(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 1 603 369 A1

(12) EUROPEAN PATENT APPLICATION
published in accordance with Art. 158(3) EPC

(43) Date of publication:
07.12.2005 Bulletin 2005/49

(21) Application number: **03712695.0**

(22) Date of filing: **13.03.2003**

(51) Int Cl.^{7}: **H05B 33/22**, H05B 33/14

(86) International application number:
PCT/JP2003/003027

(87) International publication number:
WO 2004/082338 (23.09.2004 Gazette 2004/39)

(84) Designated Contracting States:
AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HU IE IT LI LU MC NL PT RO SE SI SK TR
Designated Extension States:
AL LT LV MK

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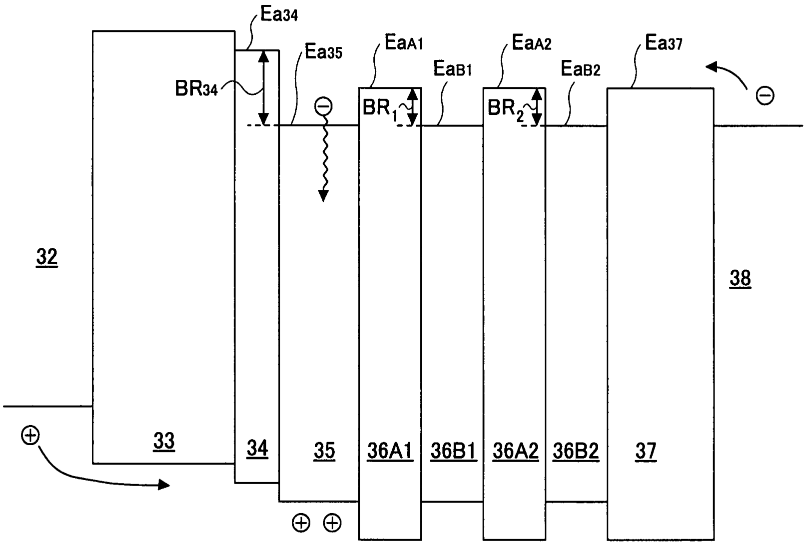
(54) ORGANIC ELECTROLUMINESCENCE DEVICE AND ORGANIC ELECTROLUMINESCENCE DISPLAY

(57) An organic EL device includes a transparent substrate (31), an anode (32), a hole injection layer (33), a hole transport layer (34), an luminescent layer (35), an electron transport multilayered body (36), a cathode-side electron transport layer (37), and a cathode (38) formed on the substrate (31) in that order. The electron transport multilayered body (36) includes alternate layered two kinds of electron transport layers (36A, 36B) having electron affinities that are different from each other. Increasing the amount of electron current injected from the cathode (38) for balancing with the amount of hole current, and enhancing luminous efficiency.

FIG.5



EP 1 603 369 A1

Printed by Jouve, 75001 PARIS (FR)

Description

TECHNICAL FIELD

[0001] The present invention generally relates to a photoelectron device and a flat panel display using a photoelectron device, and more particularly to an organic electroluminescence device and an organic electroluminescence display.

BACKGROUND ART

[0002] In recent years, demands of the market are shifting from a conventional large size and heavyweight CRT (Braun tube) display to a thin-size and light-weight flat display. As for the flat displays, there are liquid crystal displays and plasma displays which are employed, for example, as home television receivers and monitors of personal computers.

[0003] Currently, an electroluminescence display (hereinafter referred to as "EL display") and more particularly, an organic electroluminescence display is drawing attention as the next generation flat display. Ever since the report on a multilayered type element being layered with organic thin films having a hole transport property and an electron transport property (C.W.Tang and S.A.Van Slyke, Applied Physics Letters vol.51, 913 (1987), organic EL devices included in the organic EL display are gathering attention as a large area light emitting element that illuminates with a low voltage of 10 V or less, and are being researched extensively. Compared to the liquid crystal display, the organic EL display requires no backlight since the organic EL display is self-luminous, thereby yielding a thinner, simple-structured, and flexible display. Therefore, the organic EL display is expected to be widely applied in various fields. In employing the organic EL display for practical use, however, a task of obtaining a longer life-span for the organic EL display is yet to be achieved.

[0004] Fig.1 is a cross-sectional drawing showing a conventional organic EL device. As shown in Fig.1, an organic EL device 10 has a transparent anode 12, a hole injection layer 13, a hole transport layer 14, a luminescent layer 15, an electron transport layer 16, and a cathode 18 which are formed in this order on a transparent insulating substrate 11. In the organic EL device 10, holes are injected from the transparent anode 12 to the hole injection layer 13, while electrons are injected from the cathode 18. The holes and electrons are recombined in the luminescent layer 15, to thereby release energy. This energy excites, for example, an organic fluorescent part in the luminescent layer 15, thereby causing illumination. Luminance is determined by the amount of recombination of the recombining holes and electrons with respect to time. Since luminous efficiency is expressed by luminance with respect to current consumption, the luminous efficiency becomes higher when the amount of electrons and holes contributing to illumination is more balanced.

[0005] In the organic EL device 10, the transparent anode 12 is formed of ITO (Indium Tin Oxide). By oxidizing the surface of the ITO with, for example, UV ozone or oxygen plasma, the work function can be consistent with the ionization potential of the hole injection layer. This reduces the hole injection barrier between the transparent anode 12 to the hole injection layer 13 and increases the amount of hole current.

[0006] Meanwhile, metals, such as Li, Mg, or alloys such as Al-Li, Mg-Ag, which have a low electron injection barrier work function with respect to the electron transport layer 16, are used in the cathode 18. In recent years, it is found that use of a metal fluoride (e.g. LiF/Al) for the electron injection layer, even in a case of employing a simple A1 as the cathode 18, yields an electron injection performance that is equal to a device using low work function simple metals, such as Li, Mg or alloys thereof, and that device properties, such as luminous efficiency, are the same as or greater than using metals with low work function (L.S.Hung, C.W.Tang Tang, and M.G.Mason, Applied Physics Letters vol.70(2), 152 (1997).

[0007] Nevertheless, when a low work function simple metal, alloys or LiF, for example, is used in the cathode 18, the amount of electron current reaching the luminescent layer 15 becomes less than the amount of hole current, thereby causing unevenness in the amount of current between the electrons and the holes. This causes the current of the holes not contributing to illumination to become wasted and prevents a sufficient luminous efficiency from being obtained.

[0008] Furthermore, in a case where luminous efficiency is low, the amount of voltage is to be increased for obtaining a sufficient luminance and supplying a greater amount of current. However, an excess application of voltage is liable to cause a chemical reaction at interfaces between the anode 12 and the hole injection layer 13 and between the cathode 18 and the electron transport layer 16. This leads to property change and function deterioration in the hole injection layer 13 and the electron transport layer 16, and furthermore, device failure. Therefore, there is a problem in obtaining a sufficient life-span for the device.

[0009] Furthermore, Japanese Laid-Open Patent Application No.2002-43063 discloses a multilayer electron transport area provided for improving carrier injection to a luminescent layer and reducing operating voltage. However, this document does not disclose a specific configuration of the structure of the multilayer electron transport area.

(Patent Document 1) Japanese Laid-Open Patent Application No.2002-43063

(Patent Document 2) Japanese Laid-Open Patent Application No.2001-357975

DISCLOSURE OF INVENTION

[0010] Accordingly, it is an object of the present invention to provide a novel and useful organic electroluminescence device and an organic electroluminescence display that obviate the above-described problems.

[0011] A more specific object of the present invention is to provide an organic electroluminescence device that has excellent luminous efficiency and that is able attain a long life-span.

[0012] According to one aspect of the present invention, an organic electroluminescence device includes an anode, a luminescent layer formed on the anode, a carrier transport multilayered body formed on the luminescent layer, and a cathode formed on the carrier transport multilayered body,

wherein the carrier transport multilayered body is alternately layered by a first carrier transport layer and a second carrier transport layer,

wherein the first carrier transport layer and the second carrier transport layer have electron transportabilities that are different from each other.

[0013] Here, electron transportability is defined by, for example, the electron affinity, the ionization potential, or the energy gap of the organic material included in the first carrier transport layer and the second carrier transport layer.

[0014] With the present invention, the amount of electron current injected from the luminescent layer can be increased by providing the carrier transport multilayered body having alternate layers of the first carrier transport layer and the second carrier transport layer having different electron affinities between the luminescent layer and the cathode. As a result, the organic electroluminescence device of the present invention attains a high luminous efficiency and a long life-span by balancing the amount of electron current and the amount of hole current.

[0015] The first carrier transport layer and the second carrier transport layer have different electron affinities. Owing to the different electron affinities of the first carrier transport layer and the second carrier transport layer, multiple quantum wells can be formed and the amount of electron current can be increased. It is to be noted that electron affinity is expressed as an energy difference between an energy of a lower end of a conductive member of a material included in, for example, the carrier transport layer and a vacuum level, and is indicated as a positive value.

[0016] According to another aspect of the present invention, an organic electroluminescence display including any one of the aforementioned organic electroluminescence devices is provided.

[0017] With the present invention, an organic electroluminescence display having high luminous efficiency and a long life-span can be obtained.

BRIEF DESCRIPTION OF THE DRAWINGS

[0018]

Fig.1A is a cross-sectional view showing a conventional organic EL device;

Fig.2 is a cross-sectional view showing a configuration of a device body of the present invention;

Fig.3 is a characteristic diagram I-V of the configuration of the device body shown in Fig.2;

Fig.4 is a cross-sectional view showing an organic EL device according to a first embodiment of the present invention;

Fig.5 is an energy diagram of an organic EL device according to the first embodiment;

Fig.6 is a drawing for describing a method for obtaining an energy gap;

Fig.7 is a drawing for describing a method for obtaining an ionization potential;

Fig.8 is a diagram showing a characteristic value of an electron transport layer and a hole transport layer used in organic EL devices for examples and comparative examples;

Fig.9 is a diagram showing layer configuration and evaluation results of organic EL devices of first-third examples and first-second comparative examples;

Fig.10 is a diagram showing layer configuration and evaluation results of organic EL devices of fourth-fifth examples and third-fourth comparative examples; and

Fig.11 is an exploded perspective view showing an organic EL display according to a second embodiment of the present invention.

BEST MODE FOR CARRYING OUT THE INVENTION

[0019] First, the process in reaching the present invention is described below. The inventors of the present invention conducted the below-given experiments and found that injectable current density can be increased by providing a luminescent layer and a cathode having therebetween an electron transport multilayered body with alternately layered electron transport layers having different electron transportabilities.

[0020] Fig.2 is a cross-sectional drawing showing a configuration of a device body of the present invention which was used in the experiments. With reference to Fig.2, a device body 20 includes a substrate 21, an anode 22 formed on the substrate 21, an electron transport multilayered body 26 with two different electron transport layers 26A, 26B that are alternately layered on the anode 22, a cathode-side electron transport layer 27 layered on the electron transport multilayered body 26, and a cathode 28 formed on the cathode-side electron transport layer 27. A1 is used for the anode 22, and LiF/Al is used for the cathode 28. Furthermore, TYE 704 (Name of product manufactured by Toyo Ink Mfg. Co. Ltd.) is used for the cathode-side electron transport layer 27. The electron transport multilayered body 26 includes a first electron transport layer 26A1 and a second electron transport layer 26B1 layers (starting from the anode side), and further continuing in this order, a first electron transport layer 26A2 and a second electron transport layer 26B2 being layered alternately. Here, TYE 704 is used for the first electron transport layer 26A, and TYG 201 (Name of product manufactured by Toyo Ink Mfg. Co. Ltd.) is used for the second electron transport layer 26B. Although TYG 201 (Name of product manufactured by Toyo Ink Mfg. Co. Ltd.) is known as a green color luminescent material, it can also be used for an electron transport layer. Device bodies for which the layering is repeated N=1,3,4 times and a device body for comparison having a similar configuration but without an electron transport multilayered body, that is, a device body to which the layering is repeated N=0 times, are fabricated. The thicknesses of the electron transport multilayered body 26 (as indicated inside brackets {}) and the cathode-side electron transport layer 27 are shown below. It is to be noted that the cathode-side electron transport layer 26 is provided to each of the device bodies so that the condition(s) of the electron injection barrier from the anode may be uniform.

N=0: TYG 201 (80 nm)
N=1: {[TYG 201 (30 nm)/TYE 704 (30 nm)]$_1$}/TYG 201 (20 nm)
N=3: {[TYG 201 (10 nm)/TYE 704 (10 nm)]$_3$}/TYG 201 (20 nm)
N=4: {[TYG 201 (7.5 nm)/TYE 704 (7.5 nm)]$_4$}/TYG 201 (20 nm)

[0021] In order to measure the amount of current flowing in the electron transport multilayered body 26, a direct current of 0-10 V in intervals of 0.5 V is applied between the anode 22 and the cathode 28, and the amount of current flowing in the device is measured with an ampere meter.

[0022] Fig.3 is a drawing showing properties I - V of the device body shown in Fig.2. In comparing between N=0 and N=1 with reference to Fig.3, the amount of current is relatively the same, or N=1 is slightly smaller. Meanwhile, in N=3 and N=4, it is apparent that the amount of current increases considerably and thus increases as the number of repetitions becomes larger. Accordingly, the amount of electron current can be increased by repeating the layering of the two electron transport layers 25A and 25B that have electron transportabilities that are different from each other. By increasing the number of the multilayers, a sufficient amount of electron current can be provided to obtain balance with the amount of hole current. It is assumed that this owes to multiple quantum wells being formed by alternately layering electron transport layers having different electron affinities, in which the effect of the multiple quantum wells increases the amount of electron current. It is to be noted that the reason the amount of current does not increase in a case of N=1 is because it is assumed that no multiple quantum well is formed in a case of N=1.

[0023] Accordingly, the inventors have arrived at the present invention of an organic EL device having an electron transport multilayered body with layers of different electron transport layers.

(First Embodiment)

[0024] Embodiments of the present invention of an organic EL device are described with reference to the accompanying drawings.

[0025] Fig.4 is a cross-sectional view showing an organic EL device according to an embodiment of the present invention. Fig.5 is an example of an energy diagram of an organic EL device according to the embodiment shown in Fig.4. In Fig.5, "Ea" indicates electron affinity, "Eg" indicates energy gap, and "Ip" indicates ionization potential. With reference to Figs.4 and 5, according to this embodiment, an organic EL device 30 has a transparent substrate 31, an anode 32, a hole injection layer 33, a hole transport layer 34, a luminescent layer 35, an electron transport multilayered body 36, a cathode-side electron transport layer 37, and a cathode 38 which are formed in that order on the substrate 31.

[0026] For example, a transparent insulating substrate such as glass or quartz, a semiconductor substrate such as Si, a film such as PET or PEN, or a resin substrate such as PVA may be used for the substrate 31. Furthermore, TFTs (Thin Film Transistor) may be disposed in a matrix-like manner for controlling on/off of the organic EL device on the aforementioned substrates. Although the thickness of the substrate 31 may suitably selected depending on the material of the substrate, the thickness of the substrate 31 is approximately 200 μ*m-1000* μm.

[0027] The anode 32 is formed on the substrate 31 by using a conductive material such as Al and performing a vapor deposition method or a sputtering method therewith. From the aspect of hole injection property, it is preferable to use, for example, Au, Cr, or Mo, which has a large work function. However, in a case where light is emitted from the anode side, it is formed with a transparent material such as ITO or indium oxide.

[0028] The hole injection layer 33 and the hole transport layer 34 have high HOMO, that is, materials with low ionization potential are employed thereto. As a representative example, there is copper phthalocyanine (CuPc), m-MTDATA of starburst type amine, 2-TNATA, TPD, or α-NPD. It is to be noted that a hole injection layer may be provided for performing more hole injections between the anode and the hole transport layer. The aforementioned copper phthalocyanine (CuPc), m-MTDATA of starburst type amine, or 2-TNATA may be used as the hole injection layer.

[0029] Furthermore, it is preferable for the hole transport layer 34 to have a small electron affinity with respect to the luminescent layer 35. Thereby, electrons can accumulate in the luminescent layer and space density of electrons in the luminescent layer can be increased. More specifically, as shown in Fig.5, an energy barrier having a height BR_{34} (=Ea_{35}-Ea_{34}) is formed, wherein a relation between electron affinity Ea_{34} of the hole transport layer 34 and electron affinity Ea_{35} of the luminescent layer is represented as $Ea_{34}<Ea_{35}$.

[0030] It is to be noted that hole transport layers having different ionization potentials may be provided in an alternately layered manner. By forming the energy barrier against the holes, the amount of hole current can be controlled, thereby achieving balance with the amount of electron current.

[0031] A metal complex material, for example, Alq 3 (tris (8-hydroxyquinolio aluminum), Znq 2, or Balq 2, or a pigment material, for example, PZ10 or EM2 may be used for the luminescent layer 35. Furthermore, a material having a pigment such as rubrene or TPB, doped with a host material such as Alq3, may also be used.

[0032] The cathode-side electron transport layer 37 includes the same material(s) included in the below-described electron transport multilayered body 36. More particularly, it is preferable for the cathode-side electron transport layer 37 to have an energy gap that is same as or greater than the electron transport layer included in the electron transport multilayered body 36. Thereby, the cathode-side electron transport layer 37 can be prevented from illuminating.

[0033] A metal (e.g. Li) or a metal alloy (e.g. Mg-Ag, Al-Li) having a small work function is used for the cathode 38. A cathode having an electron injection layer of, for example, metal fluoride (e.g. LiF/Al) may also be used.

[0034] The electron transport multilayered body 36 has alternately layered layers including a first electron transport layer 36A and a second electron transport layer 36B having different electron transportabilities. Here, different electron transportabilities signify, for example, a difference in HOMO, LUMO (Lowest Unoccupied Molecular Orbital), or conductivity. In this embodiment, the first electron transport layer 36A and the second electron transport layer 36B are described as layers having different electron affinities with respect to each other.

[0035] A metal chelate of 8-hydroxyquinoline, a metal thioxynoid compound, an oxadiazole metal chelate, triazine, 4,4'-bis (2,2-diphenylvinyl) biphenyl, for example, may be used for the first electron transport layer 36A or the second electron transport layer 36B. A preferable material among the metal chelates of 8-hydroxyquinoline may be, for example, Alq 3 (tris (8-hydroxyquinolinate) aluminum, Balq (bis (8-hydroxyquinolato)-(4-phenylphenolato) aluminum, or bis PBD. Furthermore, a preferable material among the metal thioxynoid compounds may be, for example, bis (8-quinolinthiolato) zinc, bis (8-quinolinthilato) cadmium, tris (8-quinolinthiolato) gallium, or tris (8-quinolinthiolato) indium. Furthermore, a preferable material among the oxadiazole metal chelates may be, for example, bis [2-(2-hydroxyphenyl)-5-phenyl-1,3,4-oxadiazolato] zinc, bis [2-(2-hydroxyphenyl)-5-phenyl-1,3,4-oxadiazolato] beryllium, bis [2-(2-hydroxyphenyl)-5-(1-naphthyl)-1,3,4-oxadiazolato] zinc, or bis [2-(2-hydroxyphenyl)-5-(1-naphthyl)-1,3,4-oxadiazolato] beryllium.

[0036] The aforementioned materials for the electron transport layers are chosen as the first electron transport layer 36A and the second electron transport layer 36B so that the relation of electron affinities satisfies $Ea_A<Ea_B$ in a case where Ea_A indicates the electron affinity of the first electron transport layer 36A and Ea_B indicates the electron affinity of the second electron transport layer 36B. In choosing a material that satisfies this relation, a below-described measuring method may be employed to obtain electron affinity.

[0037] Although electrons flow from the cathode 38 toward the luminescent layer 35, in the interface between the second electron transport layer 36B2 and the first electron transport layer 36A2, for example, of the electron transport multilayered body 36, a difference Ea_{B2}-Ea_{A2} in electron affinity between the two layers causes an energy barrier BR_2 to be formed and a square-well potential to be generated. Likewise, between the second transport layer 36B1 and the first electron transport layer 36A1, an energy barrier BR_1 is formed and a square-well potential is generated. It is, therefore, assumed that multiple quantum wells are formed and the amount of electron current is increased.

[0038] The film thicknesses of the first electron transport layer 36A and the second electron transport layer 36B can be suitably selected in accordance with the number of repetitions of the layering of the first electron transport layer 36A and the second electron transport layer 36B; they are, however, set in a range of 2 nm - 50 nm (preferably 5 nm-20 nm). If the thickness is greater than 50 nm, the thickness of the entire organic EL device would be too thick. Thereby, application voltage would too large, an electrochemical reaction would more easily occur at the anode, cathode, or at its interface with respect to a contacting hole injection layer or a cathode-side electron transport layer, and the lifespan of the organic EL element would be adversely affected. Furthermore, if the thickness is less than 2 nm, a successive film would be difficult to form, and the periodicity of the square-well potential would be out of order.

[0039] The first electron transport layer 36A and the second electron transport layer 36B are set to a predetermined thicknesses within the above-described range, respectively. Accordingly, a satisfactory periodicity of the multiple quantum well can be attained. It is to be noted that the first electron transport layer 36A and the second electron transport layer 36B may be of the same thickness or different thicknesses.

[0040] Furthermore, the first electron transport layer 36A may be a thin film which is thinner than the second electron transport layer 36B. Although the first electron transport layer 36A functions as a barrier layer owing to its small electron affinity, the amount of electron current may be further increased by making the barrier layer thinner.

[0041] Furthermore, the number of repetitions of the layering of the first electron transport layer 36A and the second electron transport layer 36B are set from 2-10 (preferably 2-4). If the repetition is greater than 10, the organic EL device would be too thick. If the repetition is less than 2, the multiple quantum wells could not be formed.

[0042] It is to be noted that the energy gap, ionization potential, and electron affinity for the electron transport layer, the hole transport layer, etc. are obtained according to the below-given measuring conditions and measuring methods.

[0043] As for energy gap Eg, the optical absorption spectrum is measured, in which the energy of the long wavelength end of the optical absorption spectrum is the energy gap Eg. More specifically, with the same conditions for forming each layer of the organic EL device, each measure target (e.g. electron transport layer) is individually formed into a thin film with a thickness of approximately 50 nm. A spectrophotometric apparatus capable of measuring optical absorption spectra (Spectrophotometer U-4100: name of product manufactured by Hitachi Ltd.) is used, in which the optical absorption spectrum (wavelength dependency) is measured by irradiating ultraviolet light through visible light to a thick film in the atmosphere.

[0044] Fig.6 is a drawing showing properties of the optical absorption spectrum. With reference to Fig.6, the energy gap Eg is obtained by performing energy conversion with respect to a wavelength of an intersection point CP1 between a straight line being extrapolated by approximating a straight line portion LN1 at a skirt of the optical absorption spectrum toward a long wavelength side and a straight line being extrapolated by approximating straight line portion BG1 of the background toward a short wavelength side.

[0045] As for the ionization potential Ip, ionization potential Ip is the energy of photoemission threshold measured by an ultraviolet photoelectron analyzing method. More specifically, the number of discharged photoelectrons are measured by irradiating an ultraviolet light to a thin film in the atmosphere by using an atmosphere type ultraviolet photoelectron analyzing apparatus (AC-1: Name of product manufactured by Riken Keiki Co. Ltd.), in which the same thick film used for the energy gap Eg is employed. Thereby, a relation between the energy of the incident ultraviolet ray and the number of photoelectrons is obtained. In terms of measuring conditions, the range of energy of the incident ultraviolet ray is 3.8-6.2 eV, and the strength of the ultraviolet ray is 20 nW.

[0046] Fig.7 is a characteristic diagram showing an example of a relation between a square root of the number of photoelectrons and the energy of the incident ultraviolet ray. With reference to Fig.7, the ionization potential Ip is obtained from the energy of an intersection point CP2 between a straight line being extrapolated by approximating a straight line portion LN2 at a rising part of the characteristic line toward a low energy side and a straight line being extrapolated by approximating a straight line portion of the background toward a high energy side.

[0047] Furthermore, the electron affinity Ea is obtained by the difference between the above-obtained ionization potential Ip and the energy gap Eg (Ea=Ip-Eg).

[0048] By using these methods, the energy gap, the ionization potential, and the energy affinity of each electron transport material is measured. Thereby, selection of a combination of electron transport layers to be included in the electron transport multilayered body is enabled.

[0049] Fig.8 is a diagram showing measurement values of the energy gap, the ionization potential, the electron affinity of the electron transport layer and the hole transport layer included in the organic EL device with respect to examples which are of the present invention and comparative examples which are not of the present invention. The examples and comparative examples are described below based on the measurement values shown in Fig.8.

[Example 1]

[0050] An anode with a thickness of 150 nm is formed on a glass substrate by a sputtering method with use of ITO, and is subject to UV ozone processing in which the anode surface is irradiated with UV rays under an oxygen atmosphere for 20 minutes. Then, 2-TNATA (thickness 40 nm) serving as the hole injection layer, α-NPD (thickness 10 nm) serving as the hole transport layer, and TYG 201 (thickness 20 nm) serving as the luminescent layer, are formed in order.

[0051] Next, starting from TYE 704, the formation of a combination of TYE 704 (thickness 15 nm) and TYG 201 (thickness 15 nm), serving as the electron transport multilayered body, is repeated twice. Furthermore, a single layer of TYE 704 (thickness 20 nm) is formed on the electron transport multilayered body, and the cathode formed of LiF/Al is last formed.

[0052] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 3 V. When voltage of 10 V is applied, luminance of 913 cd/m^2 and a luminous efficiency of 8.40 cd/A are obtained.

[Second Example]

[0053] The organic EL element of this example is the same as that of the first example except for repeating the formation of a combination of TYE 704 (thickness 10 nm) and TYG 201 (thickness 10 nm) for three times.

[0054] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 3 V. When voltage of 10 V is applied, luminance of 1075 cd/m^2 and a luminous efficiency of 9.70 cd/A are obtained.

[Third Example]

[0055] The organic EL element of this example is the same as that of the first example except for repeating the formation of a combination of TYE 704 (thickness 7.5 nm) and TYG 201 (thickness 7.5 nm) for four times.

[0056] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 3 V. When voltage of 10 V is applied, luminance of 1017 cd/m^2 and a luminous efficiency of 8.89 cd/A are obtained.

[First Comparative Example]

[0057] The organic EL device in this comparative example is the same as that of the first example except for forming TYG 201 (thickness 50 nm) as the luminescent layer and forming a single layer of TYG 201 (thickness 50 nm) as the electron transport layer instead of the electron transport multilayered body.

[0058] In the organic EL device in this comparative example, a green luminescence is observed in a case where voltage is no less than 3 V. When voltage of 10 V is applied, luminance of 967 cd/m^2 and a luminous efficiency of 8.25 cd/A are obtained.

[Second Comparative Example]

[0059] The organic EL device in this comparative example is the same as that of the first example except for providing a combination of TYE 704 (thickness 30 nm) and TYG 201 (thickness 30 nm) as the electron transport multilayered body.

[0060] In the organic EL device in this comparative example, a green luminescence is observed in a case where voltage is no less than 4 V. When voltage of 10 V is applied, luminance of 750 cd/m^2 and a luminous efficiency of 7.48 cd/A are obtained.

[0061] Fig.9 shows the layer configurations and evaluation results of the first-third examples and the first-second comparative examples. With reference to Fig.9, it is apparent that luminous efficiency increases when the layering of the TYE 704 and TYG 201, which form the electron transport multilayered body, is repeated two times or more, when compared to the first comparative example in which the electron transport multilayered body is a single layer of the electron transport layer or the second comparative example in which the number of repetition is 1. Furthermore, the luminous efficiency is highest for the organic EL device of the second example. It is assumed that this owes to the balance between the amount of electron current and the amount of hole current. Furthermore, it is apparent that luminance is also highest for the organic EL device of the second example.

[0062] Next, in the TYE 704 and the TYG 201 forming the electron transport multilayered body, examples and comparative examples replacing the TYG 201 with Alq 3 is described.

[Fourth Example]

[0063] The organic EL element of this example is the same as that of the second example except for using Alq 3 (thickness 10 nm) as an alternative of TYG 201 (thickness 10 nm), in which the number of the repetitions is 3.

[0064] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 5 V. When voltage of 10 V is applied, luminance of 994 cd/m^2 and a luminous efficiency of 7.52 cd/A are obtained.

[Fifth Example]

[0065] The organic EL element of this example is the same as that of the fourth example except for forming each layer of the electron transport multilayered body with a thickness of 7.5 nm, and repeating for 4 times.

[0066] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 5 V. When voltage of 10 V is applied, luminance of 1021 cd/m^2 and a luminous efficiency of 7.44 cd/A are obtained.

[Third Comparative Example]

[0067] The organic EL element of this comparative example is the same as that of the fourth example except for employing an electron transport layer having a single layer of Alq 3 (thickness 30 nm) as an alternative of the electron transport multilayered body and employing TYG 201 (thickness 50 nm) as the electron transport layer in abutment with the cathode.

[0068] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 5 V. When voltage of 10 V is applied, luminance of 1058 cd/m^2 and a luminous efficiency of 6.68 cd/A are obtained.

[Fourth Comparative Example]

[0069] The organic EL element of this comparative example is the same as that of the fourth example except for forming each layer of the electron transport multilayered body with a thickness of 30 nm and repeating 1 time.

[0070] In the organic EL device in this example, a green luminescence is observed in a case where voltage is no less than 5 V. When voltage of 10 V is applied, luminance of 1005 cd/m^2 and a luminous efficiency of 6.75 cd/A are obtained.

[0071] Fig.10 shows the layer configurations and evaluation results of the fourth-fifth examples and the third-fourth comparative examples. With reference to Fig. 10, it is apparent that luminous efficiency increases when the layering of the TYE 704 and Alq 3, which form the electron transport multilayered body, is repeated three times or more, when compared to the third comparative example in which the electron transport multilayered body is a single layer of the electron transport layer or the fourth comparative example in which the number of repetition is 1.

[0072] Furthermore, in comparing the second-third examples with the fourth-fifth examples having the same number of repetitions, the organic EL device of the second-third examples having multilayers of the TYG 201 layer and the TYE 704 layer have a higher improvement rate of luminous efficiency than the organic EL device of the fourth-fifth examples having multilayers of the Alq3 layer and the TYE 704 layer with respect to the comparative example where its repetition number is 1 (second comparative example, fourth comparative example). The reason for this is assumed in that the difference of electron affinity between the Alq 3 layer and the TYE 704 layer is 0.10 eV whereas the difference of electron affinity between the TYG 201 layer and the TYE 704 layer is 0.23 eV, and that the combination between the TYG 201 and the TYE 704 can form multiple quantum wells more sufficiently, thereby a more significant multiple quantum well effect is generated.

(Second Embodiment)

[0073] Fig.11 is an exploded perspective view showing an organic EL display according to a second embodiment of the present invention. With reference to Fig. 11, an organic EL display 50 includes, for example, a glass substrate 51, a cathode 52 formed on the glass substrate in a stripe-like manner, an anode 54 formed perpendicularly facing the cathode 52 in a stripe-like manner, and a multilayered body 53 formed between the cathode 52 and the anode 54. Furthermore, although not illustrated, the organic EL display 50 includes, for example, a drive circuit for driving voltage applied between the cathode and the anode, and a sealing material for preventing exposure to water vapor or oxygen.

[0074] The organic EL display 50 is able to illuminate a desired area by applying voltage to the cathode 52 and the anode 54 in a desired area. The characteristic of the organic EL display 50 is that the cathode 52, the multilayered body 53, and the anode 54 are formed of the above-described organic EL device of the present invention. Therefore, an organic EL display which is able to attain excellent luminous efficiency and a long life-span can be obtained.

[0075] Further, the present invention is not limited to these embodiments, but variations and modifications may be made without departing from the scope of the present invention.

[0076] For example, in the embodiments of the present invention, the organic EL device may be formed by layering on a substrate from an anode side or from a cathode side.

Industrial Applicability

[0077] With the present invention, by providing an electron transport multilayered body having alternately layered electron transport layers having different electron transportabilities on a cathode side of a luminescent layer in an organic electroluminescence device, an organic electroluminescence device capable of attaining excellent luminous efficiency and a long life-span can be provided.

Claims

1. An organic electroluminescence device **characterized by** comprising:

    an anode;
    a luminescent layer formed on the anode;
    a carrier transport multilayered body formed on the luminescent layer; and
    a cathode formed on the carrier transport multilayered body;

    wherein the carrier transport multilayered body is alternately layered by a first carrier transport layer and a second carrier transport layer,
    wherein the first carrier transport layer and the second carrier transport layer have electron transportabilities that are different from each other.

2. The organic electroluminescence device as claimed in claim 1, **characterized in that** the first carrier transport layer and the second carrier transport layer have electron affinities that are different from each other.

3. The organic electroluminescence device as claimed in claim 1, **characterized in that** the carrier transport multilayered body includes alternate layers of the first carrier transport layer and the second carrier transport layer that are repeated in a range from 2-10 times.

4. The organic electroluminescence device as claimed in claim 1, **characterized in that** the first carrier transport layer and the second carrier transport layer have predetermined film thicknesses.

5. The organic electroluminescence device as claimed in claim 1, **characterized in that** the first

carrier transport layer has an electron affinity which is smaller than that of the second carrier transport layer, and has a thickness which is same as or smaller than that of the second carrier transport layer.

6. The organic electroluminescence device as claimed in claim 1, **characterized in that** one of the first carrier transport layer and the second carrier transport layer is of a material which is the same as that of the luminescent layer.

7. The organic electroluminescence device as claimed in claim 1, **characterized by** further comprising:

   an electron transport layer between the carrier transport multilayered body and the cathode;

   wherein the electron transport layer has an energy gap that is same as or larger than a larger one of the first carrier transport layer and the second carrier transport layer.

8. The organic electroluminescence device as claimed in claim 1, **characterized in that** the carrier transport multilayered body further includes a third carrier transport layer, and includes repeated layers of the first carrier transport layer, the second carrier transport layer, and the third carrier transport layer **in that** order.

9. The organic electroluminescence device as claimed in claim 1, **characterized by** further comprising:

   a hole transport layer between the anode and the luminescent layer;

   wherein the hole transport layer has an electron affinity which is larger than that of the luminescent layer.

10. The organic electroluminescence device as claimed in claim 9, **characterized by** further comprising:

    another hole transport layer between the anode and the luminescent layer;

    wherein the hole transport layer and the other hole transport layer are alternately layered,
    wherein the hole transport layer and the other hole transport layer have ionization potentials that are different from each other.

11. An organic electroluminescence display **characterized by** comprising:

    the organic electroluminescence device as in claim 1-10.

FIG.1

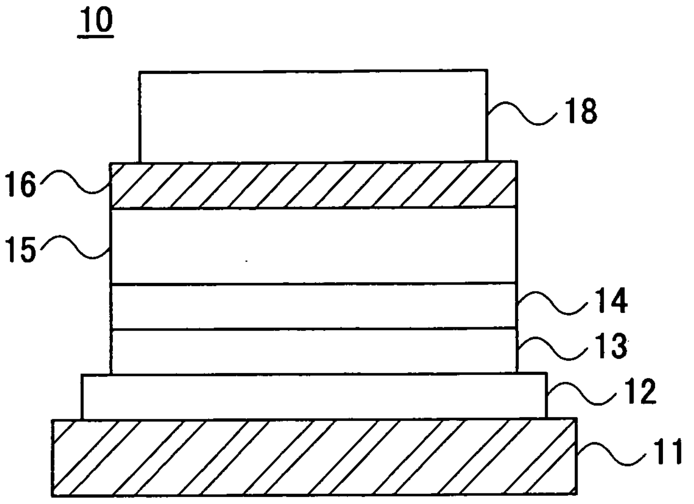


FIG.2

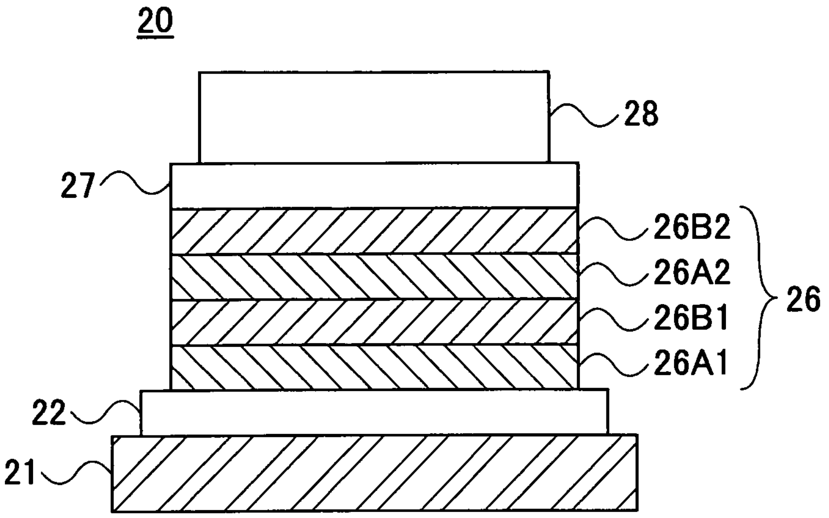


FIG.3

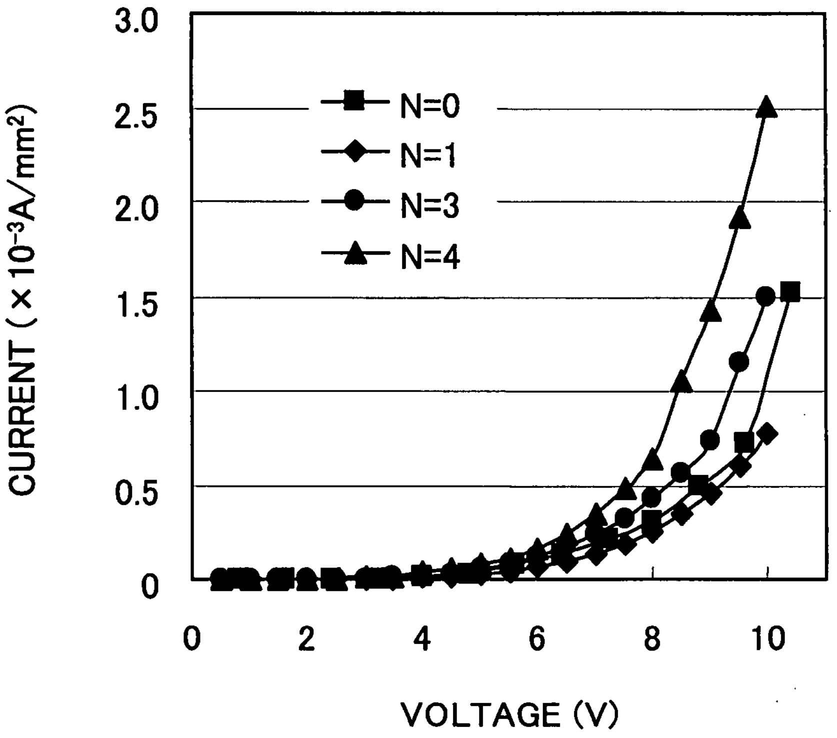

3.0
2.5
2.0
1.5
1.0
0.5
0
CURRENT (× 10-3A/mm2)
N=0
N=1
N=3
N=4
0
2
4
6
8
10
VOLTAGE (V)

FIG.4

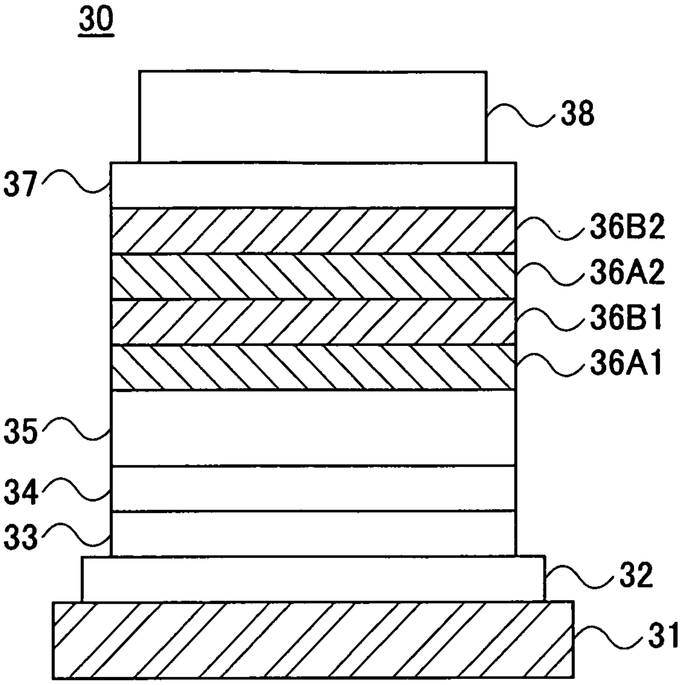


FIG.5

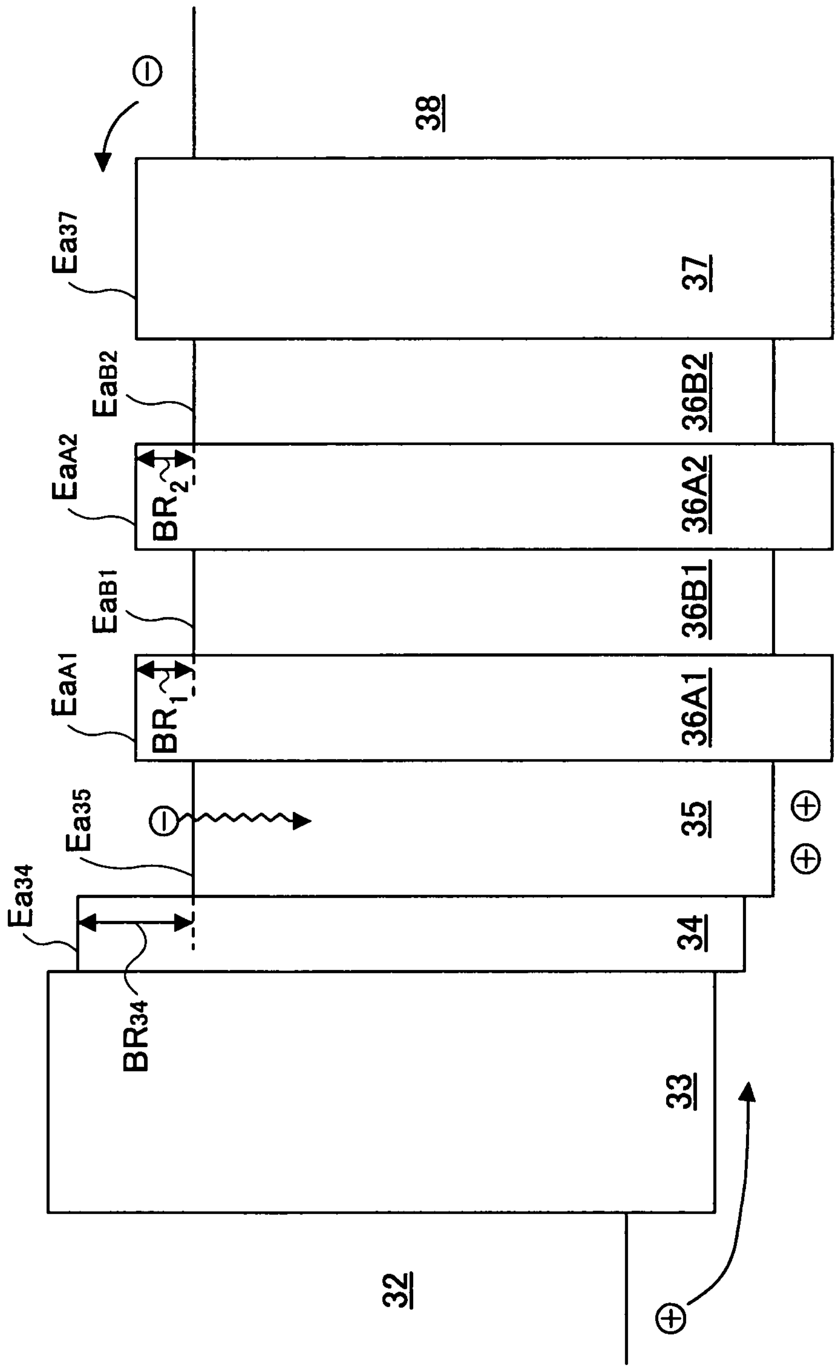

⊖
38
Ea37
37
EaB2
36B2
EaA2
BR2
36A2
EaB1
36B1
EaA1
BR1
36A1
Ea35
⊖
35
⊕
⊕
Ea34
BR34
34
33
32
⊕

FIG.6

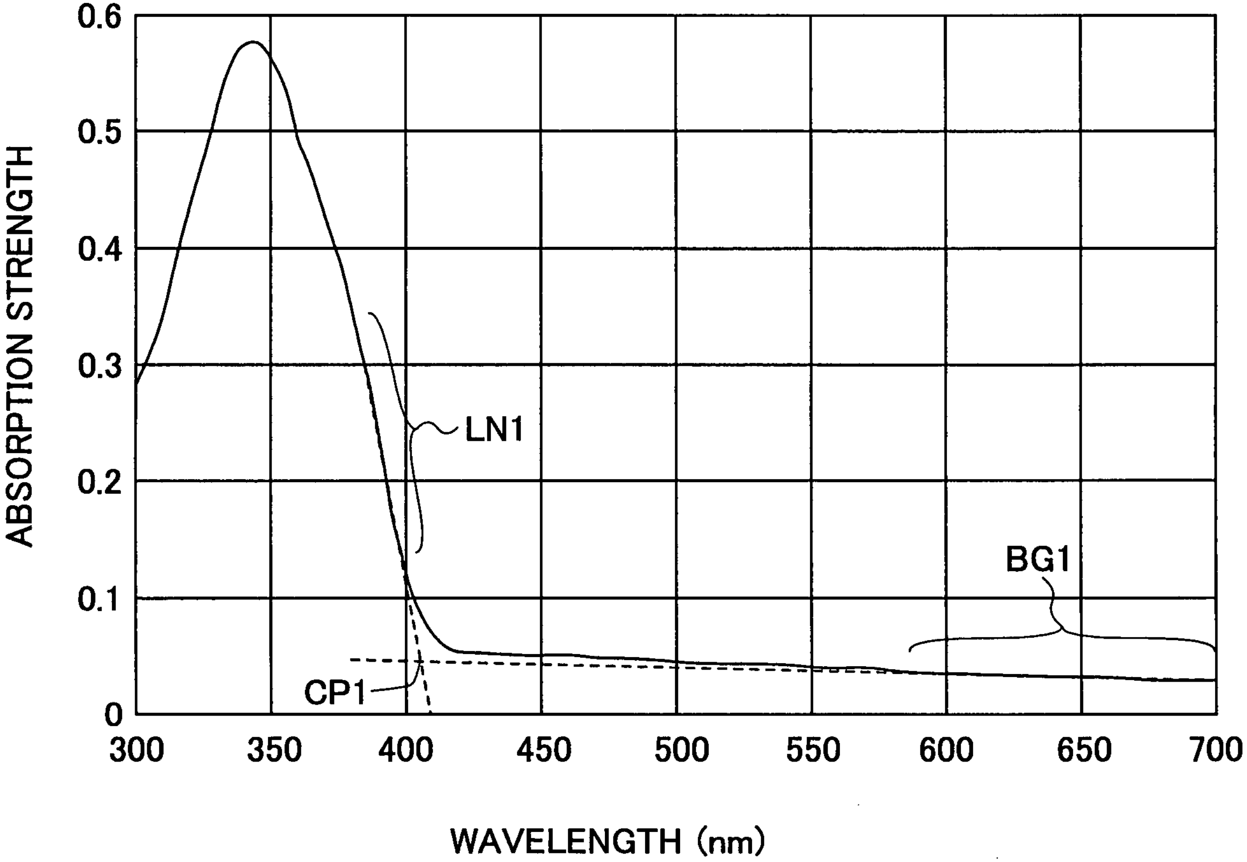

0.6
0.5
0.4
0.3
0.2
0.1
0
ABSORPTION STRENGTH
300
350
400
450
500
550
600
650
700
WAVELENGTH (nm)
LN1
CP1
BG1

FIG.7

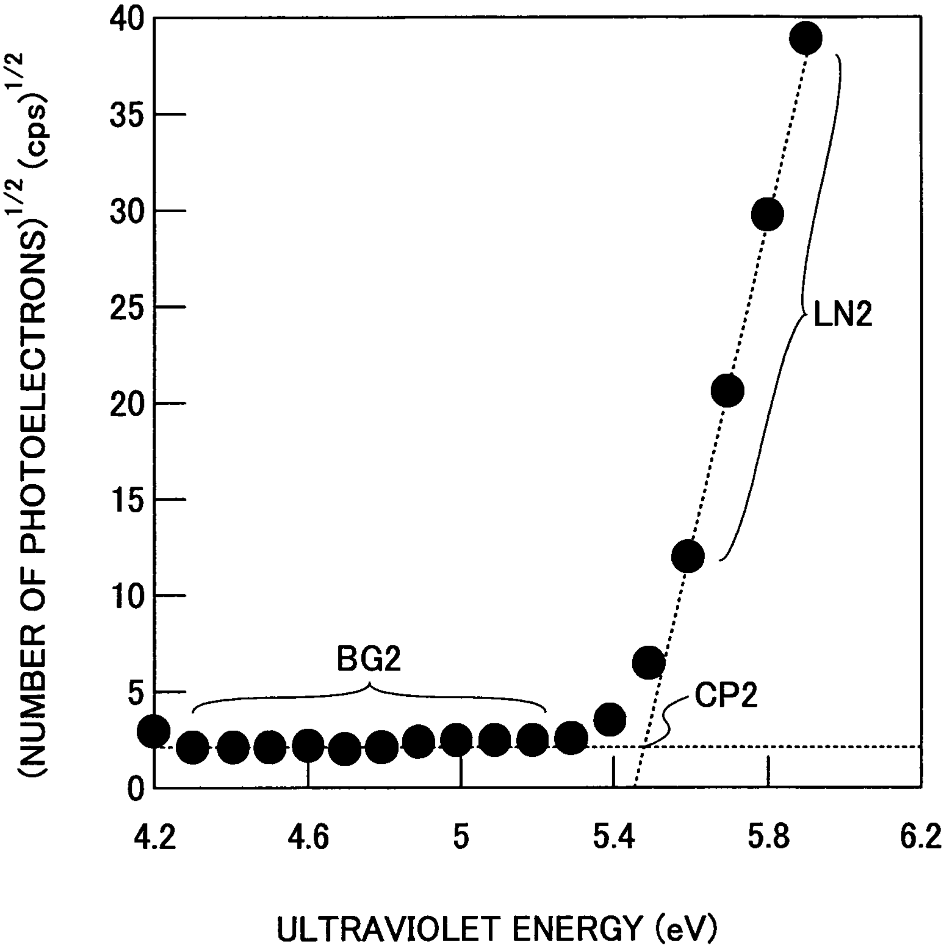

(NUMBER OF PHOTOELECTRONS)$^{1/2}$ (cps)$^{1/2}$
40
35
30
25
20
15
10
5
0
4.2
4.6
5
5.4
5.8
6.2
ULTRAVIOLET ENERGY (eV)
BG2
CP2
LN2

FIG.8

LAYER MATERIAL	ELECTRON AFFINITY Ea(eV)	ENERGY GAP Eg(eV)	IONIZATION POTENTIAL Ip(eV)
TYG-201 LAYER	3.20	2.40	5.60
TYE-704 LAYER	2.97	2.76	5.73
Alq3 LAYER	3.07	2.71	5.78
α-NPD LAYER	2.42	3.04	5.46
2-TNATA LAYER	2.19	3.00	5.19

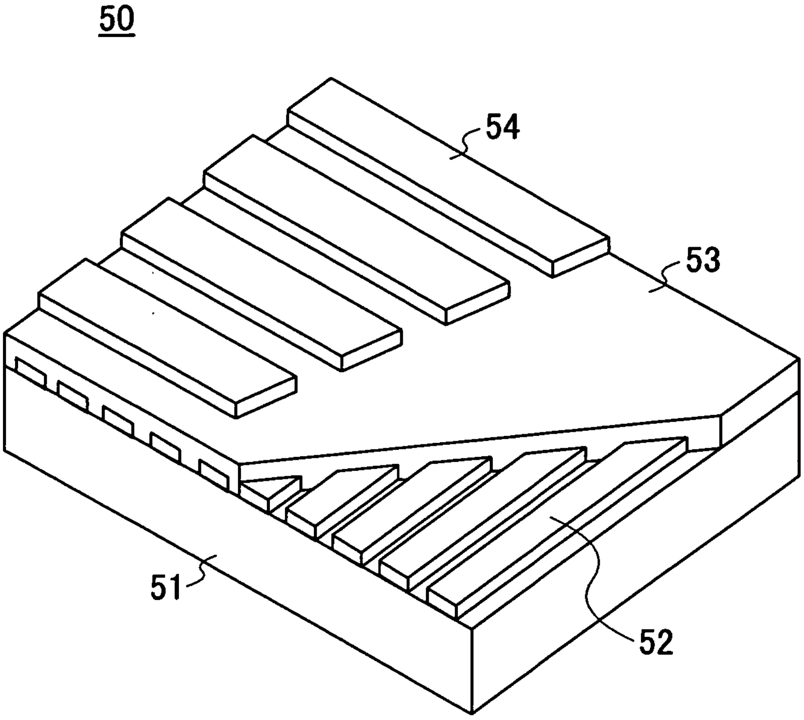
FIG.9

	FIRST EXAMPLE	SECOND EXAMPLE	THIRD EXAMPLE	FIRST COMPARATIVE EXAMPLE	SECOND COMPARATIVE EXAMPLE
CATHODE	LiF/Al	LiF/Al	LiF/Al	LiF/Al	LiF/Al
ELECTRON TRANSPORT LAYER	TYE704(20nm)	TYE704(20nm)	TYE704(20nm)	TYE704(20nm)	TYE704(20nm)
ELECTRON TRANSPORT MULTILAYER BODY	NUMBER OF REPETITION TYG201(15nm) TYE704(15nm) } 2	NUMBER OF REPETITION TYG201(10nm) TYE704(10nm) } 3	NUMBER OF REPETITION TYG201(7.5nm) TYE704(7.5nm) } 4	TYG201(50nm)	NUMBER OF REPETITION TYG201(30nm) TYE704(30nm) } 1
LUMINESCENT LAYER	TYG201(20nm)	TYG201(20nm)	TYG201(20nm)	TYG201(50nm)	TYG201(20nm)
HOLE TRANSPORT LAYER	α-NPD(10nm)	α-NPD(10nm)	α-NPD(10nm)	α-NPD(10nm)	α-NPD(10nm)
HOLE INJECTION LAYER	2-TANA(40nm)	2-TANA(40nm)	2-TANA(40nm)	2-TANA(40nm)	2-TANA(40nm)
ANODE	ITO	ITO	ITO	ITO	ITO
LUMINESCENCE THRESHOLD (V)	3	3	3	4	4
CURRENT DENSITY (mA/cm^2)	10.86	11.09	11.44	11.73	10.03
LUMINANCE (cd/m^2)	913	1075	1017	967	750
LUMINOUS EFFICIENCY (cd/m)	8.40	9.70	8.89	8.25	7.48

FIG.10

	FOURTH EXAMPLE	FIFTH EXAMPLE	THIRD COMPARATIVE EXAMPLE	FOURTH COMPARATIVE EXAMPLE
CATHODE	LiF/Al	LiF/Al	LiF/Al	LiF/Al
ELECTRON TRANSPORT LAYER	TYE704(20nm)	TYE704(20nm)	TYG201(50nm)	TYE704(20nm)
ELECTRON TRANSPORT MULTILAYER BODY	NUMBER OF REPETITION Alq3(10nm) TYE704(10nm) } 3	NUMBER OF REPETITION Alq3(7.5nm) TYE704(7.5nm) } 4	Alq3(30nm)	NUMBER OF REPETITION Alq3(30nm) TYE704(30nm) } 1
LUMINESCENT LAYER	TYG201(20nm)	TYG201(20nm)	TYG201(20nm)	TYG201(20nm)
HOLE TRANSPORT LAYER	α-NPD(10nm)	α-NPD(10nm)	α-NPD(10nm)	α-NPD(10nm)
HOLE INJECTION LAYER	2-TANA(40nm)	2-TANA(40nm)	2-TANA(40nm)	2-TANA(40nm)
ANODE	ITO	ITO	ITO	ITO
LUMINESCENCE THRESHOLD (V)	5	5	5	5
CURRENT DENSITY (mA/cm^2)	13.22	13.72	15.84	14.48
LUMINANCE (cd/m^2)	994	1021	1058	1005
LUMINOUS EFFICIENCY (cd/m)	7.52	7.44	6.68	6.75

FIG.11



INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP03/03027

A. CLASSIFICATION OF SUBJECT MATTER
Int.Cl^7 H05B33/22, H05B33/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
Int.Cl^7 H05B33/00-33/28

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

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2003
Kokai Jitsuyo Shinan Koho	1971-2003	Toroku Jitsuyo Shinan Koho	1994-2003

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

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 6-36877 A (Toshiba Corp.), 10 February, 1994 (10.02.94), Page 3, column 3, line 18 to column 4, line 35; page 4, column 5, line 41 to column 6, line 39; page 6, column 10, line 14 to page 7, column 11, line 15; Figs. 7, 12 to 13 & EP 553950 A & US 5343050 A	1-7,11
A	JP 7-188649 A (Fuji Electric Co., Ltd.), 25 July 1995 (25.07.95), Claims 3 to 5; page 4, column 6, line 37 to page 5, column 7, line 9; Fig. 1 (Family: none)	1-7,11

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E"	earlier document but published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search 28 April, 2003 (28.04.03)	Date of mailing of the international search report 13 May, 2003 (13.05.03)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (July 1998)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP03/03027

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 2000-82585 A (TDK Corp.), 21 March, 2000 (21.03.00), Page 5, column 5, lines 28 to 36 (Family: none)	1-7,11

Form PCT/ISA/210 (continuation of second sheet) (July 1998)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP03/03027

Box I Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☒ Claims Nos.: 8-10
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
The inventions of claims 8-10 are not supported by the description of the application.
The technical matter that the electron affinity of a hole transport layer (continued to extra sheet)

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest ☐ The additional search fees were accompanied by the applicant's protest.
☐ No protest accompanied the payment of additional search fees.

Form PCT/ISA/210 (continuation of first sheet (1)) (July 1998)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP03/03027

Continuation of Box No.I-2 of continuation of first sheet(1)

is "greater" than that of a luminous layer, stated in claim 9, is also not supported by the description. (Though the technical matter that the electron affinity is "smaller" than that of a luminous layer is described in the description, p.7.)

专利名称(译)	有机电致发光器件和有机电致发光显示器		
公开(公告)号	EP1603369A1	公开(公告)日	2005-12-07
申请号	EP2003712695	申请日	2003-03-13
[标]申请(专利权)人(译)	富士通株式会社		
申请(专利权)人(译)	FUJITSU LIMITED		
当前申请(专利权)人(译)	UDC IRELAND LIMITED		
[标]发明人	NAKAYAMA MASAYA FUJITSU LIMITED ITAI YUICHIRO FUJITSU LIMITED KINOSHITA MASARU FUJITSU LIMITED KODAMA JUN FUJITSU LIMITED		
发明人	NAKAYAMA, MASAYA, FUJITSU LIMITED ITAI, YUICHIRO, FUJITSU LIMITED KINOSHITA, MASARU, FUJITSU LIMITED KODAMA, JUN, FUJITSU LIMITED		
IPC分类号	H01L51/50 H01L51/00 H05B33/00 H05B33/14 H05B33/22		
CPC分类号	H01L51/5048 H01L51/0059 H01L51/006 H01L51/0077 H01L51/0078 H01L51/0081		
其他公开文献	EP1603369B1 EP1603369A4		
外部链接	Espacenet		

摘要(译)

有机EL器件包括透明基板（31），阳极（32），空穴注入层（33），空穴传输层（34），发光层（35），电子传输多层体（36），阴极侧电子传输层（37）和阴极（38）依次形成在基板（31）上。电子传输多层体（36）包括交替层叠的两种电子传输层（36A，36B），它们具有彼此不同的电子亲和力。增加从阴极（38）注入的电子流量以与空穴电流量平衡，并提高发光效率。

FIG.5