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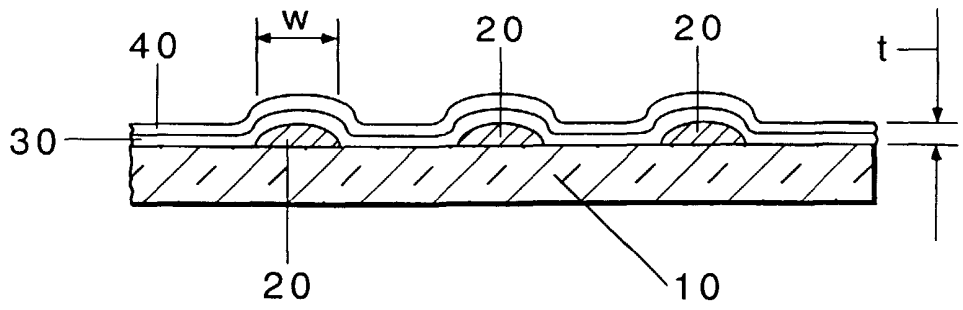
(54) Title: ELECTROLUMINESCENT DEVICE HAVING ANODE ARRAY AND REFLECTIVE CATHODE FOCUSING FEATURE




(57) Abstract: This invention is a light emitting device comprising at least one anode (20), which comprises a conductive material having a thickness greater than about 50 nm and less than about 1 mm, and a width greater than about 1 μm and less than about 1 mm; a film comprising an electroluminescent material (30) positioned next to the anode in a manner such that the film conforms to the shape of the anode, and a reflective cathode material (40) positioned on the opposite side of the film from the anode forming a concave reflective surface on the side of the film. The invention also includes a convenient method for making such a device.

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# ELECTROLUMINESCENT DEVICE HAVING ANODE ARRAY AND REFLECTIVE CATHODE FOCUSING FEATURE

This application was made with United States Government support under Cooperative Agreement No. N00421-98-C-1187. The United States Government has certain rights in the invention.

This invention relates to light emitting devices, especially electroluminescent devices having organic or polymeric light emitting layers.

Light emitting diodes typically are constructed to have an electroluminescent material (for example, inorganic phosphors or organic light emitting compounds) positioned so as to transport current between two electrodes. Two different structures have been proposed for organic light emitting devices.

In one structure, the cathode and anode are located on the same side of the light emitting material (see, for example, Smela, et al., "Planar Microfabricated Polymer Light Emitting Diodes," *Semicond. Sci. Technol.*, 13 (1998) pp. 433-439). A deficiency of this type of structure is that a negative image of the electrode structure will be seen in the emitted light from the device.

In the more conventional mode, a sandwich structure is used. This structure has a light emitting layer located between a cathode material and a transparent anode. The transparent anode is typically indium tin oxide (ITO) borne upon a transparent substrate. While ITO anodes on glass substrates function very well, such structures can be somewhat heavy depending upon the size of the structure. In addition, such structures are not flexible. Switching to a transparent plastic substrate provides a lighter and more flexible structure. Unfortunately, ITO films on plastic surfaces tend to be much rougher. Such roughness can make deposition of subsequent layers difficult and can lead to current leaks and uneven light emission in a device. Devices made with ITO can also be expensive.

Extremely thin layers (< 40 nm) of gold may also be semitransparent and, thus, have also been used (see, for example, U.S. Patent 5,965,281). Unfortunately, such very thin layers tend to have a high resistance to the current flow, which may lead either to restricting usage to very low voltage regimes or, if not, excessive operating temperatures and poorer device performance.

Thus, an approach is still desired for making inexpensive electroluminescent devices and/or flexible electroluminescent devices with reasonable device yield and performance, in which the electrode structure is not highly visible during operation.

Thus, according to a first embodiment, this invention is a light emitting device comprising

at least one anode, which comprises a conductive material having a thickness greater than about 50 nm and less than about 1 mm, and a width greater than about 1 μm and less than about 1 mm;

a film comprising an electroluminescent material positioned next to the anode in a manner such that the film conforms to the shape of the anode, and

a reflective cathode material positioned on the opposite side of the film from the anode forming a concave reflective surface on the side of the film.

Thus, although the anode may be an opaque structure, light emitted from the electroluminescent material reflects off of the concave reflective surface of the cathode, which serves as a focusing element. At a reasonable viewing distance, for example one foot or more from the device, the light converges due to the focusing action and forms a reasonably homogeneously lighted area. Varying the dimensions of the anode structure varies the optical focusing effect and will create a more or less homogeneous light-emitting area to the viewer.

According to a second embodiment, this invention is a method of making such a device comprising forming, on a substrate, an anode having a thickness in the range of about 50 nm to 1 mm and a width in the range of 1 μm to 1 mm, applying a composition comprising an electroluminescent material over the anode structure to form a film, which conforms to the shape of the anode, and applying a cathode material over the film in a manner to form a concave reflecting surface.

The file of this patent contains at least one drawing executed in color. Copies of this patent with color drawings will be provided by the Patent and Trademark Office upon request and payment of the necessary fee.

Figure 1 is a cross-section (not to scale) of a representative electroluminescent device of this invention.

Figure 2 is a top view of a representative anode pattern that may be used.

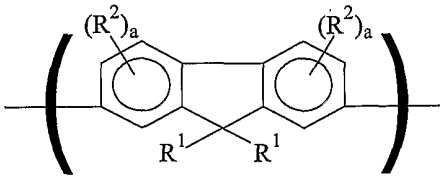
Figure 3 is a photograph of a top view of a non-optimized test device under a 4.5 V bias, which shows the focussing effect created by the topography in the device.

The devices of this invention are preferably and conveniently formed on a transparent substrate. While the substrate may be glass, polymeric substrates take full advantage of the flexibility benefits of the proposed structure, as well as the ease of making different shapes. Suitable polymeric substrates include polyesters, such as polyethylene terephthalate, polyethylenenaphthalate, polystyrene, polycarbonates, etc. Applicants contemplate that the substrate could be a temporary support, in which case it need not be transparent. For example, the anode could be formed on a surface having good release properties, such as a fluorinated polymer, the electroluminescent material and cathode material could then be applied in order over the anode. The entire structure could then be removed from the substrate and used as desired. The substrate may be of a variety of shapes, but preferably is in the form of a sheet or a slab. The sheet may have been molded, for example, into a desired shape before application of the anode and other layers.

The anode may be any suitable conductive material, preferably having a relatively high work function, that is, a work function greater than about 4.0 eV, more preferably greater than 4.5 eV, and most preferably greater than 5.0 eV. The anode structure may be a transparent material, such as ITO. However, a benefit of this invention is that transparency is not required in the anode, thus, allowing a much broader choice of anode materials. Examples of suitable materials include gold, silver, copper, nickel, platinum, etc. and various alloys of such materials. Gold is a preferred material. The anode structure could also be a composite of a conductive bus line of one material with a second material in contact with the electroluminescent film. Use of such a composite structure may enable improvement of charge injection from the anode structure into the electroluminescent film or reduction of resistance in long thin anode lines. For example, ITO could be coated over or adjacent to a conductive metal line. The dimensions of the composite anode must be adequate to provide the curvature on the reflective cathode needed to give the desired focus to diminish the appearance of the anode structure. In addition, an inherently conducting polymer or a polymer composition filled with conductive particles could be used as the anode.

The anode may be applied by any known method, such as vapor depostion, electroplating, or sputtering. The anode structure provides a base upon which other layers are built and the dimensions should be selected to give the desired focus. The anode structure has a thickness of at least 0.05 μm, preferably at least 0.1 μm, even more preferably at least 0.12 μm and most preferably at least 0.15 μm. The thickness is preferably no greater than 1000 μm, more preferably no greater than 100 μm, more preferably no greater than about 10 μm, and most preferably no greater than 1 μm. The anode structure may be of any suitable design, such as a line or an array of lines. However, the width of the anode lines is preferably no greater than 2000 μm, and more preferably no greater than 1000 μm. The width of the anode lines is preferably at least about 1 μm, more preferably at least 100 μm, and most preferably at least 200 μm.

The film comprising an electroluminescent material is applied over the anode structure in a manner such that the topography is still discernible, that is, the electroluminescent film does not planarize the surface. The electroluminescent material may be any known electroluminescent material, but is preferably an organic material, and more preferably a polymeric material. Examples of small molecule organic materials include materials that include, for example, tertiary aromatic amines, metal complexes of 8-hydroxyquinoline, diarylbutadienes, and stilbenes, as disclosed in U.S. Patent 4,769,292 (Tang, et al.) and Tang, Information Display, October (1996), pp. 16-19. Examples of suitable polymeric materials include polyarylene vinylenes and polyfluorene (see, for example, Kraft, et al., *Angew. Chem. Int. Ed.*, Vol. 37, pp. 402-428 (1998); U.S. Patents 5,247,190; 5,708,130; 4,885,221 5,728,801, and 6,169,163; WO 97/33193 and WO 99/54385; and U.S. Application, Serial Number 08/289,344). Polyfluorene based materials are most preferred. Specifically, polymers are preferred which have groups of the formula:




wherein $R^1$ is independently, in each occurrence, hydrogen, a $C_{1-20}$ hydrocarbyl or $C_{1-20}$ hydrocarbyl containing one or more S, N, O, P or Si atoms, $C_{4-16}$ hydrocarbyl carbonyloxy, $C_{4-16}$ aryl(trialkylsiloxy) or both $R^1$ may form with the 9-carbon on the

fluorene ring a $C_{5-20}$ ring structure or a $C_{4-20}$ ring structure containing one or more heteroatoms of S, N or O;

$R^2$ is independently, in each occurrence, $C_{1-20}$ hydrocarbyl, $C_{1-20}$ hydrocarbyloxy, $C_{1-20}$ thioether, $C_{1-20}$ hydrocarbylcarbonyloxy or cyano; and

a is independently, in each occurrence, 0 or 1. Preferably, substantially all the fluorene groups are connected through the 2 and 7 carbon atoms as shown. There are at least 10 fluorene groups and the polymer has a polydispersity of less than 5.

The film comprising the electroluminescent material may also comprise other materials blended with the electroluminescent material as may be known in the art, such as stabilizers, adhesion promoters, fillers, hole transport materials, electron transport materials and matrix materials. The film, optionally, may also comprise more than one layer of various materials. These additional layers may be (a) other electroluminescent materials, which may be chosen, for example, to yield a desired color output, (b) materials selected to enhance charge injection from the anode or cathode, for example, a hole transport layer made of a conductive polymer, such as polyaniline, polythiophene, polypyrrole or a mixture thereof doped with a polymeric acid, or (c) materials, such as those which provide barrier or protective properties, provided such materials do not unduly inhibit the light emission or current generating properties of the film.

The total thickness of the film is preferably at least 50 nm, preferably at least 100 nm. The total thickness of the film is preferably no greater than 1000, more preferably 700 and most preferably 500, nm. The film may be applied by any known method for reliably coating such thin films. Spin coating and ink jet printing are two preferred methods. If a multilayer film is used, more than one coating step may be required.

On top of the film, a cathode material is applied. Suitable cathode materials are typically low work function metals, such as lithium, calcium, ytterbium, and alloys and blends of such metals with metals of higher work function. These materials may be applied by any known method, such as sputtering or vapor deposition. The cathode material may be applied as a substantially continuous sheet or may be applied according to a pattern. The cathode material is preferably 5 nm to 1 μm thick.

Protective layers and interconnects should be used as known in the art and as desired, (see, for example, WO 00/06665 for a method of providing protective packaging for the device).

Referring now to Fig. 1, which shows a cross-section of a representative device, the anode 20, for example, gold, is applied to the substrate 10. The anode 20 has a thickness t and a width w. The electroluminescent film 30 is applied over anode 20 to form an arch pattern or "pocket" around the anode 20. The cathode 40, in this case a continuous sheet of material, is applied over the film 30. Light emitted from the film 30 reflects from the cathode 40 and each "pocket" of anode/film/cathode becomes a miniature lamp with shaped reflector geometry.

The anode structure may advantageously be arranged as a network of connected conducting lines. One example of such a structure is shown in Figure 2. Anode lines 20 are separated by gaps 21 where no anode material has been applied to the substrate. If a substantially continuous patch or region of light is desired, the gaps between the anode lines should be kept in proportion to the dimensions of the anode and the thickness of the light emitting film. The ratio of gap width to width of wires, in the range of 0.5:1 to 50:1, may be desirable.

Example

A grid of 200 nm thick gold lines were applied to a glass substrate, as shown in Figure 2, by thermal deposition in a vacuum using a mask to control where the substrate becomes coated. A hole transport layer comprised of polyethylene dioxythiophene was spin coated over the grid to give a film thickness of about 50 nm. A polymer composition comprising a blend of a fluorene containing copolymers was spin coated over the hole transport layer. Finally, a 300 nm layer of calcium was vacuum deposited over the electroluminescent film layer. The gold lines, via a back and forth pattern, as shown in Figure 2, covered an area about 7 mm by 8 mm. The ratio of the gap between the gold lines to the width of the lines was about 1:1. When a 4.5 V bias was applied to this non-optimized test device, the topography of the cathode metal over the gold lines provided an image in which the anode pattern is observable but substantially diffused, as shown in Figure 3. With optimization, the anode pattern should become less visible during operation of the device.

WHAT IS CLAIMED IS:

1. A light emitting device comprising

at least one anode structure, which comprises a conductive material having a thickness in the range of 0.05 to 1000 μm, and a width in the range 1 μm to 2000 μm,

a film comprising an electroluminescent material positioned next to the anode in a manner such that the film conforms to the shape of the anode, and

a reflective cathode material positioned on the opposite side of the film from the anode forming a concave reflective surface on the side adjacent to the film.

2. The device of Claim 1 wherein the anode is positioned on a transparent substrate.

3. The device of Claim 2 wherein the substrate is a polymer.

4. The device of Claim 2 wherein the substrate is flexible.

5. The device of Claim 1 wherein the anode structure comprises an opaque conducting material.

6. The device of Claim 1 wherein the anode structure comprises a metal selected from gold, platinum, silver, copper, nickel, and alloys of such materials.

7. The device of Claim 1 wherein the electroluminescent material is organic.

8. The device of Claim 1 wherein the electroluminescent material is a polymer.

9. The device of Claim 8 wherein the electroluminescent polymer is selected from polyarylene vinylenes and fluorene based polymers.

10. The device of Claim 9 wherein the electroluminescent polymer is a fluorene based polymer or copolymer.

11. The device of Claim 8 wherein the film comprises a hole transport layer adjacent to the anode and an electroluminescent layer over the hole transport layer.

12. The device of Claim 1 wherein the thickness of the anode is from 0.1 μm to 100 μm.

13. The device of Claim 12 wherein the thickness of the anode is less than 10 μm.

14. The device of Claim 12 wherein the width of the anode is in the range of 100 to 1000 μm.

15. The device of Claim 1 wherein the thickness of the film is in the range of 50 to 1000 nm.

16. The device of Claim 1 wherein the cathode is selected from ytterbium, calcium, lithium, and alloys of such materials with other low work function metals.

17. The device of Claim 1 wherein the anode structure comprises a conductive metal and a layer of indium tin oxide over or adjacent to the conductive metal.

18. The method of Claim 2 wherein the substrate has a surface curved surface.

19. A method of making the device of Claim 1 comprising

forming an elevated anode structure on a substrate,

forming the electroluminescent film over the anode structure in a manner such that the film conforms to the anode structure and the substrate, and

forming the cathode over the electroluminescent film to form a concavity around the anode structure.

20. The method of Claim 19 further comprising removing the substrate.

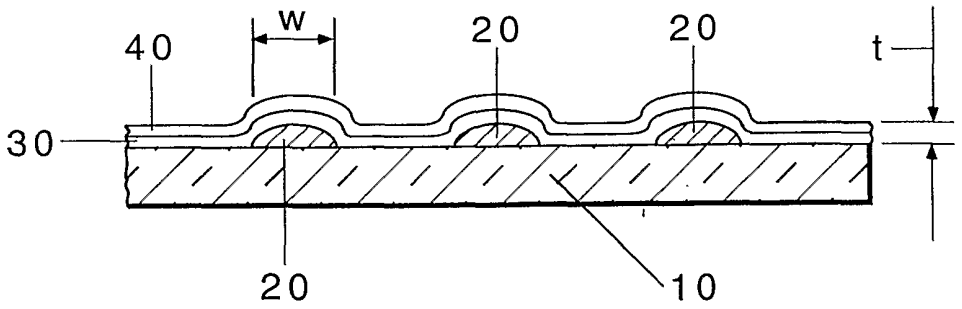



Fig. 1

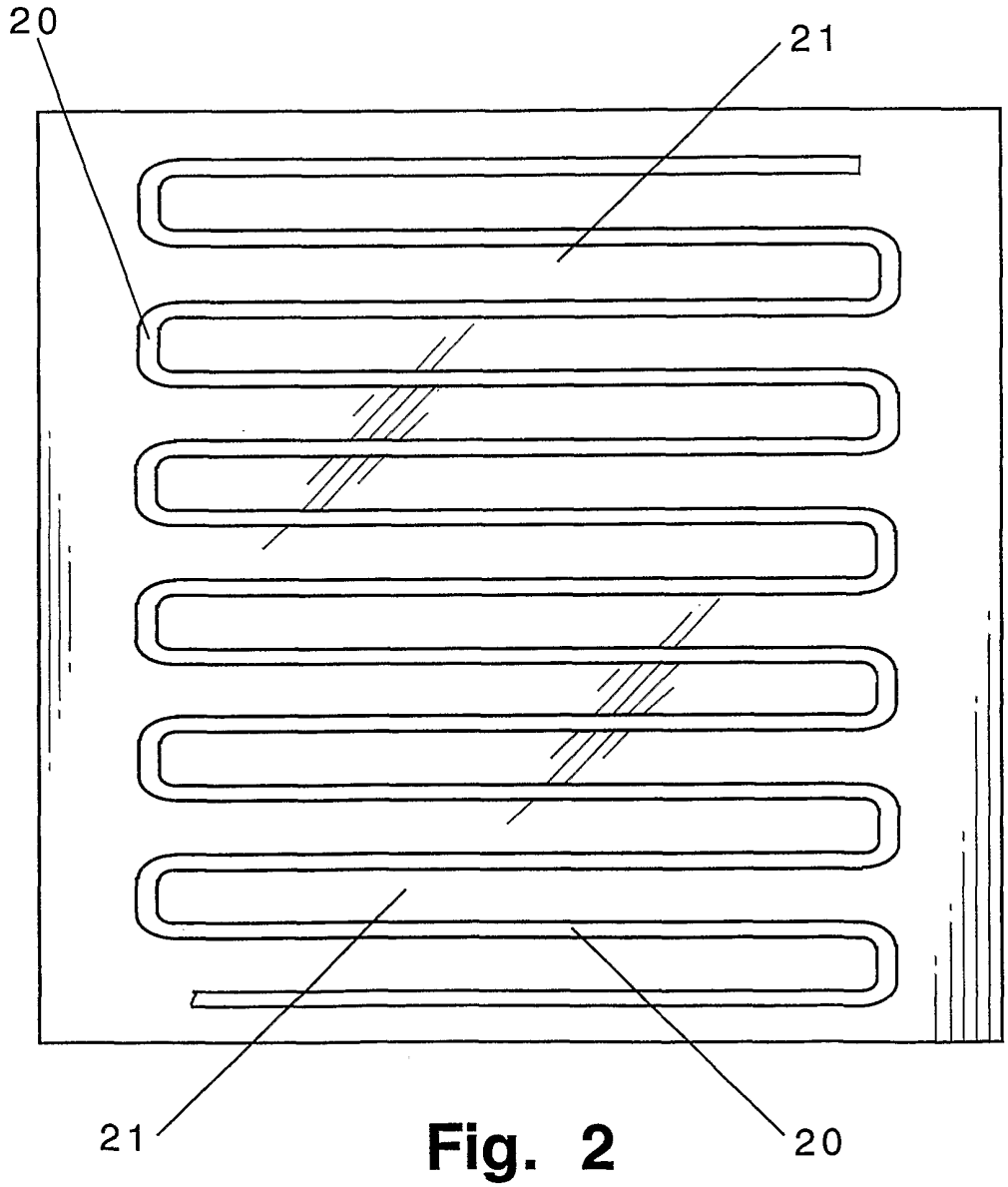
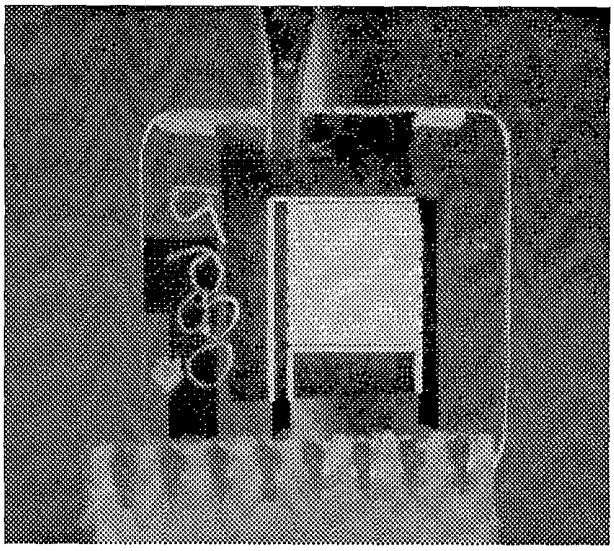



Fig. 2

FIG. 3



# INTERNATIONAL SEARCH REPORT

rnational Application No
PCT/US 01/49228

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01L51/20

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)
IPC 7 H01L

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

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

PAJ, WPI Data, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category ° | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|---|---|---|
| X | PATENT ABSTRACTS OF JAPAN<br>vol. 1999, no. 13,<br>30 November 1999 (1999-11-30)<br>-& JP 11 214162 A (NEC CORP),<br>6 August 1999 (1999-08-06)<br>abstract<br>-& US 6 396 208 B1 (ATUSHI ODA ET AL)<br>28 May 2002 (2002-05-28)<br>--- | 1,2,5-7,<br>11-13,<br>15,19 |
| X | PATENT ABSTRACTS OF JAPAN<br>vol. 1999, no. 13,<br>30 November 1999 (1999-11-30)<br>-& JP 11 214163 A (NEC CORP),<br>6 August 1999 (1999-08-06)<br>abstract<br>& US 6 396 208 B1 (ATUSHI ODA ET AL)<br>28 May 2002 (2002-05-28)<br>---<br>-/-- | 1,2,5-7,<br>11-13,<br>15,19 |

[X] Further documents are listed in the continuation of box C.

[ ] Patent family members are listed in annex.

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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|---|---|---|
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**Information on patent family members**

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