value of the energy level, the LUMO level of the substance having an electron-transport property in the electron-relay layer 118 is preferably higher than or equal to -5.0 eV, further preferably higher than or equal to -5.0 eV and lower than or equal to -3.0 eV. Note that as the substance having an electron-transport property in the electron-relay layer 118, a phthalocyanine-based material or a metal complex having a metal-oxygen bond and an aromatic ligand is preferably used.

The electron-injection buffer layer 119 can be formed using a substance having a high electron-injection property, e.g., an alkali metal, an alkaline earth metal, a rare earth metal, or a compound thereof (an alkali metal compound (including an oxide such as lithium oxide, a halide, and a carbonate such as lithium carbonate or cesium carbonate), an alkaline earth metal compound (including an oxide, a halide, and a carbonate), or a rare earth metal compound (including an oxide, a halide, and a carbonate)).

In the case where the electron-injection buffer layer 119 contains the substance having an electron-transport property and a donor substance, an organic compound such as tetrathianaphthacene (abbreviation: TTN), nickelocene, or decamethylnickelocene can be used as the donor substance, as well as an alkali metal, an alkaline earth metal, a rare earth metal, a compound thereof (e.g., an alkali metal compound (including an oxide such as lithium oxide, a halide, lithium carbonate, or a carbonate such as cesium carbonate), an alkaline earth metal compound (including an oxide, a halide, and a carbonate), or a rare earth metal compound (including an oxide, a halide, and a carbonate)).

As the substance having an electron-transport property, a material similar to the above-described material for the electron-transport layer 114 can be used. Since the above-described material is an organic compound having a low refractive index, the use of the material for the electron-injection buffer layer 119 can offer a light-emitting device with high external quantum efficiency.

For the second electrode 102, a metal, an alloy, an electrically conductive compound, or a mixture thereof each having a low work function (specifically, lower than or equal to 3.8 eV) or the like can be used. Specific examples of such a cathode material are elements belonging to Group 1 or 2 of the periodic table, such as alkali metals (e.g., lithium (Li) or cesium (Cs)), magnesium (Mg), calcium (Ca), and strontium (Sr), alloys containing these elements (e.g., MgAg and AlLi), rare earth metals such as europium (Eu) and ytterbium (Yb), and alloys containing these rare earth metals. However, when the electron-injection layer is provided between the second electrode 102 and the electron-transport layer, a variety of conductive materials such as Al, Ag, ITO, or indium oxide-tin oxide containing silicon or silicon oxide can be used for the second electrode 102 regardless of the work function.

When the second electrode 102 is formed using a material that transmits visible light, the light-emitting device can emit light from the cathode side as illustrated in FIG. 1D.

When the first electrode 101 is formed on the substrate side, the light-emitting device can be what is called a top-emission light-emitting device.

Films of these conductive materials can be deposited by a dry process such as a vacuum evaporation method or a sputtering method, an ink-jet method, a spin coating method, or the like. Alternatively, a wet process using a sol-gel method or a wet process using a paste of a metal material may be employed.

Any of a variety of methods can be used for forming the EL layer 103, regardless of a dry method or a wet method. For example, a vacuum evaporation method, a gravure printing method, an offset printing method, a screen printing method, an ink-jet method, a spin coating method, or the like may be used.

Different methods may be used to form the electrodes or the layers described above.

The structure of the layers provided between the first electrode 101 and the second electrode 102 is not limited to the above-described structure. Preferably, a light-emitting region where holes and electrons recombine is positioned away from the first electrode 101 and the second electrode 102 so as to inhibit quenching due to the proximity of the light-emitting region and a metal used for electrodes or carrier-injection layers.

Furthermore, in order that transfer of energy from an exciton generated in the light-emitting layer can be inhibited, preferably, the hole-transport layer or the electron-transport layer, which is in contact with the light-emitting layer 113, particularly a carrier-transport layer closer to the recombination region in the light-emitting layer 113, is preferably formed using a substance having a wider band gap than the light-emitting material of the light-emitting layer or the light-emitting material included in the light-emitting layer.

Next, an embodiment of a light-emitting device with a structure in which a plurality of light-emitting units are stacked (this type of light-emitting device is also referred to as a stacked or tandem device) is described. This light-emitting device includes a plurality of light-emitting units between an anode and a cathode. One light-emitting unit has substantially the same structure as the EL layer 103 illustrated in FIG. 1A. In other words, the light-emitting device illustrated in FIG. 1A or 1B includes a single light-emitting unit, and the tandem device includes a plurality of light-emitting units.

In the tandem device, a first light-emitting unit and a second light-emitting unit are stacked between an anode and a cathode, and a charge-generation layer is provided between the first light-emitting unit and the second light-emitting unit. The anode and the cathode correspond, respectively, to the first electrode 101 and the second electrode 102 illustrated in FIG. 1A, and the materials given in the description for FIG. 1A can be used. Furthermore, the first light-emitting unit and the second light-emitting unit may have the same structure or different structures.

The charge-generation layer in the tandem device has a function of injecting electrons into one of the light-emitting units and injecting holes into the other of the light-emitting units when voltage is applied between the anode and the cathode. That is, the charge-generation layer injects electrons into the first light-emitting unit and holes into the second light-emitting unit when voltage is applied such that the potential of the anode becomes higher than the potential of the cathode.

The charge-generation layer preferably has a structure similar to that of the charge-generation layer 116 described

with reference to FIG. 1B. A composite material of an organic compound and a metal oxide has an excellent carrier-injection property and an excellent carrier-transport property; thus, low-voltage driving and low-current driving can be achieved. In the case where the anode-side surface of a light-emitting unit is in contact with the charge-generation layer, the charge-generation layer can also function as a hole-injection layer of the light-emitting unit; therefore, a hole-injection layer is not necessarily provided in the light-emitting unit.

In the case where the charge-generation layer of the tandem device includes the electron-injection buffer layer 119, the electron-injection buffer layer 119 functions as the electron-injection layer in the light-emitting unit on the anode side; thus, an electron-injection layer is not necessarily formed in the light-emitting unit on the anode side.

The tandem device having two light-emitting units is described above; one embodiment of the present invention can also be applied to a tandem device in which three or more light-emitting units are stacked. With a plurality of light-emitting units partitioned by the charge-generation layer between a pair of electrodes, it is possible to provide a long-life element that can emit light with high luminance at a low current density. A light-emitting apparatus that can be driven at a low voltage and has low power consumption can be provided.

When the emission colors of the light-emitting units are different, light emission of a desired color can be obtained from the light-emitting device as a whole. For example, in a light-emitting device having two light-emitting units, the emission colors of the first light-emitting unit may be red and green and the emission color of the second light-emitting unit may be blue, so that the light-emitting device can emit white light as the whole.

The above-described layers or electrodes such as the EL layer 103, the first light-emitting unit, the second light-emitting unit, and the charge-generation layer can be formed by a method such as an evaporation method (including a vacuum evaporation method), a droplet discharge method (also referred to as an ink-jet method), a coating method, or a gravure printing method. A low molecular material, a middle molecular material (including an oligomer and a dendrimer), or a high molecular material may be included in the layers or electrodes.

This embodiment can be freely combined with any of the other embodiments.

## Embodiment 2

In this embodiment, a light-emitting apparatus including the light-emitting device described in Embodiment 1 is described.

In this embodiment, an active matrix light-emitting apparatus manufactured using the light-emitting device described in Embodiment 1 is described with reference to FIGS. 2A and 2B. Note that FIG. 2A is a top view of the light-emitting apparatus and FIG. 2B is a cross-sectional view taken along the dashed-dotted lines A-B and C-D in FIG. 2A. This light-emitting apparatus includes a driver circuit portion (source line driver circuit) 601, a pixel portion 602, and a driver circuit portion (gate line driver circuit) 603, which are to control light emission of a light-emitting device and illustrated with dotted lines. Reference numeral 604 denotes a sealing substrate; 605, a sealing material; and 607, a space surrounded by the sealing material 605.

Reference numeral 608 denotes a lead wiring for transmitting signals to be input to the source line driver circuit

601 and the gate line driver circuit 603 and receiving signals such as a video signal, a clock signal, a start signal, and a reset signal from a flexible printed circuit (FPC) 609 serving as an external input terminal. Although only the FPC is illustrated here, a printed wiring board (PWB) may be attached to the FPC. The light-emitting apparatus in the present specification includes, in its category, not only the light-emitting apparatus itself but also the light-emitting apparatus provided with the FPC or the PWB.

Next, a cross-sectional structure is described with reference to FIG. 2B. The driver circuit portions and the pixel portion are formed over an element substrate 610; here, the source line driver circuit 601, which is a driver circuit portion, and one pixel in the pixel portion 602 are illustrated.

The element substrate 610 may be a substrate containing glass, quartz, an organic resin, a metal, an alloy, a semiconductor, or the like or a plastic substrate formed of fiber reinforced plastic (FRP), poly(vinyl fluoride) (PVF), polyester, acrylic resin, or the like.

The structure of transistors used in pixels or driver circuits is not particularly limited. For example, inverted staggered transistors may be used, or staggered transistors may be used. Furthermore, top-gate transistors or bottom-gate transistors may be used. A semiconductor material used for the transistors is not particularly limited, and for example, silicon, germanium, silicon carbide, gallium nitride, or the like can be used. Alternatively, an oxide semiconductor containing at least one of indium, gallium, and zinc, such as an In—Ga—Zn-based metal oxide, may be used.

There is no particular limitation on the crystallinity of a semiconductor material used for the transistors, and an amorphous semiconductor or a semiconductor having crystallinity (a microcrystalline semiconductor, a polycrystalline semiconductor, a single crystal semiconductor, or a semiconductor partly including crystal regions) may be used. A semiconductor having crystallinity is preferably used because deterioration of the transistor characteristics can be inhibited.

Here, an oxide semiconductor is preferably used for semiconductor devices such as the transistors provided in the pixels or driver circuits and transistors used for touch sensors described later, and the like. In particular, an oxide semiconductor having a wider band gap than silicon is preferably used. When an oxide semiconductor having a wider band gap than silicon is used, the off-state current of the transistors can be reduced.

The oxide semiconductor preferably contains at least indium (In) or zinc (Zn). Further preferably, the oxide semiconductor contains an oxide represented by an In-M-Zn-based oxide (M represents a metal such as Al, Ti, Ga, Ge, Y, Zr, Sn, La, Ce, or Hf).

As a semiconductor layer, it is particularly preferable to use an oxide semiconductor film including a plurality of crystal parts whose c-axes are aligned perpendicular to a surface on which the semiconductor layer is formed or the top surface of the semiconductor layer and in which the adjacent crystal parts have no grain boundary.

The use of such materials for the semiconductor layer makes it possible to provide a highly reliable transistor in which a change in the electrical characteristics is inhibited.

Charge accumulated in a capacitor through a transistor including the above-described semiconductor layer can be held for a long time because of the low off-state current of the transistor. When such a transistor is used in a pixel, operation of a driver circuit can be stopped while a gray scale of an image displayed in each display region is

maintained. As a result, an electronic device with extremely low power consumption can be obtained.

For stable characteristics of the transistor, a base film is preferably provided. The base film can be formed with a single layer or stacked layers using an inorganic insulating film such as a silicon oxide film, a silicon nitride film, a silicon oxynitride film, or a silicon nitride oxide film. The base film can be formed by a sputtering method, a chemical vapor deposition (CVD) method (e.g., a plasma CVD method, a thermal CVD method, or a metal organic CVD (MOCVD) method), an atomic layer deposition (ALD) method, a coating method, a printing method, or the like. Note that the base film is not necessarily provided.

Note that an FET 623 is illustrated as a transistor formed in the driver circuit portion 601. The driver circuit may be formed with any of a variety of circuits such as a CMOS circuit, a PMOS circuit, or an NMOS circuit. Although a driver integrated type in which the driver circuit is formed over the substrate is illustrated in this embodiment, the driver circuit is not necessarily formed over the substrate, and the driver circuit can be formed outside, not over the substrate.

The pixel portion 602 includes a plurality of pixels including a switching FET 611, a current controlling FET 612, and a first electrode 613 electrically connected to a drain of the current controlling FET 612. One embodiment of the present invention is not limited to the structure. The pixel portion 602 may include three or more FETs and a capacitor in combination.

Note that an insulator 614 is formed to cover an end portion of the first electrode 613. Here, the insulator 614 can be formed using a positive photosensitive acrylic resin film.

In order to improve the coverage with an EL layer or the like which is formed later, the insulator 614 is formed to have a curved surface with curvature at its upper or lower end portion. For example, in the case where a positive photosensitive acrylic resin is used as a material of the insulator 614, only the upper end portion of the insulator 614 preferably has a curved surface with a curvature radius (0.2 μm to 3 μm). As the insulator 614, either a negative photosensitive resin or a positive photosensitive resin can be used.

An EL layer 616 and a second electrode 617 are formed over the first electrode 613. Here, the first electrode 613 serves as an anode. A material having a high work function is preferably used as a material for the anode. For example, a single-layer film of an ITO film, an indium tin oxide film containing silicon, an indium oxide film containing zinc oxide at 2 wt % to 20 wt %, a titanium nitride film, a chromium film, a tungsten film, a Zn film, a Pt film, or the like, a stack of any of these films and a film containing silver as its main component, a stack of a titanium nitride film and a film containing aluminum as its main component, a stack of three layers of a titanium nitride film, a film containing aluminum as its main component, and a titanium nitride film, or the like can be used. The stacked-layer structure enables low wiring resistance, favorable ohmic contact, and a function as an anode.

The EL layer 616 is formed by any of a variety of methods such as an evaporation method using an evaporation mask, an inkjet method, and a spin coating method. The EL layer 616 has the structure described in Embodiment 1.

As a material used for the second electrode 617, which is formed over the EL layer 616, a material having a low work function (e.g., Al, Mg, Li, and Ca, or an alloy or a compound thereof, such as MgAg, MgIn, and AlLi) is preferably used. In the case where light generated in the EL layer 616 passes

through the second electrode 617, a stack of a thin metal or alloy film and a transparent conductive film (e.g., ITO, indium oxide containing zinc oxide at 2 wt % to 20 wt %, indium tin oxide containing silicon, or zinc oxide (ZnO)) is preferably used for the second electrode 617.

Note that the light-emitting device is formed with the first electrode 613, the EL layer 616, and the second electrode 617. The light-emitting device is the light-emitting device described in Embodiment 1. In the light-emitting apparatus 10 of this embodiment, the pixel portion, which includes a plurality of light-emitting devices, may include both the light-emitting device described in Embodiment 1 and a light-emitting device having a different structure. In that case, in the light-emitting apparatus of one embodiment of 15 the present invention, a common hole-transport layer can be used for light-emitting devices that emit light with different wavelengths, allowing the light-emitting apparatus to be manufactured in a simple process at low costs.

The sealing substrate 604 is attached to the element 20 substrate 610 with the sealing material 605, so that a light-emitting device 618 is provided in the space 607 surrounded by the element substrate 610, the sealing substrate 604, and the sealing material 605. The space 607 may be filled with a filler, or may be filled with an inert gas (such as nitrogen or argon), or the sealing material. It is preferable that the sealing substrate be provided with a recessed portion and a drying agent be provided in the recessed portion, in which case deterioration due to influence of moisture can be inhibited.

30 An epoxy-based resin or glass frit is preferably used for the sealing material 605. It is preferable that such a material transmit moisture or oxygen as little as possible. As the sealing substrate 604, a glass substrate, a quartz substrate, or a plastic substrate formed of fiber reinforced plastic (FRP), poly(vinyl fluoride) (PVF), polyester, acrylic resin, or the like can be used.

Although not illustrated in FIGS. 2A and 2B, a protective film may be provided over the cathode. As the protective film, an organic resin film or an inorganic insulating film 40 may be formed. The protective film may be formed so as to cover an exposed portion of the sealing material 605. The protective film may be provided so as to cover surfaces and side surfaces of the pair of substrates and exposed side surfaces of a sealing layer, an insulating layer, and the like.

45 The protective film can be formed using a material that does not easily transmit an impurity such as water. Thus, diffusion of an impurity such as water from the outside into the inside can be effectively inhibited.

As a material of the protective film, an oxide, a nitride, a 50 fluoride, a sulfide, a ternary compound, a metal, a polymer, or the like can be used. For example, the material may contain aluminum oxide, hafnium oxide, hafnium silicate, lanthanum oxide, silicon oxide, strontium titanate, tantalum oxide, titanium oxide, zinc oxide, niobium oxide, zirconium oxide, tin oxide, yttrium oxide, cerium oxide, scandium oxide, erbium oxide, vanadium oxide, indium oxide, aluminum nitride, hafnium nitride, silicon nitride, tantaluminitride, titaniumnitride, niobiumnitride, molybdenumnitride, zirconiumnitride, gallium nitride, a nitride containing titanium and aluminum, an oxide containing titanium and aluminum, an oxide containing aluminum and zinc, a sulfide containing manganese and zinc, a sulfide containing cerium and strontium, an oxide containing erbium and aluminum, an oxide containing yttrium and zirconium, or the like.

60 The protective film is preferably formed using a deposition method with favorable step coverage. One such method is an atomic layer deposition (ALD) method. A material that

can be deposited by an ALD method is preferably used for the protective film. A dense protective film having reduced defects such as cracks or pinholes or a uniform thickness can be formed by an ALD method. Furthermore, damage caused to a process member in forming the protective film can be reduced.

By an ALD method, a uniform protective film with few defects can be formed even on, for example, a surface with a complex uneven shape or upper, side, and lower surfaces of a touch panel.

As described above, the light-emitting apparatus manufactured using the light-emitting device described in Embodiment 1 can be obtained.

The light-emitting apparatus in this embodiment is manufactured using the light-emitting device described in Embodiment 1 and thus can have favorable characteristics. Specifically, since the light-emitting device described in Embodiment 1 has high emission efficiency, the light-emitting apparatus can achieve low power consumption.

FIGS. 3A and 3B each illustrate an example of a light-emitting apparatus that includes coloring layers (color filters) and the like to improve color purity. FIG. 3A illustrates a substrate 1001, a base insulating film 1002, a gate insulating film 1003, gate electrodes 1006, 1007, and 1008, a first interlayer insulating film 1020, a second interlayer insulating film 1021, a peripheral portion 1042, a pixel portion 1040, a driver circuit portion 1041, anodes 1024R, 1024G, and 1024B of light-emitting devices, a partition 1025, an EL layer 1028, a second electrode 1029 of the light-emitting devices, a sealing substrate 1031, a sealing material 1032, and the like.

In FIG. 3A, coloring layers (a red coloring layer 1034R, a green coloring layer 1034G, and a blue coloring layer 1034B) are provided on a transparent base material 1033. A black matrix 1035 may be additionally provided. The transparent base material 1033 provided with the coloring layers and the black matrix is aligned and fixed to the substrate 1001. Note that the coloring layers and the black matrix 1035 are covered with an overcoat layer 1036.

FIG. 3B shows an example in which the coloring layers (the red coloring layer 1034R, the green coloring layer 1034G, and the blue coloring layer 1034B) are provided between the gate insulating film 1003 and the first interlayer insulating film 1020. As in the structure, the coloring layers may be provided between the substrate 1001 and the sealing substrate 1031.

The above-described light-emitting apparatus has a structure in which light is extracted from the substrate 1001 side where FETs are formed (a bottom emission structure), but may have a structure in which light is extracted from the sealing substrate 1031 side (a top emission structure). FIG. 4 is a cross-sectional view of a light-emitting apparatus having a top emission structure. In this case, a substrate that does not transmit light can be used as the substrate 1001. The process up to the step of forming a connection electrode which connects the FET and the anode of the light-emitting device is performed in a manner similar to that of the light-emitting apparatus having a bottom emission structure. Then, a third interlayer insulating film 1037 is formed to cover the electrode 1022. This insulating film may have a planarization function. The third interlayer insulating film 1037 can be formed using a material similar to that of the second interlayer insulating film 1021, and can alternatively be formed using any of other known materials.

The anodes 1024R, 1024G, and 1024B of the light-emitting devices may each serve as a cathode. Furthermore, in the case of a light-emitting apparatus having a top

emission structure as illustrated in FIG. 4, the anodes are preferably reflective electrodes. The EL layer 1028 is formed to have a structure similar to the structure of the EL layer 103 described in Embodiment 1.

In the case of a top emission structure as illustrated in FIG. 4, sealing can be performed with the sealing substrate 1031 on which the coloring layers (the red coloring layer 1034R, the green coloring layer 1034G, and the blue coloring layer 1034B) are provided. The sealing substrate 1031 may be provided with the black matrix 1035 which is positioned between pixels. The coloring layers (the red coloring layer 1034R, the green coloring layer 1034G, and the blue coloring layer 1034B) or the black matrix 1035 may be covered with the overcoat layer 1036. Note that a light-transmitting substrate is used as the sealing substrate 1031.

In the light-emitting apparatus having a top emission structure, a microcavity structure can be favorably employed. A light-emitting device with a microcavity structure is formed with the use of an electrode including a reflective electrode as one of the electrodes and a transflective electrode as the other electrode. At least an EL layer is positioned between the reflective electrode and the transflective electrode, and the EL layer includes at least a light-emitting layer serving as a light-emitting region.

Note that the reflective electrode has a visible light reflectivity of 40% to 100%, preferably 70% to 100%, and a resistivity of  $1 \times 10^{-2}$   $\Omega\text{cm}$  or lower. In addition, the transflective electrode has a visible light reflectivity of 20% to 80%, preferably 40% to 70%, and a resistivity of  $1 \times 10^{-2}$   $\Omega\text{cm}$  or lower.

Light emitted from the light-emitting layer included in the EL layer is reflected and resonated by the reflective electrode and the transflective electrode.

In the light-emitting device, by changing thicknesses of the transparent conductive film, the composite material, the carrier-transport material, or the like, the optical path length between the reflective electrode and the transflective electrode can be changed. Thus, light with a wavelength that is resonated between the reflective electrode and the transflective electrode can be intensified while light with a wavelength that is not resonated therebetween can be attenuated.

Note that light that is reflected back by the reflective electrode (first reflected light) considerably interferes with light that directly enters the transflective electrode from the light-emitting layer (first incident light). For this reason, the optical path length between the reflective electrode and the light-emitting layer is preferably adjusted to  $(2n-1)\lambda/4$  ( $n$  is a natural number of 1 or larger and  $\lambda$  is a wavelength of light to be amplified). By adjusting the optical path length, the phases of the first reflected light and the first incident light can be aligned with each other and the light emitted from the light-emitting layer can be further amplified.

Note that in the above structure, the EL layer may include a plurality of light-emitting layers or may include a single light-emitting layer. The tandem light-emitting device described above may be combined with a plurality of EL layers; for example, a light-emitting device may have a structure in which a plurality of EL layers are provided with a charge-generation layer provided therebetween, and each EL layer includes a plurality of light-emitting layers or a single light-emitting layer.

With the microcavity structure, emission intensity with a specific wavelength in the front direction can be increased, whereby power consumption can be reduced. Note that in the case of a light-emitting apparatus that displays images with subpixels of four colors, red, yellow, green, and blue,

the light-emitting apparatus can have favorable characteristics because the luminance can be increased owing to yellow light emission and each subpixel can employ a microcavity structure suitable for wavelengths of the corresponding color.

The light-emitting apparatus in this embodiment is manufactured using the light-emitting device described in Embodiment 1 and thus can have favorable characteristics. Specifically, since the light-emitting device described in Embodiment 1 has high emission efficiency, the light-emitting apparatus can achieve low power consumption.

The active matrix light-emitting apparatus is described above, whereas a passive matrix light-emitting apparatus is described below. FIGS. 5A and 5B illustrate a passive matrix light-emitting apparatus manufactured using the present invention. Note that FIG. 5A is a perspective view of the light-emitting apparatus, and FIG. 5B is a cross-sectional view taken along the line X-Y in FIG. 5A. In FIGS. 5A and 5B, over a substrate 951, an EL layer 955 is provided between an electrode 952 and an electrode 956. An end portion of the electrode 952 is covered with an insulating layer 953. A partition layer 954 is provided over the insulating layer 953. The sidewalls of the partition layer 954 are aslope such that the distance between both sidewalls is gradually narrowed toward the surface of the substrate. In other words, a cross section taken along the direction of the short side of the partition layer 954 is trapezoidal, and the lower side (a side of the trapezoid that is parallel to the surface of the insulating layer 953 and is in contact with the insulating layer 953) is shorter than the upper side (a side of the trapezoid that is parallel to the surface of the insulating layer 953 and is not in contact with the insulating layer 953). The partition layer 954 thus provided can prevent defects in the light-emitting device due to static electricity or others. The passive matrix light-emitting apparatus also includes the light-emitting device described in Embodiment 1; thus, the light-emitting apparatus can have high reliability or low power consumption.

In the light-emitting apparatus described above, many minute light-emitting devices arranged in a matrix can each be controlled; thus, the light-emitting apparatus can be suitably used as a display device for displaying images.

This embodiment can be freely combined with any of the other embodiments.

### Embodiment 3

#### [Light-Emitting Apparatus]

Another example of the light-emitting apparatus of one embodiment of the present invention using the above light-emitting device, and a manufacturing method thereof will be described below.

FIG. 14A illustrates a schematic top view of a light-emitting apparatus 450 of one embodiment of the present invention. The light-emitting apparatus 450 includes a plurality of light-emitting devices 110R emitting red light, a plurality of light-emitting devices 110G emitting green light, and a plurality of light-emitting devices 110B emitting blue light. In FIG. 14A, light-emitting regions of the light-emitting devices are denoted by R, G, and B to easily differentiate the light-emitting devices.

The light-emitting devices 110R, the light-emitting devices 110G, and the light-emitting devices 110B are arranged in a matrix. FIG. 14A shows what is called a stripe arrangement, in which the light-emitting devices of the same color are arranged in one direction. Note that the arrange-

ment of the light-emitting devices is not limited thereto; another arrangement such as a delta, zigzag, or PenTile pattern may also be used.

The light-emitting device 110R, the light-emitting device 110G, and the light-emitting device 110B are arranged in the X direction. The light-emitting devices of the same color are arranged in the Y direction intersecting with the X direction.

The light-emitting device 110R, the light-emitting device 110G, and the light-emitting device 110B have the above structure.

FIG. 14B is a cross-sectional schematic view taken along the dashed-dotted line A1-A2 in FIG. 14A. FIG. 14C is a cross-sectional schematic view taken along the dashed-dotted line B1-B2 in FIG. 14A.

FIG. 14B shows cross sections of the light-emitting device 110R, the light-emitting device 110G, and the light-emitting device 110B. The light-emitting device 110R includes a first electrode 101R, an EL layer 120R, an EL layer 121, and the second electrode 102. The light-emitting device 110G includes a first electrode 101G, an EL layer 120G, the EL layer (electron-injection layer) 115, and the second electrode 102. The light-emitting device 110B includes a first electrode 101B, an EL layer 120B, the EL layer 121, and the second electrode 102. The EL layer 121 and the second electrode 102 are provided in common to the light-emitting device 110R, the light-emitting device 110G, and the light-emitting device 110B. The EL layer 121 can also be referred to as a common layer.

The EL layer 120R included in the light-emitting device 110R contains a light-emitting organic compound that emits light with intensity at least in a red wavelength range. The EL layer 120G included in the light-emitting device 110G contains a light-emitting organic compound that emits light with intensity at least in a green wavelength range. The EL layer 120B included in the light-emitting device 110B contains a light-emitting organic compound that emits light with intensity at least in a blue wavelength range. At least one of the light-emitting device 110R, the light-emitting device 110G, and the light-emitting device 110B is preferably the light-emitting device of one embodiment of the present invention, and the light-emitting device of one embodiment of the present invention is preferably the light-emitting device 110B.

Each of the EL layer 120R, the EL layer 120G, and the EL layer 120B includes at least a light-emitting layer and a hole-transport layer, and may further include one or more of an electron-injection layer, an electron-transport layer, a hole-injection layer, a carrier-blocking layer, an exciton-blocking layer, and the like. The EL layer 121 does not necessarily include the light-emitting layer. The EL layer 121 is preferably the electron-injection layer. In the case where the electron-transport layer also serves as the electron-injection layer, the EL layer 121 may be omitted.

The first electrode 101R, the first electrode 101G, and the first electrode 101B are provided for the respective light-emitting devices. The second electrode 102 and the EL layer 121 are each provided as a layer common to the light-emitting devices. The hole-transport layers in the EL layer 120R, the EL layer 120G, and the EL layer 120B, which are separated between the light-emitting devices with different emission colors, preferably have the same structure.

A conductive film that transmits visible light is used for either the first electrode 101 or the second electrode 102, and a reflective conductive film is used for the other. When the first electrode 101 is a light-transmitting electrode and the second electrode 102 is a reflective electrode, a bottom-emission display device is obtained. When the first electrode

**101** is a reflective electrode and the second electrode **102** is a light-transmitting electrode, a top-emission display device is obtained. Note that when both the first electrode **101** and the second electrode **102** transmit light, a dual-emission display device can be obtained. The light-emitting device of one embodiment of the present invention is suitable for a top-emission light-emitting device.

An insulating layer **124** is provided to cover end portions of the first electrode **101R**, the first electrode **101G**, and the first electrode **101B**. The end portions of the insulating layer **124** are preferably tapered. Note that the insulating layer **124** is not necessarily provided.

The EL layer **120R**, the EL layer **120G**, and the EL layer **120B** each include a region in contact with a top surface of a pixel electrode and a region in contact with a surface of the insulating layer **124**. End portions of the EL layer **120R**, the EL layer **120G**, and the EL layer **120B** are positioned over the insulating layer **124**.

As shown in FIG. 14B, there is a gap between the EL layers of two light-emitting devices with different colors. The EL layer **120R**, the EL layer **120G**, and the EL layer **120B** are thus preferably provided so as not to be in contact with each other. This effectively prevents unintentional light emission from being caused by current flowing through two adjacent EL layers. As a result, the contrast can be increased to achieve a display device with high display quality.

FIG. 14C shows an example in which the EL layer **120R** is formed in a band shape so as to be continuous in the Y direction. When the EL layer **120R** and the like are formed in a band shape, no space for dividing the layer is needed to reduce a non-light-emitting area between the light-emitting devices, resulting in a higher aperture ratio. FIG. 14C shows the cross section of the light-emitting device **110R** as an example; the light-emitting device **110G** and the light-emitting device **110B** can have a similar shape. Note that the EL layer may be divided for the light-emitting devices in the Y direction.

A protective layer **131** is provided over the second electrode **102** so as to cover the light-emitting device **110R**, the light-emitting device **110G**, and the light-emitting device **110B**. The protective layer **131** has a function of preventing diffusion of impurities such as water into each light-emitting device from the above.

The protective layer **131** can have, for example, a single-layer structure or a stacked-layer structure at least including an inorganic insulating film. Examples of the inorganic insulating film include an oxide film or a nitride film such as a silicon oxide film, a silicon oxynitride film, a silicon nitride oxide film, a silicon nitride film, an aluminum oxide film, an aluminum oxynitride film, or a hafnium oxide film. Alternatively, a semiconductor material such as indium gallium oxide or indium gallium zinc oxide may be used for the protective layer **131**.

As the protective layer **131**, a stacked film of an inorganic insulating film and an organic insulating film can be used. For example, a structure in which an organic insulating film is sandwiched between a pair of inorganic insulating films is preferable. Furthermore, it is preferable that the organic insulating film function as a planarization film. With this structure, the top surface of the organic insulating film can be flat, and accordingly, coverage with the inorganic insulating film over the organic insulating film is improved, leading to an improvement in barrier properties. Moreover, since the top surface of the protective layer **131** is flat, a preferable effect can be obtained; when a component (e.g., a color filter, an electrode of a touch sensor, a lens array, or

the like) is provided above the protective layer **131**, the component is less affected by an uneven shape caused by the lower structure.

FIG. 14A also illustrates a connection electrode **101C** that is electrically connected to the second electrode **102**. The connection electrode **101C** is supplied with a potential (e.g., an anode potential or a cathode potential) that is to be supplied to the second electrode **102**. The connection electrode **101C** is provided outside a display region where the light-emitting devices **110R** and the like are arranged. In FIG. 14A, the second electrode **102** is denoted by a dashed line.

The connection electrode **101C** can be provided along the outer periphery of the display region. For example, the connection electrode **101C** may be provided along one side of the outer periphery of the display region or two or more sides of the outer periphery of the display region. That is, in the case where the display region has a rectangular top surface, the top surface of the connection electrode **101C** can have a band shape, an L shape, a square bracket shape, a quadrangular shape, or the like.

FIG. 14D is a cross-sectional schematic view taken along the dashed-dotted line C1-C2 in FIG. 14A. FIG. 14D illustrates a connection portion **130** at which the connection electrode **101C** is electrically connected to the second electrode **102**. In the connection portion **130**, the second electrode **102** is provided on and in contact with the connection electrode **101C** and the protective layer **131** is provided to cover the second electrode **102**. In addition, the insulating layer **124** is provided to cover end portions of the connection electrode **101C**.

#### [Manufacturing Method Example]

An example of a method for manufacturing the display device of one embodiment of the present invention is described below with reference to the drawings. Here, description is made with use of the light-emitting apparatus **450** shown in the above structure example. FIGS. 15A to 15F are cross-sectional schematic views of steps in a manufacturing method of a display device described below. In FIG. 15A and the like, the cross-sectional schematic views of the connection portion **130** and the periphery thereof are also illustrated on the right side.

Note that thin films included in the display device (e.g., insulating films, semiconductor films, or conductive films) can be formed by any of a sputtering method, a chemical vapor deposition (CVD) method, a vacuum evaporation method, a pulsed laser deposition (PLD) method, an atomic layer deposition (ALD) method, and the like. Examples of the CVD method include a plasma-enhanced chemical vapor deposition (PECVD) method and a thermal CVD method. An example of a thermal CVD method is a metal organic CVD (MOCVD) method.

Alternatively, thin films included in the display device (e.g., insulating films, semiconductor films, and conductive films) can be formed by a method such as spin coating, dipping, spray coating, ink-jetting, dispensing, screen printing, or offset printing or with a doctor knife, a slit coater, a roll coater, a curtain coater, or a knife coater.

Thin films included in the display device can be processed by a photolithography method or the like. Besides, a nano-imprinting method, a sandblasting method, a lift-off method, or the like may be used to process thin films. Alternatively, island-shaped thin films may be directly formed by a film formation method using a shielding mask such as a metal mask.

There are two typical examples of photolithography methods. In one of the methods, a resist mask is formed over a

thin film that is to be processed, the thin film is processed by etching or the like, and then the resist mask is removed. In the other method, a photosensitive thin film is formed and then processed into a desired shape by light exposure and development.

As light for exposure in a photolithography method, light with an i-line (with a wavelength of 365 nm), light with a g-line (with a wavelength of 436 nm), light with an h-line (with a wavelength of 405 nm), or light in which the i-line, the g-line, and the h-line are mixed can be used. Alternatively, ultraviolet light, KrF laser light, ArF laser light, or the like can be used. Exposure may be performed by liquid immersion exposure technique. As the light for exposure, extreme ultraviolet (EUV) light or X-rays may also be used. Furthermore, instead of the light used for the exposure, an electron beam can also be used. It is preferable to use EUV, X-rays, or an electron beam because extremely minute processing can be performed. Note that a photomask is not needed when exposure is performed by scanning with a beam such as an electron beam.

For etching of thin films, a dry etching method, a wet etching method, a sandblast method, or the like can be used.

[Preparation for Substrate 100]

A substrate that has heat resistance high enough to withstand at least heat treatment performed later can be used as the substrate 100. When an insulating substrate is used as the substrate 100, a glass substrate, a quartz substrate, a sapphire substrate, a ceramic substrate, an organic resin substrate, or the like can be used. Alternatively, a semiconductor substrate can be used. For example, a single crystal semiconductor substrate or a polycrystalline semiconductor substrate of silicon, silicon carbide, or the like; a compound semiconductor substrate of silicon germanium or the like; an SOI substrate; or the like can be used.

As the substrate 100, it is particularly preferable to use the semiconductor substrate or the insulating substrate over which a semiconductor circuit including a semiconductor element such as a transistor is formed. The semiconductor circuit preferably forms a pixel circuit, a gate line driver circuit (a gate driver), a source line driver circuit (a source driver), or the like. In addition to the above, an arithmetic circuit, a memory circuit, or the like may be formed.

[Formation of first electrodes 101R, 101G, and 101B, and connection electrode 101C]

Next, the first electrodes 101R, 101G, and 101B, and the connection electrode 101C are formed over the substrate 100. First, a conductive film to be an anode (a pixel electrode) is formed, a resist mask is formed by a photolithography method, and an unnecessary portion of the conductive film is removed by etching. After that, the resist mask is removed to form the first electrodes 101R, 101G, and 101B.

In the case where a conductive film that reflects visible light is used as each pixel electrode, it is preferable to use a material (e.g., silver or aluminum) having reflectance as high as possible in the whole wavelength range of visible light. This can increase both light extraction efficiency of the light-emitting devices and color reproducibility. In the case where a conductive film that reflects visible light is used as each pixel electrode, what is called a top-emission light-emitting apparatus in which light is extracted in the direction opposite to the substrate can be obtained. In the case where a conductive film that transmits light is used as each pixel electrode, what is called a bottom-emission light-emitting apparatus in which light is extracted in the direction of the substrate can be obtained.

[Formation of Insulating Layer 124]

Then, the insulating layer 124 is provided to cover end portions of the first electrode 101R, the first electrode 101G, and the first electrode 101B (FIG. 15A). An organic insulating film or an inorganic insulating film can be used as the insulating layer 124. The end portions of the insulating layer 124 are preferably tapered to improve step coverage with an EL film to be formed later. In particular, when an organic insulating film is used, a photosensitive material is preferably used so that the shape of the end portions can be easily controlled by the conditions of light exposure and development. In the case where the insulating layer 124 is not provided, the distance between the light-emitting devices can be further reduced to offer a light-emitting apparatus with higher resolution.

[Formation of EL film 120Rb]

Subsequently, the EL film 120Rb, which is to be the EL layer 120R, is formed over the first electrode 101R, the first electrode 101G, the first electrode 101B, and the insulating layer 124.

The EL film 120Rb includes at least a light-emitting layer containing a light-emitting material and a hole-transport layer. The EL film 120Rb may have a structure in which one or more films functioning as an electron-injection layer, an electron-transport layer, a charge-generation layer, and a hole-injection layer are further stacked. The EL film 120Rb can be formed by, for example, an evaporation method, a sputtering method, an inkjet method, or the like. Without limitation to this, the above-described film-formation method can be used as appropriate.

For example, the EL film 120Rb is preferably a stacked film in which a hole-injection layer, a hole-transport layer, a light-emitting layer, and an electron-transport layer are stacked in this order. In that case, a film including the electron-injection layer 115 can be used as the EL layer formed later. In the light-emitting apparatus of one embodiment of the present invention, the electron-transport layer is provided to cover the light-emitting layer, which can inhibit the light-emitting layer from being damaged by a subsequent photolithography step or the like, so that a highly reliable light-emitting device can be fabricated.

The EL film 120Rb is preferably formed so as not to overlap with the connection electrode 101C. For example, in the case where the EL film 120Rb is formed by an evaporation method (or a sputtering method), it is preferable that the EL film 120Rb be formed using a shielding mask so as not to be formed over the connection electrode 101C, or the EL film 120Rb be removed in a later etching step.

[Formation of Sacrificial Film 144a]

Then, the sacrificial film 144a is formed to cover the EL film 120Rb. The sacrificial film 144a is provided in contact with a top surface of the connection electrode 101C.

As the sacrificial film 144a, it is possible to use a film highly resistant to etching treatment performed on various EL films such as the EL film 120Rb, i.e., a film having high etching selectivity with respect to the EL film. Furthermore, as the sacrificial film 144a, it is possible to use a film having high etching selectivity with respect to a protective film such as a protective film 146a described later. Moreover, as the sacrificial film 144a, it is possible to use a film that can be removed by a wet etching method less likely to cause damage to the EL film.

The sacrificial film 144a can be formed using an inorganic film such as a metal film, an alloy film, a metal oxide film, a semiconductor film, or an inorganic insulating film, for example. The sacrificial film 144a can be formed by any of

a variety of film formation methods such as a sputtering method, an evaporation method, a CVD method, and an ALD method.

The sacrificial film 144a can be formed using a metal material such as gold, silver, platinum, magnesium, nickel, tungsten, chromium, molybdenum, iron, cobalt, copper, palladium, titanium, aluminum, yttrium, zirconium, or tantalum or an alloy material containing the metal material. It is particularly preferable to use a low-melting-point material such as aluminum or silver.

Alternatively, the sacrificial film 144a can be formed using a metal oxide such as an indium-gallium-zinc oxide (In—Ga—Zn oxide, also referred to as IGZO). It is also possible to use indium oxide, indium zinc oxide (In—Zn oxide), indium tin oxide (In—Sn oxide), indium titanium oxide (In—Ti oxide), indium tin zinc oxide (In—Sn—Zn oxide), indium titanium zinc oxide (In—Ti—Zn oxide), indium gallium tin zinc oxide (In—Ga—Sn—Zn oxide), or the like. Indium tin oxide containing silicon, or the like can also be used.

An element M (M is one or more of aluminum, silicon, boron, yttrium, copper, vanadium, beryllium, titanium, iron, nickel, germanium, zirconium, molybdenum, lanthanum, cerium, neodymium, hafnium, tantalum, tungsten, and magnesium) may be used instead of gallium. In particular, M is preferably one or more of gallium, aluminum, and yttrium.

Alternatively, the sacrificial film 144a can be formed using an inorganic insulating material such as aluminum oxide, hafnium oxide, or silicon oxide.

The sacrificial film 144a is preferably formed using a material that can be dissolved in a solvent chemically stable with respect to at least the uppermost film of the EL film 120Rb. Specifically, a material that will be dissolved in water or alcohol can be suitably used for the sacrificial film 144a. In formation of the sacrificial film 144a, it is preferable that application of such a material dissolved in a solvent such as water or alcohol be performed by a wet process and followed by heat treatment for evaporating the solvent. At this time, the heat treatment is preferably performed under a reduced-pressure atmosphere, in which case the solvent can be removed at a low temperature in a short time and thermal damage to the EL film 120Rb can be accordingly minimized.

The sacrificial film 144a can be formed by spin coating, dipping, spray coating, ink-jetting, dispensing, screen printing, or offset printing, or with a doctor knife, a slit coater, a roll coater, a curtain coater, or a knife coater, for example.

The sacrificial film 144a can be formed using an organic material such as polyvinyl alcohol (PVA), polyvinylbutyral, polyvinylpyrrolidone, polyethylene glycol, polyglycerin, pullulan, water-soluble cellulose, or an alcohol-soluble polyamide resin.

#### [Formation of Protective Film 146a]

Next, the protective film 146a is formed over the sacrificial film 144a (FIG. 15B).

The protective film 146a is a film used as a hard mask when the sacrificial film 144a is etched later. In a later step of processing the protective film 146a, the sacrificial film 144a is exposed. Thus, the combination of films having high etching selectivity therebetween is selected for the sacrificial film 144a and the protective film 146a. It is thus possible to select a film that can be used for the protective film 146a depending on an etching condition of the sacrificial film 144a and an etching condition of the protective film 146a.

For example, in the case where dry etching using a gas containing fluorine (also referred to as a fluorine-based gas) is performed for the etching of the protective film 146a, the

protective film 146a can be formed using silicon, silicon nitride, silicon oxide, tungsten, titanium, molybdenum, tantalum, tantalum nitride, an alloy containing molybdenum and niobium, an alloy containing molybdenum and tungsten, or the like. Here, a metal oxide film using IGZO, ITO, or the like is given as a film having high etching selectivity (that is, enabling low etching rate) in dry etching using the fluorine-based gas, and such a film can be used as the sacrificial film 144a.

Without being limited to the above, a material of the protective film 146a can be selected from a variety of materials depending on etching conditions of the sacrificial film 144a and the protective film 146a. For example, any of the films that can be used for the sacrificial film 144a can be used.

As the protective film 146a, a nitride film can be used, for example. Specifically, a nitride such as silicon nitride, aluminum nitride, hafnium nitride, titanium nitride, tantalum nitride, tungsten nitride, gallium nitride, or germanium nitride can be used.

As the protective film 146a, an oxide film can also be used. Typically, it is possible to use a film of an oxide or an oxynitride such as silicon oxide, silicon oxynitride, aluminum oxide, aluminum oxynitride, hafnium oxide, or hafnium oxynitride.

Alternatively, as the protective film 146a, an organic film that can be used for the EL film 120Rb or the like can be used. For example, the protective film 146a can be formed using the organic film that is used for the EL film 120Rb or an EL film 120Gb. Use of such an organic film is preferable because the same film-formation apparatus can be used for formation of the EL film 120Rb or the like.

#### [Formation of Resist Mask 143a]

Then, the resist mask 143a is formed in positions over the protective film 146a that overlap with the first electrode 101R and the connection electrode 101C (FIG. 15C).

For the resist mask 143a, a resist material containing a photosensitive resin such as a positive type resist material or a negative type resist material can be used.

On the assumption that the resist mask 143a is formed over the sacrificial film 144a without the protective film 146a therebetween, there is a risk of dissolving the EL film 120Rb due to a solvent of the resist material if a defect such as a pinhole exists in the sacrificial film 144a. Such a defect can be prevented by using the protective film 146a.

In the case where a film that is unlikely to cause a defect such as a pinhole is used as the sacrificial film 144a, the resist mask 143a may be formed directly on the sacrificial film 144a without the protective film 146a therebetween.

#### [Etching of Protective Film 146a]

Next, part of the protective film 146a that is not covered with the resist mask 143a is removed by etching, so that a band-shaped protective layer 147a is formed. At that time, the protective layer 147a is formed also over the connection electrode 101C.

In the etching of the protective film 146a, an etching condition with high selectively is preferably employed so that the sacrificial film 144a is not removed by the etching. Either wet etching or dry etching can be performed for the etching of the protective film 146a. With use of dry etching, a reduction in a processing pattern of the protective film 146a can be inhibited.

#### [Removal of Resist Mask 143a]

Then, the resist mask 143a is removed (FIG. 15D). The removal of the resist mask 143a can be performed by wet etching or dry etching. It is particularly preferable to

perform dry etching (also referred to as plasma ashing) using an oxygen gas as an etching gas to remove the resist mask 143a.

At this time, the removal of the resist mask 143a is performed in a state where the EL film 120Rb is covered with the sacrificial film 144a; thus, the EL film 120Rb is less likely to be affected by the removal. In particular, when the EL film 120Rb is exposed to oxygen, the electrical characteristics of the light-emitting device are adversely affected in some cases. Therefore, it is preferable that the EL film 120Rb be covered by the sacrificial film 144a when etching using an oxygen gas, such as plasma ashing, is performed.

[Etching of Sacrificial Film 144a]

Next, part of the sacrificial film 144a that is not covered with the protective layer 147a is removed by etching with use of the protective layer 147a as a mask, so that a band-shaped sacrificial layer 145a is formed (FIG. 15E). At that time, the sacrificial layer 145a is formed also over the connection electrode 101C.

Either wet etching or dry etching can be performed for the etching of the sacrificial film 144a. With use of dry etching, a reduction in a processing pattern of the sacrificial film 144a can be inhibited.

[Etching of EL Film 120Rb and Protective Layer 147a]

Next, the protective layer 147a and part of the EL film 120Rb that is not covered with the sacrificial layer 145a are removed by etching at the same time, so that the band-shaped EL layer 120R is formed (FIG. 15F). At that time, the protective layer 147a over the connection electrode 101C is also removed.

The EL film 120Rb and the protective layer 147a are preferably etched by the same treatment so that the process can be simplified to reduce the fabrication cost of the display device.

For the etching of the EL film 120Rb, it is particularly preferable to perform dry etching using an etching gas that does not contain oxygen as its main component. This is because the alteration of the EL film 120Rb is inhibited, and a highly reliable display device can be achieved. Examples of the etching gas that does not contain oxygen as its main component include CF<sub>4</sub>, C<sub>4</sub>F<sub>8</sub>, SF<sub>6</sub>, CHF<sub>3</sub>, Cl<sub>2</sub>, H<sub>2</sub>O, BC<sub>1</sub><sub>3</sub>, or a rare gas such as H<sub>2</sub> or He. Alternatively, a mixed gas of the above gas and a dilution gas that does not contain oxygen can be used as the etching gas.

Note that the etching of the EL film 120Rb and the etching of the protective layer 147a may be performed separately. In that case, either the etching of the EL film 120Rb or the etching of the protective layer 147a may be performed first.

At this step, the EL layer 120R and the connection electrode 101C are covered with the sacrificial layer 145a.

[Formation of EL Film 120Gb]

Subsequently, the EL film 120Gb, which is to be the EL layer 120G, is formed over the sacrificial layer 145a, the insulating layer 124, the first electrode 101G, and the first electrode 101B. In that case, similarly to the EL film 120Rb, the EL film 120Gb is preferably not provided over the connection electrode 101C.

For the formation method of the EL film 120Gb, the above description of the EL film 120Rb can be referred to.

[Formation of Sacrificial Film 144b]

Then, the sacrificial film 144b is formed over the EL film 120Gb. The sacrificial film 144b can be formed in a manner similar to that for the sacrificial film 144a. In particular, the sacrificial film 144b and the sacrificial film 144a are preferably formed using the same material.

At that time, the sacrificial film 144a is formed also over the connection electrode 101C so as to cover the sacrificial layer 145a.

[Formation of Protective Film 146b]

Next, the protective film 146b is formed over the sacrificial film 144b. The protective film 146b can be formed in a manner similar to that for the protective film 146a. In particular, the protective film 146b and the protective film 146a are preferably formed using the same material.

[Formation of Resist Mask 143b]

Then, the resist mask 143b is formed in positions over the protective film 146b that overlap with the first electrode 101G and the connection electrode 101C (FIG. 16A).

The resist mask 143b can be formed in a manner similar to that for the resist mask 143a.

[Etching of Protective Film 146b]

Next, part of the protective film 146b that is not covered with the resist mask 143b is removed by etching, so that a band-shaped protective layer 147b is formed (FIG. 16B). At that time, the protective layer 147b is formed also over the connection electrode 101C.

For the etching of the protective film 146b, the above description of the protective film 146a can be referred to.

[Removal of Resist Mask 143b]

Then, the resist mask 143b is removed. For the removal of resist mask 143b, the above description of the resist mask 143a can be referred to.

[Etching of Sacrificial Film 144b]

Next, part of the sacrificial film 144b that is not covered with the protective layer 147b is removed by etching with use of the protective layer 147b as a mask, so that a band-shaped sacrificial layer 145b is formed. At that time, the sacrificial layer 145b is formed also over the connection electrode 101C. The sacrificial layer 145a and the sacrificial layer 145b are stacked over the connection electrode 101C.

For the etching of the sacrificial film 144b, the above description of the sacrificial film 144a can be referred to.

[Etching of EL Film 120Gb and Protective Layer 147b]

Next, the protective layer 147b and part of the EL film 120Gb that is not covered with the sacrificial layer 145b are removed by etching at the same time, so that the band-shaped EL layer 120G is formed (FIG. 16C). At that time, the protective layer 147b over the connection electrode 101C is also removed.

For the etching of the EL film 120Gb and the protective layer 147b, the above description of the EL film 120Rb and the protective layer 147a can be referred to.

At this time, the EL layer 120R is protected by the sacrificial layer 145a, and thus can be prevented from being damaged in the etching step of the EL film 120Gb.

In the above manner, the band-shaped EL layer 120R and the band-shaped EL layer 120G can be separately formed with highly accurate alignment.

[Formation of EL Layer 120B]

The above steps are performed on an EL film 120Bb (not illustrated), whereby the island-shaped EL layer 120B and an island-shaped sacrificial layer 145c can be formed (FIG. 16D).

That is, after the formation of the EL layer 120G, the EL film 120Bb, a sacrificial film 144c, a protective film 146c, and a resist mask 143c (each of which is not illustrated) are sequentially formed. After that, the protective film 146c is etched to form a protective layer 147c (not illustrated); then, the resist mask 143c is removed. Subsequently, the sacrificial film 144c is etched to form the sacrificial layer 145c. Then, the protective layer 147c and the EL film 120Bb are etched to form the band-shaped EL layer 120B.

After the EL layer 120B is formed, the sacrificial layer 145c is also formed over the connection electrode 101C. The sacrificial layer 145a, the sacrificial layer 145b, and the sacrificial layer 145c are stacked over the connection electrode 101C.

[Removal of Sacrificial Layer]

Next, the sacrificial layer 145a, the sacrificial layer 145b, and the sacrificial layer 145c are removed, whereby top surfaces of the EL layer 120R, the EL layer 120G, and the EL layer 120B are exposed (FIG. 16E). At that time, the top surface of the connection electrode 101C is also exposed.

The sacrificial layer 145a, the sacrificial layer 145b, and the sacrificial layer 145c can be removed by wet etching or dry etching. At this time, a method that causes damage to the EL layer 120R, the EL layer 120G, and the EL layer 120B as little as possible is preferably employed. In particular, a wet etching method is preferably used. For example, wet etching using a tetramethyl ammonium hydroxide (TMAH) solution, diluted hydrofluoric acid, oxalic acid, phosphoric acid, acetic acid, nitric acid, or a mixed solution thereof is preferably performed.

Alternatively, the sacrificial layer 145a, the sacrificial layer 145b, and the sacrificial layer 145c are preferably removed by being dissolved in a solvent such as water or alcohol. Examples of the alcohol in which the sacrificial layer 145a, the sacrificial layer 145b, and the sacrificial layer 145c can be dissolved include ethyl alcohol, methyl alcohol, isopropyl alcohol (IPA), and glycerin.

After the sacrificial layer 145a, the sacrificial layer 145b, and the sacrificial layer 145c are removed, drying treatment is preferably performed in order to remove water contained in the EL layer 120R, the EL layer 120G, and the EL layer 120B and water adsorbed on the surfaces of the EL layer 120R, the EL layer 120G, and the EL layer 120B. For example, heat treatment is preferably performed in an inert gas atmosphere or a reduced-pressure atmosphere. The heat treatment can be performed at a substrate temperature higher than or equal to 50°C. and lower than or equal to 200°C., preferably higher than or equal to 60°C. and lower than or equal to 150°C., and further preferably higher than or equal to 70°C. and lower than or equal to 120°C. The heat treatment is preferably performed in a reduced-pressure atmosphere because drying at a lower temperature is possible.

In the above manner, the EL layer 120R, the EL layer 120G, and the EL layer 120B can be separately formed.

[Formation of Electron-Injection Layer 115]

Then, the electron-injection layer 115 is formed to cover the EL layer 120R, the EL layer 120G, and the EL layer 120B.

The electron-injection layer 115 can be formed in a manner similar to that for the EL film 120Rb or the like. In the case where the electron-injection layer 115 is formed by an evaporation method, the electron-injection layer 115 is preferably formed using a shielding mask so as not to be formed over the connection electrode 101C.

[Formation of Second Electrode 102]

Then, the second electrode 102 is formed to cover the electron-injection layer 115 and the connection electrode 101C (FIG. 16F).

The second electrode 102 can be formed by a method such as an evaporation method or a sputtering method. Alternatively, a film formed by an evaporation method and a film formed by a sputtering method may be stacked. In that case, the second electrode 102 is preferably formed so as to cover a region where the electron-injection layer 115 is formed. That is, a structure in which end portions of the electron-

injection layer 115 overlap with the second electrode 102 can be obtained. The second electrode 102 is preferably formed using a shielding mask.

The second electrode 102 is electrically connected to the connection electrode 101C outside a display region.

[Formation of Protective Layer]

Then, a protective layer is formed over the second electrode 102. An inorganic insulating film used for the protective layer is preferably formed by a sputtering method, a PECVD method, or an ALD method. In particular, an ALD method is preferable because a film deposited by ALD has good step coverage and is less likely to cause a defect such as pinhole. An organic insulating film is preferably formed by an inkjet method because a uniform film can be formed in a desired area.

In the above manner, the light-emitting apparatus of one embodiment of the present invention can be manufactured.

Although the second electrode 102 and the electron-injection layer 115 are formed so as to have different top surface shapes, they may be formed in the same region.

#### Embodiment 4

In this embodiment, an example in which the light-emitting device described in Embodiment 1 is used for a lighting device will be described with reference to FIGS. 6A and 6B. FIG. 6B is a top view of the lighting device, and FIG. 6A is a cross-sectional view taken along the line e-f in FIG. 6B.

In the lighting device in this embodiment, an anode 401 is formed over a substrate 400 which is a support with a light-transmitting property. The anode 401 corresponds to the first electrode 101 in Embodiment 1. When light is extracted through the anode 401, the anode 401 is formed using a material having a light-transmitting property.

A pad 412 for applying voltage to a cathode 404 is provided over the substrate 400.

An EL layer 403 is formed over the anode 401. The structure of the EL layer 403 corresponds to, for example, the structure of the EL layer 103 in Embodiment 1. Refer to the descriptions for the structure.

The cathode 404 is formed to cover the EL layer 403. The cathode 404 corresponds to the second electrode 102 in Embodiment 1. The cathode 404 is formed using a material having high reflectance when light is extracted through the anode 401. The cathode 404 is connected to the pad 412, thereby receiving voltage.

As described above, the lighting device described in this embodiment includes a light-emitting device including the anode 401, the EL layer 403, and the cathode 404. Since the light-emitting device is a light-emitting device with high emission efficiency, the lighting device in this embodiment can have low power consumption.

The substrate 400 provided with the light-emitting device having the above structure is fixed to a sealing substrate 407 with sealing materials 405 and 406 and sealing is performed, whereby the lighting device is completed. It is possible to use only either the sealing material 405 or the sealing material 406. The inner sealing material 406 (not illustrated in FIG. 6B) can be mixed with a desiccant which enables moisture to be adsorbed, increasing reliability.

When parts of the pad 412 and the anode 401 are extended to the outside of the sealing materials 405 and 406, the extended parts can serve as external input terminals. An IC chip 420 mounted with a converter or the like may be provided over the external input terminals.

The lighting device described in this embodiment includes as an EL element the light-emitting device described in Embodiment 1; thus, the lighting device can have low power consumption.

This embodiment can be freely combined with any of the other embodiments.

### Embodiment 5

In this embodiment, examples of electronic devices each including the light-emitting device described in Embodiment 1 will be described. The light-emitting device described in Embodiment 1 has high emission efficiency and low power consumption. As a result, the electronic devices described in this embodiment can each include a light-emitting portion with low power consumption.

Examples of the electronic device including the above light-emitting device include television devices (also referred to as TV or television receivers), monitors for computers and the like, digital cameras, digital video cameras, digital photo frames, cellular phones (also referred to as mobile phones or mobile phone devices), portable game machines, portable information terminals, audio playback devices, and large game machines such as pachinko machines. Specific examples of these electronic devices are shown below.

FIG. 7A shows an example of a television device. In the television device, a display portion 7103 is incorporated in a housing 7101. Here, the housing 7101 is supported by a stand 7105. Images can be displayed on the display portion 7103, and in the display portion 7103, the light-emitting devices described in Embodiment 1 are arranged in a matrix.

The television device can be operated with an operation switch of the housing 7101 or a separate remote controller 7110. With operation keys 7109 of the remote controller 7110, channels or volume can be controlled and images displayed on the display portion 7103 can be controlled. Furthermore, the remote controller 7110 may be provided with a display portion 7107 for displaying data output from the remote controller 7110. The light-emitting devices described in Embodiment 1 may also be arranged in a matrix in the display portion 7107.

Note that the television device is provided with a receiver, a modem, and the like. With the use of the receiver, a general television broadcast can be received. Moreover, when the television device is connected to a communication network with or without wires via the modem, one-way (from a sender to a receiver) or two-way (between a sender and a receiver or between receivers) data communication can be performed.

FIG. 7B1 illustrates a computer, which includes a main body 7201, a housing 7202, a display portion 7203, a keyboard 7204, an external connection port 7205, a pointing device 7206, and the like. Note that this computer is manufactured using the light-emitting devices described in Embodiment 1 and arranged in a matrix in the display portion 7203. The computer illustrated in FIG. 7B1 may have a structure illustrated in FIG. 7B2. A computer illustrated in FIG. 7B2 is provided with a display portion 7210 instead of the keyboard 7204 and the pointing device 7206. The display portion 7210 is a touch panel, and input operation can be performed by touching display for input on the display portion 7210 with a finger or a dedicated pen. The display portion 7210 can also display images other than the display for input. The display portion 7203 may also be a touch panel. Connecting the two screens with a hinge can

prevent troubles; for example, the screens can be prevented from being cracked or broken while the computer is being stored or carried.

FIG. 7C shows an example of a portable terminal. A 5 cellular phone is provided with a display portion 7402 incorporated in a housing 7401, operation buttons 7403, an external connection port 7404, a speaker 7405, a microphone 7406, and the like. Note that the cellular phone has the display portion 7402 in which the light-emitting devices described in Embodiment 1 are arranged in a matrix.

When the display portion 7402 of the portable terminal illustrated in FIG. 7C is touched with a finger or the like, data can be input to the portable terminal. In this case, operations such as making a call and creating an e-mail can be performed by touching the display portion 7402 with a finger or the like.

The display portion 7402 has mainly three screen modes. The first mode is a display mode mainly for displaying images. The second mode is an input mode mainly for inputting data such as text. The third mode is a display-and-input mode in which the two modes, the display mode and the input mode, are combined.

For example, in the case of making a call or creating an 20 e-mail, a text input mode mainly for inputting text is selected for the display portion 7402 so that text displayed on the screen can be input. In this case, it is preferable to display a keyboard or number buttons on almost the entire screen of the display portion 7402.

When a sensing device including a sensor such as a 25 gyroscope sensor or an acceleration sensor for detecting inclination is provided inside the portable terminal, display on the screen of the display portion 7402 can be automatically changed in direction by determining the orientation of the portable terminal (whether the portable terminal is placed horizontally or vertically).

The screen modes are switched by touching the display portion 7402 or operating the operation buttons 7403 of the 30 housing 7401. Alternatively, the screen modes can be 40 switched depending on the kind of images displayed on the display portion 7402. For example, when a signal of an image displayed on the display portion is a signal of moving image data, the screen mode is switched to the display mode. When the signal is a signal of text data, the screen mode is 45 switched to the input mode.

Moreover, in the input mode, when input by touching the display portion 7402 is not performed for a certain period while a signal sensed by an optical sensor in the display portion 7402 is sensed, the screen mode may be controlled 50 so as to be switched from the input mode to the display mode.

The display portion 7402 may also function as an image sensor. For example, an image of a palm print, a fingerprint, or the like is taken when the display portion 7402 is touched 55 with the palm or the finger, whereby personal authentication can be performed. Furthermore, by providing a backlight or a sensing light source which emits near-infrared light in the display portion, an image of a finger vein, a palm vein, or the like can be taken.

Note that the structure described in this embodiment can 60 be combined with any of the structures described in Embodiments 1 to 4 as appropriate.

As described above, the application range of the light-emitting apparatus having the light-emitting device described in Embodiment 1 or 2 is so wide that this light-emitting apparatus can be used in electronic devices in a variety of fields. By using the light-emitting device

described in Embodiment 1 or 2, an electronic device with low power consumption can be obtained.

FIG. 8A is a schematic view showing an example of a cleaning robot.

A cleaning robot 5100 includes a display 5101 on its top surface, a plurality of cameras 5102 on its side surface, a brush 5103, and operation buttons 5104. Although not illustrated, the bottom surface of the cleaning robot 5100 is provided with a tire, an inlet, and the like. Furthermore, the cleaning robot 5100 includes various sensors such as an infrared sensor, an ultrasonic sensor, an acceleration sensor, a piezoelectric sensor, an optical sensor, and a gyroscope sensor. The cleaning robot 5100 has a wireless communication means.

The cleaning robot 5100 is self-propelled, detects dust 5120, and sucks up the dust through the inlet provided on the bottom surface.

The cleaning robot 5100 can determine whether there is an obstacle such as a wall, furniture, or a step by analyzing images taken by the cameras 5102. When the cleaning robot 5100 detects an object that is likely to be caught in the brush 5103 (e.g., a wire) by image analysis, the rotation of the brush 5103 can be stopped.

The display 5101 can display the remaining capacity of a battery, the amount of collected dust, and the like. The display 5101 may display a path on which the cleaning robot 5100 has run. The display 5101 may be a touch panel, and the operation buttons 5104 may be provided on the display 5101.

The cleaning robot 5100 can communicate with a portable electronic device 5140 such as a smartphone. Images taken by the cameras 5102 can be displayed on the portable electronic device 5140. Accordingly, an owner of the cleaning robot 5100 can monitor his/her room even when the owner is not at home. The owner can also check the display on the display 5101 by the portable electronic device 5140 such as a smartphone.

The light-emitting apparatus of one embodiment of the present invention can be used for the display 5101.

A robot 2100 illustrated in FIG. 8B includes an arithmetic device 2110, an illuminance sensor 2101, a microphone 2102, an upper camera 2103, a speaker 2104, a display 2105, a lower camera 2106, an obstacle sensor 2107, and a moving mechanism 2108.

The microphone 2102 has a function of detecting a speaking voice of a user, an environmental sound, and the like. The speaker 2104 has a function of outputting sound. The robot 2100 can communicate with a user using the microphone 2102 and the speaker 2104.

The display 2105 has a function of displaying various kinds of information. The robot 2100 can display information desired by a user on the display 2105. The display 2105 may be provided with a touch panel. Moreover, the display 2105 may be a detachable information terminal, in which case charging and data communication can be performed when the display 2105 is set at the home position of the robot 2100.

The upper camera 2103 and the lower camera 2106 each have a function of taking an image of the surroundings of the robot 2100. The obstacle sensor 2107 can detect an obstacle in the direction where the robot 2100 advances with the moving mechanism 2108. The robot 2100 can move safely by recognizing the surroundings with the upper camera 2103, the lower camera 2106, and the obstacle sensor 2107. The light-emitting apparatus of one embodiment of the present invention can be used for the display 2105.

FIG. 8C shows an example of a goggle-type display. The goggle-type display includes, for example, a housing 5000, a first display portion 5001, a speaker 5003, an LED lamp 5004 (including a power switch or an operation switch), a connection terminal 5006, a sensor 5007 (a sensor having a function of measuring force, displacement, position, speed, acceleration, angular velocity, rotational frequency, distance, light, liquid, magnetism, temperature, chemical substance, sound, time, hardness, electric field, current, voltage, electric power, radiation, flow rate, humidity, gradient, oscillation, odor, or infrared ray), a microphone 5008, a second display portion 5002, a support 5012, and an earphone 5013.

The light-emitting apparatus of one embodiment of the present invention can be used for the first display portion 5001 and the second display portion 5002.

FIG. 9 shows an example in which the light-emitting device described in Embodiment 1 is used for a table lamp which is a lighting device. The table lamp illustrated in FIG. 9 includes a housing 2001 and a light source 2002, and the lighting device described in Embodiment 4 may be used for the light source 2002.

FIG. 10 shows an example in which the light-emitting device described in Embodiment 1 is used for an indoor lighting device 3001. Since the light-emitting device described in Embodiment 1 has high emission efficiency, the lighting device can have low power consumption. Furthermore, since the light-emitting device described in Embodiment 1 can have a large area, the light-emitting device can be used for a large-area lighting device. Furthermore, since the light-emitting device described in Embodiment 1 is thin, the light-emitting device can be used for a lighting device having a reduced thickness.

The light-emitting device described in Embodiment 1 can also be used for an automobile windshield, an automobile dashboard, or the like. FIG. 11 illustrates one mode in which the light-emitting devices described in Embodiment 1 are used for an automobile windshield and an automobile dashboard. Display regions 5200 to 5203 each include the light-emitting device described in Embodiment 1.

The display regions 5200 and 5201 are display devices which are provided in the automobile windshield and include the light-emitting device described in Embodiment 1. The light-emitting device described in Embodiment 1 can be formed into what is called a see-through display device, through which the opposite side can be seen, by including an anode and a cathode formed of light-transmitting electrodes. Such see-through display devices can be provided even in the automobile windshield without hindering the view. In the case where a driving transistor or the like is provided, a transistor having a light-transmitting property, such as an organic transistor including an organic semiconductor material or a transistor including an oxide semiconductor, is preferably used.

The display region 5202 is a display device which is provided in a pillar portion and includes the light-emitting device described in Embodiment 1. The display region 5202 can compensate for the view hindered by the pillar by displaying an image taken by an imaging unit provided in the car body. Similarly, the display region 5203 provided in the dashboard portion can compensate for the view hindered by the car body by displaying an image taken by an imaging unit provided on the outside of the automobile. Thus, blind areas can be eliminated to enhance the safety. Images that compensate for the areas which a driver cannot see enable the driver to ensure safety easily and comfortably.

The display region 5203 can provide a variety of kinds of information by displaying navigation data, a speedometer, a

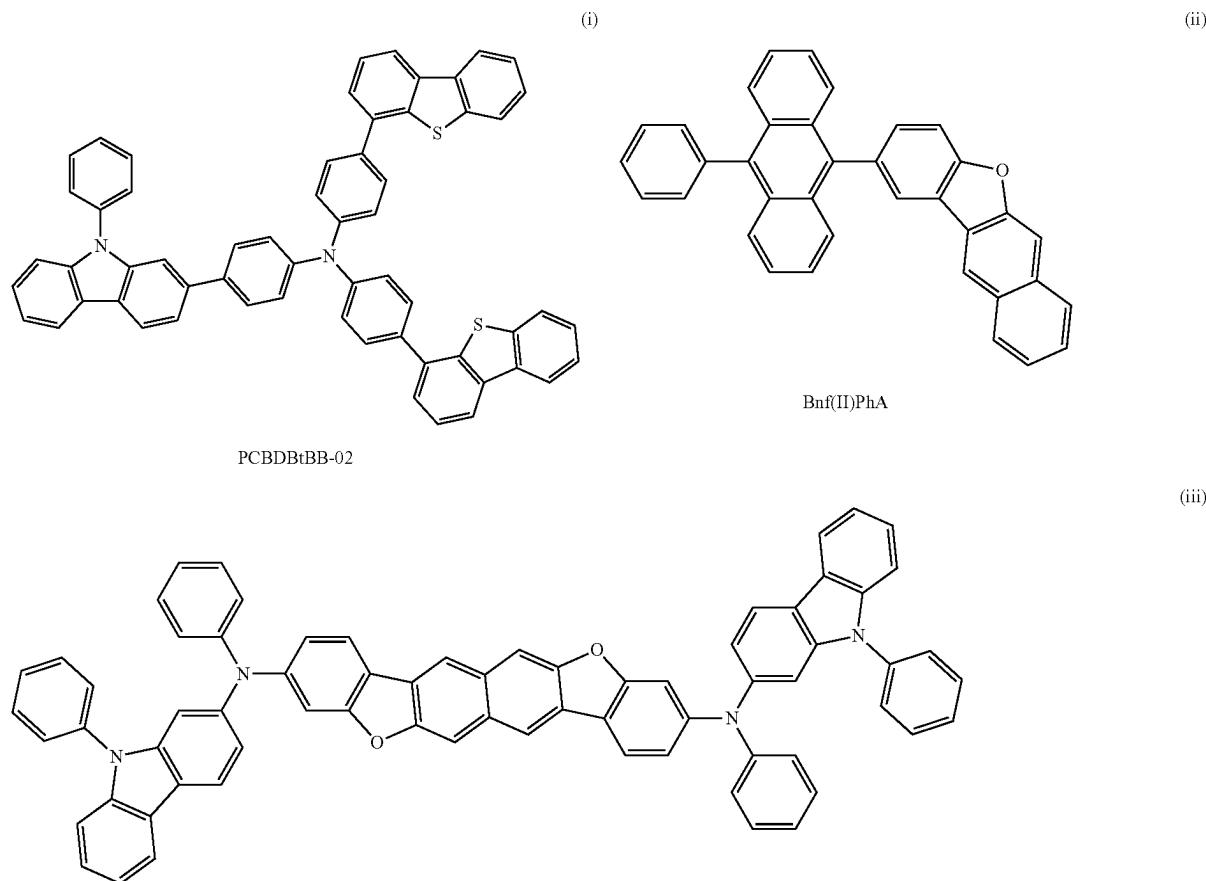
tachometer, air-condition setting, and the like. The content or layout of the display can be changed freely by a user as appropriate. Note that such information can also be displayed on the display regions 5200 to 5202. The display regions 5200 to 5203 can also be used as lighting devices.

FIGS. 12A and 12B illustrate a foldable portable information terminal 5150. The foldable portable information terminal 5150 includes a housing 5151, a display region 5152, and a bend portion 5153. FIG. 12A illustrates the portable information terminal 5150 that is opened. FIG. 12B illustrates the portable information terminal that is folded. Despite its large display region 5152, the portable information terminal 5150 is compact in size and has excellent portability when folded.

The display region 5152 can be folded in half with the bend portion 5153. The bend portion 5153 includes a flexible member and a plurality of supporting members. When the display region is folded, the flexible member expands and the bend portion 5153 has a radius of curvature greater than or equal to 2 mm, preferably greater than or equal to 3 mm.

Note that the display region 5152 may be a touch panel (an input/output device) including a touch sensor (an input device). The light-emitting apparatus of one embodiment of the present invention can be used for the display region 5152.

[Chemical Formula 14]

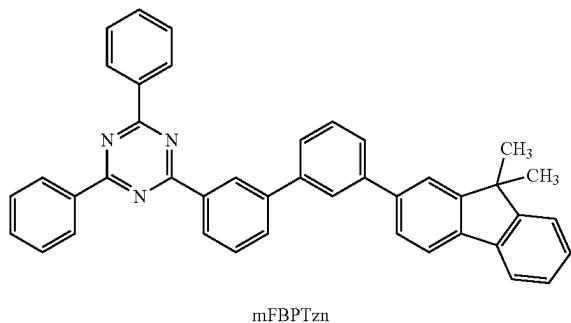


FIGS. 13A to 13C illustrate a foldable portable information terminal 9310. FIG. 13A illustrates the portable information terminal 9310 that is opened. FIG. 13B illustrates the portable information terminal 9310 that is being opened or being folded. FIG. 13C illustrates the portable information terminal 9310 that is folded. The portable information terminal 9310 is highly portable when folded. The portable information terminal 9310 is highly browsable when opened because of a seamless large display region.

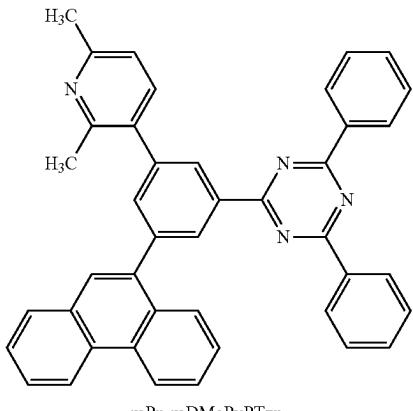
A display panel 9311 is supported by three housings 9315 joined together by hinges 9313. Note that the display panel 9311 may be a touch panel (an input/output device) including a touch sensor (an input device). By folding the display panel 9311 at the hinges 9313 between two housings 9315, the portable information terminal 9310 can be reversibly changed in shape from the opened state to the folded state. The light-emitting apparatus of one embodiment of the present invention can be used for the display panel 9311.

#### Example 1

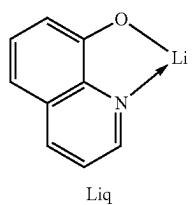
Described in this example are light-emitting devices (a light-emitting device 1 to a light-emitting device 8), each of which includes a hole-transport region provided with at least three layers having different combinations of refractive indices. Structural formulae of organic compounds used in this example are shown below.



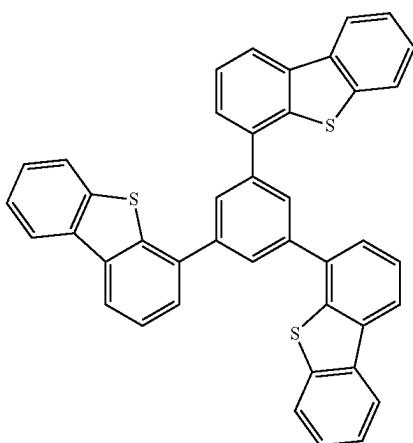
-continued  
(iv)



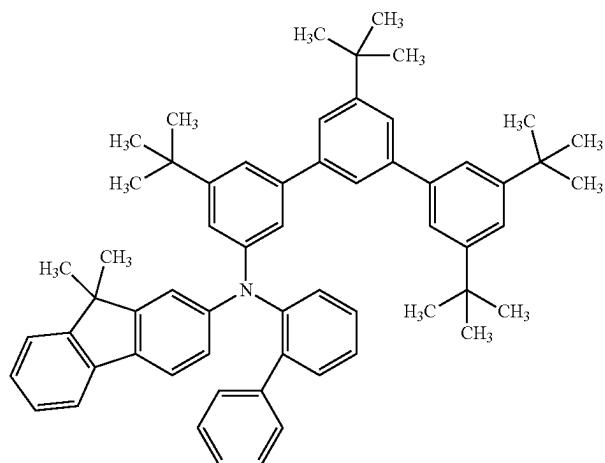
(v)



(vi)



(vii)



(viii)

#### (Fabrication Method of Light-Emitting Device 1)

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 10 nm by a sputtering method, whereby the first electrode **101** was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode **101**

60 is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode **101**.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was 65 washed with water and baked at 200°C. for 1 hour, and then UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to approximately  $10^{-4}$  Pa, vacuum baking was performed at 170° C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

Next, the substrate provided with the first electrode **101** was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode **101** was formed faced downward. Then, 4,4'-bis(dibenzothiophen-4-yl)-4"--(9-phenyl-9H-carbazol-2-yl)triphenylamine (abbreviation: PCBD<sub>B</sub>tBB-02) represented by Structural Formula (i) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the first electrode **101** to a thickness of 10 nm by co-evaporation such that the weight ratio of PCBD<sub>B</sub>tBB-02 to OCHD-003 was 1:0.1, whereby the hole-injection layer **111** was formed.

Over the hole-injection layer **111**, N-(1,1'-biphenyl-2-yl)-N-(3,3",5",5"-tetra-t-butyl-1,1':3',1"-terphenyl-5-yl)-9,9-di-methyl-9H-fluoren-2-amine (abbreviation: mm<sub>T</sub>BumTPoFBi-02) represented by Structural Formula (viii) was deposited by evaporation to a thickness of 35 nm as a first layer, PCBD<sub>B</sub>tBB-02 was deposited by evaporation to a thickness of 40 nm as a second layer, and then mm<sub>T</sub>BumTPoFBi-02 was deposited by evaporation to a thickness of 35 nm as a third layer, whereby the hole-transport layer **112** was formed.

Subsequently, over the hole-transport layer **112**, PCBD<sub>B</sub>tBB-02 was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer was formed.

Then, 2-(10-phenyl-9-anthracenyl)-benzo[b]naphtho[2,3-d]furan (abbreviation: Bnf(II)PhA) represented by Structural Formula (ii) and 3,10-bis[N-(9-phenyl-9H-carbazol-2-yl)-N-phenylamino]naphtho[2,3-b;6,7-b']bisbenzofuran (abbreviation: 3,10PCA2Nbf(IV)-02) represented by Structural Formula (iii) were deposited to a thickness of 25 nm by co-evaporation such that the weight ratio of Bnf(II)PhA to 3,10PCA2Nbf(IV)-02 was 1:0.015, whereby the light-emitting layer **113** was formed.

After that, 2-[3'-(9,9-dimethyl-9H-fluoren-2-yl)-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation:

mFBPTzn) represented by Structural Formula (iv) was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer was formed. Then, 2-[3-(2,6-dimethyl-3-pyridinyl)-5-(9-phenanthrenyl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mPn-mDMePyPTzn) represented by Structural Formula (v) and 8-hydroxyquinolinato-lithium (abbreviation: Liq) represented by Structural Formula (vi) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of mPn-mDMePyPTzn to Liq was 1:1, whereby the electron-transport layer **114** was formed.

After the electron-transport layer **114** was formed, lithium fluoride (LiF) was deposited to a thickness of 1 nm to form the electron-injection layer **115**, and lastly silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode **102**, whereby the light-emitting device **1** was fabricated. The second electrode **102** is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission device in which light is extracted through the second electrode **102**. Over the second electrode **102**, 4,4",4"- (benzene-1,3,5-triyl) tri(dibenzothiophene) (abbreviation: DBT3P-II) represented by Structural Formula (vii) was deposited by evaporation to a thickness of 70 nm so that light extraction efficiency can be improved.

(Fabrication Method of Comparative Light-Emitting Device **1**)

The comparative light-emitting device **1** was fabricated in a manner similar to that for the light-emitting device **1** except that the first layer and the third layer in the hole-transport layer were each formed using PCBD<sub>B</sub>tBB-02 to a thickness of 30 nm.

(Fabrication Method of Comparative Light-Emitting Device **2**)

The comparative light-emitting device **2** was fabricated in a manner similar to that for the light-emitting device **1** except that the second layer in the hole-transport layer was formed using mm<sub>T</sub>BumTPoFBi-02 to a thickness of 60 nm.

The structures of the light-emitting device **1**, the comparative light-emitting device **1**, and the comparative light-emitting device **2** are listed in the following table.

TABLE 1

		Light-emitting device 1	Comparative light-emitting device 1	Comparative light-emitting device 2
Cap layer		70 nm	DBT3P-II	
Cathode		15 nm	Ag:Mg (10:1)	
Electron-injection layer		1 nm	LiF	
Electron-transport layer		20 nm	mPn-mDMePyPTzn:Liq (1:1)	
Hole-blocking layer		10 nm	mFBPTzn	
Light-emitting layer		25 nm	Bnf(II)PhA:3,10PCA2Nbf(IV)-02 (1:0.015)	
Electron-blocking layer		10 nm	PCBD <sub>B</sub> tBB-02	
Hole- transport layer	3 2 1	Another table	L H L	H H H
Hole-injection layer		10 nm	PCBD <sub>B</sub> tBB-02:OCHD-003 (1:0.1)	L
Anode		10 nm	ITSO	L
Reflective electrode		100 nm	Ag	

L: mm<sub>T</sub>BumTPoFBi-02,  
H: PCBD<sub>B</sub>tBB-02

75

TABLE 2

	H	L
Hole-transport layer	3 2 1	30 nm 40 nm 30 nm
	35 nm 60 nm 35 nm	

FIG. 17 shows the measurement results of ordinary refractive indices of PCBDBtBB-02 and mmtBumTPoFBi-02. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A. Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

The graph shows that PCBDBtBB-02 and mmtBumTPoFBi-02 are a combination having a difference in ordinary refractive index greater than or equal to 0.2 and less than 0.5 in the wavelength range of 450 nm to 650 nm.

The HOMO levels of PCBDBtBB-02 and mmtBumTPoFBi-02 that were calculated from cyclic voltammetry (CV) measurement results were -5.51 eV and -5.43 eV, respectively, with N,N-dimethylformamide (DMF) used as a solvent. This indicates that the difference between the HOMO levels of PCBDBtBB-02 and mmtBumTPoFBi-02

shows the current density-voltage characteristics thereof. FIG. 22 shows the blue index-luminance characteristics thereof. FIG. 23 shows the emission spectra thereof. Table 3 shows the main characteristics of the light-emitting device 1 and the comparative light-emitting devices 1 and 2 at a luminance of about 1000 cd/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectra were measured at normal temperature with a spectroradiometer (SR-UL1R manufactured by TOPCON TECHNOHOUSE CORPORATION).

Note that the blue index (BI) is a value obtained by dividing current efficiency (cd/A) by chromaticity y (cd/A/y), and is one of the indicators of characteristics of blue light emission. As the chromaticity y is smaller, the color purity of blue light emission tends to be higher. With high color purity, a wide range of blue can be expressed even with a small number of luminance components; thus, using blue light emission with high color purity reduces the luminance needed for expressing blue, leading to lower power consumption. Thus, BI that is based on chromaticity y, which is one of the indicators of color purity of blue, is suitably used as a mean for showing efficiency of blue light emission. The light-emitting device with higher BI can be regarded as a blue light-emitting device having higher efficiency for a display.

TABLE 3

Voltage (V)	Current (mA)	Current		Current		BI (cd/A/y)
		density (mA/cm <sup>2</sup> )	Chromaticity x	Chromaticity y	efficiency (cd/A)	
Light-emitting device 1	4.4	0.44	10.9	0.14	0.05	9.3
Comparative light-emitting device 1	4.0	0.54	13.5	0.14	0.05	8.1
Comparative light-emitting device 2	4.4	0.43	10.8	0.14	0.05	8.2

is less than or equal to 0.1 eV. Note that the CV measurement was performed with an electrochemical analyzer (ALS model 600A or 600C manufactured by BAS Inc.) as a measurement apparatus, and the measurement was performed on a solution in which a material to be measured was dissolved in a solvent.

The light-emitting device and the comparative light-emitting devices were sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the elements, only the UV curable sealing material was irradiated with UV while the light-emitting devices were prevented from being irradiated with the UV, and heat treatment was performed at 80° C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting devices were measured.

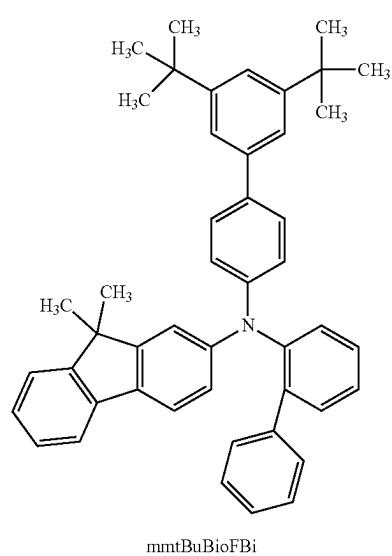
FIG. 18 shows the luminance-current density characteristics of the light-emitting device 1 and the comparative light-emitting devices 1 and 2. FIG. 19 shows the luminance-voltage characteristics thereof. FIG. 20 shows the current efficiency-luminance characteristics thereof. FIG. 21

FIGS. 18 to 23 and Table 3 show that the light-emitting device 1 of one embodiment of the present invention is a light-emitting device that has higher current efficiency and blue index (BI) than the other light-emitting devices. In particular, with an extremely high BI of 200 (cd/A/y) or more, the light-emitting device 1 can be said to have favorable BI. Thus, one embodiment of the present invention is particularly suitable for a light-emitting device used in a display.

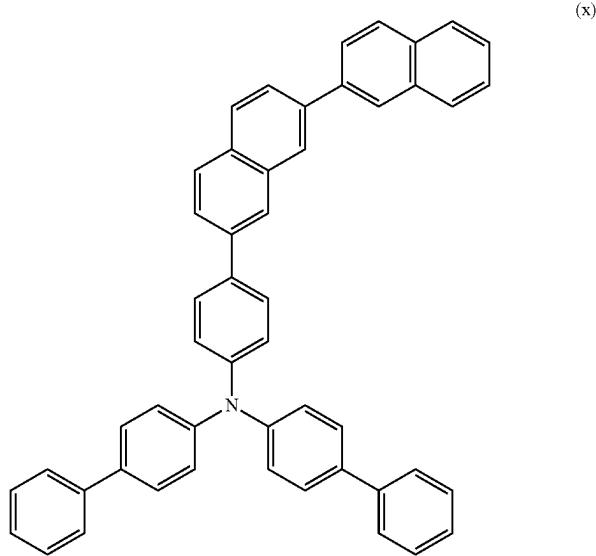
## Example 2

In this example, a light-emitting device 10 of one embodiment of the present invention and a comparative light-emitting device 10 which is a comparative light-emitting device are described. Structural formulae of organic compounds used in this example are shown below.

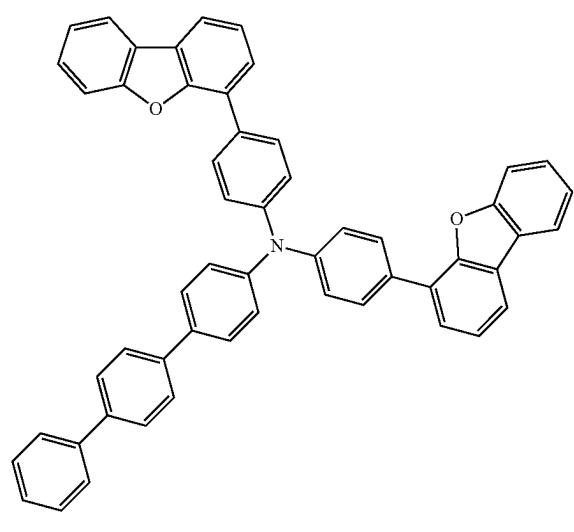
[Chemical Formula 15]



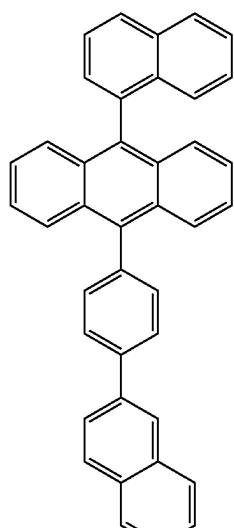
(ix)



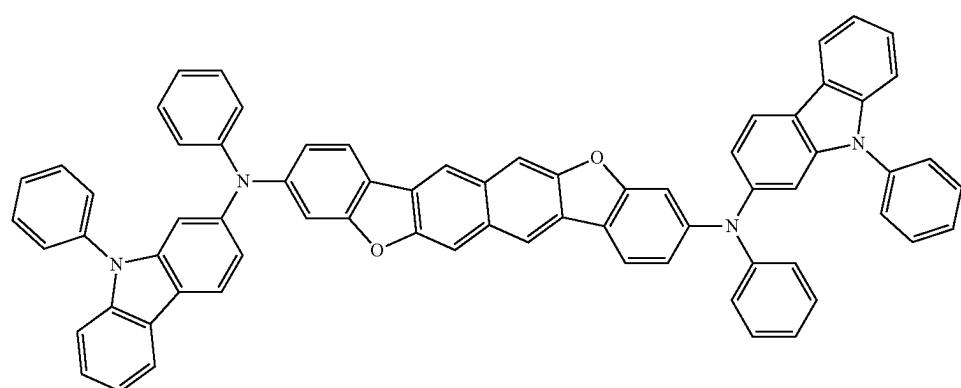
(x)



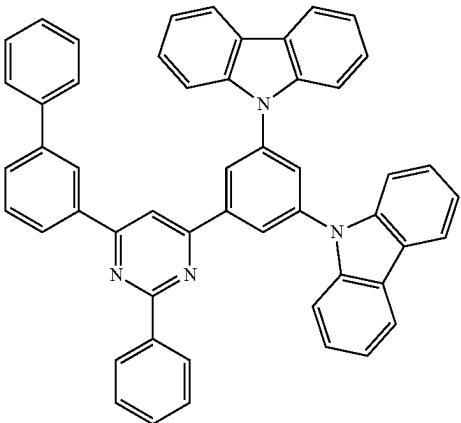
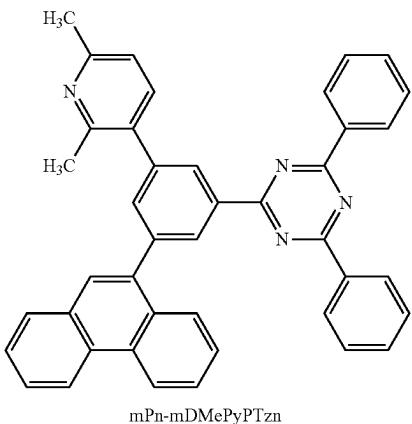
(xi)



(xii)

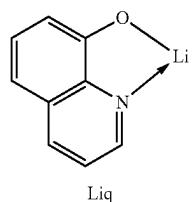


(iii)

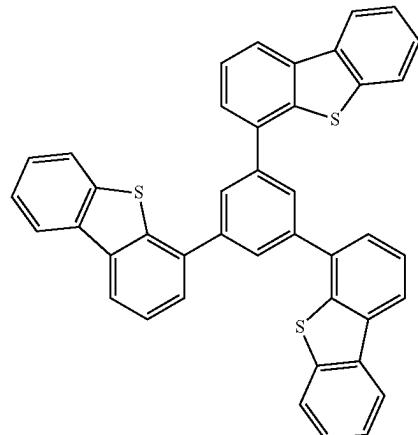


(xiv)

(xv)

-continued  
(v)

(vi)



(vii)

## (Fabrication Method of Light-Emitting Device 10)

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 10 nm by a sputtering method, whereby the first electrode **101** was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode **101** is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode **101**.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was washed with water and baked at 200°C. for 1 hour, and then UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to approximately 10<sup>-4</sup> Pa, vacuum baking was performed at 170°C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

Next, the substrate provided with the first electrode **101** was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode **101** was formed faced downward. Then,

<sup>40</sup> N-3',5'-ditertiarybutyl-1,1'-biphenyl-4-yl-N-1,1'-biphenyl-2-yl-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmTBuBioFBi) represented by Structural Formula (ix) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the <sup>45</sup> first electrode **101** to a thickness of 10 nm by co-evaporation such that the weight ratio of mmTBuBioFBi to OCHD-003 was 1:0.1, whereby the hole-injection layer **111** was formed.

Over the hole-injection layer **111**, mmTBuBioFBi was <sup>50</sup> deposited by evaporation to a thickness of 30 nm as a first layer, 4,4'-diphenyl-4"--(7;2'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA(βN2)B-03) represented by Structural Formula (x) was deposited by evaporation to a thickness of 50 nm as a second layer, and then mmTBuBioFBi was <sup>55</sup> deposited by evaporation to a thickness of 40 nm as a third layer, whereby the hole-transport layer **112** was formed.

Subsequently, over the hole-transport layer **112**, N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBfBB1TP) represented by Structural Formula (xi) was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer was formed.

Then, 9-(1-naphthyl)-10-[4-(2-naphthyl)phenyl]anthracene (abbreviation: αN-βNPAnth) represented by Structural Formula (xii) and 3,10-bis[N-(9-phenyl-9H-carbazol-2-yl)-N-phenylamino]naphtho[2,3-b;6,7-b']bisbenzofuran (abbreviation:

viation: 3,10PCA2Nbf(IV)-02) represented by Structural Formula (iii) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of  $\alpha$ N- $\beta$ NPAanth to 3,10PCA2Nbf(IV)-02 was 1:0.015, whereby the light-emitting layer 113 was formed.

After that, 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm) represented by Structural Formula (xiv) was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer was formed. Then, 2-[3-(2,6-dimethyl-3-pyridinyl)-5-(9-phenanthrenyl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mPn-mDMePyPTzn) represented by Structural Formula (v) and 8-hydroxyquinolinato-lithium (abbreviation: Liq) represented by Structural Formula (vi) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of mPn-mDMePyPTzn to Liq was 1:1, whereby the electron-transport layer 114 was formed.

After the electron-transport layer 114 was formed, lithium fluoride (LiF) was deposited to a thickness of 1 nm to form the electron-injection layer 115, and lastly silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode 102, whereby the light-emitting device 10 was fabricated. The second electrode 102 is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission device in which light is extracted through the second electrode 102. Over the second electrode 102, 4,4',4''-(benzene-1,3,5-triyl)tri(dibenzothiophene) (abbreviation: DBT3P-II) represented by Structural Formula (vii) was deposited by evaporation to a thickness of 70 nm so that light extraction efficiency can be improved.

#### (Fabrication Method of Comparative Light-Emitting Device 10)

The comparative light-emitting device 10 was fabricated in a manner similar to that for the light-emitting device 10 except that the thickness of the first layer in the hole-transport layer was set to 35 nm and the second layer was formed using mmTBuBioFBi.

The structures of the light-emitting device 10 and the comparative light-emitting device 10 are listed in the following table.

TABLE 4

		Light-emitting device 10	Comparative light-emitting device 10
Cap layer	70 nm		DBT3P-II
Cathode	15 nm		Ag:Mg (10:1)
Electron-injection layer	1 nm		LiF
Electron-transport layer	20 nm	mPn-mDMePyPTzn:Liq (1:1)	
Hole-blocking layer	10 nm	6mBP-4CzP2Pm	
Light-emitting layer	20 nm	$\alpha$ N- $\beta$ NPAanth:3,10PCA2Nbf(IV)-02 (1:0.015)	
Electron-blocking layer	10 nm		DBTB1TP
Hole-transport layer	3 40 nm	mmTBuBioFBi	
	2 50 nm	BBA( $\beta$ N2)B-03	mmTBuBioFBi
	1 *3	mmTBuBioFBi	
Hole-injection layer	10 nm	mmTBuBioFBi:OCHD-003 (1:0.1)	

TABLE 4-continued

		Light-emitting device 10	Comparative light-emitting device 10
Anode	10 nm	ITSO	
Reflective electrode	100 nm	Ag	

\*3 Light-emitting device 10:30 nm,  
Comparative light-emitting device 10:35 nm

FIG. 24 shows the measurement results of ordinary refractive indices of BBA( $\beta$ N2)B-03 and mmTBuBioFBi. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A. Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

The graph shows that BBA( $\beta$ N2)B-03 and mmTBuBioFBi are a combination having a difference in ordinary refractive index greater than or equal to 0.2 and less than 0.5 in the wavelength range of 450 nm to 650 nm.

The HOMO levels of BBA( $\beta$ N2)B-03 and mmTBuBioFBi that were calculated from cyclic voltammetry (CV) measurement results were -5.47 eV and -5.42 eV, respectively, with N,N-dimethylformamide (DMF) used as a solvent. This indicates that the difference between the HOMO levels of BBA( $\beta$ N2)B-03 and mmTBuBioFBi is less than or equal to 0.1 eV. Note that the CV measurement was performed with an electrochemical analyzer (ALS model 600A or 600C manufactured by BAS Inc.) as a measurement apparatus, and the measurement was performed on a solution in which a material to be measured was dissolved in a solvent.

The light-emitting device and the comparative light-emitting device were sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the elements, only the UV curable sealing material was irradiated with UV while the light-emitting devices were prevented from being irradiated with the UV, and heat treatment was performed at 80° C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting devices were measured.

FIG. 25 shows the luminance-current density characteristics of the light-emitting device 10 and the comparative light-emitting device 10. FIG. 26 shows the luminance-voltage characteristics thereof. FIG. 27 shows the current efficiency-luminance characteristics thereof. FIG. 28 shows the current density-voltage characteristics thereof. FIG. 29 shows the blue index-current density characteristics thereof. FIG. 30 shows the emission spectra thereof. FIG. 31 shows the viewing angle characteristics thereof. Table 5 shows the main characteristics of the light-emitting device 10 and the comparative light-emitting device 10 at a luminance of about 1000 cd/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectra were measured at normal temperature with a spectroradiometer (SR-ULIR manufactured by TOPCON TECHNOHOUSE CORPORATION). To measure the viewing angle characteristics, given that the direction perpendicular to a light-emitting surface of the light-emitting device was 0°, emission spectra were measured at points at every 10° from -80° to 80°; then, the photon intensity ratio of the light-emitting device at each angle was obtained. For the measurement, a multi-channel spectrometer (PMA-12 produced by Hamamatsu Photonics K.K.) was used.

TABLE 5

	Voltage (V)	Current (mA)	Current density (mA/cm <sup>2</sup> )	Chromaticity x	Chromaticity y	Current efficiency (cd/A)	BI (cd/A/y)
Light-emitting device 10	4.6	0.39	9.8	0.14	0.04	8.5	201
Comparative light-emitting device 10	4.4	0.58	14.4	0.14	0.04	7.3	176

FIGS. 25 to 31 and Table 5 show that the light-emitting device 10 of one embodiment of the present invention is a light-emitting device that has high current efficiency and blue index (BI). In particular, with an extremely high BI of 200 (cd/A/y) or more, the light-emitting device 10 can be said to have favorable BI. Thus, one embodiment of the present invention is particularly suitable for a light-emitting device used in a display.

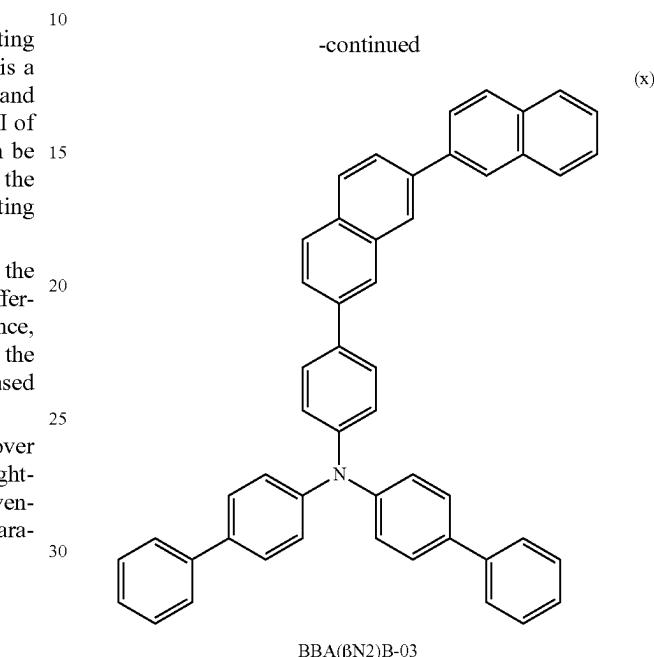
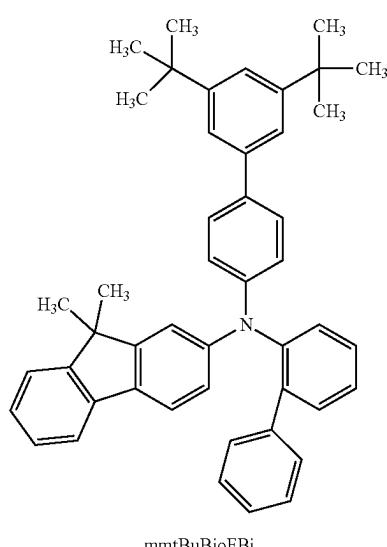
It is also found that the light-emitting device 10 and the comparative light-emitting device 10 have almost no difference in viewing angle characteristics in FIG. 31. Hence, according to one embodiment of the present invention, the efficiency of the light-emitting device can be increased without a change in viewing angle characteristics.

FIG. 32 is a graph showing a change in luminance over driving time at a current density of 50 mA/cm<sup>2</sup>. The light-emitting device 10 of one embodiment of the present invention was found to have higher reliability than the comparative light-emitting device 10.

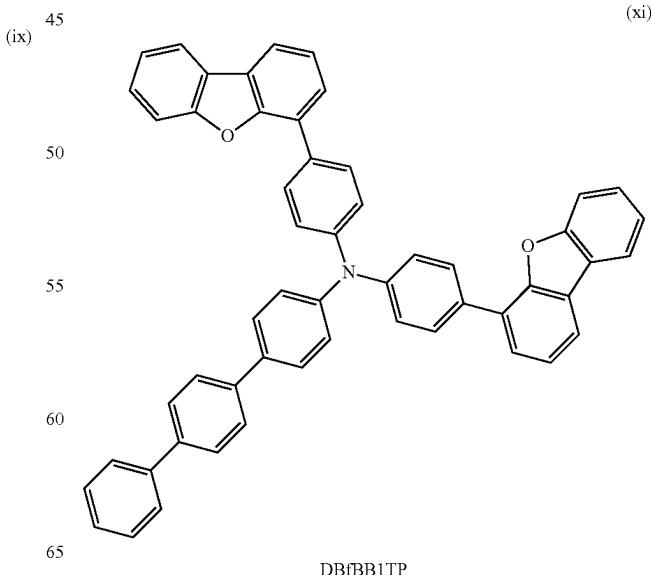
### Example 3

In this example, a light-emitting device 20 of one embodiment of the present invention is described. Structural formulae of organic compounds used in this example are shown below.

[Chemical Formula 16]

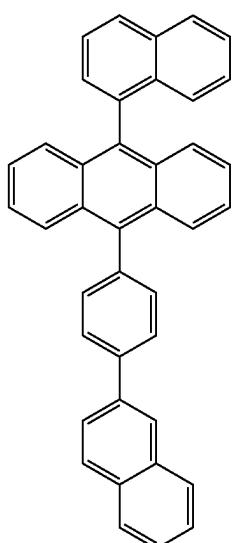
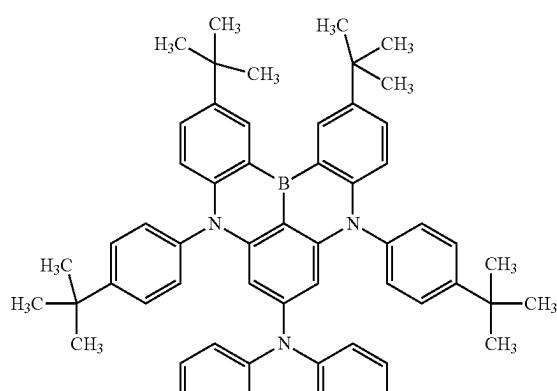
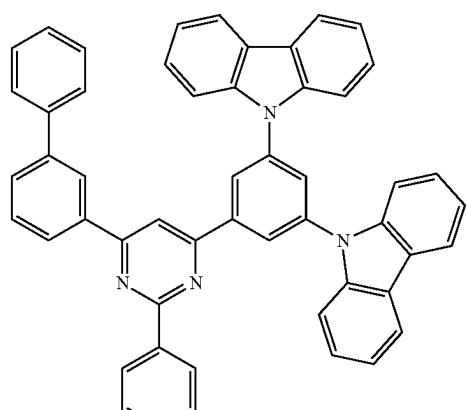


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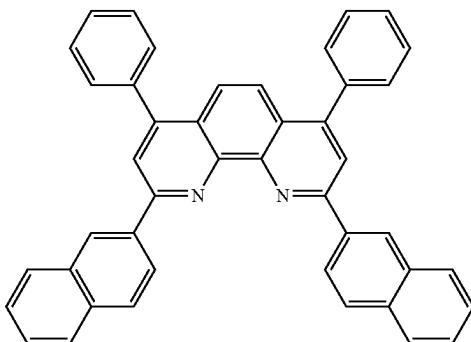
**85**

-continued

**αN-βNPAnth****DPhA-tBu<sub>4</sub>DABNA****6mBP-4Cz2PPm****86**

-continued

(xii)



5

10

15

20

(xiii)

25

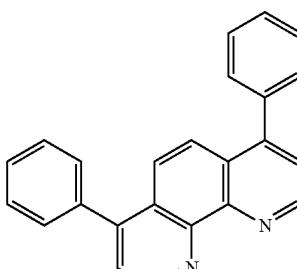
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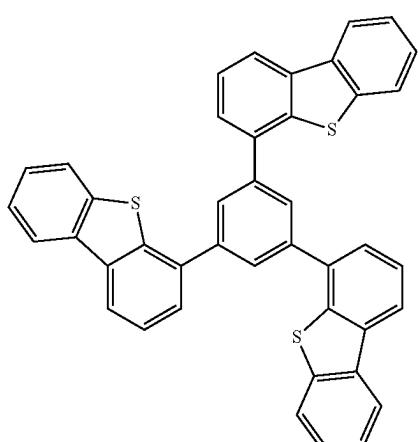
45

(xiv)

**NBPhen**

(xv)

(xvi)



(vii)

**DBT3P-II**

50

(Fabrication Method of Light-Emitting Device **20**)

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, 55 indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 10 nm by a sputtering method, whereby the first electrode **101** was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode **101** is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode **101**.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was washed with water and baked at 200°C. for 1 hour, and then 65 UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to

approximately  $10^{-4}$  Pa, vacuum baking was performed at 170° C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

Next, the substrate provided with the first electrode 101 was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode 101 was formed faced downward. Then, N-3',5'-diteriarybutyl-1,1'-biphenyl-4-yl-N-1,1'-biphenyl-2-yl-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmtBuBioFBi) represented by Structural Formula (ix) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the first electrode 101 to a thickness of 10 nm by co-evaporation such that the weight ratio of mmtBuBioFBi to OCHD-003 was 1:0.04, whereby the hole-injection layer 111 was formed.

Over the hole-injection layer 111, mmtBuBioFBi was deposited by evaporation to a thickness of 35 nm as a first layer, 4,4'-diphenyl-4''-(7,2'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA(βN2)B-03) represented by Structural Formula (x) was deposited by evaporation to a thickness of 50 nm as a second layer, and then mmtBuBioFBi was

evaporation to a thickness of 5 nm, whereby the electron-transport layer 114 was formed.

After the electron-transport layer 114 was formed, bathophenanthroline (abbreviation: BPhen) represented by Structural Formula (xvi) and lithium fluoride (LiF) were deposited to a thickness of 27 nm by co-evaporation such that the volume ratio of BPhen to LiF was 1:3, whereby the electron-injection layer 115 was formed. Lastly, silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode 102, whereby the light-emitting device 20 was fabricated. The second electrode 102 is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission device in which light is extracted through the second electrode 102. Over the second electrode 102, 4,4',4''-(benzene-1,3,5-triyl)tri(dibenzothiophene) (abbreviation: DBT3P-II) represented by Structural Formula (vii) was deposited by evaporation to a thickness of 70 nm so that light extraction efficiency can be improved.

The structures of the light-emitting device 20 are listed in the following table.

TABLE 6

Structure	Film thickness	Material
Cap layer	70 nm	DBT3P-II
Cathode	15 nm	Ag:Mg (10:0.5)
Electron-injection layer	27 nm	LiF:BPhen (3:1)
Electron-transport layer	5 nm	NBPhen
Hole-blocking layer	5 nm	6mBP-4Cz2PPm
Light-emitting layer	20 nm	αN-βNPAnth:DPhA-tBu4DABNA (1:0.015)
Electron-blocking layer	10 nm	DBFBB1TP
Hole-transport layer	40 nm	mmtBuBioFBi
Second hole-transport layer	50 nm	BBA(βN2)B-03
First hole-transport layer	35 nm	mmtBuBioFBi
Hole-injection layer	10 nm	mmtBuBioFBi:OCHD-003 (1:0.04)
Anode	10 nm	ITSO
Reflective electrode	100 nm	Ag

deposited by evaporation to a thickness of 40 nm as a third layer, whereby the hole-transport layer 112 was formed.

Subsequently, over the hole-transport layer 112, N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBfBB1TP) represented by Structural Formula (xi) was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer was formed.

Then, 9-(1-naphthyl)-10-[4-(2-naphthyl)phenyl]anthracene (abbreviation: αN-βNPAnth) represented by Structural Formula (xii) and 2,12-di(tert-butyl)-5,9-di(4-tert-butylphenyl)-N,N-diphenyl-5H,9H-[1,4]benzazaborino[2,3,4-kl]phenazaborin-7-amine (abbreviation: DPhA-tBu4DABNA) represented by Structural Formula (xiii) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of αN-βNPAnth to DPhA-tBu4DABNA was 1:0.015, whereby the light-emitting layer 113 was formed.

After that, 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm) represented by Structural Formula (xiv) was deposited by evaporation to a thickness of 5 nm, whereby a hole-blocking layer was formed. Then, 2,9-di(2-naphthyl)-4,7-diphenyl-1,10-phenanthroline (abbreviation: NBPhen) represented by Structural Formula (xv) was deposited by

FIG. 24 shows the measurement results of ordinary refractive indices of BBA(βN2)B-03 and mmtBuBioFBi. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A. Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

The graph shows that BBA(βN2)B-03 and mmtBuBioFBi are a combination having a difference in ordinary refractive index greater than or equal to 0.2 and less than 0.5 in the wavelength range of 450 nm to 650 nm.

The HOMO levels of BBA(βN2)B-03 and mmtBuBioFBi that were calculated from cyclic voltammetry (CV) measurement results were -5.47 eV and -5.42 eV, respectively, with N,N-dimethylformamide (DMF) used as a solvent. This indicates that the difference between the HOMO levels of BBA(βN2)B-03 and mmtBuBioFBi is less than or equal to 0.1 eV. Note that the CV measurement was performed with an electrochemical analyzer (ALS model 600A or 600C manufactured by BAS Inc.) as a measurement apparatus, and the measurement was performed on a solution in which a material to be measured was dissolved in a solvent.

The light-emitting device was sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the element, only the UV curable sealing material was irradiated with UV while the light-emitting device was prevented from being irradiated with the UV, and heat treatment was performed at 80°C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting device were measured.

FIG. 33 shows the luminance-current density characteristics of the light-emitting device 20. FIG. 34 shows the

luminance-voltage characteristics thereof. FIG. 35 shows the current efficiency-luminance characteristics thereof. FIG. 36 shows the current density-voltage characteristics thereof. FIG. 37 shows the blue index-luminance characteristics thereof. FIG. 38 shows the emission spectrum thereof. Table 7 shows the main characteristics of the light-emitting device 20 at a luminance of about 1000 cd/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectra were measured at normal temperature with a spectroradiometer (SR-ULIR manufactured by TOPCON TECHNOHOUSE CORPORATION).

TABLE 7

Voltage (V)	Current (mA)	Current density (mA/cm <sup>2</sup> )	Chromaticity x	Chromaticity y	Current efficiency (cd/A)	BI (cd/A/y)
4.8	0.54	13.6	0.15	0.03	8.1	253

20

FIGS. 33 to 38 and Table 7 show that the light-emitting device 20 of one embodiment of the present invention is a light-emitting device that has extremely high current efficiency and blue index (BI). In particular, with an extremely high BI of 250 (cd/A/y) or more, the light-emitting device 20 can be said to have favorable BI. Thus, one embodiment of the present invention is particularly suitable for a light-emitting device used in a display.

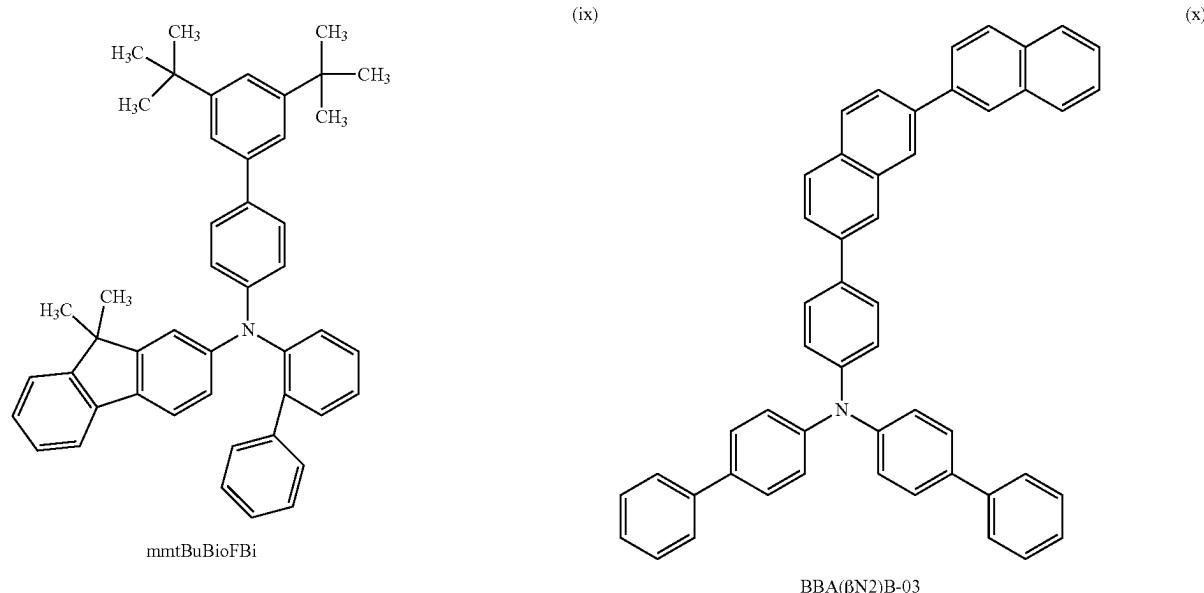
FIG. 39 is a graph showing a change in luminance over driving time at a current density of 50 mA/cm<sup>2</sup>. The light-emitting device 20 of one embodiment of the present invention was found to have high reliability.

## Example 4

35

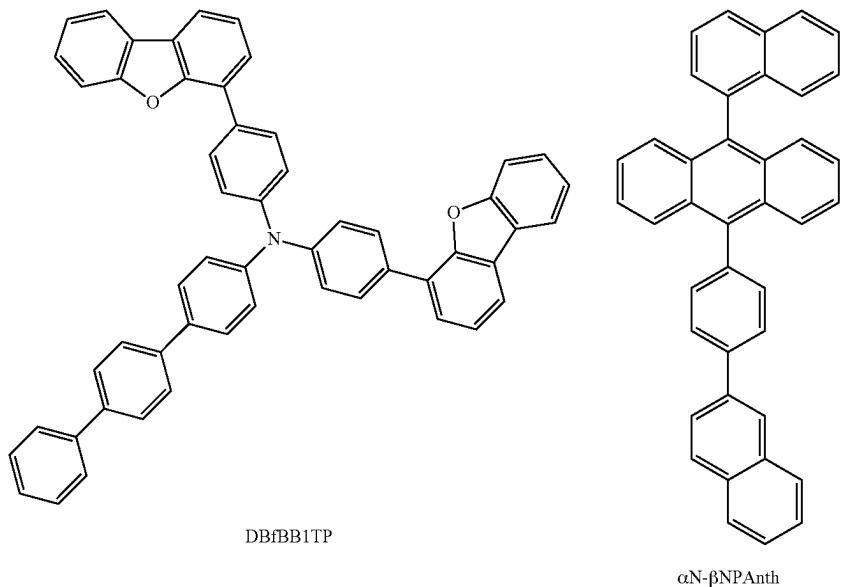
In this example, a light-emitting device 30 of one embodiment of the present invention is described. Structural formulae of organic compounds used in this example are shown below.

[Chemical Formula 17]

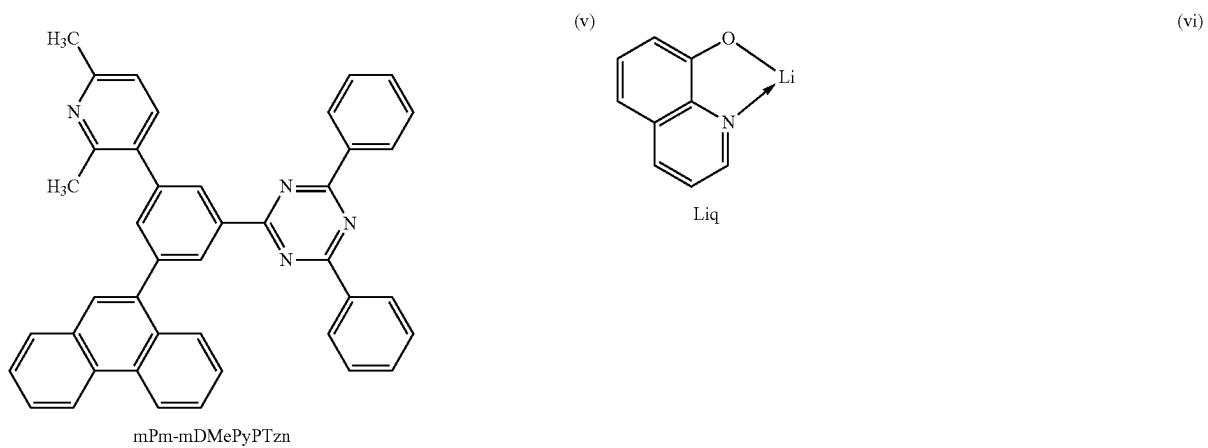
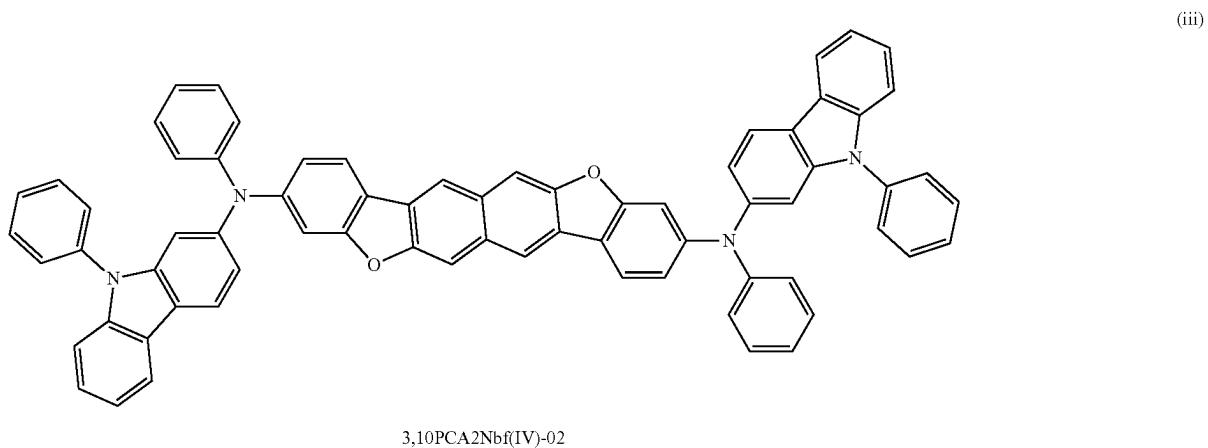


**91****92**

-continued  
(xi)

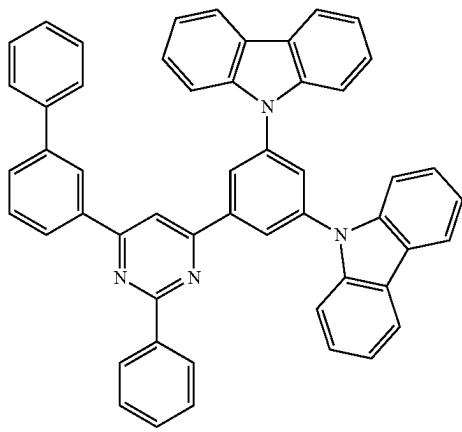


(xii)



-continued

(xiv)



## (Fabrication Method of Light-Emitting Device 30)

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 5 nm by a sputtering method, whereby the first electrode **101** was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode **101** is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode **101**.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was washed with water and baked at 200°C. for 1 hour, and then UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to approximately 10<sup>-4</sup> Pa, vacuum baking was performed at 170°C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

Next, the substrate provided with the first electrode **101** was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode **101** was formed faced downward. Then, N-3',5'-ditertiarybutyl-1,1'-biphenyl-4-yl-N-1,1'-biphenyl-2-yl-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmt-BuBioFBi) represented by Structural Formula (ix) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the first electrode **101** to a thickness of 10 nm by co-evaporation such that the weight ratio of mmtBuBioFBi to OCHD-003 was 1:0.1, whereby the hole-injection layer **111** was formed.

Over the hole-injection layer **111**, mmtBuBioFBi was deposited by evaporation to a thickness of 42.5 nm as a first layer, 4,4'-diphenyl-4''-(7,2'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA(βN2)B-03) represented by Structural Formula (x) was deposited by evaporation to a thickness of 50 nm as a second layer, and then mmtBuBioFBi was deposited by evaporation to a thickness of 40 nm as a third layer, whereby the hole-transport layer **112** was formed.

Subsequently, over the hole-transport layer **112**, N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBfBB1TP) represented by Structural Formula (xi)

was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer was formed.

Then, 9-(1-naphthyl)-10-[4-(2-naphthyl)phenyl]anthracene (abbreviation: αN-βNPAnth) represented by Structural Formula (xii) and 3,10-bis[N-(9-phenyl-9H-carbazol-2-yl)-N-phenylamino]naphtho[2,3-b;6,7-b']bisbenzofuran (abbreviation: 3,10PCA2Nbf(IV)-02) represented by Structural Formula (iii) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of αN-βNPAnth to 3,10PCA2Nbf(IV)-02 was 1:0.015, whereby the light-emitting layer **113** was formed.

After that, 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm) represented by Structural Formula (xiv) was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer was formed. Then, 2-[3-(2,6-dimethyl-3-pyridinyl)-5-(9-phenanthrenyl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mPn-mDMePyPTzn) represented by Structural Formula (v) and 8-hydroxyquinolinato-lithium (abbreviation: Liq) represented by Structural Formula (vi) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of mPn-mDMePyPTzn to Liq was 0.4:0.6, whereby the electron-transport layer **114** was formed.

After the electron-transport layer **114** was formed, lithium fluoride (LiF) was deposited to a thickness of 1 nm to form the electron-injection layer **115**, and lastly silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode **102**, whereby the light-emitting device **30** was fabricated. The second electrode **102** is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission device in which light is extracted through the second electrode **102**. Over the second electrode **102**, an organic compound was deposited by evaporation to a thickness of 65 nm as a cap layer so that light extraction efficiency can be improved.

The structures of the light-emitting device **30** are listed in the following table.

TABLE 8

		Light-emitting device 30
Cathode	15 nm	Ag:Mg (10:1)
Electron-injection layer	1 nm	LiF
Electron-transport layer	20 nm	mPn-mDMePyPTzn:Liq (0.4:0.6)
Hole-blocking layer	10 nm	6mBP-4CzP2Pm
Light-emitting layer	20 nm	$\alpha$ N- $\beta$ NP Anth:3,10PCA2NbF(IV)-02 (1:0.015)
Electron-blocking layer	10 nm	DBFB1TP
Hole-transport layer	3 40 nm	mmtBuBioFBi
	2 50 nm	BBA( $\beta$ N2)B-03
	1 42.5 nm	mmtBuBioFBi
Hole-injection layer	10 nm	mmtBuBioFBi:OCHD-003 (1:0.1)
Anode	5 nm	ITSO
Reflective electrode	100 nm	Ag

the UV curable sealing material was irradiated with UV while the light-emitting device was prevented from being irradiated with the UV, and heat treatment was performed at 80° C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting device were measured.

FIG. 40 shows the luminance-current density characteristics of the light-emitting device 30. FIG. 41 shows the luminance-voltage characteristics thereof. FIG. 42 shows the current efficiency-luminance characteristics thereof. FIG. 43 shows the current density-voltage characteristics thereof. FIG. 44 shows the blue index-luminance characteristics thereof. FIG. 45 shows the emission spectrum thereof. Table 9 shows the main characteristics of the light-emitting device 30 at a luminance of about 1000 cd/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectra were measured at normal temperature with a spectroradiometer (SR-ULIR manufactured by TOPCON TECHNOHOUSE CORPORATION).

TABLE 9

	Voltage (V)	Current (mA)	Current density (mA/cm <sup>2</sup> )	Chromaticity x	Chromaticity y	Current efficiency (cd/A)	Blue index (cd/A/y)
Light-emitting device 30	4.6	0.36	8.9	0.14	0.04	9.2	216

FIG. 24 shows the measurement results of ordinary refractive indices of BBA( $\beta$ N2)B-03 and mmtBuBioFBi. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A. Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

The graph shows that BBA( $\beta$ N2)B-03 and mmtBuBioFBi are a combination having a difference in ordinary refractive index greater than or equal to 0.2 and less than 0.5 in the wavelength range of 450 nm to 650 nm.

The HOMO levels of BBA( $\beta$ N2)B-03 and mmtBuBioFBi that were calculated from cyclic voltammetry (CV) measurement results were -5.47 eV and -5.42 eV, respectively, with N,N-dimethylformamide (DMF) used as a solvent. This indicates that the difference between the HOMO levels of BBA( $\beta$ N2)B-03 and mmtBuBioFBi is less than or equal to 0.1 eV. Note that the CV measurement was performed with an electrochemical analyzer (ALS model 600A or 600C manufactured by BAS Inc.) as a measurement apparatus, and the measurement was performed on a solution in which a material to be measured was dissolved in a solvent.

The light-emitting device was sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the element, only

FIGS. 40 to 45 and Table 9 show that the light-emitting device 30 of one embodiment of the present invention is a light-emitting device that has high current efficiency and blue index (BI). In particular, with an extremely high BI of 200 (cd/A/y) or more, the light-emitting device 30 can be said to have favorable BI. Thus, one embodiment of the present invention is particularly suitable for a light-emitting device used in a display.

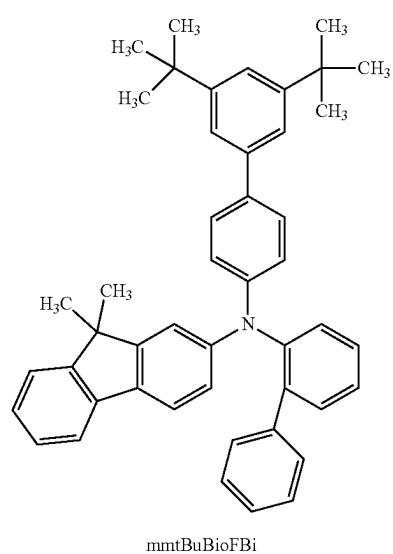
FIG. 46 is a graph showing a change in luminance over driving time at a current density of 50 mA/cm<sup>2</sup>. The light-emitting device 30 of one embodiment of the present invention was found to have high reliability.

#### Example 5

Described in this example are a light-emitting device 40 and a light-emitting device 41, which are different in order of values of GSP in a first hole-transport layer, a second hole-transport layer, and a third hole-transport layer. Structural formulae of organic compounds used in this example are shown below.

**97**

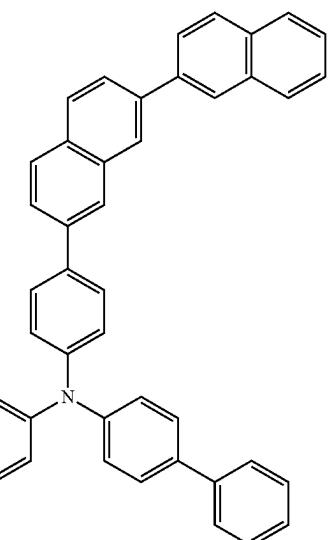
[Chemical Formula 18]



mmtBuBioFBi

**98**

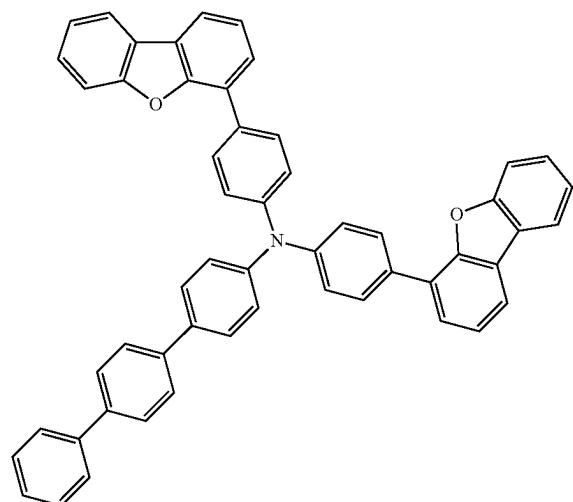
(ix)



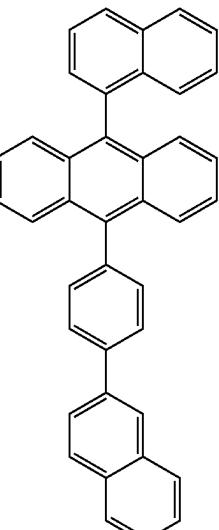
BBA(βN2)B-03

(x)

(xi)



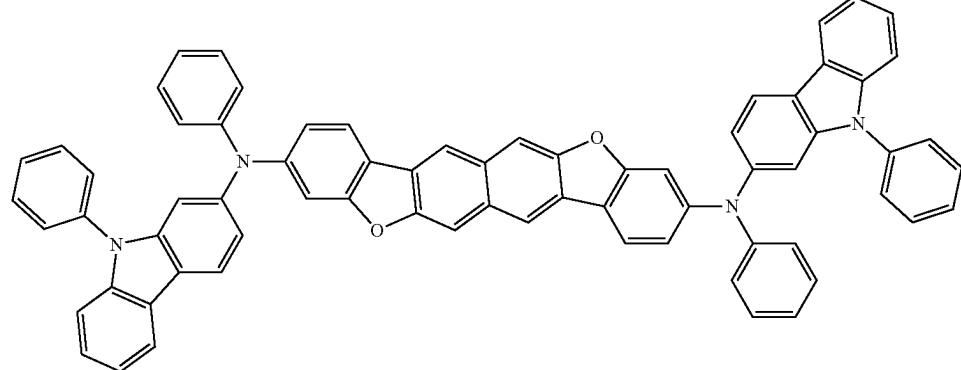
DBfBB1TP



αN-βNPAanth

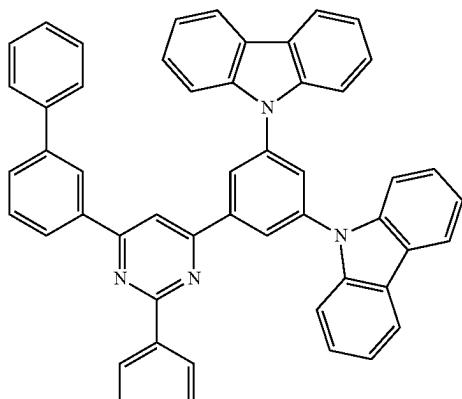
(xii)

(iii)

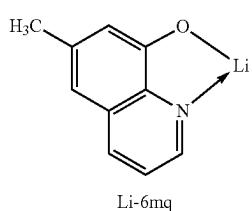
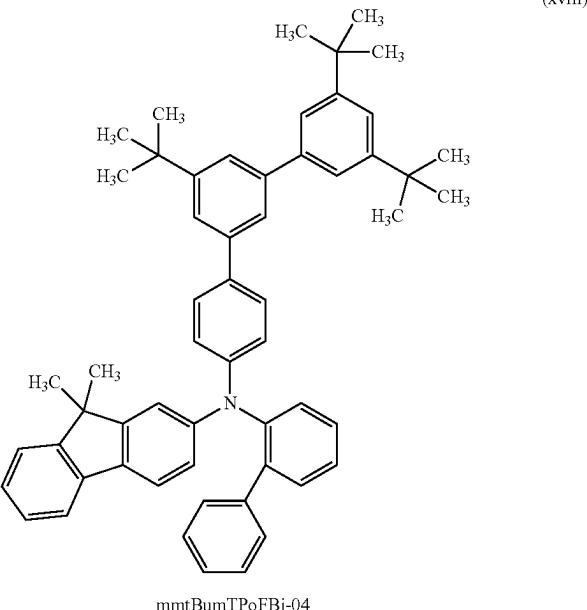


3,10PCA2Nbf(IV)-02

99

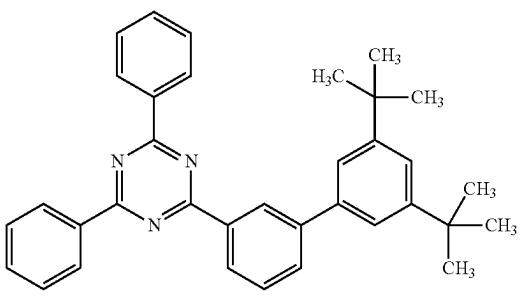
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(xiv)

100



(xx)

(xix)

**(Fabrication Method of Light-Emitting Device 40)**

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 5 nm by a sputtering method, whereby the first electrode **101** was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode **101** is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode **101**.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was washed with water and baked at 200°C. for 1 hour, and then UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to approximately 10<sup>-4</sup> Pa, vacuum baking was performed at 170°C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

Next, the substrate provided with the first electrode **101** was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode **101** was formed faced downward. Then, N-3',5'-ditertiarybutyl-1,1'-biphenyl-4-yl-N-1,1'-biphenyl-

2-yl-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmTBuBioFBi) represented by Structural Formula (ix) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the first electrode **101** to a thickness of 10 nm by co-evaporation such that the weight ratio of mmTBuBioFBi to OCHD-003 was 1:0.1, whereby the hole-injection layer **111** was formed.

Over the hole-injection layer **111**, mmTBuBioFBi was deposited by evaporation to a thickness of 40 nm as a first layer, 4,4'-diphenyl-4''-(7,2'-binaphthyl-2-yl)triphenylamine (abbreviation: BBA(βN2)B-03) represented by Structural Formula (x) was deposited by evaporation to a thickness of 50 nm as a second layer, and then mmTBuBioFBi was deposited by evaporation to a thickness of 40 nm as a third layer, whereby the hole-transport layer **112** was formed.

Subsequently, over the hole-transport layer **112**, N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBfBB1TP) represented by Structural Formula (xi) was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer was formed.

Then, 9-(1-naphthyl)-10-[4-(2-naphthyl)phenyl]anthracene (abbreviation: αN-βNPAnth) represented by Structural Formula (xii) and 3,10-bis[N-(9-phenyl-9H-carbazol-2-yl)-N-phenylamino]naphtho[2,3-b;6,7-b']bisbenzofuran (abbreviation: 3,10PCA2Nbf(IV)-02) represented by Structural Formula (iii) were deposited to a thickness of 20 nm by

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co-evaporation such that the weight ratio of  $\alpha$ -N- $\beta$ NPAanth to 3,10PCA2NbF(IV)-02 was 1:0.015, whereby the light-emitting layer **113** was formed.

After that, 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm) represented by Structural Formula (xiv) was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer was formed. Then, 2-{(3',5'-di-tert-butyl)-1,1'-biphenyl-3-yl}-4,6-diphenyl-1,3,5-triazine (ab-

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The comparative light-emitting device **40** was fabricated in a manner similar to that for the light-emitting device **40** or the light-emitting device **41** except that the hole-transport layer **112** was formed to a thickness of 130 nm using mmtBuBioFBi.

The structures of the light-emitting device **40**, the light-emitting device **41**, and the comparative light-emitting device **40** are listed in the following table.

TABLE 10

	Film thickness	Light-emitting device 40	Light-emitting device 41	Comparative light-emitting device 40
Cathode	15 nm		Ag:Mg (10:1) LiF	
Electron-injection layer	1 nm			
Electron-transport layer	20 nm		mmtBumBPTzn:Li-6mq (1:1)	
Hole-blocking layer	10 nm		6mBP-4Cz2PPm	
Light-emitting layer	20 nm		$\alpha$ N- $\beta$ NPAanth:3,10PCA2NbF(IV)-02 (1:0.015)	
Electron-blocking layer	10 nm		DBfBB1TP	
Hole-transport layer	3	Another table	L1	L1
	2		H	H
	1		L1	L2
Hole-injection layer	10 nm	mmtBuBioFBi:OCHD-003 (1:0.1)	mmtBumTPoFBi-04:OCHD-003 (1:0.1)	mmtBuBioFBi:OCHD-003 (1:0.1)
Anode	5 nm		ITSO	
Reflective electrode	100 nm		Ag	

L1: mmtBuBioFBi,

H: BBA( $\beta$ N2)B-03,

L2: mmtBumTPoFBi-04

breviation: mmtBumBPTzn) represented by Structural Formula (xix) and 6-methyl-8-quinolinolato-lithium (abbreviation: Li-6mq) represented by Structural Formula (xx) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of mmtBumBPTzn to Li-6mq was 1:1, whereby the electron-transport layer **114** to **114** was formed.

After the electron-transport layer **114** was formed, lithium fluoride (LiF) was deposited to a thickness of 1 nm to form the electron-injection layer **115**, and lastly silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode **102**, whereby the light-emitting device **40** was fabricated. The second electrode **102** is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission device in which light is extracted through the second electrode **102**. Over the second electrode **102**, an organic compound was deposited by evaporation to a thickness of 65 nm as a cap layer so that light extraction efficiency can be improved. (Fabrication Method of Light-Emitting Device **41**)

The light-emitting device **41** was fabricated in a manner similar to that for the light-emitting device **40** except that the first layer in the hole-transport layer **112** was formed using N-(3",5",5"-tri-tert-butyl-1,1':3",1"-terphenyl-4-yl)-N-(1,1'-biphenyl-2-yl)-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmtBumTPoFBi-04) represented by Structural Formula (xviii), and the thickness thereof was set to 52.5 nm. (Fabrication Method of Comparative Light-Emitting Device **40**)

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TABLE 11

		Light-emitting device 40	Light-emitting device 41	Comparative light-emitting device 40
40	Hole-transport layer	3 2 1	40 nm 50 nm 40 nm	130 nm 52.5 nm

45 FIGS. 47A and 47B show the measurement results of ordinary refractive indices of mmtBuBioFBi, BBA( $\beta$ N2)B-03, and mmtBumTPoFBi-04. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A. Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

50 55 The graph shows that a combination of BBA( $\beta$ N2)B-03 and mmtBuBioFBi, and a combination of BBA( $\beta$ N2)B-03 and mmtBumTPoFBi-04 each have a difference in ordinary refractive index greater than or equal to 0.2 and less than 0.5 in the wavelength range of 450 nm to 650 nm.

The HOMO levels of BBA( $\beta$ N2)B-03, mmtBuBioFBi, and mmtBumTPoFBi-04 that were calculated from cyclic voltammetry (CV) measurement results were -5.47 eV, -5.42 eV, and -5.42 eV, respectively, with N,N-dimethylformamide (DMF) used as a solvent. This indicates that the difference between the HOMO levels of BBA( $\beta$ N2)B-03 and mmtBuBioFBi is less than or equal to 0.1 eV and the difference between the HOMO levels of BBA( $\beta$ N2)B-03 and mmtBumTPoFBi-04 is less than or equal to 0.1 eV. Note that the CV measurement was performed with an electro-

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chemical analyzer (ALS model 600A or 600C manufactured by BAS Inc.) as a measurement apparatus, and the measurement was performed on a solution in which a material to be measured was dissolved in a solvent.

GSP of each of mmtBuBioFBi, BBA( $\beta$ N2)B-03, and mmtBumTPoFBi-04 is listed in the following table. The table shows that the light-emitting device 40 is a device fabricated without consideration of GSP, whereas the light-emitting device 41 is a device fabricated in consideration of GSP in which the third layer has the largest GSP and the value obtained by subtracting GSP of the second layer from GSP of the first layer is less than or equal to 10 (mV/nm).

TABLE 12

	mmtBuBioFBi	H:BBA( $\beta$ N2)B-03	mmtBumTPoFBi-04
GSP (mV/nm)	25.5	10.6	16.7

The light-emitting devices were sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the elements, only the UV curable sealing material was irradiated with UV while the light-emitting devices were prevented from being irradiated with the UV, and heat treatment was performed at 80° C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting devices were measured.

FIG. 48 shows the luminance-current density characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40. FIG. 49 shows the current efficiency-luminance characteristics thereof. FIG. 50 shows the luminance-voltage characteristics thereof. FIG. 51 shows the current density-voltage characteristics thereof. FIG. 52 shows the blue index-luminance characteristics thereof. FIG. 53 shows the power efficiency-luminance characteristics thereof. FIG. 54 shows the emission spectra thereof. Table 13 shows the main characteristics of the light-emitting device 40, the light-emitting device 41, and the comparative light-emitting device 40 at a luminance of about 1000 cd/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectra were measured at normal temperature with a spectroradiometer (SR-UL1R manufactured by TOPCON TECHNOHOUSE CORPORATION). The power efficiency is calculated under assumption of Lambertian.

TABLE 13

	Voltage (V)	Current (mA)	Current		Chromaticity x	Chromaticity y	Current efficiency (cd/A)	Power efficiency (lm/W)	BI (cd/A/y)
			density (mA/cm <sup>2</sup> )						
Light-emitting device 40	5.0	0.48	11.9		0.14	0.04	9.2	5.8	215
Light-emitting device 41	4.2	0.46	11.4		0.14	0.04	9.3	7.0	226
Comparative light-emitting device 40	4.6	0.56	14.0		0.14	0.04	7.9	5.4	200

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FIGS. 48 to 54 and Table 13 show that the light-emitting devices 40 and 41 of one embodiment of the present invention, and the comparative light-emitting device 40 have favorable characteristics. In particular, the light-emitting devices 40 and 41 having the structure of the present invention each include the hole-transport layer consisting of the first to third layers, and thus have high current efficiency and BI.

The comparison between the light-emitting device 40 and the comparative light-emitting device 40 reveals that the driving voltage of the light-emitting device 40 is increased by the influence of interface resistance due to the three-layer structure of the hole-transport layer 112, or the like. However, this structure has a significant effect of improving current efficiency; thus, the power efficiency of the light-emitting device 40 is higher than that of the comparative light-emitting device 40 even when the driving voltage thereof increases.

In contrast, the light-emitting device 41, which includes the hole-transport layer 112 consisting of the first to third layers formed in consideration of GSP, exhibits lower driving voltage than the comparative light-emitting device 40 including a single hole-transport layer. As a result, the light-emitting device 41 was found to exhibit extremely high power efficiency.

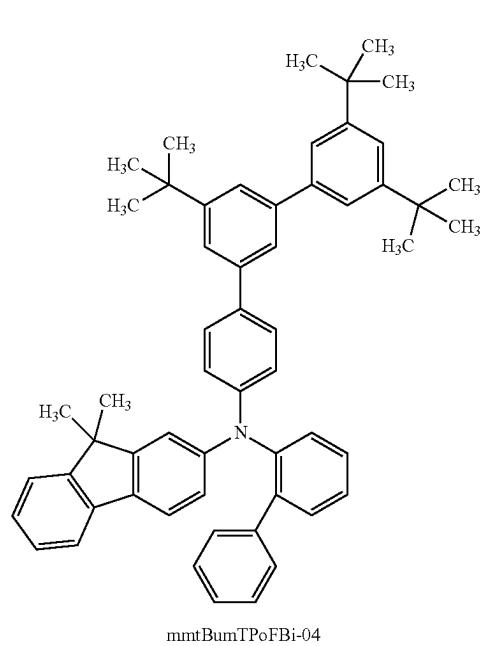
FIG. 55 is a graph showing a change in luminance over driving time at a current density of 50 mA/cm<sup>2</sup>. The light-emitting devices 40 and 41 of one embodiment of the present invention were found to have higher reliability than the comparative light-emitting device 40.

#### Example 6

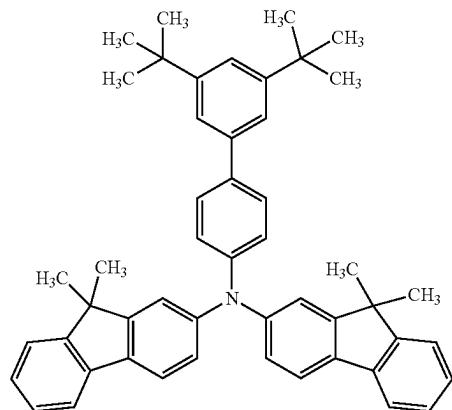
Described in this example are a light-emitting device 50 and a light-emitting device 51, which are different in order of values of GSP in a first hole-transport layer, a second hole-transport layer, and a third hole-transport layer. Structural formulae of organic compounds used in this example are shown below.

**105**

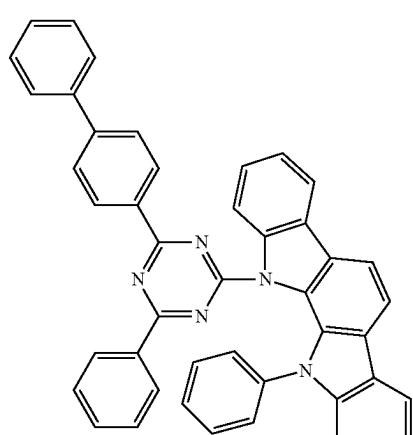
[Chemical Formula 19]



mmtBuTPoFBi-04



mmtBuBiFF

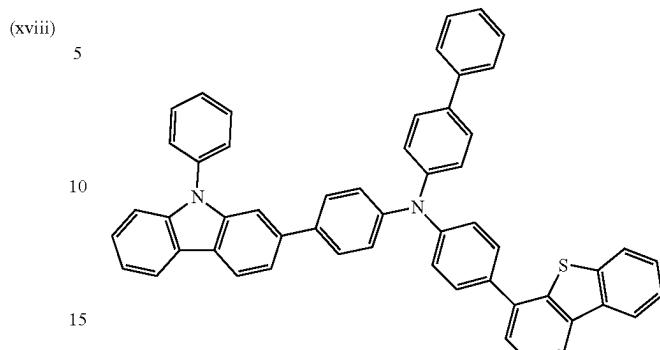


BP-Icz(II)Tzn

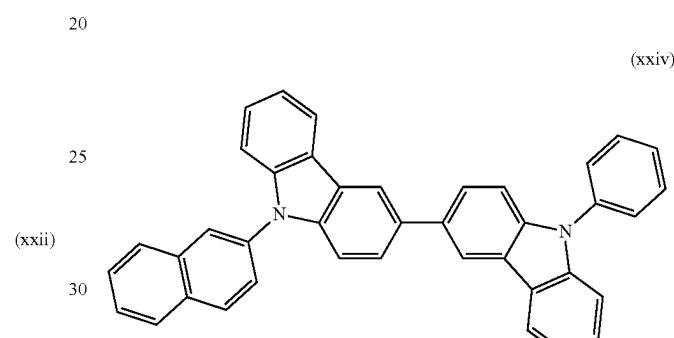
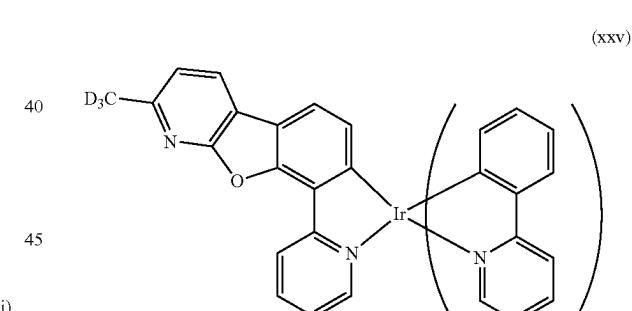
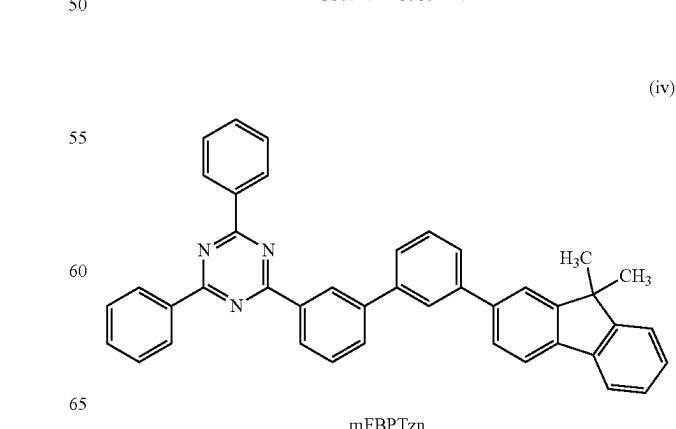
**106**

-continued

(xxi)



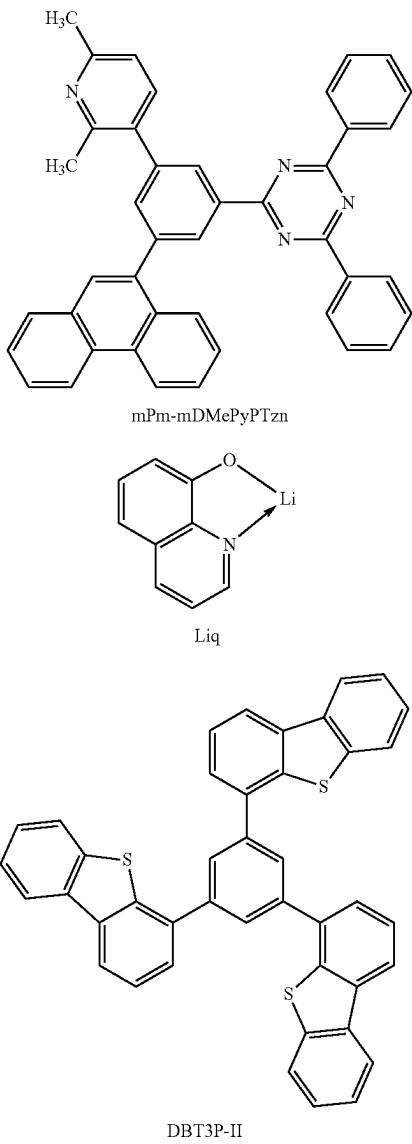
PCBBiPDBt-02

 $\beta$ NCCP $\text{Ir}(\text{ppy})_2(\text{mbfpypy-d3})$ 

mFBPTzn

**107**

-continued

(Fabrication Method of Light-Emitting Device **50**)

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 10 nm by a sputtering method, whereby the first electrode **101** was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode **101** is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode **101**.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was washed with water and baked at 200°C. for 1 hour, and then UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to approximately 10<sup>-4</sup> Pa, vacuum baking was performed at 170°C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

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Next, the substrate provided with the first electrode **101** was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode **101** was formed faced downward. Then, N-(3",5",5"-tri-tert-butyl-1,1'-3",1"-terphenyl-4-yl)-N-(1,1'-biphenyl-2-yl)-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmtBumTPoFBi-04) represented by Structural Formula (xviii) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the first electrode **101** to a thickness of 10 nm by co-evaporation such that the weight ratio of mmtBumTPoFBi-04 to OCHD-003 was 1:0.1, whereby the hole-injection layer **111** was formed.

Over the hole-injection layer **111**, mmtBumTPoFBi-04 was deposited by evaporation to a thickness of 50 nm as a first layer, 3-(dibenzothiophen-4-yl)-4'-phenyl-4"--(phenyl-9H-carbazol-2-yl)triphenylamine (abbreviation: PCB-BiPDDBt-02) represented by Structural Formula (xxi) was deposited by evaporation to a thickness of 50 nm as a second layer, and then N-(3',5',-di-tert-butyl-1,1'-biphenyl-4-yl)-bis(9,9-dimethyl-9H-fluoren)-2,2'-amine (abbreviation: mmtBuBiFF) was deposited by evaporation to a thickness of 50 nm as a third layer, whereby the hole-transport layer **112** was formed.

Then, 11-(4-[1,1'-diphenyl]-4-yl-6-phenyl-1,3,5-triazin-2-yl)-11,12-dihydro-12-phenyl-indolo[2,3-a]carbazole (abbreviation: BP-Icz(II)Tzn) represented by Structural Formula (xxiii), 9-(2-naphthyl)-9'-phenyl-9H,9'H-3,3'-bicarbazole (abbreviation: βNCCP) represented by Structural Formula (xxiv), and [2-d3-methyl-(2-pyridinyl-κN)benzofuro[2,3-b]pyridine-κC]bis[2-(2-pyridinyl-κN)phenyl-KC]iridium(III) (abbreviation: Ir(ppy)<sub>2</sub>(mbfpypy-d3)) represented by Structural Formula (xxv) were deposited to a thickness of 40 nm by co-evaporation such that the weight ratio of BP-Icz(II)Tzn to βNCCP and Ir(ppy)<sub>2</sub>(mbfpypy-d3) was 0.5:0.5:0.1, whereby the light-emitting layer **113** was formed.

After that, 2-[3'-(9,9-dimethyl-9H-fluoren-2-yl)-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mFBPTzn) represented by Structural Formula (iv) was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer was formed. Then, 2-[3-(2,6-dimethyl-3-pyridinyl)-5-(9-phenanthrenyl)phenyl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mPn-mDMePyPTzn) represented by Structural Formula (v) and 8-hydroxyquinolinato-lithium (abbreviation: Liq) represented by Structural Formula (vi) were deposited to a thickness of 25 nm by co-evaporation such that the weight ratio of mPn-mDMePyPTzn to Liq was 1:1, whereby the electron-transport layer **114** was formed.

After the electron-transport layer **114** was formed, lithium fluoride (LiF) was deposited to a thickness of 1 nm to form the electron-injection layer **115**, and lastly silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode **102**, whereby the light-emitting device **50** was fabricated. The second electrode **102** is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission device in which light is extracted through the second electrode **102**. Over the second electrode **102**, 4,4',4"--(benzene-1,3,5-triyl)tri(dibenzothiophene) (abbreviation: DBT3P-II) represented by Structural Formula (vii) was deposited by evaporation to a thickness of 70 nm so that light extraction efficiency can be improved.

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(Fabrication Method of Comparative Light-Emitting Device 50)

The comparative light-emitting device 50 was fabricated in a manner similar to that for the light-emitting device 50 except that mmtBumTPoFBi-04 and PCBBiPDBt-02 were replaced with each other.

The structures of the light-emitting device 50 and the comparative light-emitting device 50 are listed in the following table.

TABLE 14

	Film thickness	Light-emitting device 50	Comparative light-emitting device 50
Cap layer	70 nm	DBT3P-II	
Cathode	15 nm	Ag:Mg (10:1)	
Electron-injection layer	1 nm	LiF	
Electron-transport layer	25 nm	mPn-mDMePyPTzn:Liq (1:1)	
Hole-blocking layer	10 nm	mFBPTzn	
Light-emitting layer	40 nm	BP-Icz(II)Tzn:BNCCP:Ir(ppy) <sub>2</sub> (mbfpypy-d3) (0.5:0.5:0.1)	
Hole-transport layer	50 nm	mmtBuBiFF	
Anode	10 nm	PCBBiPDBt-02	mmtBumTPoFBi-04
Reflective electrode	50 nm	mmtBumTPoFBi-04	PCBBiPDBt-02
Hole-injection layer	10 nm	mmtBumTPoFBi-04;OCHD-003	PCBBiPDBt-02;OCHD-003
Anode	10 nm	ITSO	
Reflective electrode	100 nm	Ag	

FIGS. 62A and 62B show the measurement results of ordinary refractive indices of mmtBumTPoFBi-04, PCBBiPDBt-02, and mmtBuBiFF. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A. Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

The graphs indicate that in the light-emitting device 50, the second layer of the hole-transport layer has a high refractive index and the first and third layers have a low refractive index. In the comparative light-emitting device 50, the first layer of the hole-transport layer has a high refractive index and the second and third layers have a low refractive index.

The HOMO levels of mmtBumTPoFBi-04, PCBBiPDBt-02, and mmtBuBiFF that were calculated from cyclic voltammetry (CV) measurement results were -5.42 eV, -5.48 eV, and -5.33 eV, respectively, with N,N-dimethylformamide (DMF) used as a solvent. This indicates that the difference between the HOMO levels of mmtBumTPoFBi-04 and PCBBiPDBt-02 is less than or equal to 0.2 eV, the difference between the HOMO levels of PCBBiPDBt-02 and mmtBuBiFF is less than or equal to 0.2 eV, and the difference between the HOMO levels of mmtBumTPoFBi-04 and mmtBuBiFF is less than or equal to 0.2 eV. Note that the CV measurement was performed with an electrochemical analyzer (ALS model 600A or 600C manufactured by BAS Inc.) as a measurement apparatus, and the measurement was performed on a solution in which a material to be measured was dissolved in a solvent.

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GSP of each of mmtBumTPoFBi-04, PCBBiPDBt-02, and mmtBuBiFF is listed in the following table. The table shows that the light-emitting device 50 is a device fabricated in consideration of GSP, in which the third layer has the largest GSP and the first layer has the smallest GSP. In contrast, the comparative light-emitting device 50 is a device in which the third layer has the largest GSP and the value obtained by subtracting GSP of the second layer from GSP of the first layer is greater than or equal to 10 (mV/nm).

TABLE 15

	mmtBumTPoFBi-04	PCBBiPDBt-02	mmtBuBiFF
GSP (mV/nm)	16.7	28.3	39.5

The light-emitting devices were sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the elements, only the UV curable sealing material was irradiated with UV while the light-emitting devices were prevented from being irradiated with the UV, and heat treatment was performed at 80° C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting devices were measured.

FIG. 56 shows the luminance-current density characteristics of the light-emitting device 50 and the comparative light-emitting device 50. FIG. 57 shows the luminance-voltage characteristics thereof. FIG. 58 shows the current efficiency-luminance characteristics thereof. FIG. 59 shows the current density-voltage characteristics thereof. FIG. 60 shows the power efficiency-luminance characteristics thereof. FIG. 61 shows the emission spectrum thereof. Table 16 shows the main characteristics of the light-emitting device 50 and the comparative light-emitting device 50 at a luminance of about 1000 cd/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectra were measured at normal temperature with a spectroradiometer (SR-UL1R manufactured by TOPCON TECHNOHOUSE CORPORATION). The power efficiency is calculated under assumption of Lambertian.

TABLE 16

	Voltage (V)	Current (mA)	Current density (mA/cm <sup>2</sup> )	Chromaticity x	Chromaticity y	Current efficiency (cd/A)	Power efficiency (lm/W)
Light-emitting device 50	2.6	0.03	0.7	0.22	0.73	185	223
Comparative light-emitting device 50	3.5	0.02	0.5	0.25	0.72	173	155

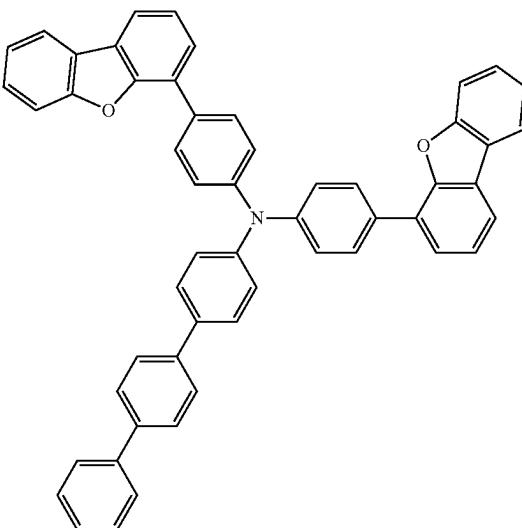
FIGS. 56 to 61 and Table 16 show that the light-emitting device 50 of one embodiment of the present invention and the comparative light-emitting device 50 have favorable characteristics. In particular, the light-emitting device 50 having the structure of the present invention includes the hole-transport layer consisting of the first to third layers as described in Embodiment 1, and thus has high current efficiency. Furthermore, the first to third layers in the light-emitting device 50 have a GSP relationship of the first layer<the second layer<the third layer, achieving a light-emitting device with a low driving voltage and an extremely high power efficiency.

## Example 7

In this example, a light-emitting device 60 of one embodiment of the present invention is described. Note that the light-emitting device 60 is a tandem light-emitting device including a plurality of light-emitting layers with a charge-generation layer therebetween. The Structural formulae of organic compounds used in this example are shown below.

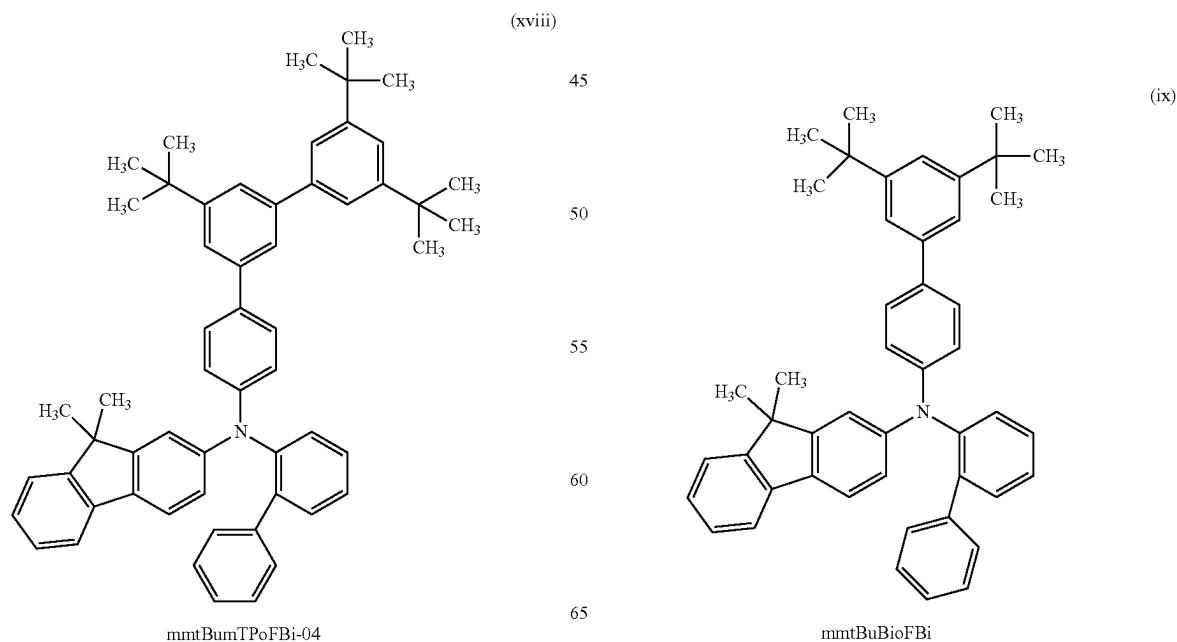
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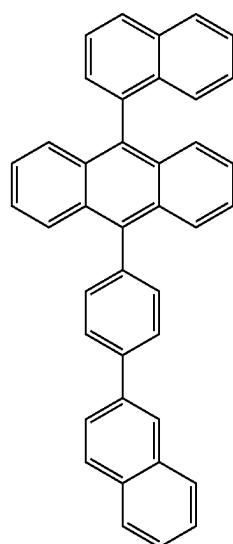
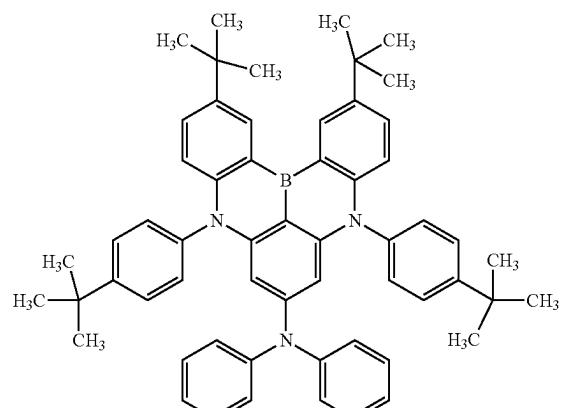


DBfBB1TP

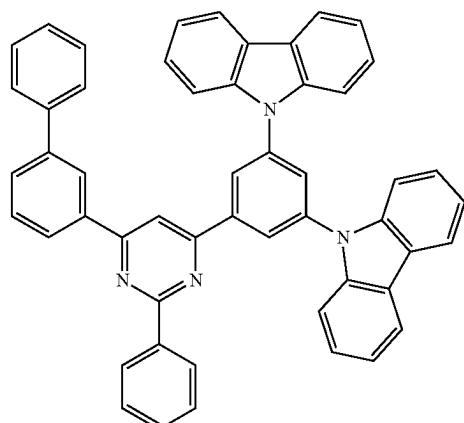
[Chemical Formula 20]



**113**  
-continued

 $\alpha$ N- $\beta$ NPAanth

DPhA-tBu4DABNA



6mBP-4Cz2PPm

(xii)

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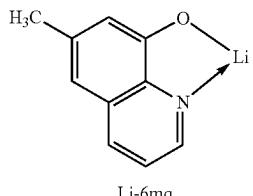
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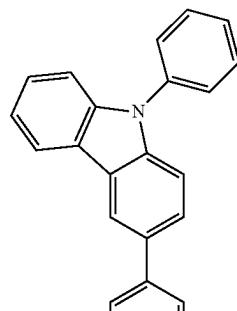
**114**  
-continued



Li-6mq

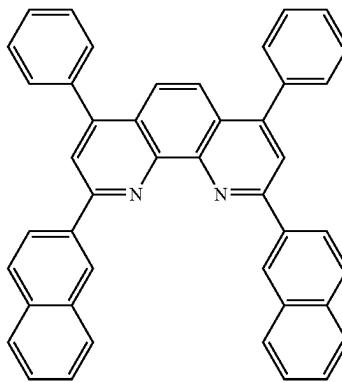
(xx)

(xxvii)



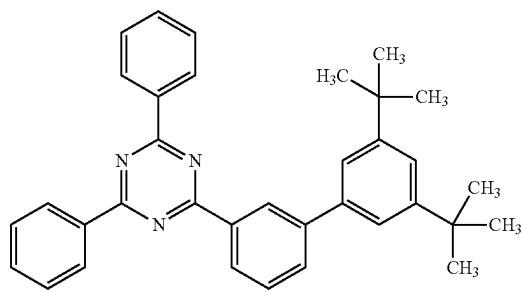
PCBBiF

(xv)



NBPhen

(xix)

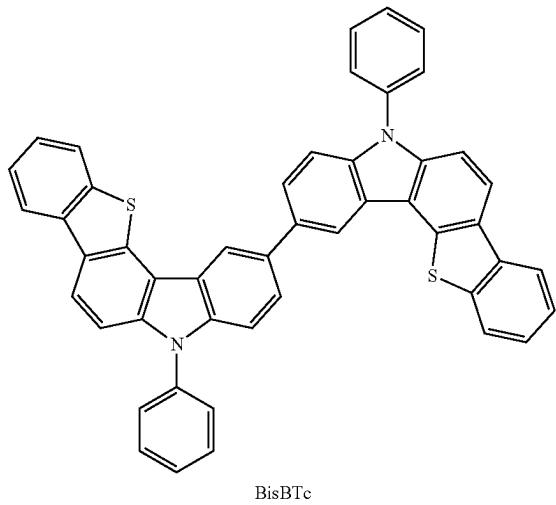


mmtBumBPTzn

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-continued

(xxvi)



## (Fabrication Method of Light-Emitting Device 60)

First, as a reflective electrode, silver (Ag) was deposited over a glass substrate to a thickness of 100 nm by a sputtering method, and then, as a transparent electrode, indium tin oxide containing silicon oxide (ITSO) was deposited to a thickness of 5 nm by a sputtering method, whereby the first electrode 101 was formed. The electrode area was set to 4 mm<sup>2</sup> (2 mm×2 mm). Note that the first electrode 101 is a transparent electrode and the transparent electrode and the reflective electrode can be collectively regarded as the first electrode 101.

Next, in pretreatment for forming the light-emitting device over the substrate, a surface of the substrate was washed with water and baked at 200°C. for 1 hour, and then UV ozone treatment was performed for 370 seconds.

After that, the substrate was transferred into a vacuum evaporation apparatus where the pressure was reduced to approximately 10<sup>-4</sup> Pa, vacuum baking was performed at 170°C. for 30 minutes in a heating chamber of the vacuum evaporation apparatus, and then the substrate was cooled down for approximately 30 minutes.

Next, the substrate provided with the first electrode 101 was fixed to a substrate holder provided in the vacuum evaporation apparatus such that the surface on which the first electrode 101 was formed faced downward. Then, N-(3",5",5"-tri-tert-butyl-1,1":3",1"-terphenyl-4-yl)-N-(1,1'-biphenyl-2-yl)-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmtBumTPoFBi-04) represented by Structural Formula (xviii) and a fluorine-containing electron acceptor material with a molecular weight of 672 (OCHD-003) were deposited on the first electrode 101 to a thickness of 10 nm by co-evaporation such that the weight ratio of mmtBumTPoFBi-04 to OCHD-003 was 1:0.1, whereby a hole-injection layer was formed.

Over the hole-injection layer, mmtBumTPoFBi-04 was deposited by evaporation to a thickness of 35 nm as a first layer, N,N-bis[4-(dibenzofuran-4-yl)phenyl]-4-amino-p-terphenyl (abbreviation: DBfBB1TP) represented by Structural Formula (xi) was deposited by evaporation to a thickness of 50 nm as a second layer, and then N-3",5"-ditertiarybutyl-1,1'-biphenyl-4-yl-N-1,1'-biphenyl-2-yl-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmtBuBioFBi) was deposited by evaporation to a thickness of 40 nm as a third layer, whereby a hole-transport layer A was formed.

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After that, over the hole-transport layer A, DBfBB1TP was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer A was formed.

Then, 9-(1-naphthyl)-10-[4-(2-naphthyl)phenyl]anthracene (abbreviation:  $\alpha$ N- $\beta$ NPAanth) represented by Structural Formula (xii) and 2,12-di(tert-butyl)-5,9-di(4-tert-butylphenyl)-N,N-diphenyl-5H,9H-[1,4]benzazaborino[2,3,4-kl]phenazaborin-7-amine (abbreviation: DPhA-tBu4DABNA) represented by Structural Formula (xiii) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of  $\alpha$ N- $\beta$ NPAanth to DPhA-tBu4DABNA was 1:0.015, whereby a light-emitting layer A was formed.

After that, 6-(1,1'-biphenyl-3-yl)-4-[3,5-bis(9H-carbazol-9-yl)phenyl]-2-phenylpyrimidine (abbreviation: 6mBP-4Cz2PPm) represented by Structural Formula (xiv) was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer A was formed. Then, 2,9-di(2-naphthyl)-4,7-diphenyl-1,10-phenanthroline (abbreviation: NBPhen) represented by Structural Formula (xv) was deposited by evaporation to a thickness of 20 nm, whereby an electron-transport layer A was formed.

After the electron-transport layer A was formed, lithium oxide (Li<sub>2</sub>O) was deposited by evaporation to a thickness of 0.1 nm, and N-(1,1'-biphenyl-4-yl)-N-[4-(9-phenyl-9H-carbazol-3-yl)phenyl]-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: PCBBiF) represented by Structural Formula (xxvii) and OCHD-003 were deposited to a thickness of 10 nm by co-evaporation such that the weight ratio of PCBBiF to OCHD-003 was 1:0.1, whereby a charge-generation layer was formed.

Over the charge-generation layer, PCBBiF was deposited by evaporation to a thickness of 40 nm, whereby a hole-transport layer B was formed.

After that, over the hole-transport layer B, DBfBB1TP was deposited by evaporation to a thickness of 10 nm, whereby an electron-blocking layer B was formed.

Then,  $\alpha$ N- $\beta$ NPAanth and DPhA-tBu4DABNA were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of  $\alpha$ N- $\beta$ NPAanth to DPhA-tBu4DABNA was 1:0.015, whereby a light-emitting layer B was formed.

After that, 6mBP-4Cz2PPm was deposited by evaporation to a thickness of 10 nm, whereby a hole-blocking layer B was formed. Then, 2-[{3',5'-di-tert-butyl}-1,1'-biphenyl-3-yl]-4,6-diphenyl-1,3,5-triazine (abbreviation: mmtBumBPTzn) represented by Structural Formula (xix) and 6-methyl-8-quinolinolato-lithium (abbreviation: Li-6mq) represented by Structural Formula (xx) were deposited to a thickness of 20 nm by co-evaporation such that the weight ratio of mmtBumBPTzn to Li-6mq was 1:1, whereby an electron-transport layer B was formed.

After the electron-transport layer B was formed, lithium fluoride (LiF) was deposited to a thickness of 2 nm to form the electron-injection layer 115, and lastly silver (Ag) and magnesium (Mg) were deposited to a thickness of 15 nm by co-evaporation such that the volume ratio of Ag to Mg was 1:0.1 to form the second electrode 102, whereby the light-emitting device 60 was fabricated.

The second electrode 102 is a transreflective electrode having a function of reflecting light and a function of transmitting light; thus, the light-emitting device of this example is a top emission tandem device in which light is extracted through the second electrode 102. Over the second electrode 102, 5,5'-diphenyl-2,2'-di-5H-[1]benzothieno[3,2-c]carbazole (abbreviation: BisBTc) represented by Struc-

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tural Formula (xxvi) was deposited by evaporation to a thickness of 65 nm as a cap layer so that light extraction efficiency can be improved.

The structures of the light-emitting device **60** are listed in the following table.

TABLE 17

	Film thickness	Light-emitting device <b>60</b>
Cap layer	65 nm	BisBTc
Cathode	15 nm	Ag:Mg (10:1)
Electron-injection layer	2 nm	LiF
Electron-transport layer B	20 nm	mmtBumBPTzn:Li-6mq (1:1)
Hole-blocking layer B	10 nm	6mBP-4CzP2Pm
Light-emitting layer B	20 nm	$\alpha$ N- $\beta$ NPAanth:DhA-tBu4DABNA (1:0.015)
Electron-blocking layer B	10 nm	DBfBB1TP
Hole-transport layer B	40 nm	PCBBiF
Charge-generation layer	10 nm	PCBBiF: OCHD-003 (1:0.1)
Electron-transport layer A	0.1 nm	Li <sub>2</sub> O
Hole-blocking layer A	20 nm	NBPhen
Light-emitting layer A	10 nm	6mBP-4CzP2Pm
Electron-blocking layer A	20 nm	$\alpha$ N- $\beta$ NPAanth:DhA-tBu4DABNA (1:0.015)
Hole-transport layer A	3	40 nm
	2	50 nm
	1	35 nm
Hole-injection layer	10 nm	mmtBumTPoFBi-04 mmtBumTPoFBi-04:OCHD-003 (1:0.1)
Anode	5 nm	ITSO
Reflective electrode	100 nm	Ag

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FIG. 65 shows the emission spectrum of DPhA-tBu4DABNA as a light-emitting substance in a toluene solution. According to FIG. 65, the half-width of the emission spectrum of DPhA-tBu4DABNA is 22 nm, the spectral width at 1/e of the maximum intensity is 29 nm, and the values obtained by converting these widths into energy are respectively 0.13 eV and 0.17 eV, which means that DPhA-tBu4DABNA is a light-emitting substance that emits light with a narrow spectral width.

The light-emitting device **60** was sealed using a glass substrate in a glove box containing a nitrogen atmosphere so as not to be exposed to the air. Specifically, a UV curable sealing material was applied to surround the element, only the UV curable sealing material was irradiated with UV while the light-emitting device was prevented from being irradiated with the UV, and heat treatment was performed at 80° C. under an atmospheric pressure for one hour. Then, the initial characteristics of the light-emitting device were measured.

FIG. 66 shows the luminance-current density characteristics of the light-emitting device **60**. FIG. 67 shows the current efficiency-luminance characteristics thereof. FIG. 68 shows the luminance-voltage characteristics thereof. FIG. 69 shows the current density-voltage characteristics thereof. FIG. 70 shows the blue index-luminance characteristics thereof. FIG. 71 shows the emission spectrum thereof. Table 18 shows the main characteristics of the light-emitting device **60** at a current density of about 10 mA/m<sup>2</sup>. Luminance, CIE chromaticity, and emission spectrum were measured at normal temperature with a spectroradiometer (SR-ULIR manufactured by TOPCON TECHNOHOUSE CORPORATION).

TABLE 18

	Voltage (V)	Current (mA)	Current density (mA/cm <sup>2</sup> )	Chromaticity x	Chromaticity y	Current efficiency (cd/A)	BI (cd/A/y)	BI MAX (cd/A/y)
Light-emitting device <b>60</b>	7.8	0.31	7.7	0.15	0.03	13.2	458	460

FIGS. 63 and 64 show the measurement results of refractive indices of mmtBumTPoFBi-04, DBfBB1TP, mmtBuBioFBi, mmtBumBPTzn, Li-6mq, and BisBTc. The measurement was performed with a spectroscopic ellipsometer (M-2000U, produced by J.A Woollam Japan Corp.). As a sample used for the measurement, a film was formed to a thickness of approximately 50 nm with the material of each layer over a quartz substrate by a vacuum evaporation method.

FIGS. 63 and 64 indicate that mmtBumTPoFBi-04, mmtBuBioFBi, mmtBumBPTzn, and Li-6mq are low refractive index materials having an ordinary refractive index greater than or equal to 1.50 and less than or equal to 1.75 in the whole wavelength range of 455 nm to 465 nm, and DBfBB1TP and BisBTc are high refractive index materials having an ordinary refractive index greater than or equal to 1.90 and less than or equal to 2.40 in the whole wavelength range of 455 nm to 465 nm. Note that the light-emitting device **60** emits blue light.

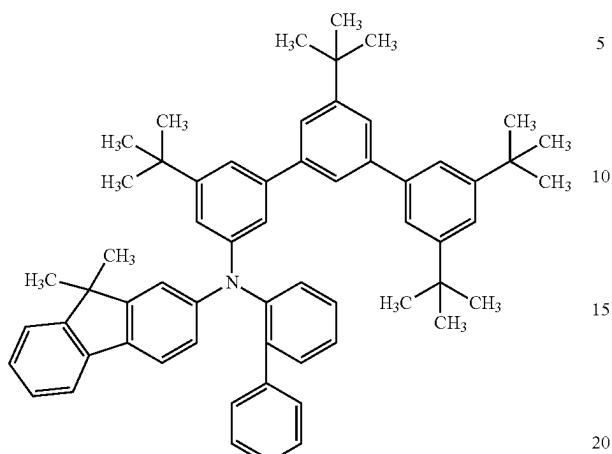
FIGS. 66 to 71 and Table 18 show that the light-emitting device **60** has an extremely high BI of 458 (cd/A/y) or more, and the maximum BI thereof is 460 (cd/A/y). Thus, one embodiment of the present invention is particularly suitable for a light-emitting device used in a display.

## Reference Synthesis Example 1

Described in this synthesis example is a method for synthesizing N-(1,1'-biphenyl-2-yl)-N-(3,3",5"-tetra-tert-butyl-1,1':3',1"-terphenyl-5-yl)-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmtBumTPoFBi-02) used in Example 1. The structure of mmtBumTPoFBi-02 is shown below.

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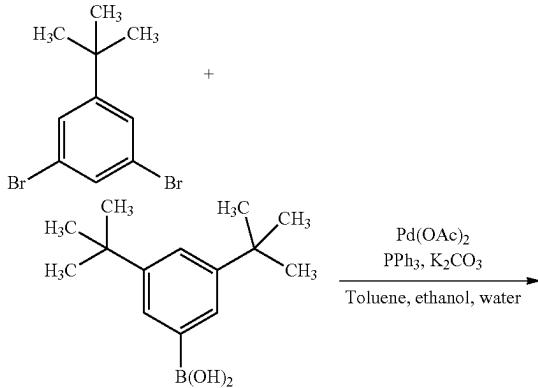
[Chemical Formula 21]



### Step 1: Synthesis of 3-bromo-3',5,5'-tri-tert-butylbiphenyl

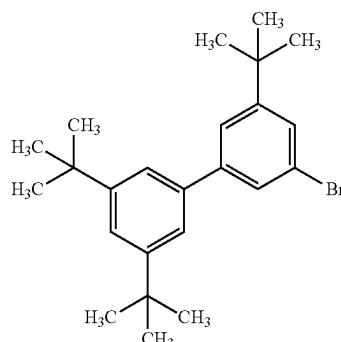
In a three-neck flask were put 37.2 g (128 mmol) of 1,3-dibromo-5-tert-butylbenzene, 20.0 g (85 mmol) of 3,5-di-tert-butylphenylboronic acid, 35.0 g (255 mmol) of potassium carbonate, 570 mL of toluene, 170 mL of ethanol, and 130 mL of tap water. The mixture was degassed under reduced pressure, and then the air in the flask was replaced with nitrogen. Then, 382 mg (1.7 mmol) of palladium acetate (abbreviation: Pd(OAc)<sub>2</sub>) and 901 mg (3.4 mmol) of triphenylphosphine were added, and the mixture was heated at 40° C. for approximately 5 hours. After that, the temperature of the flask was lowered to room temperature, and the mixture was separated into an organic layer and an aqueous layer. Magnesium sulfate was added to this organic layer to eliminate moisture, whereby the organic layer was concentrated. The obtained solution was purified by silica gel column chromatography, whereby 21.5 g of a target colorless oily substance was obtained in a yield of 63%. The synthesis scheme of Step 1 is shown below.

[Chemical Formula 22]



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-continued

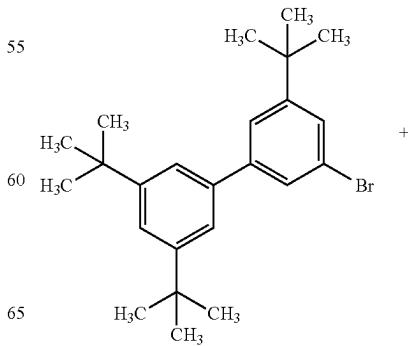


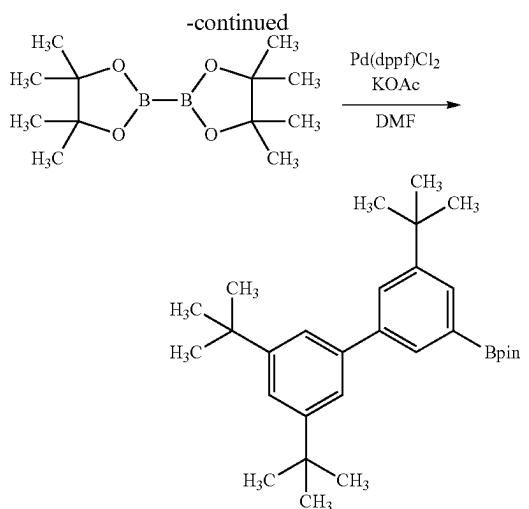
Step 2: Synthesis of 2-(3',5',5'-tri-tert-butyl[1,1'-biphenyl]-3-yl)-4,4,5,5-tetramethyl-1,3,2-dioxaborolane

In a three-neck flask were put 15.0 g (38 mmol) of 3-bromo-3',5,5'-tri-tert-butylbiphenyl obtained in Step 1, 25 10.5 g (41 mmol) of 4,4,4',4',5,5,5',5'-octamethyl-2,2'-bi-1, 3,2-dioxaborolane, 11.0 g (113 mmol) of potassium acetate, and 125 mL of N,N-dimethylformamide (abbreviation: DMF). The mixture was degassed under reduced pressure, the air in the flask was replaced with nitrogen, 1.5 g (1.9 30 mmol) of [1,1'-bis(diphenylphosphino)ferrocene]dichloropalladium(II) (abbreviation: Pd(dppf)Cl<sub>2</sub>) was added thereto, and the mixture was heated at 100° C. for approximately 3 hours. Then, the temperature of the flask was 35 lowered to room temperature, the mixture was separated into an organic layer and an aqueous layer, and extraction was performed with ethyl acetate. Magnesium sulfate was added to the solution of the extract to eliminate moisture, whereby the solution of the extract was concentrated. A toluene 40 solution of the obtained mixture was purified by silica gel column chromatography, and the resulting solution was concentrated to give a condensed toluene solution. Ethanol was added to this toluene solution and the toluene solution was concentrated under reduced pressure, whereby an ethanol 45 suspension was obtained. The precipitate was filtrated at approximately 20° C., and the obtained solid was dried at approximately 80° C. under reduced pressure, whereby 13.6 g of a target white solid was obtained in a yield of 81%. The synthesis scheme of Step 2 is shown below.

50

[Chemical Formula 23]

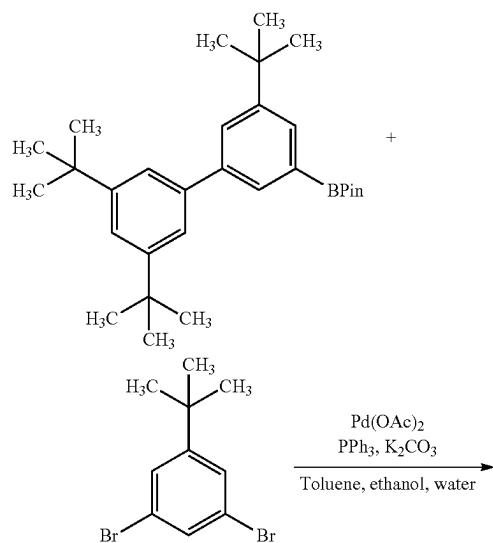
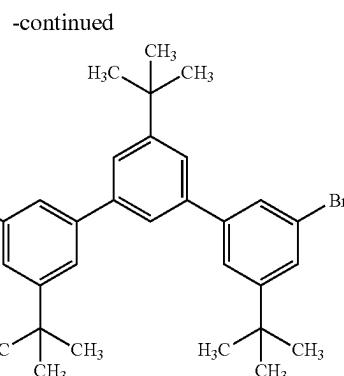


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**Step 3: Synthesis of 3-bromo-3",5,5',5"-tetra-tert-butyl-1,1':3',1"-terphenyl**

In a three-neck flask were put 5.0 g (11.1 mmol) of 2-(3',5,5'-tri-tert-butyl[1,1'-biphenyl]-3-yl)-4,4,5,5-tetramethyl-1,3,2-dioxaborolane, 4.8 g (16.7 mmol) of 1,3-dibromo-5-tert-butylbenzene, 4.6 g (33.3 mmol) of potassium carbonate, 56 mL of toluene, 22 mL of ethanol, and 17 mL of tap water. The mixture was degassed under reduced pressure, and then the air in the flask was replaced with nitrogen. Then, 50 mg (0.22 mmol) of palladium acetate and 116 mg (0.44 mmol) of triphenylphosphine were added, and the mixture was heated at 80° C. for approximately 10 hours. After that, the temperature of the flask was lowered to room temperature, and the mixture was separated into an organic layer and an aqueous layer. Magnesium sulfate was added to this solution to eliminate moisture, whereby this solution was concentrated. The obtained hexane solution was purified by silica gel column chromatography, whereby 3.0 g of a target white solid was obtained in a yield of 51.0%. The synthesis scheme of 3-bromo-3",5,5',5"-tetra-tert-butyl-1,1':3',1"-terphenyl of Step 3 is shown below.

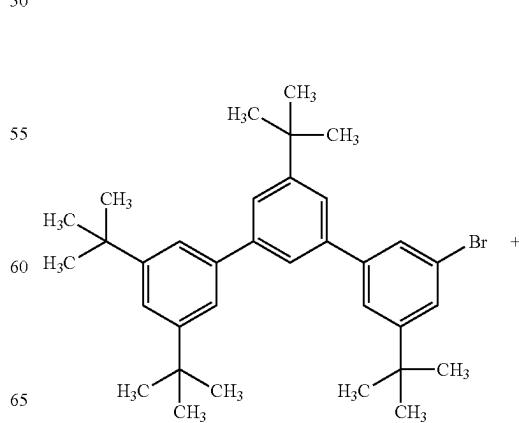
[Chemical Formula 24]

**122**

**Step 4: Synthesis of mmmtBumTPoFBi-02**

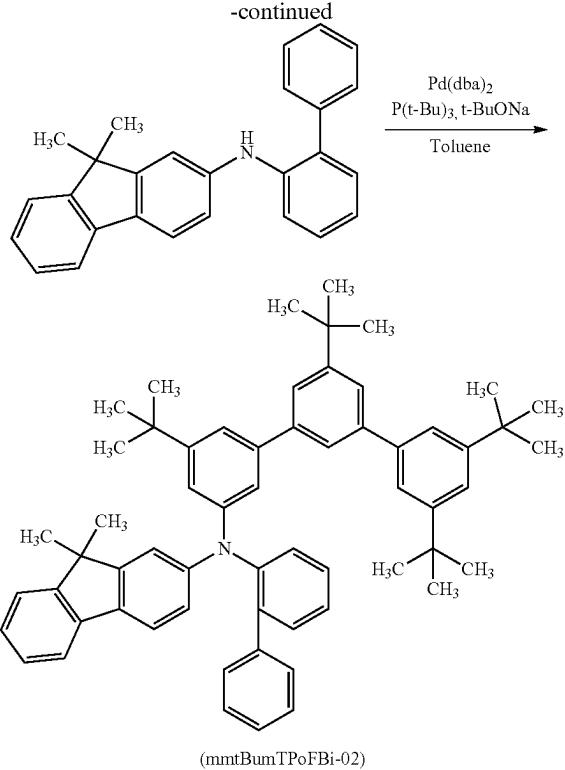
In a three-neck flask were put 5.8 g (10.9 mmol) of 3-bromo-3",5,5',5"-tetra-tert-butyl-1,1':3',1"-terphenyl obtained in Step 3, 3.9 g (10.9 mmol) of N-(1,1'-biphenyl-4-yl)-N-phenyl-9,9-dimethyl-9H-fluoren-2-amine, 3.1 g (32.7 mmol) of sodium-tert-butoxide, and 55 mL of toluene. The mixture was degassed under reduced pressure, the air in the flask was replaced with nitrogen, 64 mg (0.11 mmol) of bis(dibenzylideneacetone)palladium(0)(abbreviation: Pd(dba)2) and 132 mg (0.65 mmol) of tri-tert-butylphosphine were added thereto, and the mixture was heated at 80° C. for approximately 2 hours. After that, the temperature of the flask was lowered to approximately 60° C., approximately 1 mL of water was added, a precipitated solid was separated by filtration, and the solid was washed with toluene. The filtrate was concentrated, and the obtained toluene solution was purified by silica gel column chromatography. The obtained solution was concentrated to give a condensed toluene solution. Ethanol was added to this toluene solution and the toluene solution was concentrated under reduced pressure, whereby an ethanol suspension was obtained. The precipitate was filtrated at approximately 20° C., and the obtained solid was dried at approximately 80° C. under reduced pressure, whereby 8.1 g of a target white solid was obtained in a yield of 91%. The synthesis scheme of mmmtBumTPoFBi-02 is shown below.

[Chemical Formula 25]



**123**

-continued



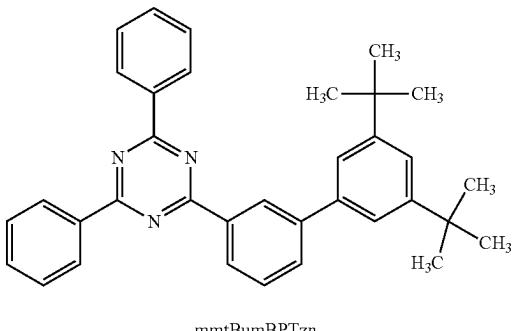
Analysis results by nuclear magnetic resonance ( $^1\text{H}$ -NMR) spectroscopy of the white solid obtained in the above step are shown below. The results show that mmTBumTPoFBi-02 was synthesized in this synthesis example.

$^1\text{H-NMR}$ .  $\delta$  (CDCl<sub>3</sub>): 7.56 (d, 1H, J=7.4 Hz), 7.50 (dd, 1H, J=1.7 Hz), 7.33-7.46 (m, 11H), 7.27-7.29 (m, 2H), 7.22 (dd, 1H, J=2.3 Hz), 7.15 (d, 1H, J=6.9 Hz), 6.98-7.07 (m, 7H), 6.93 (s, 1H), 6.84 (d, 1H, J=6.3 Hz), 1.38 (s, 9H), 1.37 (s, 18H), 1.31 (s, 6H), 1.20 (s, 9H).

#### Reference Synthesis Example 2

Described in this synthesis example is a method for synthesizing 2-{(3',5'-di-tert-butyl)-1,1'-biphenyl-3-yl}-4,6-diphenyl-1,3,5-triazine (abbreviation: mmTBumBPTzn) used in Example 1. The structure of mmTBumBPTzn is shown below.

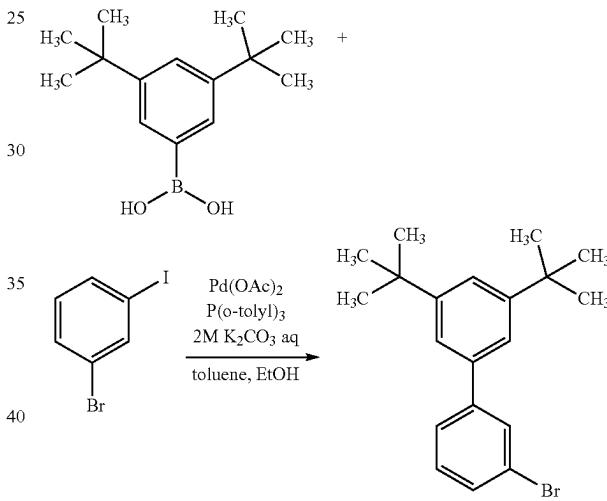
[Chemical Formula 26]

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#### Step 1: Synthesis of 3-bromo-3',5'-di-tert-butylbiphenyl

First, 1.0 g (4.3 mmol) of 3,5-di-tert-butylphenylboronic acid, 1.5 g (5.2 mmol) of 1-bromo-3-iodobenzene, 4.5 mL of 2 mol/L aqueous solution of potassium carbonate, 20 mL of toluene, and 3 mL of ethanol were put into a three-neck flask and stirred under reduced pressure to be degassed. Furthermore, 52 mg (0.17 mmol) of tris(2-methylphenyl)phosphine (abbreviation: P(o-tolyl)<sub>3</sub>) and 10 mg (0.043 mmol) of palladium(II) acetate were added to this mixture, and reacted under a nitrogen atmosphere at 80°C. for 14 hours. After the reaction, extraction with toluene was performed and the resulting organic layer was dried with magnesium sulfate. This mixture was subjected to gravity filtration and the filtrate was purified by silica gel column chromatography (the developing solvent: hexane) to give 1.0 g of a target white solid (yield: 68%). The synthesis scheme of Step 1 is shown below.

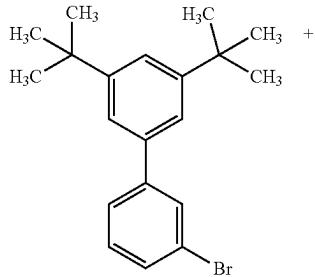
[Chemical Formula 27]



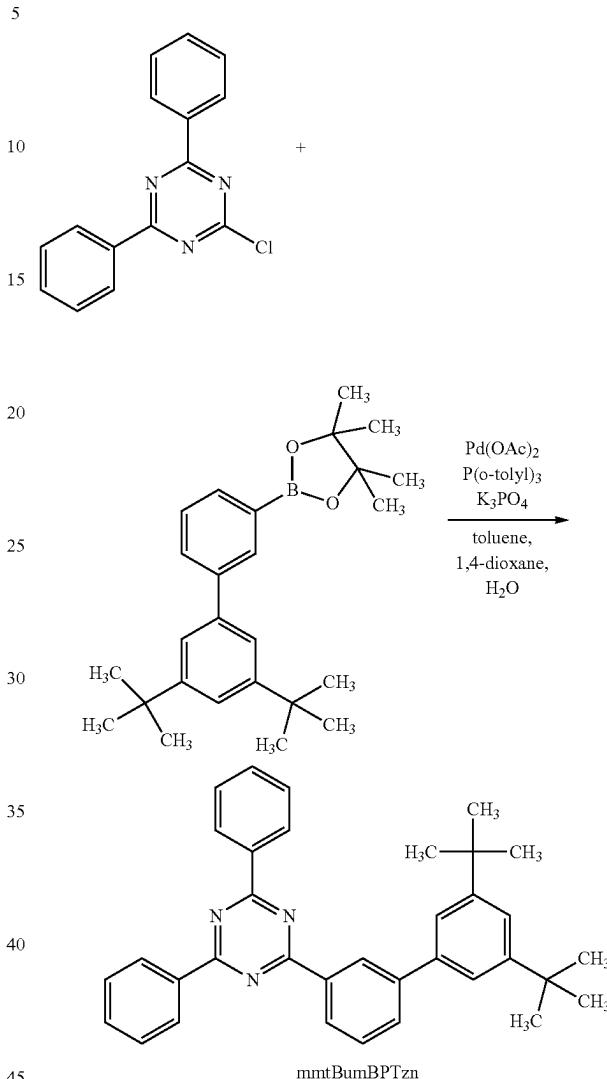
#### Step 2: Synthesis of 2-(3',5'-di-tert-butylbiphenyl-3-yl)-4,4,5,5-tetramethyl-1,3,2-dioxaborolane

First, 1.0 g (2.9 mmol) of 3-bromo-3',5'-di-tert-butylbiphenyl, 0.96 g (3.8 mmol) of bis(pinacolato)diboron, 0.94 g (9.6 mmol) of potassium acetate, and 30 mL of 1,4-dioxane were put into a three-neck flask and stirred under reduced pressure to be degassed. Furthermore, 0.12 g (0.30 mmol) of 2-dicyclohexylphosphino-2',6'-dimethoxybiphenyl and 0.12 g (0.15 mmol) of [1,1'-bis(diphenylphosphino)ferrocene] palladium(II) dichloride dichloromethane adduct (abbreviation: Pd(dppf)<sub>2</sub>Cl<sub>2</sub>·CH<sub>2</sub>Cl<sub>2</sub>) were added to this mixture, and reacted under a nitrogen atmosphere at 110°C. for 24 hours. After the reaction, extraction with toluene was performed and the resulting organic layer was dried with magnesium sulfate. This mixture was subjected to gravity filtration and the filtrate was purified by silica gel column chromatography (the developing solvent: toluene) to give 0.89 g of a target yellow oil (yield: 78%). The synthesis scheme of Step 2 is shown below.

[Chemical Formula 28]



[Chemical Formula 29]



## Step 3: Synthesis of mmmtBumBPTzn

First, 1.5 g (5.6 mmol) of 4,6-diphenyl-2-chloro-1,3,5-triazine, 2.4 g (6.2 mmol) of 2-(3',5'-di-tert-butylphenyl)-3-yI)-4,4,5,5-tetramethyl-1,3,2-dioxaborolane, 2.4 g (11 mmol) of tripotassium phosphate, 10 mL of water, 28 mL of toluene, and 10 mL of 1,4-dioxane were put into a three-neck flask and stirred under reduced pressure to be degassed. Furthermore, 13 mg (0.056 mmol) of palladium(II) acetate and 34 mg (0.11 mmol) of tris(2-methylphenyl)phosphine were added to this mixture, and heated and refluxed under a nitrogen atmosphere for 14 hours. After the reaction, extraction with ethyl acetate was performed and the resulting organic layer was removed with magnesium sulfate. This mixture was subjected to gravity filtration and the filtrate was purified by silica gel column chromatography (the developing solvent, chloroform:hexane=1:5 changed to 1:3). The obtained solid was recrystallized with hexane to give 2.0 g of a target white solid (yield: 51%). The synthesis scheme of Step 3 is shown below.

By a train sublimation method, 2.0 g of the obtained white solid was purified by sublimation under an argon gas stream at a pressure of 3.4 Pa and a temperature of 220°C. The solid was heated. After the sublimation purification, 1.8 g of a target white solid was obtained at a collection rate of 80%.

Analysis results by nuclear magnetic resonance spectroscopy ( $^1\text{H-NMR}$ ) of the white solid obtained in Step 3 are shown below. The results reveal that mmmtBumBPTzn was obtained in this example.

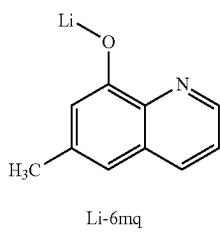
$^1\text{H NMR}$  ( $\text{CDCl}_3$ , 300 MHz):  $\delta = 1.44$  (s, 18H), 7.51-7.68 (m, 10H), 7.83 (d, 1H), 8.73-8.81 (m, 5H), 9.01 (s, 1H).

## Reference Synthesis Example 3

Described in this example is a method for synthesizing 6-methyl-8-quinolinolato-lithium (abbreviation: Li-6mq) used in Example 1. The structural formula of Li-6mq is shown below.

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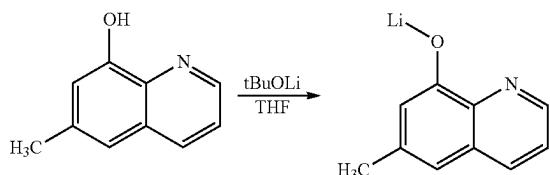
[Chemical Formula 30]



Li-6mq

First, 2.0 g (12.6 mmol) of 8-hydroxy-6-methylquinoline and 130 mL of dehydrated tetrahydrofuran (abbreviation: THF) were put into a three-neck flask and stirred. Then, 10.1 mL (10.1 mmol) of 1M THF solution of lithium tert-butoxide (abbreviation: tBuOLi) was added to this solution and stirred at room temperature for 47 hours. The reacted solution was concentrated to give a yellow solid. Acetonitrile was added to this solid and subjected to ultrasonic irradiation and filtration, so that a pale yellow solid was obtained. This washing step was performed twice. The obtained residue was 1.6 g of pale yellow solid of Li-6mq (yield: 95%). This synthesis scheme is shown below.

[Chemical Formula 31]



Next, the absorption and emission spectra of Li-6mq in a dehydrated acetone solution were measured. The absorption spectrum was measured with an ultraviolet-visible light spectrophotometer (V550, manufactured by JASCO Corporation), and the spectrum of dehydrated acetone alone in a quartz cell was subtracted. The emission spectrum was measured with a fluorescence spectrophotometer (FP-8600, produced by JASCO Corporation).

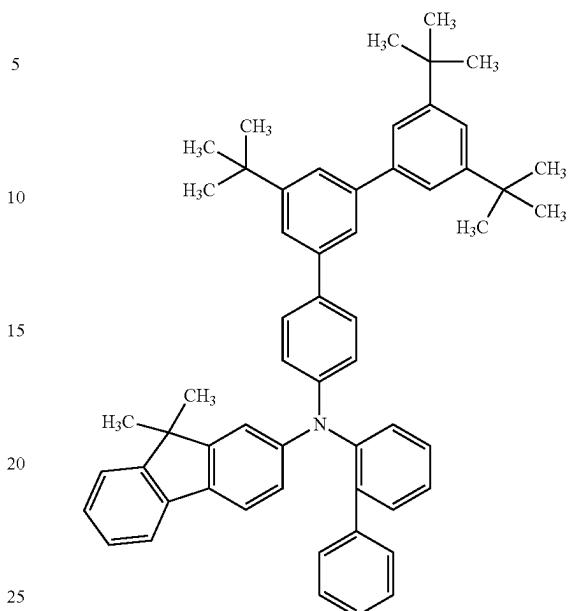
As a result, Li-6mq in the dehydrated acetone solution has an absorption peak at 390 nm, and an emission wavelength peak at 540 nm (excitation wavelength: 385 nm).

## Reference Synthesis Example 4

Described in this example is a method for synthesizing N-(3",5",5"-tri-tert-butyl-1,1': 3',1"-terphenyl-4-yl)-N-(1,1'-biphenyl-2-yl)-9,9-dimethyl-9H-fluoren-2-amine (abbreviation: mmTBuTPoFBi-04). The structure of mmTBuTPoFBi-04 is shown below.

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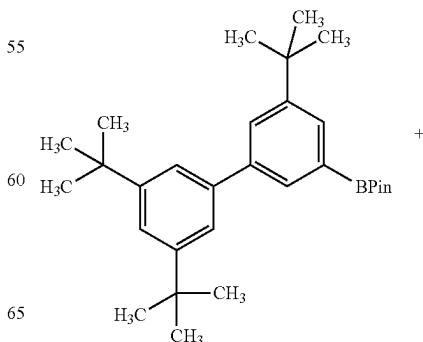
[Chemical Formula 32]



Step 1: Synthesis of 4-bromo-3",5',5"-tri-tert-butyl-1,1':3',1"-terphenyl

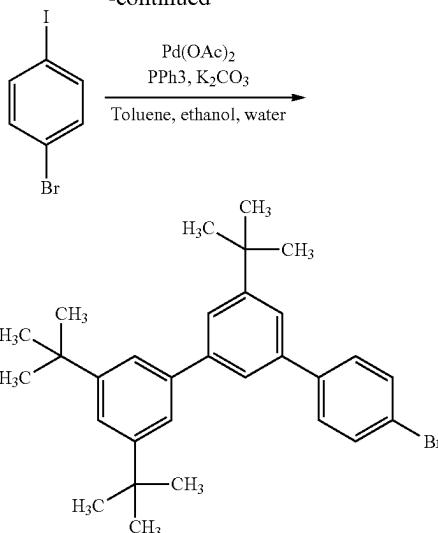
In a three-neck flask were put 9.0 g (20.1 mmol) of 2-(3',5,5'-tri-tert-butyl[1,1'-biphenyl]-3-yl)-4,4,5,5-tetramethyl-1,3,2-dioxaborolane, 6.8 g (24.1 mmol) of 1-bromo-4-iodobenzene, 8.3 g (60.3 mmol) of potassium carbonate, 100 mL of toluene, 40 mL of ethanol, and 30 mL of tap water. The mixture was degassed under reduced pressure, and then the air in the flask was replaced with nitrogen. Then, 91 mg (0.40 mmol) of palladium acetate and 211 mg (0.80 mmol) of triphenylphosphine were added, and the mixture was heated at 80° C. for approximately 4 hours. After that, the temperature of the flask was lowered to room temperature, and the mixture was separated into an organic layer and an aqueous layer. Magnesium sulfate was added to this solution to eliminate moisture, whereby this solution was concentrated. The obtained hexane solution was purified by silica gel column chromatography, whereby 6.0 g of a target white solid was obtained in a yield of 62.5%. The synthesis scheme of Step 1 is shown below.

[Chemical Formula 33]

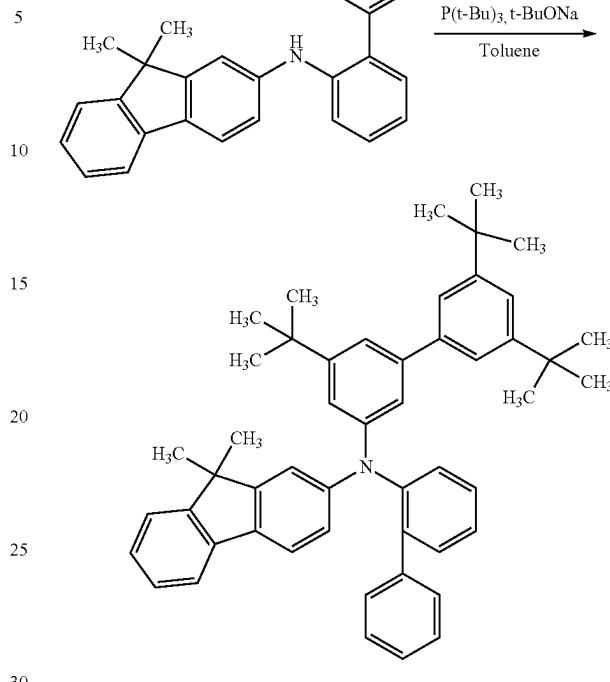


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-continued

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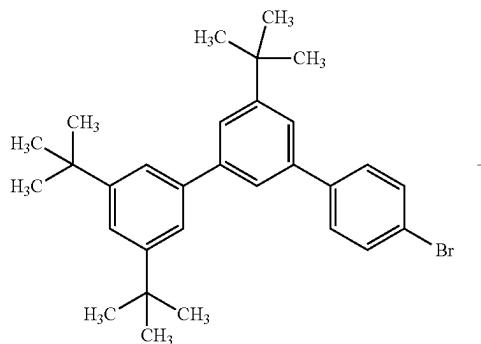
-continued



## Step 2: Synthesis of mmTBumTPoFBi-04

In a three-neck flask were put 3.0 g (6.3 mmol) of 4-bromo-3", 5", 5"-tri-tert-butyl-1,1":3", 1"-terphenyl, 2.3 g (6.3 mmol) of N-(1,1'-biphenyl-4-yl)-N-phenyl-9,9-dimethyl-9H-fluoren-2-amine, 1.8 g (18.9 mmol) of sodium-tert-butoxide, and 32 mL of toluene. The mixture was degassed under reduced pressure, the air in the flask was replaced with nitrogen, 72 mg (0.13 mmol) of bis(dibenzylideneacetone)palladium(0) and 76 mg (0.38 mmol) of tri-tert-butylphosphine were added thereto, and the mixture was heated at 120° C. for approximately 8 hours. After that, the temperature of the mixture was lowered to approximately 60° C., approximately 1 mL of water was added, a precipitated solid was separated by filtration, and the solid was washed with toluene. The filtrate was concentrated, and the obtained toluene solution was purified by silica gel column chromatography. The obtained solution was concentrated to give a condensed toluene solution. Ethanol was added to this toluene solution and the toluene solution was concentrated under reduced pressure, whereby an ethanol suspension was obtained. The precipitate was filtrated at approximately 20° C., and the obtained solid was dried at approximately 80° C. under reduced pressure, whereby 3.6 g of a target white solid was obtained in a yield of 75%. The synthesis scheme of Step 2 is shown below.

[Chemical Formula 34]



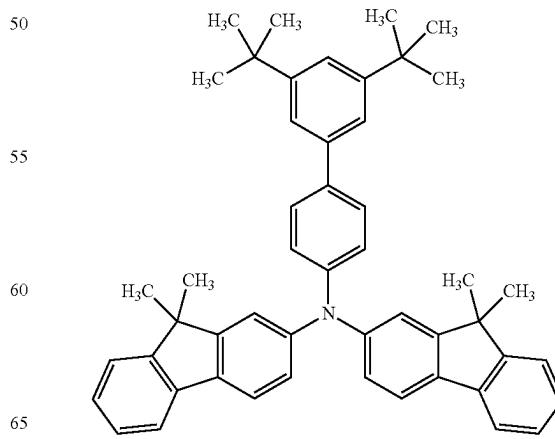
Analysis results by nuclear magnetic resonance spectroscopy (<sup>1</sup>H-NMR) of the white solid obtained in Step 2 are shown below. The results reveal that mmTBumTPoFBi-04 was obtained in this example.

<sup>1</sup>H-NMR δ (CDCl<sub>3</sub>): 7.54-7.56 (m, 1H), 7.53 (dd, 1H, J=1.7 Hz), 7.50 (dd, 1H, J=1.7 Hz), 7.27-7.47 (m, 13H), 7.23 (dd, 1H, J=6.3 Hz, 1.2 Hz), 7.18-7.19 (m, 2H), 7.08-7.00 (m, 5H), 6.88 (d, 1H, J=1.7 Hz) 6.77 (dd, 1H, J=8.0 Hz, 2.3 Hz), 1.42 (s, 9H), 1.39 (s, 18H), 1.29 (s, 6H).

## Reference Synthesis Example 5

Described in this example is a method for synthesizing N-(3", 5", -di-tert-butyl-1,1":3", 1"-biphenyl-4-yl)-bis(9,9-dimethyl-9H-fluoren-2,2'-amine (abbreviation: mmTBuBiFF). The structure of mmTBuBiFF is shown below.

[Chemical Formula 35]

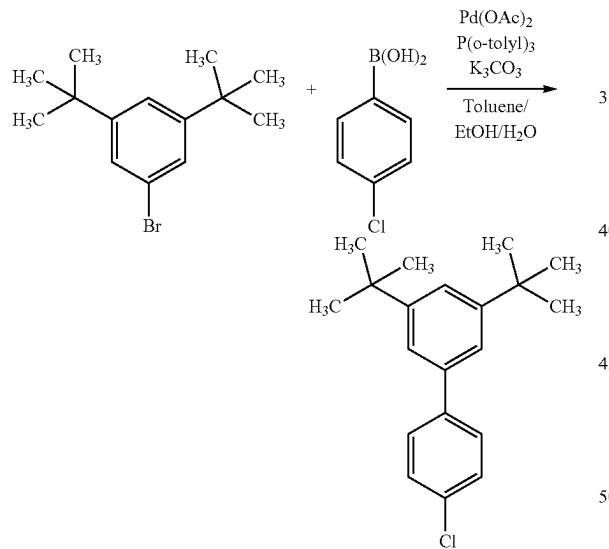


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Step 1: Synthesis of 3',5'-di-tert-butyl-4-chloro-1,1'-biphenyl

In a three-neck flask were put 30.0 g (114 mmol) of 3,5-di-tert-butyl-1-bromobenzene, 19.2 g (123 mmol) of 4-chlorophenylboronic acid, 46.1 g (334 mmol) of potassium carbonate, 550 mL of toluene, 140 mL of ethanol, and 160 mL of water. The mixture was degassed under reduced pressure, and then the air in the flask was replaced with nitrogen. Then, 251 mg (1.12 mmol) of palladium acetate and 695 mg (2.28 mmol) of tris(2-methylphenyl)phosphine were added, and the mixture was heated and refluxed at 90° C. for approximately 5 hours. After that, the temperature of the flask was lowered to room temperature, and the mixture was separated into an organic layer and an aqueous layer. Magnesium sulfate was added to this organic layer to eliminate moisture, and then a solution separated by filtration was concentrated to give a condensed brown solution. The obtained solution was purified by silica gel column chromatography. The obtained solution was concentrated and dried for hardening. After that, hexane was added for recrystallization. The mixed solution in which a white solid was precipitated was cooled with ice and filtrated. The obtained solid was dried at approximately 100° C. in a vacuum, whereby 29.6 g of a target white solid was obtained in a yield of 88%. The synthesis scheme of Step 1 is shown below.

[Chemical Formula 36]



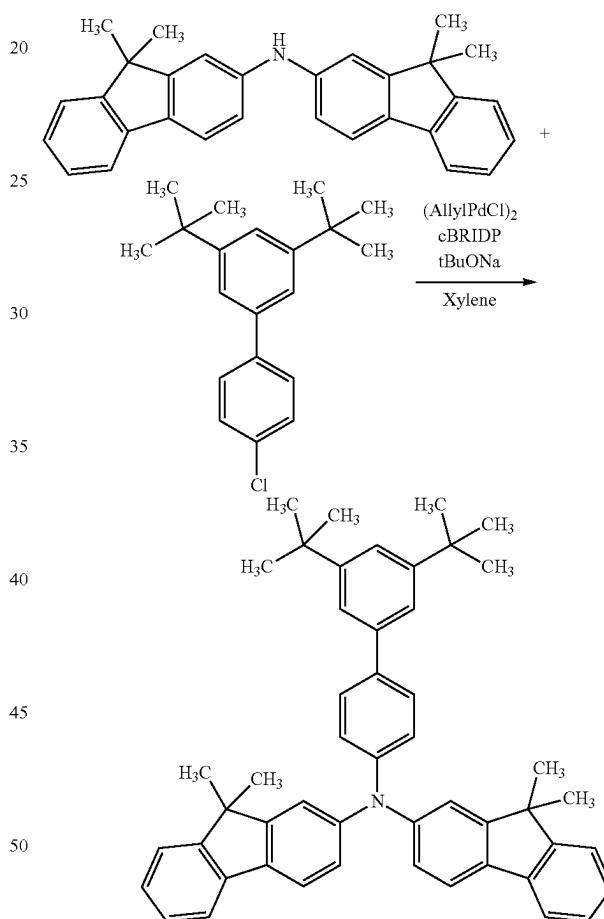
## Step 2: Synthesis of mmTBuBiFF

In a three-neck flask were put 30.4 g (75.7 mmol) of bis(9,9-dimethyl-9H-fluoren-2-yl)amine, 22.8 g (75.8 mmol) of 3',5'-di-tert-butyl-4-chloro-1,1'-biphenyl, 21.9 g (228 mmol) of sodium-tert-butoxide, and 380 mL of xylene. The mixture was degassed under reduced pressure, and then the air in the flask was replaced with nitrogen. The mixture was stirred while being heated to approximately 60° C. Then, 283 mg (0.773 mmol) of allylpalladium(II) chloride dimer (abbreviation: (AllylPdCl)<sub>2</sub>) and 1.05 g (2.98 mmol) of di-tert-butyl(1-methyl-2,2-diphenylcyclopropyl)phosphine (abbreviation: cBRIDP (registered trademark)) were

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added, and the mixture was heated at 100° C. for approximately 5 hours. After that, the temperature of the mixture was lowered to approximately 60° C., and approximately 2 mL of water was added to the mixture, so that a solid was precipitated. The precipitated solid was separated by filtration to give a filtrate. The filtrate was concentrated, and the obtained solution was purified by silica gel column chromatography. The obtained solution was concentrated to give a condensed toluene solution. The toluene solution was dropped into ethanol for reprecipitation. The precipitate was collected by filtration at approximately 10° C. and the obtained solid was dried at approximately 100° C. under reduced pressure, whereby 44.2 g of a target white solid was obtained in a yield of 88%. The synthesis scheme of Step 2 is shown below.

[Chemical Formula 37]



Analysis results by nuclear magnetic resonance spectroscopy (<sup>1</sup>H-NMR) of the white solid obtained in Step 2 are shown below. The results reveal that N-(3',5'-di-tert-butyl-1,1'-biphenyl-4-yl)-bis(9,9-dimethyl-9H-fluoren)-2,2'-amine (abbreviation: mmTBuBiFF) was obtained in this example.

<sup>1</sup>H-NMR (500 MHz DMSO-d<sub>6</sub>):  $\delta=7.75$  (t, 4H,  $J=7.8$  Hz), 7.62 (d, 2H,  $J=9.0$  Hz), 7.51 (d, 2H,  $J=5.0$  Hz), 7.42 (s, 2H), 7.38 (s, 1H), 7.34-7.25 (m, 6H), 7.18 (d, 2H,  $J=8.0$  Hz, 2.0 Hz), 7.03 (dd, 2H,  $J=8.0$  Hz, 2.0 Hz), 1.37 (s, 12H), 1.34 (s, 18H).

This application is based on Japanese Patent Application Serial No. 2021-028509 filed with Japan Patent Office on

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Feb. 25, 2021, Japanese Patent Application Serial No. 2021-079545 filed with Japan Patent Office on May 10, 2021, and Japanese Patent Application Serial No. 2021-170317 filed with Japan Patent Office on Oct. 18, 2021, the entire contents of which are hereby incorporated by reference.

What is claimed is:

1. A light-emitting device comprising:  
a first electrode;  
a second electrode; and  
an Electro-Luminescent (EL) layer positioned between the first electrode and the second electrode,  
wherein the EL layer comprises at least a light-emitting layer, a first layer, a second layer, and a third layer,  
wherein the first layer is positioned between the first electrode and the light-emitting layer,  
wherein the third layer is positioned between the first layer and the light-emitting layer,  
wherein the second layer is positioned between the first layer and the third layer,  
wherein the first layer comprises a first organic compound,  
wherein the second layer comprises a second organic compound,  
wherein the third layer comprises a third organic compound,  
wherein an ordinary refractive index of the second organic compound with respect to light with any of wavelengths greater than or equal to 450 nm and less than or equal to 650 nm is higher than an ordinary refractive index of the first organic compound with respect to light with any of the wavelengths and an ordinary refractive index of the third organic compound with respect to light with any of the wavelengths, and  
wherein a value obtained by subtracting a Giant Surface Potential (GSP) (mV/nm) of the third layer from a GSP (mV/nm) of the first layer is less than or equal to 10 (mV/nm).

2. The light-emitting device according to claim 1, wherein an ordinary refractive index of the second organic compound with respect to light with a wavelength of 633 nm is higher than an ordinary refractive index of the first organic compound with respect to light with the wavelength of 633 nm and an ordinary refractive index of the third organic compound with respect to light with the wavelength of 633 nm.

3. The light-emitting device according to claim 1, wherein a difference between a Highest Occupied Molecular Orbit (HOMO) level of the first organic compound and a HOMO level of the third organic compound is less than or equal to 0.2 eV.

4. The light-emitting device according to claim 1, wherein the first organic compound and the third organic compound are the same organic compound.

5. The light-emitting device according to claim 1, wherein a difference between a Highest Occupied Molecular Orbit (HOMO) level of the first organic compound and a HOMO level of the second organic compound is less than or equal to 0.2 eV.

6. The light-emitting device according to claim 1, wherein the third layer does not comprise fluorine.

7. The light-emitting device according to claim 1, wherein the first layer to the third layer each have a thickness larger than or equal to 25 nm and smaller than or equal to 70 nm.

8. The light-emitting device according to claim 1, wherein the thickness of the second layer is larger than a thickness of the first layer and the thickness of the third layer.

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9. The light-emitting device according to claim 1, wherein the first layer is in contact with the second layer, and  
wherein the second layer is in contact with the third layer.

10. The light-emitting device according to claim 1, wherein the first electrode comprises a reflective electrode.

11. The light-emitting device according to claim 1, wherein a value obtained by subtracting the GSP (mV/nm) of the third layer from a GSP (mV/nm) of the second layer is less than or equal to 10 (mV/nm).

12. The light-emitting device according to claim 1, wherein the value obtained by subtracting the GSP (mV/nm) of the third layer from the GSP (mV/nm) of the first layer is less than or equal to 0 (mV/nm).

13. The light-emitting device according to claim 1, wherein the GSP (mV/nm) of the third layer is larger than a GSP (mV/nm) of the second layer.

14. The light-emitting device according to claim 1, wherein the GSP (mV/nm) of the third layer is larger than the GSP (mV/nm) of the first layer.

15. The light-emitting device according to claim 1, wherein a GSP (mV/nm) of the second layer is larger than the GSP (mV/nm) of the first layer.

16. The light-emitting device according to claim 1, wherein the first organic compound and the third organic compound are different organic compounds.

17. A display device comprising:  
a first light-emitting device; and  
a second light-emitting device,  
wherein the first light-emitting device comprises the light-emitting device according to claim 1,

wherein the second light-emitting device comprises an Electro-Luminescent (EL) layer between a third electrode and the second electrode,

wherein the EL layer of the second light-emitting device comprises at least a light-emitting layer, a fourth layer, a fifth layer, and a sixth layer,

wherein the fourth layer, the fifth layer, and the sixth layer respectively have the same structure as the first layer, the second layer, and the third layer, and

wherein a peak wavelength of light emitted from a light-emitting material comprised in the light-emitting layer of the first light-emitting device is different from a peak wavelength of light emitted from a light-emitting material comprised in the light-emitting layer of the second light-emitting device by more than or equal to 30 nm.

18. A display device comprising:  
a first light-emitting device; and  
a second light-emitting device,  
wherein the first light-emitting device comprises the light-emitting device according to claim 1,

wherein the second light-emitting device comprises an Electro-Luminescent (EL) layer between a third electrode and the second electrode,

wherein the EL layer of the second light-emitting device comprises at least a light-emitting layer, a fourth layer, a fifth layer, and a sixth layer,

wherein the fourth layer, the fifth layer, and the sixth layer respectively have the same thickness as the first layer, the second layer, and the third layer, and

wherein a peak wavelength of light emitted from a light-emitting material comprised in the light-emitting layer of the first light-emitting device is different from a peak wavelength of light emitted from a light-emitting material comprised in the light-emitting layer of the second light-emitting device by more than or equal to 30 nm.

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- 19.** A display device comprising:  
the light-emitting device according to claim 1; and  
at least one of a sensor, an operation button, a speaker, and  
a microphone.
- 20.** A light-emitting apparatus comprising:  
the light-emitting device according to claim 1; and  
at least one of a transistor and a substrate.
- 21.** A lighting device comprising:  
the light-emitting device according to claim 1; and  
a housing.
- 22.** A light-emitting device comprising:  
a first electrode;  
a second electrode; and  
an Electro-Luminescent (EL) layer positioned between  
the first electrode and the second electrode,  
wherein the EL layer comprises at least a light-emitting  
layer, a first layer, a second layer, and a third layer,  
wherein the first layer is positioned between the first  
electrode and the light-emitting layer,  
wherein the third layer is positioned between the first  
layer and the light-emitting layer,  
wherein the second layer is positioned between the first  
layer and the third layer,  
wherein the first layer comprises a first organic com-  
pound,  
wherein the second layer comprises a second organic  
compound,  
wherein the third layer comprises a third organic com-  
pound,  
wherein an ordinary refractive index of the second  
organic compound with respect to light with any of  
wavelengths greater than or equal to 455 nm and less  
than or equal to 465 nm is higher than an ordinary  
refractive index of the first organic compound with  
respect to light with any of the wavelengths and an  
ordinary refractive index of the third organic compound  
with respect to light with any of the wavelengths, and  
wherein a value obtained by subtracting a Giant Surface  
Potential (GSP) (mV/nm) of the third layer from a GSP  
(mV/nm) of the first layer is less than or equal to 10  
(mV/nm).
- 23.** A light-emitting device comprising:  
a first electrode;  
a second electrode; and  
an Electro-Luminescent (EL) layer positioned between  
the first electrode and the second electrode,

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- wherein the EL layer comprises at least a light-emitting  
layer, a first layer, a second layer, and a third layer,  
wherein the first layer is positioned between the first  
electrode and the light-emitting layer,  
wherein the third layer is positioned between the first  
layer and the light-emitting layer,  
wherein the second layer is positioned between the first  
layer and the third layer,  
wherein the light-emitting layer comprises a light-emit-  
ting material that emits light with a peak wavelength,  
wherein the first layer comprises a first organic com-  
pound,  
wherein the second layer comprises a second organic  
compound,  
wherein the third layer comprises a third organic com-  
pound,  
wherein an ordinary refractive index of the second  
organic compound with respect to light with the peak  
wavelength is higher than an ordinary refractive index  
of the first organic compound with respect to light with  
the peak wavelength and an ordinary refractive index of  
the third organic compound with respect to light with  
the peak wavelength, and  
wherein a value obtained by subtracting a Giant Surface  
Potential (GSP) (mV/nm) of the third layer from a GSP  
(mV/nm) of the first layer is less than or equal to 10  
(mV/nm).
- 24.** The light-emitting device according to claim 23,  
wherein a difference between the ordinary refractive indices  
of the second organic compound and the first organic  
compound with respect to light with the peak wavelength  
and a difference between the ordinary refractive indices of  
the second organic compound and the third organic com-  
pound with respect to light with the peak wavelength are  
each greater than or equal to 0.2 and less than 0.5.
- 25.** The light-emitting device according to claim 24,  
wherein a difference between the ordinary refractive indices  
of the first organic compound and the third organic com-  
pound with respect to light with the peak wavelength is less  
than or equal to 0.1.
- 26.** The light-emitting device according to claim 24,  
wherein a product of a thickness of the second layer and the  
ordinary refractive index of the second organic compound  
with respect to the peak wavelength is greater than or equal  
to 70% and less than or equal to 130% of  $\frac{1}{4}$  of the peak  
wavelength.

\* \* \* \* \*