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(54) ORGANIC ELECTROLUMINESCENT DEVICES AND DISPLAY DEVICE EMPLOYING THE SAME

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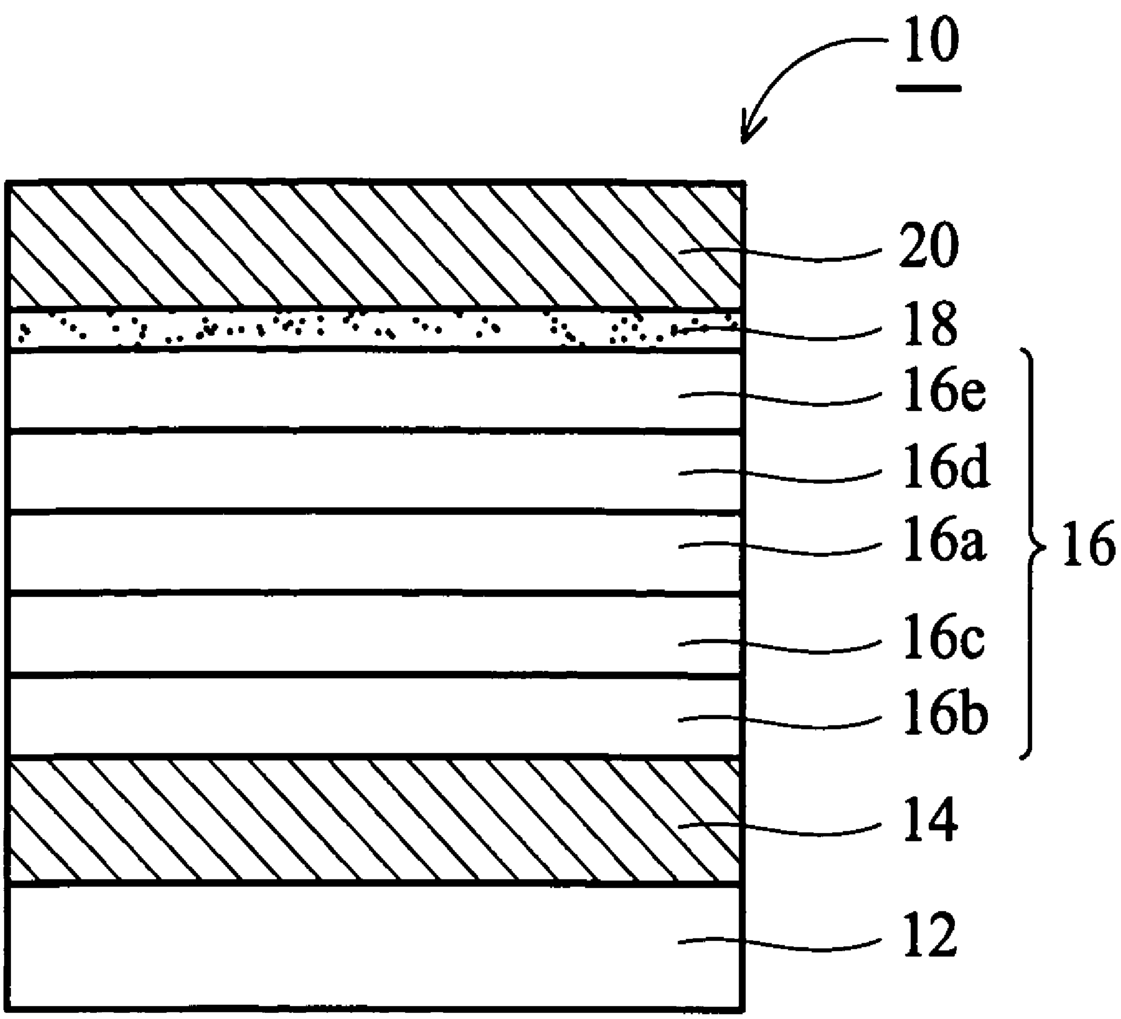
(57) **ABSTRACT**

An organic electroluminescent device and a display device including the same. The organic electroluminescent device can be a top-emission or dual emission organic electroluminescent device and comprises at least one substrate, an anode electrode on the substrate, an electroluminescent material layer on the anode, a buffer layer on the electroluminescent material layer, and a transparent cathode electrode on the buffer layer, wherein the buffer layer comprises n-type semiconductor material.



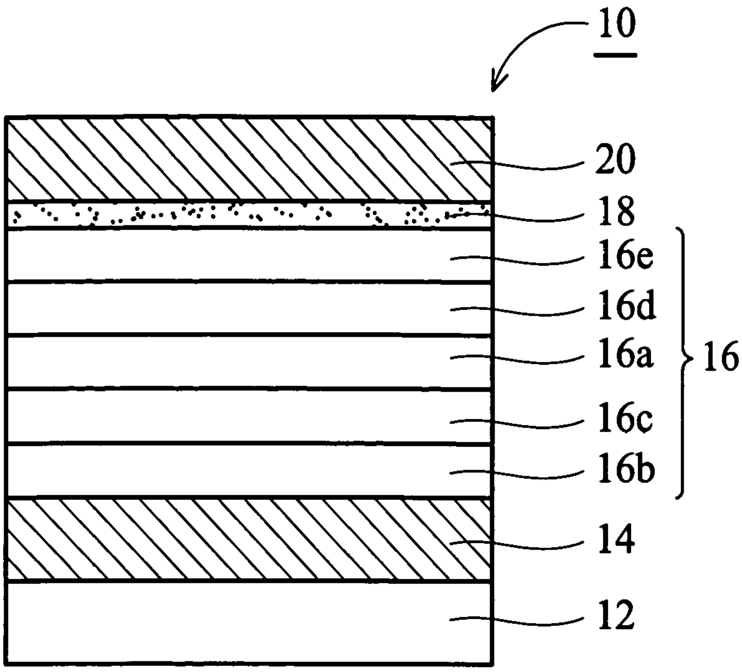

10
20
18
16e
16d
16a
16c
16b
16
14
12

FIG. 1

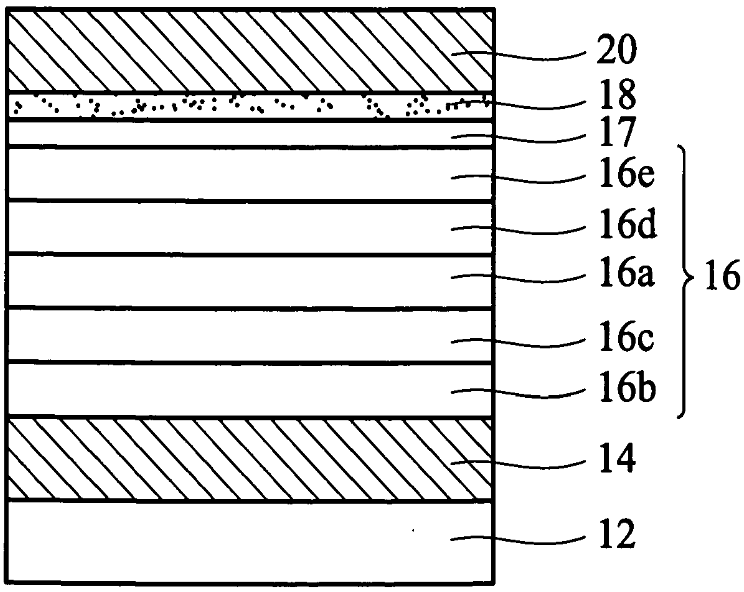

20
18
17
16e
16d
16a
16c
16b
16
14
12

FIG. 2

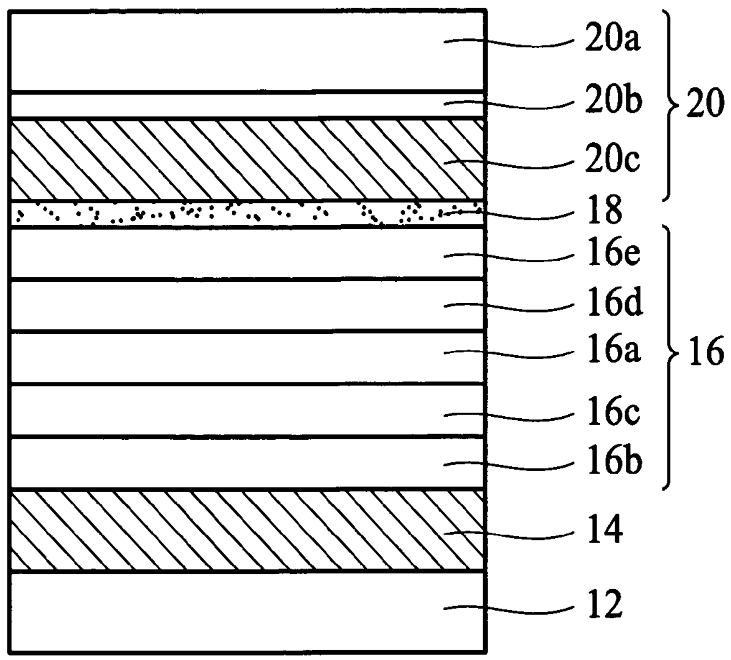

20a
20b
20c
20
18
16e
16d
16a
16c
16b
16
14
12

FIG. 3

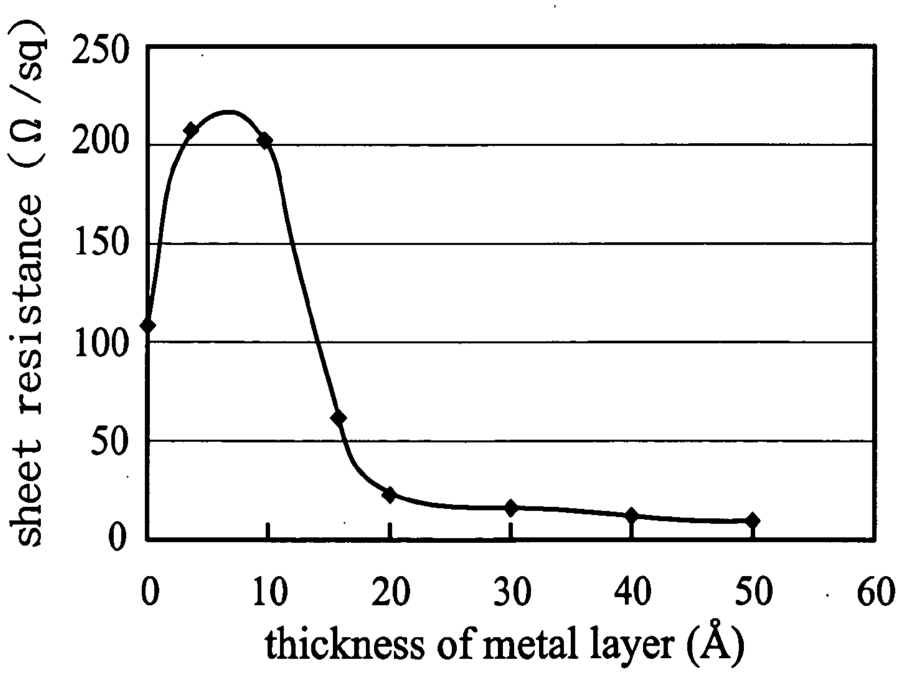
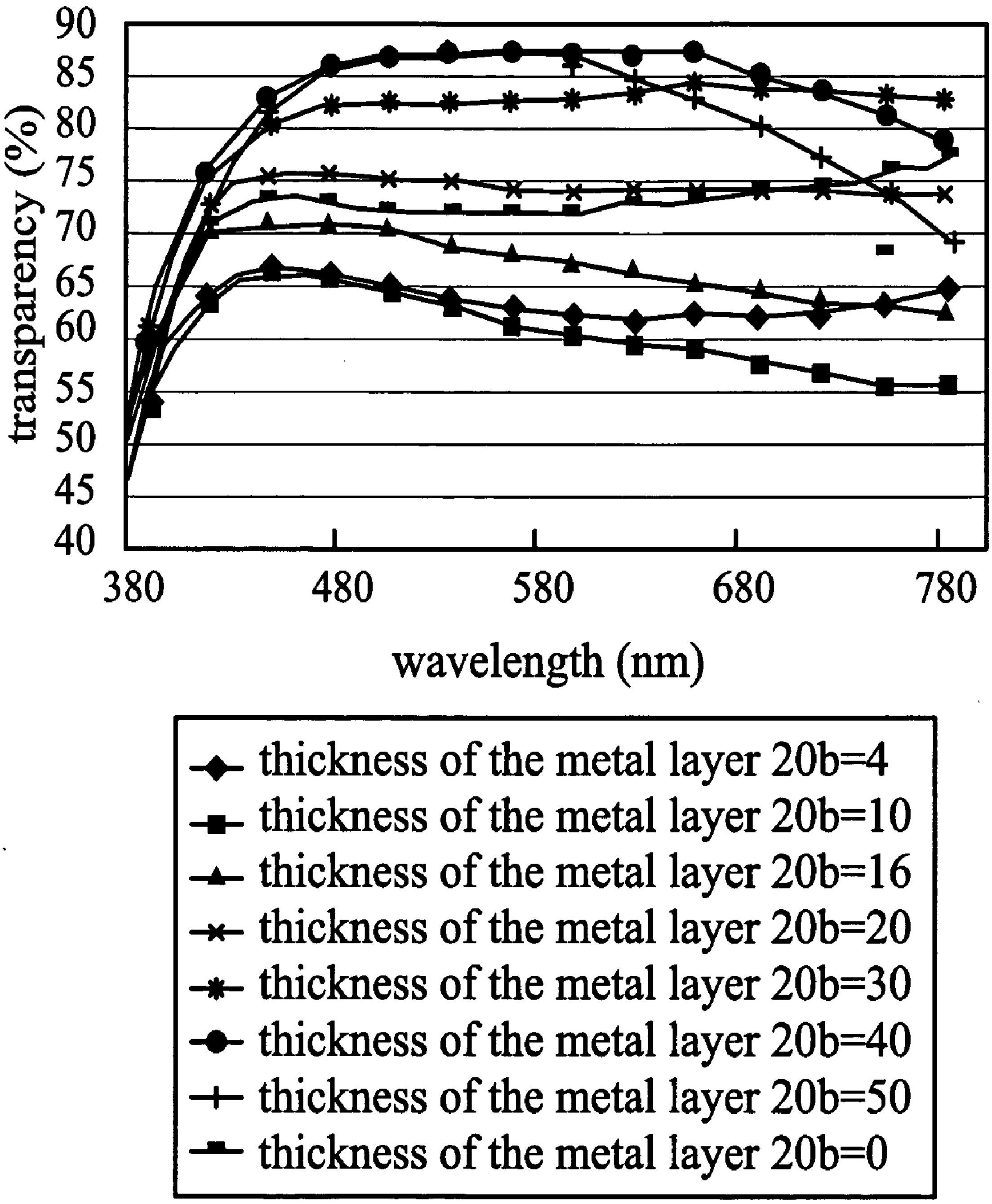

sheet resistance (Ω/sq)
250
200
150
100
50
0
0
10
20
30
40
50
60
thickness of metal layer (Å)

FIG. 4



90
85
80
75
70
65
60
55
50
45
40
transparency (%)
380
480
580
680
780
wavelength (nm)
thickness of the metal layer 20b=4
thickness of the metal layer 20b=10
thickness of the metal layer 20b=16
thickness of the metal layer 20b=20
thickness of the metal layer 20b=30
thickness of the metal layer 20b=40
thickness of the metal layer 20b=50
thickness of the metal layer 20b=0

FIG. 5

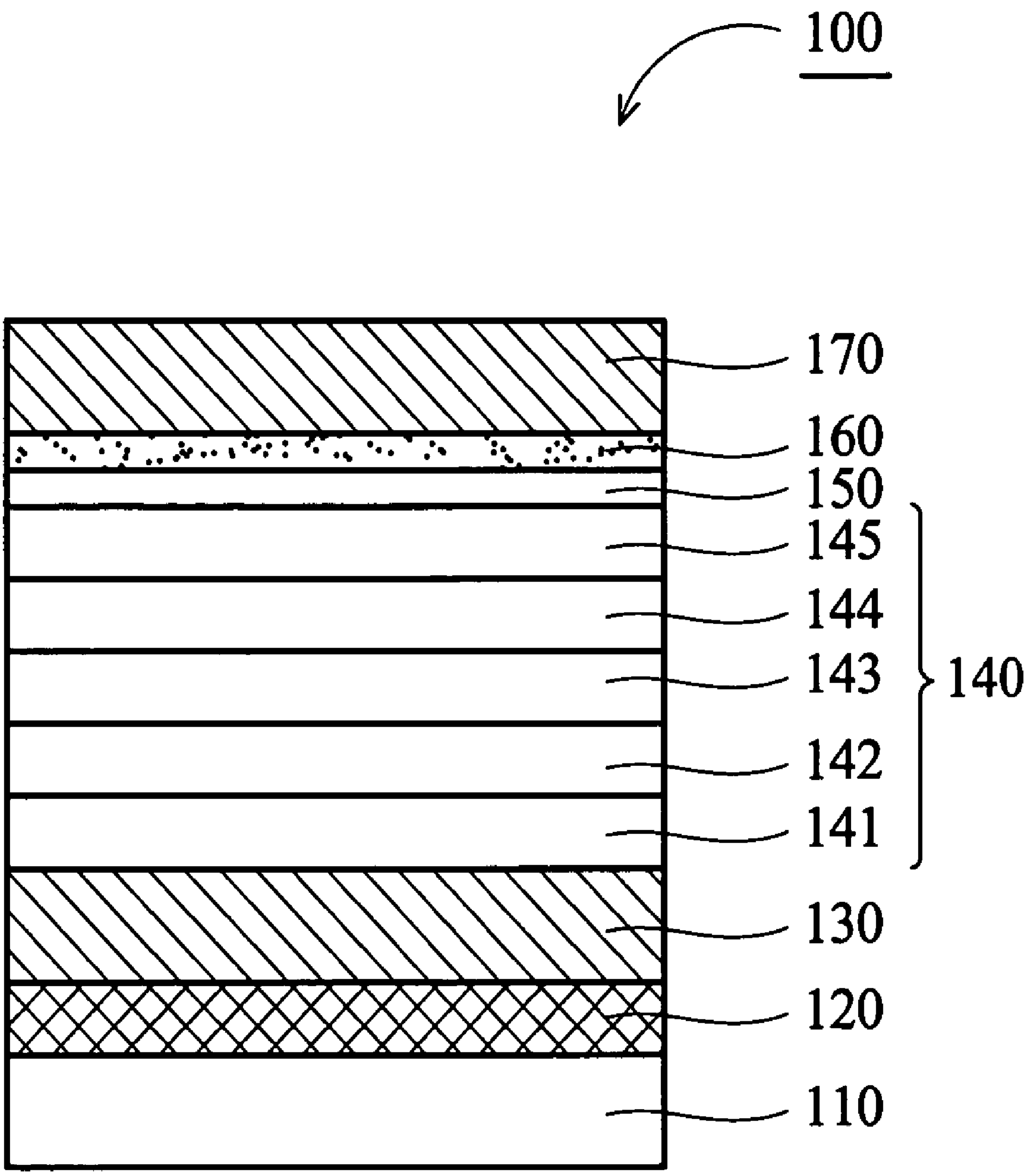


FIG. 6

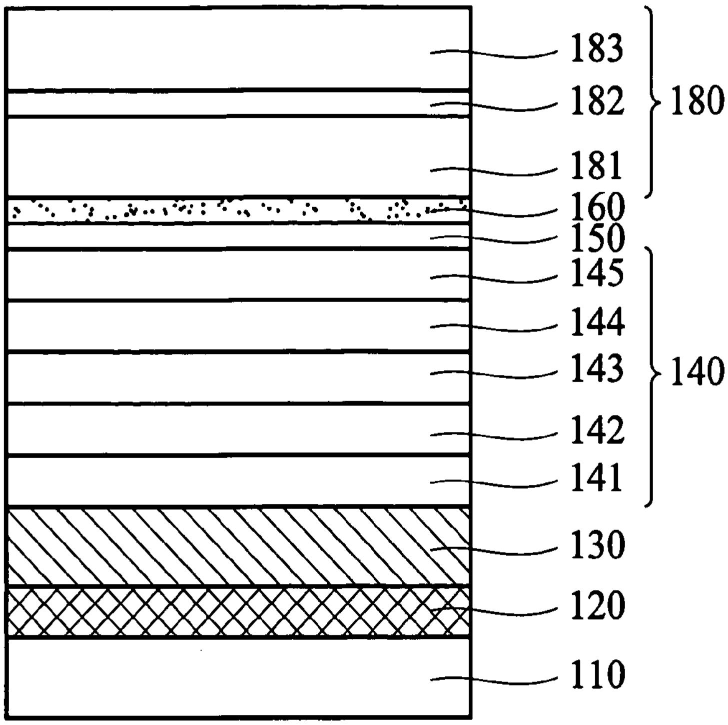

183
182
180
181
160
150
145
144
143
140
142
141
130
120
110

FIG. 7

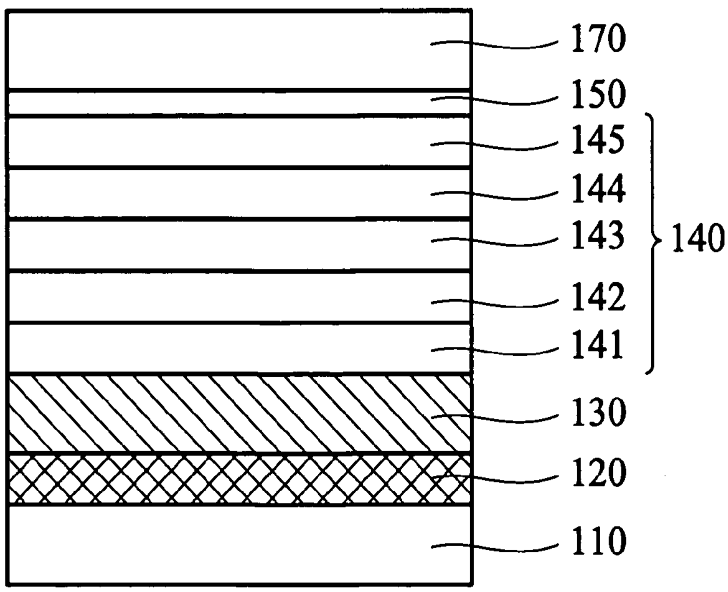

170
150
145
144
143
140
142
141
130
120
110

FIG. 8

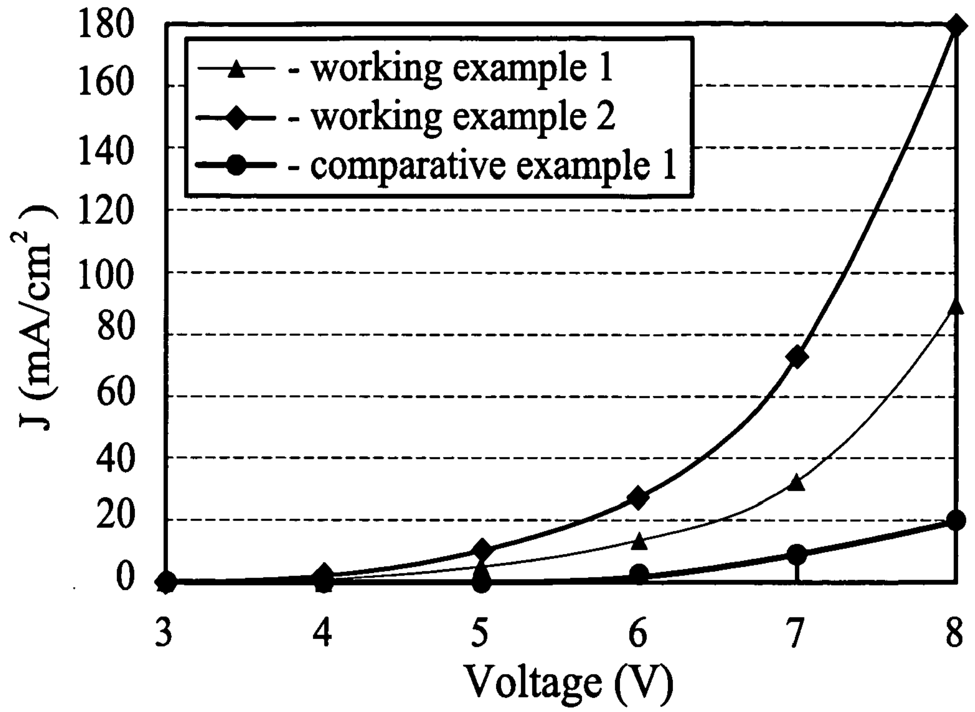

180
160
140
120
100
80
60
40
20
0
J (mA/cm²)
- working example 1
- working example 2
- comparative example 1
3
4
5
6
7
8
Voltage (V)

FIG. 9

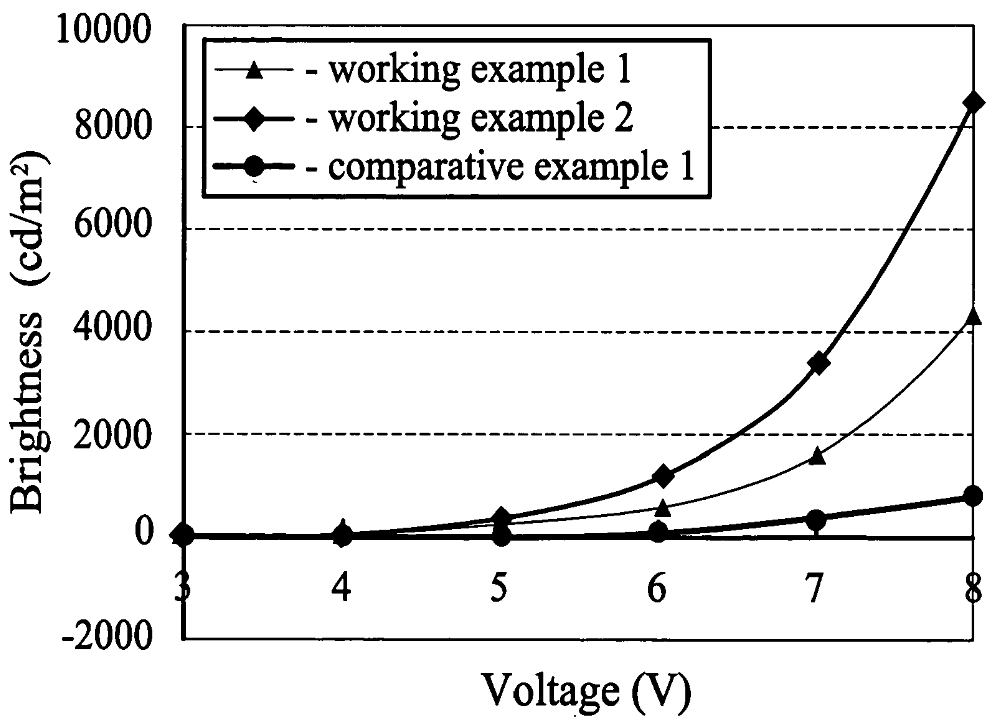

10000
8000
6000
4000
2000
0
-2000
Brightness (cd/m²)
- working example 1
- working example 2
- comparative example 1
3
4
5
6
7
8
Voltage (V)

FIG. 10

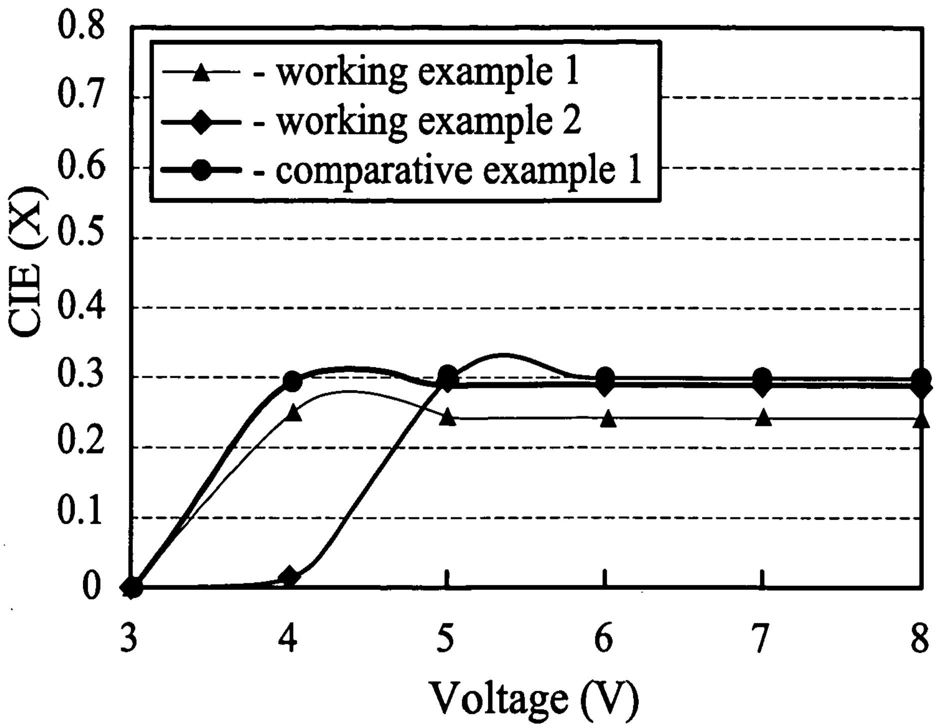

0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
CIE (X)
- working example 1
- working example 2
- comparative example 1
3
4
5
6
7
8
Voltage (V)

FIG. 11

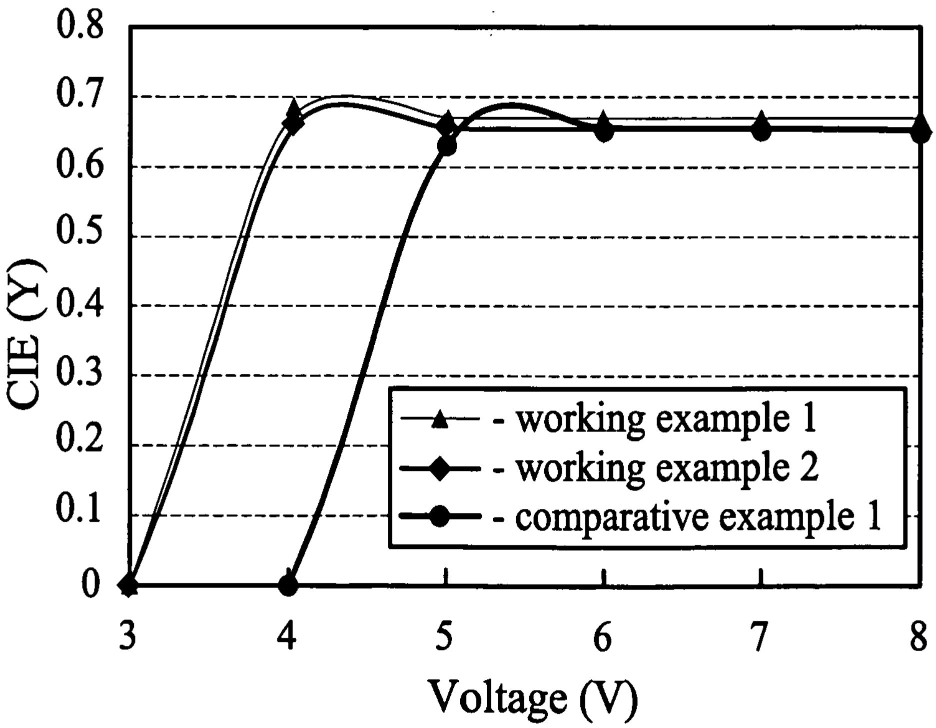

0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
CIE (Y)
- working example 1
- working example 2
- comparative example 1
3
4
5
6
7
8
Voltage (V)

FIG. 12

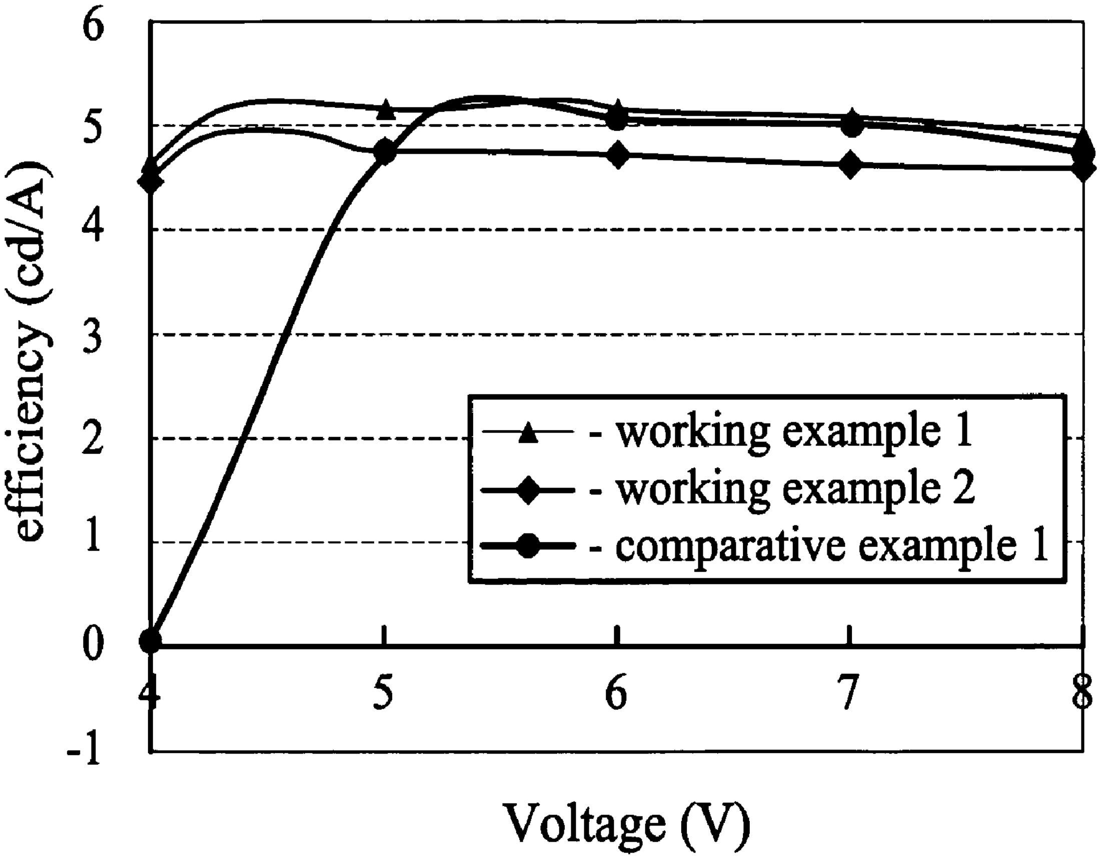

6
5
4
3
2
1
0
-1
efficiency (cd/A)
4
5
6
7
8
Voltage (V)
- working example 1
- working example 2
- comparative example 1

FIG. 13

ORGANIC ELECTROLUMINESCENT DEVICES AND DISPLAY DEVICE EMPLOYING THE SAME

BACKGROUND

[0001] The present invention relates to an organic electroluminescent device and, more particularly, to a top-emission or dual emission organic electroluminescent device.

[0002] Recently, with the development and wide application of electronic products, such as mobile phones, PDA, and notebook computers, there has been increasing demand for flat display elements which consume less electric power and occupy less space. Among flat panel displays, organic electroluminescent devices are self-emitting, and highly luminous, with wider viewing angle, faster response, and a simple fabrication process, making them the industry display of choice.

[0003] An organic light-emitting diode (OLED) is an increasingly popular light-emitting diode that uses an organic electroluminescent layer. According to the direction from which the light is obtained, organic electroluminescent devices are classified as bottom-emission, top-emission, or dual emission organic electroluminescent devices.

[0004] Top-emission and dual emission organic electroluminescent devices comprise a transparent cathode and an anode and organic electroluminescent layers, wherein light emitted by the organic electroluminescent devices passes through the transparent cathode outward. In general, methods for fabricating the transparent cathode comprise forming a thin metal layer, such as Mg, Ag, or Al, by thermal evaporation or forming a transparent conductive layer, such as ITO or IZO, by sputtering. Since the thin metal layers formed by thermal evaporation have inferior adhesion to electroluminescent materials and lower transparency, ITO or IZO electrode layers formed by sputtering are widely used due to higher transparency.

[0005] In sputtering of a transparent conductive layer, the top surface of the underlying electroluminescent layer oxidizes, deteriorates, and roughens by ion bombardment during the sputter deposition. Thus, the energy barriers of the interfaces between the transparent cathode and the electroluminescent layers increase, and the carrier movement between layers is less likely to occur, resulting in a higher operating voltage and shorter lifetime.

[0006] Accordingly, an organic electroluminescent device having an organic material or polymer layer, formed on the underlying electroluminescent layers as buffer layer has been developed to prevent damage to the underlying electroluminescent layers and solve problems of conventional technology. For example, U.S. Pat. No. 6,402,579 discloses a MEH-PPV layer and U.S. Pat. No. 6,420,031 discloses a CuPc layer serving as buffer layer. The aforementioned method avoids the underlying electroluminescent layer deterioration. However, roughness of the interface between the buffer layer and the transparent cathode is still increased.

[0007] In general, after sputtering, the transparent cathode is subjected to an annealing process to reduce sheet resistance to 30 Ω/sq. Since the electroluminescent layers are formed before the transparent cathode in the fabrication process of top-emission and dual emission organic electroluminescent devices, the annealing process is inhibitive, to prevent damage to the electroluminescent layers. Thus, the transparent cathode has a sheet resistance of 100 Ω/sq, resulting in a higher operating voltage and reduced luminance efficiency.

[0008] Therefore, it is necessary to develop organic electroluminescent devices with novel structure and low operating voltage in order to accommodate in to practical use.

SUMMARY

[0009] Embodiments of the invention provide an organic electroluminescent device, comprising a substrate, a first electrode such as an anode, an electroluminescent layer, a buffer layer, and a second electrode such as a transparent cathode, wherein the buffer layer comprises an n-type semiconductor material. The n-type semiconductor material with hole-transport properties prevents damage to the underlying electroluminescent layers. Furthermore, due to the sufficient rigidity of the n-type semiconductor material, the roughness of the interface between the n-type semiconductor buffer layer and the transparent cathode is minimized enough to avoid large leakage current or point discharge causing pixel defects. The organic electroluminescent devices can be top-emission or dual emission organic electroluminescent device.

[0010] According to some embodiment of the invention, the transparent cathode comprises a transparent electrode layer, a metal layer, and a protection layer in sequence. The provided transparent cathode has low sheet resistance, reducing the bias voltage of common drain electrode (transparent cathode) of a display panel.

[0011] Further provided is a display device, such as an organic electroluminescent display device, comprising the disclosed organic electroluminescent device and a power source element, wherein the power source element electrically couples to the organic electroluminescent device.

[0012] A detailed description is given in the following with reference to the accompanying drawing.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013] The invention can be more fully understood by reading the subsequent detailed description in conjunction with the examples and references made to the accompanying drawings, wherein:

[0014] **FIG. 1** is a cross section of an organic electroluminescent device according to an embodiment of the invention.

[0015] **FIG. 2** is a cross section of an organic electroluminescent device according to embodiments of the invention.

[0016] **FIG. 3** is a cross section of an organic electroluminescent device according to embodiments of the invention.

[0017] **FIG. 4** is a graph plotting thickness of the metal layer **20***b* shown in **FIG. 3** against sheet resistance of the cathode electrode **20** shown in **FIG. 3**.

[0018] **FIG. 5** is a graph plotting thickness of the metal layer **20***b* shown in **FIG. 3** against transparency of the cathode electrode **20** shown in **FIG. 3**.

[0019] **FIG. 6** is a cross section of an organic electroluminescent device according to Working Example 1.

[0020] **FIG. 7** is a cross section of an organic electroluminescent device according to Working Example 2.

[0021] **FIG. 8** is a cross section of an organic electroluminescent device according to Comparative Example 1.

[0022] **FIG. 9** is a graph plotting operating voltage against current density of organic electroluminescent devices as disclosed in Working Example 1, Working Example 2, and Comparative Example 1.

[0023] **FIG. 10** is a graph plotting operating voltage against brightness of organic electroluminescent devices as disclosed in Working Example 1, Working Example 2, and Comparative Example 1.

[0024] **FIG. 11** is a graph plotting operating voltage against CIE chromaticity coordinates (X axis) of organic electroluminescent devices as disclosed in Working Example 1, Working Example 2, and Comparative Example 1.

[0025] **FIG. 12** is a graph plotting operating voltage against CIE chromaticity coordinates (Y axis) of organic electroluminescent devices as disclosed in Working Example 1, Working Example 2, and Comparative Example 1.

[0026] **FIG. 13** is a graph plotting operating voltage against luminant efficiency of organic electroluminescent devices as disclosed in Working Example 1, Working Example 2, and Comparative Example 1.

DETAILED DESCRIPTION

[0027] One feature of the invention is use of a combination of an electroluminescent layer, a transparent electrode, and a n-type semiconductor buffer layer formed between the two layers. Organic electroluminescent devices of embodiments comprise at least a substrate, an anode electrode on the substrate, an electroluminescent material layer on the anode, a buffer layer on the electroluminescent material layer, and a transparent cathode electrode on the buffer layer, wherein the buffer layer comprises a n-type semiconductor material.

[0028] A method of fabricating an organic electroluminescent device according to an embodiment of the invention follows.

[0029] As shown in **FIG. 1**, a substrate **12** is provided, of an insulating material such as glass, plastic, or ceramic. Further, the substrate **12** can be a semiconductor substrate, transparent or optionally opaque, specifically a transparent substrate when the organic electroluminescent device **10** is a dual emission organic electroluminescent device, and an opaque substrate when the organic electroluminescent device **10** is a top-emission organic electroluminescent device.

[0030] A first electrode such as an anode electrode **14** is formed on the substrate **12**, and can be a transparent electrode, metal electrode, or combinations thereof, comprising indium tin oxide (ITO), indium zinc oxide (IZO), aluminum zinc oxide (AZO), zinc oxide (ZnO), Li, Mg, Ca, Al, Ag, In, Au, Ni, Pt, or alloys thereof, formed by a method such as sputtering, electron beam evaporation, thermal evaporation, or chemical vapor deposition. In an embodiment of the invention, a reflective layer is formed between the substrate **12** and the anode electrode **14**.

[0031] An electroluminescent layer **16** is formed on the anode electrode **14**, wherein the electroluminescent layer **16** at least comprises a light emitting layer **16*a***, and can further comprises a hole injection layer **16*b***, a hole transport layer **16*c***, an electron transport layer **16*d***, and an electron injection layer **16*e***, as shown in **FIG. 1**. The electroluminescent layer **16** is organic semiconductor material such as small molecule material, polymer, or organometallic complex, and can be formed by thermal vacuum evaporation, spin coating, dip coating, roll-coating, injection-fill, embossing, stamping, physical vapor deposition, or chemical vapor deposition. The emitting layer **16*a*** comprises a light-emitting material and an electroluminescent dopant doped into the light-emitting material and can perform energy transfer or carrier trapping under electron-hole recombination in the emitting layer. The light-emitting material can be fluorescent or phosphorescent.

[0032] A buffer layer **18** is formed on the electroluminescent layer **16**, comprising an n-type semiconductor material, such as fullerene. The n-type semiconductor material has an energy gap of more than 1.0 eV. The thickness of the buffer layer is 10~2000 Å, preferably 50~1500 Å. Referring to **FIG. 2**, organic electroluminescent devices of embodiments further comprise a metal conductive layer **17** formed between the electroluminescent layer **16** and the buffer layer **18**. The conductive layer, such as Al, can have a thickness of 10~500Å.

[0033] A transparent electrode, such as a transparent cathode electrode **20**, is formed on the buffer layer **18**. It should be noted that transparent cathode electrode **20** is formed directly on the buffer layer **18**. The transparent cathode electrode **20** can comprise ITO, IZO, AZO, ZnO, GaN(gallium nitride), GaInN (gallium indium nitride), CdS(cadmium sulfide), ZnS (zinc sulfide), CdSe (cadmium selenide), or ZnSe (zinc selenide).

[0034] Furthermore, according to some embodiments of the invention, the transparent cathode electrode **20** can be a composite structure and comprise a transparent electrode **20*a*** on the buffer layer **18**, a metal layer on **20*b*** the transparent layer **20*a***, and a protective layer **20*c*** on the metal layer **20*b***. The transparent electrode **20**a can comprise ITO, IZO, AZO, ZnO, GaN, GaInN, CdS, ZnS, CdSe, or ZnSe. The protective layer **20*c*** can be transparent conductive material, conductive polymer material, or semiconductor material with wide energy gap, such as ITO, IZO, AZO, ZnO, GaN, GaInN, CdS, ZnS, CdSe, ZnSe, polypyrrole, polyaniline, or polythiophene. In order to reduce sheet resistance of the transparent cathode electrode **20**, the metal layer **20*b*** preferably has electrical conductivity exceeding 105 $cm^{-1}\Omega^{31\ 1}$. For example, the metal layer **20*b*** can be made of Ag. The relationship between thickness of the metal layer **20*b*** and sheet resistance of the cathode electrode **20** is shown in **FIG. 4**, and the relationship between thickness of the metal layer **20*b*** and transparency of the cathode electrode **20** is shown in **FIG. 5**. Accordingly, a metal layer **20*b*** with a thickness of 20~50 Å exhibits superior performance.

WORKING EXAMPLE 1

[0035] As shown in **FIG. 6**, the organic electroluminescent device **100** used here was a top-emission organic electroluminescent device. A reflective layer **120** was formed on a glass substrate **110**, of Ti with a thickness of 500 Å. An ITO electrode **130** with a thickness of 750 Å, a hole injection layer **141**, a hole transport layer **142**, an emitting layer **143**, an electron transport layer **144**, an electron injection layer **145**, a thin conductive layer **150**, a buffer layer **160**, and a transparent cathode electrode **170** were all formed subsequently on the reflection layer **120**. The a hole injection layer **141**, hole transport layer **142**, emitting layer **143**, electron transport layer **144**, and electron injection layer **145** comprise an electroluminescent layer **140**.

[0036] For purposes of clarity, materials and layers formed therefrom are described as follows.

[0037] The hole injection layer **141**, at a thickness of 200 Å, consisted of CuPc (copper phthalocyanine). The hole transport mixed layer **142**, at a thickness of 400 Å, consisted of NPB (N,N'-di-1-naphthyl-N,N'-diphenyl-1,1'-biphenyl-1, 1'-biphenyl-4,4'-diamine). The emitting layer **143**, at thickness of 300 Å, consisted of C545T (10-(2-Benzothiazolyl)-2,3,6,7-tetrahydro-1,1,7,7-tetramethyl-1H,5H,11H-(1)-benzopyropyrano(6,7-8-i,j)quinolizin-11-one) as dopant, and NPB and Alq_3(tris (8-hydroxyquinoline) aluminum) as light-emitting materials, wherein the weight ratio between NBP and Alq_3 was 1:1 and the dopant amount of C545T 1.1% by weight. The electron transport layer **144**, at a thickness of 400 Å, consisted of Alq_3. The electron injection layer **145**, at a thickness of 10 Å, consisted of LiF (lithium fluoride). The thin conductive layer **150**, at a thickness of 20 Å, consisted of Al. The buffer layer **160**, at a thickness of 50 Å, consisted of fullerene. The tansparent cathode electrode **170**, at a thickness of 800 Å, consisted of IZO.

[0038] The structure of the organic electroluminescent device **100** was:

[0039] Ti 500 Å/ITO 750 Å/CuPc 200 Å/NPB 400 Å/Alq_3:NPB=1:1):C545T1.1% 300 Å/Alq_3 400 Å/LiF 10 Å/Al 20 Å/fullerene 50 Å/IZO 800 Å

[0040] The measured results of optical properties for the oganic electroluminescent device, as described in Working Eample 1, are shown in Table 1.

TABLE 1

Optical Properties for Working Example 1

Voltage (V)	Current Density (mA/cm^2)	Brightness (cd/m^2)	CIE chromaticity coordinates (X axis)	CIE chromaticity coordinates (Y axis)	Peak Wave-length (nm)
1	0	0	0	0	0
2	0	0	0	0	0
3	0.04	0	0	0	0
4	0.51	24.59	0.252	0.675	520
5	2.66	137.4	0.251	0.675	520
6	9.96	516.3	0.251	0.675	520
7	31.98	1586	0.25	0.673	520
8	87.78	4277	0.25	0.672	520

WORKING EXAMPLE 2

[0041] Working Example 2 was executed as Working Example 1 except for substitution of a composite transparent cathode electrode **180** for the transparent cathode electrode **170**. The composite transparent cathode electrode **180** comprised a transparent conductive layer **181** on the buffer layer **170**, a metal layer **182** on the transparent conductive layer **181**, and a protective layer **183** on the metal layer **182**. The transparent conductive layer **181**, at a thickness of 400 Å, consisted of IZO. The metal layer **182**, at a thickness of 20 Å, consisted of Ag. The protective layer **183**, at a thickness of 400 Å, consisted of IZO.

[0042] The structure of the organic electroluminescent device was:

[0043] Ti 500 Å/ITO 750 Å/CuPc 200 Å/NPB 400 Å/(Alq_3:NPB=1:1):C545T1.1% 300 Å/Alq_3 400 Å/LiF 10 Å/Al 20 Å/fullerene 50 Å/IZO 400 Å/Ag20 Å/IZO400 Å

[0044] The measured results of optical properties for the organic electroluminescent device, as described in Working Example 2, are shown in Table 2.

TABLE 2

Optical Properties for Working Example 1

Voltage (V)	Current Density (mA/cm^2)	Brightness (cd/m^2)	CIE chromaticity coordinates (X axis)	CIE chromaticity coordinates (Y axis)	Peak Wave-length (nm)
1	0	0	0	0	0
2	0	0	0	0	0
3	0.07	0	0	0	0
4	1.11	50.1	0.301	0.656	524
5	6.34	298.4	0.3	0.658	524
6	23.99	1124	0.299	0.658	524
7	72.05	3278	0.298	0.657	524
8	187.91	8638	0.298	0.657	524

COMPARATIVE EXAMPLE 1

[0045] Comparative Example 1 was executed as Working Example 1 except for removal of the buffer layer **160**, referring to **FIG. 8**. The structure of the organic electroluminescent device was:

[0046] Ti 500 Å/ITO 750 Å/CuPc 200 Å/NPB 400 Å/(Alq_3:NPB=1:1):C545T1.1% 300 Å/Alq_3 400 Å/LiF 10 Å/Al 20 Å/IZO 800 Å

[0047] The measured results of optical properties for the oganic electroluminescent device, as described in Comparative Example 1, are shown in Table 3.

TABLE 3

Optical Properties for Working Example 1

Voltage (V)	Current Density (mA/cm^2)	Brightness (cd/m^2)	CIE chromaticity coordinates (X axis)	CIE chromaticity coordinates (Y axis)	Peak Wave-length (nm)
1	0	0	0	0	0
2	0	0	0	0	0
3	0	0	0	0	0
4	0.05	0	0	0	0
5	0.39	18.92	0.312	0.649	528
6	1.83	91.12	0.310	0.654	528
7	6.3	310	0.309	0.655	528
8	16.79	790.2	0.308	0.655	528

[0048] FIGS. **9~13** also illustrate the differences between properties for the organic electroluminescent devices as described respectively in Working Example 1, Working Example 2, and Comparative Example 1. In FIGS. **9~10** and Table 4, the organic electroluminescent devices disclosed in Working Examples 1 and 2 have lower operating voltages compared with the conventional organic electroluminescent device disclosed in Comparative Example 1. In Working Examples 1 and 2, the n-type semiconductor buffer layer not only prevents the underlying layers form damage by sputtering, but is also rigid enough to avoid deterioration or erosion causing ion bombardment. Further, as described in Working Example 2, since the composite transparent cathode electrode **180** exhibits sheet resistance less than 30 Ω/sq, the operating voltage thereof is reduced and efficiency increased.

TABLE 4

	Under 2000nits			
	voltage(v)	efficiency (cd/A)	Power efficiency (lm/w)	CIE chromaticity coordinates
Examples 1	7.2	4.9	2.2	(0.3, 0.65)
Examples 2	6.5	4.6	2.3	(0.25, 0.67)
Comparative Examples 1	9.3	4.1	1.5	(0.3, 0.65)

[0049] In conclusion, compared with conventional top-emission or dual emission organic electroluminescent devices, stability, luminescent efficiency, and operating voltage of the organic electroluminescent devices disclosed are all significantly improved.

[0050] Moreover, since composite transparent cathode electrodes with low sheet resistance are employed, high bias-voltage problems caused by conventional organic electroluminescent devices are solved thereby.

[0051] While the invention has been described by way of example and in terms of preferred embodiment, it is to be understood that the invention is not limited thereto. It is therefore intended that the following claims be interpreted as covering all such alteration and modifications as fall within the true spirit and scope of the invention.

What is claimed is:

1. An organic electroluminescent device, comprising:

a substrate;

a first electrode on the substrate;

an electroluminescent layer disposed on the first electrode;

a buffer layer disposed on the electroluminescent layer, comprising an n-type semiconductor material; and

a second electrode formed over the buffer layer.

2. The device as claimed in claim 1, wherein the second electrode is formed in contact with the buffer layer.

3. The device as claimed in claim 1, wherein the buffer layer has a thickness of 10~2000 Å.

4. The device as claimed in claim 1, wherein the first electrode layer comprises a metal electrode or a transparent electrode.

5. The device as claimed in claim 1, wherein the substrate comprises a glass substrate, a plastic substrate, a ceramic substrate or a semiconductor substrate.

6. The device as claimed in claim 1, wherein the electroluminescent layer comprises a hole transport layer, emitting layer, electron transport layer, or combination thereof.

7. The device as claimed in claim 1, wherein the electroluminescent layer comprises small molecule or polymer material.

8. The device as claimed in claim 1, wherein the buffer layer comprises fullerene.

9. The device as claimed in claim 1, wherein the n-type semiconductor material has an energy gap exceeding 1.0 eV.

10. The device as claimed in claim 1, further comprising a conductive layer disposed between the electroluminescent layer and the buffer layer.

11. The device as claimed in claim 10, wherein the conductive layer has a thickness of 10~500 Å.

12. The device as claimed in claim 1, wherein the second electrode comprises ITO, IZO, AZO, ZnO, GaN, GaInN, CdS, ZnS, CdSe, or ZnSe.

13. The device as claimed in claim 1, wherein the second electrode comprises:

a transparent electrode disposed on the buffer layer;

a metal layer disposed on the transparent layer; and

a protective layer disposed on the metal layer.

14. The device as claimed in claim 13, wherein the transparent electrode comprises ITO, IZO, AZO, ZnO, GaN, GaInN, CdS, ZnS, CdSe, or ZnSe.

15. The device as claimed in claim 13, wherein the protective layer comprises transparent conductive material, conductive polymer material, or semiconductor material with an energy gap exceeding 1.0 eV.

16. The device as claimed in claim 13, wherein the protective layer comprises ITO, IZO, AZO, ZnO, GaN, GaInN, CdS, ZnS, CdSe, or ZnSe.

17. The device as claimed in claim 13, wherein the protective layer comprises polypyrrole, polyaniline, or polythiophene.

18. The device as claimed in claim 13, wherein the metal layer has a thickness of 10~500 Å.

19. The device as claimed in claim 13, wherein the metal layer has electric conductivity exceeding 10^5 $cm^{-1}\Omega^{-1}$.

20. The device as claimed in claim 13, wherein the metal layer comprises Ag.

21. The device as claimed in claim 13, wherein the second electrode has sheet resistance less than 30 Ω/sq.

22. A display device, comprising

an organic electroluminescent device as claimed in claim 1; and

a power source element, wherein the power source element electrically couples to the organic electroluminescent device.

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专利名称(译)	有机电致发光器件和采用该器件的显示器件		
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摘要(译)

一种有机电致发光器件和包括该器件的显示装置。有机电致发光器件可以是顶部发光或双发射有机电致发光器件，并且包括至少一个基板，基板上的阳极电极，阳极上的电致发光材料层，电致发光材料层上的缓冲层和透明的缓冲层上的阴极电极，其中缓冲层包括n型半导体材料。