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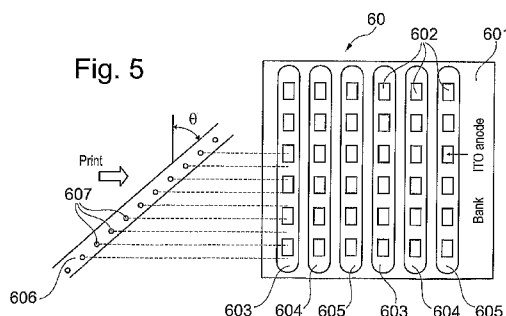
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(54) Title: METHOD FOR INK JET PRINTING ORGANIC ELECTRONIC DEVICES



(57) Abstract: A method of fabricating an organic electronic device by ink-jet printing into channels, comprises using a print head to deposit an ink into a channel, wherein the print, head travels in a direction substantially transverse, perpendicular, or orthogonal to the length of the channel. The channel may be a single or dual bank channel of 90µm pitch which may be defined by either a positive or negative photo-resist material. The ink may be an organic conducting material such as PEDOT, or PEDOTPSS. The ink may also be combined with a solvent such as toluene or xylene for example. The method can also be used to fabricate active or passive matrix displays. The print head may also be further rotated by an angle of θ , which allows the reduction of the printer's dot pitch by a factor of $\cos \theta$.



METHOD FOR INK JET PRINTING ORGANIC ELECTRONIC DEVICES

5 This invention generally relates to the deposition of material for organic electronic devices, particularly organic light emitting diodes, by an ink jet-type process. The invention is particularly concerned with depositing an ink material into channel banks.

Background of the Invention

10 Electronic flat screen display devices now pervade every area of contemporary life. From the mobile telephones to handheld computer gaming devices, flat screen computer displays and flat-screen television sets. In terms of ergonomics, power consumption and weight, present Organic Light-Emitting Diode (OLED), Liquid Crystal Display (LCD) or Thin-Film Transistor (TFT)
15 designs offer enormous improvements over the long-standing Cathode-Ray Tube (CRT) display technology. At the heart of the recent improvements in visual performance and availability of so-called flat-screen displays is the manufacturing process.

20 Flat-screen displays are manufactured using a grid of picture elements (pixels) at the screen surface which display the output image. The pixels are typically arranged in a rectilinear grid with an arranged pattern of red, green and blue pixels used to achieve colour displays. As a result, the materials which make up the red, green or blue pixels can be laid down according to the pattern of
25 the coloured pixels.

There is shown in Figure 1 a vertical cross-section through an example of an OLED device 100 as is known in the art. In an active matrix display part of the area of a pixel is occupied by associated drive circuitry (not shown in Figure 1).
30 The structure of the OLED device 100 is somewhat simplified for the purposes of illustration.

The OLED device 100 comprises a substrate 102, typically 0.7 mm or 1.1 mm glass but optionally clear plastic, on which an anode layer 106 has been

deposited. The anode layer 106 typically comprises around 150 nm thickness of ITO (indium tin oxide), over which is provided a metal contact layer, typically around 500nm of aluminium, sometimes referred to as anode metal. Glass substrates coated with indium tin oxide (ITO) and contact metal may be purchased from Corning, USA. The contact metal (and optionally the ITO) is patterned as desired, and so that it does not obscure the display, by a conventional process of photolithography followed by etching.

A substantially transparent hole injection layer 108a is provided over the anode metal, followed by an electroluminescent layer 108b. Barriers 112 may be formed on the substrate 102, for example from positive or negative photoresist material, to define wells 114 into which these active organic layers may be selectively deposited, for example by a droplet deposition or inkjet printing technique. The wells 114 thus define the light emitting areas or pixels of the display. A multicoloured display may be constructed using groups of red, green, and blue emitting pixels.

A cathode layer 110 is then applied by, say, physical vapour deposition. A cathode layer 110 typically comprises a low work function metal such as calcium or barium covered with a thicker, capping layer of aluminium and optionally including an additional layer immediately adjacent the electroluminescent layer 108b, such as a layer of lithium fluoride, for improved electron energy level matching.

The device described above is a so-called "bottom-emitting" device in which light is emitted through the substrate, however it may be beneficial (especially in the case of active matrix displays) to provide for emission through a transparent cathode ("top emitting" device), in which case the substrate and anode may be transparent, opaque or reflective.

In the case of passive matrix displays comprising a plurality of orthogonal anodes and cathodes, mutual electrical isolation of cathode lines may be achieved through the use of cathode separators 112. Typically a number of displays are fabricated on a single substrate and at the end of the fabrication process the

substrate is scribed, and the displays separated before an encapsulating can is attached to each to inhibit oxidation and moisture ingress.

Organic LEDs of this general type may be fabricated using a range of materials including polymers, dendrimers, and so-called small molecules, to emit over a range of wavelengths at varying drive voltages and efficiencies. Examples of polymer-based OLED materials are described in WO90/13148, WO95/06400 and WO99/48160; examples of dendrimer-based materials are described in WO 99/21935 and WO 02/067343; and examples of small molecule OLED materials are described in US 4,539,507. The electroluminescent layer 108b may comprise, for example, around 70nm (dry) thickness of PPV (poly(p-phenylenevinylene)) and the hole injection layer 108a, which helps match the hole energy levels of the anode layer and of the electroluminescent layer, may comprise, for example, around 50-200 nm, preferably around 150 nm (dry) thickness of PEDOT:PSS (polystyrene-sulphonate-doped polyethylenedioxythiophene).

Techniques for the deposition of material for organic light emitting diodes (OLEDs) using ink jet printing techniques are described in a number of documents including, for example, Y. Yang, "Review of Recent Progress on Polymer Electroluminescent Devices," SPIE Photonics West: Optoelectronics '98, Conf. 3279, San Jose, Jan., 1998; EP O 880 303. Ink jet techniques can be used to deposit materials for both small molecule and polymer LEDs. The use of the term "ink" in the following disclosure is taken to mean a dissolved molecular electronic material, which can include semiconductor material, Light Emitting Polymers (LEP) or small molecules.

When depositing materials for molecular electronic devices such as OLEDs, there is a need for both high resolution – generally than better than that required for the best high resolution graphics – and accurate control of the volume of material deposited. For graphics applications it is drop placement that is significant and volume variations of 5 to 10% are acceptable. However when constructing molecular electronic devices it is deposited "ink" volume which is important since this will determine the eventual film thickness which,

for an OLED, impacts upon brightness and hence drive current and device lifetime. Thus it is desirable to achieve a volume variation of better than 2%, preferably better than 1%, across an entire OLED display.

5 To deposit a molecular electronic material a volatile solvent such as toluene or xylene is employed with between 0.5% to 4% dissolved solvent material. This can take anything between a few seconds and a few minutes to dry and results in a relatively thin film in comparison with the initial "ink" volume. This drying time is dependent upon the solvent mix and the atmosphere above the
10 substrate. Often multiple drops are deposited to provide sufficient thickness of dry material, although it is strongly preferable all the drops comprising material which are eventually to make up a pixel are deposited before drying begins. Solvents which may be used include alkylated benzenes, in particular toluene, xylene or cyclohexylbenzene; others are described in WO 00/59267, WO
15 01/16251 and WO 02/18513; a solvent comprising a blend of these may also be employed.

Further, when materials are deposited on a substrate to form OLED devices, variations in the volume of ink will result in unwanted uneven emission
20 characteristics across the device. Even despite the best methods, a given nozzle may fail entirely when in use, leaving a whole line of pixels unusable or visibly weaker than the rest of the display. Where the pixels have been printed by running nozzles along the length of a channel, the effect in a colour display is diminished red, green or blue throughout one axis of the display.

25 Precision ink jet printers such as machines from Litrex Corporation of California, USA are used; suitable print heads are available from Xaar of Cambridge, UK and Spectra, Inc. of NH, USA. Such a print head usually has between 80 and 128 nozzles and the diameter of each print nozzle is usually
30 between 10µm and 100µm, the pitch between adjacent nozzles is usually between 50µm and 100µm.

Inkjet printing has many advantages for the deposition of materials for molecular electronic devices but there are also some drawbacks associated

with the technique. In practice drying is complicated by other effects such as the coffee ring – effect. With this effect because the thickness of solution is less at the edge of a drop than in the centre, as the edge dries the concentration of dissolved material there increases. Because the edge tends to be pinned solution then flows from the centre of the drop towards the edge to reduce the concentration gradient. This effect can result in dissolved material tending to be deposited in a ring rather than uniformly. The physics of the interactions of a drying solution with a surface are extremely complicated and a complete theory still awaits development.

Figure 2, which is taken from WO2005/076386 (herein incorporated by reference), shows a view from above (that is, not through the substrate) of a portion of a three-colour active matrix pixellated OLED display 200 after deposition of one of the active colour layers. The figure shows an array of barriers 112 and wells 114 defining pixels of the display 200. The wells 114 are formed as apertures in a continuous layer or sheet.

WO2008/035094 teaches the passage of sweeping an ink-jet print over a number of pixels of a portion of a colour OLED display. Figures in the document shows diagrammatically deposited droplets in place in those pixels and the presence of a ring-bank. In particular, the red (R), green (G) and blue (B) sub-pixels are each shown having a separate well with anode metal at the base. Also in WO2008/035094 is an arrangement in which the ring-banks define longitudinal channels, each holding material for a plurality of colour sub-pixels, the sub-pixels themselves being defined by anode metal. In particular, the anode islands may be separated by an underlying passivation layer such as silicon oxide or nitride or SOG (spin-on-glass). No part of the ring-bank of one pixel is shared with another pixel.

Figure 3a shows a cross-section through a portion of a display 1100 such as a passive matrix OLED display as known in the art in which a layer of insulating material 1102 is provided over portions of the anode metal in order to insulate this from later-deposited cathode material. This is seen more clearly in Figure 3b where it can be seen that were insulator 1102 not to be present, when cathode metal was deposited on the structure (to provide electrodes at right angles to the anode metal electrodes) the cathode and anode electrodes would

short out. In particular in Figure 3b, the image wells 106 are clearly identified. The insulator may comprise a conventional insulating material such as oxide, nitride or SOG or it may comprise a resist material. Where insulator 1102 comprises a resist material preferably the insulator is one of positive or negative resist, for example positive resist and the bank material is formed of the other type of resist, for example negative resist. Where insulator 1102 comprises resist material, this is preferably not fluorinated (so that the bank resist adheres well to the underlying resist). Suitable resist materials comprise the "ELX" and "WIX" series resists from Zeon Corporation, Japan.

A further problem with inkjet deposition arises when filling wells which are large compared with the size of an inkjet droplet. One way around this problem is to sufficiently over fill the well such that the dissolved material is pushed into the well corners. This can be achieved by using a large number of dilute droplets and a high barrier around the well. Techniques for depositing large volumes of liquid are described in WO 03/065474 which describes the use of very high barriers (for example at page 8 lines 8 to 20) to allow the wells to hold a large volume of liquid without the liquid overflowing to adjacent wells. However such structures cannot easily be formed by photolithography and instead a plastic substrate is embossed or injection moulded.

One solution is to lower the boundaries between pixels so that ink droplets may coalesce. As a result, ink droplets need not be as accurately placed as before. Such a manufacturing process is said to have "channel banks" of pixels.

Figures 4a and 4b show OLED pixels in a channel bank layout, as known in the field of the invention and taught in Samsung patent US7091660. In particular, Figure 4a shows an active matrix bank layout and 4b shows a passive matrix bank layout. Where appropriate, the same reference numerals are used and common to both Figures 4a and 4b are substrates 501, red colour banks 503, blue colour banks 504 and green colour banks 505. Figure 4a has active matrix display 50 and pixel anodes 502. Figure 4b has passive matrix display 55, substrate 501, red pixel banks 503, blue pixel banks 504 and green pixel banks 505. In the active matrix display 50, the pixel anodes 502 are placed one per image pixel. The pixel anodes are controlled by circuitry local to each image pixel. In the passive matrix display 55, the controlling circuitry is distant

from the image pixels and as a result the anode connections are overlaid across the banks. Also shown is a print head 507, which is usually positioned at an angle θ to the X-direction, which allows the reduction of the printer's dot pitch by a factor of $\cos \theta$.

5

US 2005/133802 discloses a channel bank structure formed by patterning a layer of a photosensitive material, wherein the channel bank has a structure selected so as to deal with the problem of inaccurate inkjet droplet deposition. The channel bank structure is designed to contain inkjet drops that are deposited between the wells rather than in the wells. The practice of filling banks or channels of same-coloured pixels is an improvement over using individual pits because it allows for easier control of the printing head and consequently an increase in the speed at which the print head moves and, as a result, a decrease in the production time for ink-jet printed display screens.

15

When filling channels to make pixels, the intuitive approach would use a single nozzle per channel. This has benefits to the print speed. Also, using continuous inkjet printing to fill the holes also yields improvements to the manufacturing process. However, an ongoing problem is that of the consistency of the nozzles from which material is deposited. The variations in the nozzle-to-nozzle volume of ink deposited on a substrate by an ink jet print head can vary substantially across a print head due to variations in the ink polymer, dried out blockages to the flow, failures within the electronics controlling the jet as well as other causes.

25

Further, the size or volume distribution of drops is non-uniform, increasing or falling off at nozzles at the edge of the print head (that is, near an end of a row of nozzles), and further non-uniformity arise from small variations in nozzle heights. Prior art print heads are limited to a drop rate between 6-8 kHz and must drop ink onto pixel anodes placed 9 μ m apart. These physical factors therefore put a restraint the speed at which the print head can move when printing into channel banks.

30

Summary of the Invention

5 The present invention therefore seeks to provide a method of ink-jet printing flat-screen displays which overcomes the above-mentioned problems of the prior art.

10 Accordingly, in a first aspect, the invention provides a method of fabricating an organic electronic device by ink-jet printing into channels as specified in claim 1.

15 Preferably the channel may be a single or dual bank channel of between 50 μ m and 100 μ m pitch, preferably approximately 90 μ m pitch, which may be defined by either a positive or negative photo-resist material.

20 Further preferably, the print head is arranged to make an oblique angle θ to the direction perpendicular to the direction of motion of the print head. θ is preferably between 0 and 90 or between 90 and 180, more preferably between 45 and 90 $^\circ$). The print head is also arranged to make an oblique angle to the longest dimension of the channels. The print head may comprise between 80 and 128 nozzles. Further, the nozzle diameter may be between 10 μ m and 100 μ m and the pitch between adjacent nozzles may be between 50 μ m and 100 μ m. The print head speed may be between 20 m/s up to 250m/s, preferably either 24m/s or 240m/s.

25 Preferably, the ink is a conductive material, or conductive organic material such as LEP, Interlayer, PEDOT or PEDOT:PSS. Also, the ink may further comprise a solvent such as cyclohexylbenzene, an alkylated benzene, toluene or xylene.

30 According to a second aspect, the invention provides a method of fabricating an OLED display in accordance with the first aspect. Preferably wherein the display may be a monochrome display, a red, blue, green (RGB) display, or a RGB rectilinear anode layout display. Further, the display may be an active matrix or a passive matrix display.

According to a third aspect, the invention provides an OED, wherein the OED is fabricated in accordance with the first aspect and preferably has a volume variation of printed material across all channels of less than 2%, preferably less than 1%.

According to a fourth aspect, the invention provides an OLED display, wherein the OLED display is fabricated in accordance with the first aspect and preferably has a volume variation of printed material across all channels of less than 2%, preferably less than 1%.

The advantages of the present invention are many, some of which are summarised here.

One advantage is that the printer can move much faster, up to 10 times faster because the area being printed by each nozzle is $90\mu\text{m}$ as compared to $9\mu\text{m}$. This actually brings the printer to the edge of their capabilities, but is achievable in practice. This results in lower per-unit manufacturing costs.

Another advantage is that if one nozzle breaks, the channel is still filled by the other nozzles, this therefore reduces nozzle to nozzle variation as droplets coalesce.

Further, the method of the present invention can be used with single bank channels (that is channels with negative or positive photo resist banks the run the length of channel and separate each channel one from another. It can also be used with dual bank channel, that is where PEDOT (or another suitable material) is constrained in each pixel by banks running transverse to the channel as well as banks that run longitudinally to the channel.

The method of the present invention can also be used to the both active and passive matrix displays.

Brief Description of the Drawings

An embodiment of the invention will now be more fully described, by way of example only, with reference to the drawings, of which:

- 5 Figure 1 is a diagram showing a vertical cross-section through an OLED device as is known in the art;
- Figure 2 is a diagram showing a view from above of a portion of a three colour pixelated OLED display known in the art;
- Figures 3a and 3b are diagrams showing, respectively, a well of a
10 passive matrix OLED display in vertical cross-section and 3D view, as is known in the art;
- Figures 4a and 4b are diagrams known in the art showing OLED pixels in a bank layout, with Figure 4a showing an active matrix bank layout and Figure 4b showing a passive matrix bank layout; and
- 15 Figure 5 is a diagram showing an ink-jet print head printing a bank layout while moving transverse to that bank layout, according to one embodiment of the present invention.

20 Detailed Description

In prior art methods of printing, for example as described with reference to Figures 3a and 3b, the print head moves prints into pixel wells. In this case, the print head location needs to be very accurate in both the x and y directions in relation to the location of the pixel well.

25

In prior art methods of printing into channels, such as described with reference to Figures 4a and 4b, the print head moves in the direction of the channel and the ink may be allowed to run down the channel to coalesce. This resolves the problem with the accuracy of the print head in the x direction, but there still
30 remain a problem with the accuracy of the print head in the y-direction. Therefore there may still be nozzle to nozzle variation from channel to channel.

The present invention provides a method of printing which requires, much less accuracy in both x and y directions, by orthogonally, or transversely, printing to the direction of the channel.

5 Figure 5 shows an ink-jet print head printing a bank layout while moving transverse to that bank layout, according to a first embodiment of the present invention. There is shown a display 60 which comprises a substrate 601, pixel
anodes 602, red pixel banks 603, blue pixel banks 604 and green pixel banks
605. A print head 606 is also shown and which comprises nozzles 607 from
10 which an ink material is printed. When printing the ink material, the print head 607 moves in a substantially transverse direction to that of the red, green and blue pixel banks (603, 604, and 605, respectively) and delivers ink from the nozzles 607 into the pixel banks 603; 604; 605. The pixel banks 603; 604; 605 have 90µm pitch comprising a 70µm channel abutted by 20µm channel walls.

15 In prior art methods, when a print head prints along the length of a channel, the nozzles 607 must not be allowed to drop ink on the 9µm gaps between anodes (not shown). In order to accommodate this constraint, as well as take into account the chemical properties and fluid dynamics of the ink, the control
20 frequency of the print head 606 is usually configured to be in the region of 6-8 kHz. This results in the print head moving at about 24m/s.

According to the present invention, the print head 606 prints transverse to the pixel banks 603; 604; 605, each of which is 90µm wide and so the print head
25 606 is configured to move a factor of 10 times faster, that is up to 240m/s. The increase in speed causes an increase in the operating vibration of the printing apparatus. However, this is not outside the operating parameters of prior art print heads.

30 The present invention provides the advantage that that if one nozzle breaks, the channel is still filled by the other nozzles, this therefore reduces nozzle to nozzle variation as droplets coalesce.

Also, even if a nozzle is out at the top or bottom of the channel banks, then the other nozzles can compensate as the PEDOT coalesces into each channel. The top and bottom locations in each channel are also not usually used and are considered in the art as “dummies”.

5

There may still be drying effects within the channels, but with the use of PEDOT this should not be so bad and PEDOT dries slowly enough to allow the material to coalesce within the channel before drying.

10

Measures may be implemented to further reduce or eliminate display artefacts resulting from such drying effects, in particular visible “swathes joins” wherein the display has a striped appearance due to non-uniform drying of material deposited in different passes (“swathes”) of the print-head.

15

Such measures include:

20

- i) Printing swathes close together if it is found that material deposited in different swathes are failing to coalesce.
- ii) Reducing the number of drops printed per swathe by one or more nozzles compared to the other nozzles. For example, if it is found that there is a problem of increased thickness at a swathe join then one or more nozzles printing at the swathe join may be adjusted to print fewer drops than other nozzles, or indeed no drops at all.

25

- iii) Adjusting nozzles so that not all nozzles print, and print overlapping swathes wherein at least some of the nozzles that do not print in one swathe do print a subsequent swathe (“interlacing”).

30

As most substrates are rectangular in size and shape, then it will be necessary in practice to reconfigure the printer set-up to allow the banks to be printed transversely in accordance with the present invention. Depending upon the printer used, this may in some cases mean small configuration changes, but in most cases will mean very large configuration changes.

Following printing of the layers described in the examples above, fabrication of the organic light emitting diode is completed by depositing a cathode. The cathode may be transparent or opaque, for which a wide range of suitable materials and deposition techniques are known to the skilled person. For light
5 emission through the anode side, the cathode may be opaque. However, in the case of active matrix devices in particular, it is preferred that light from the device is emitted through a transparent cathode. In this case, the anode is preferably formed from a reflective material, or is a transparent material such as ITO provided with an underlying layer of reflective material.

10 The organic light emitting diode is preferably encapsulated to avoid degradation caused by ingress of moisture and oxygen into the device. Suitable encapsulants include glass or metal cans, or a barrier stack comprising alternating layers of polymer and dielectric material.

15 The skilled person will recognised that the above described techniques are not limited to use in the fabrication of organic light emitting diodes (small molecules or polymer) but may be employed in the fabrication of any type of molecular electronic device in which material is dissolved in a solvent and deposited by a
20 droplet deposition technique. No doubt many effective alternatives will occur to the skilled person and it will be understood that the invention is not limited to the described embodiments encompasses modifications apparent to those skilled in the art lying within the scope of the claims appended hereto.

25

CLAIMS

1. A method of fabricating an organic electronic device by ink-jet printing into channels, comprising:
5 using a print head to deposit an ink into a channel, wherein the print head travels in a direction substantially transverse to the length of the channel.
2. A method according to claim 1, wherein the print head is rotated by an angle θ .
3. A method according to claim 2, wherein θ is between 0 and 90° or 90° and 180° degrees.
10
4. A method according to claim 3, wherein θ is between 45 and 90°.
5. A method according to any preceding claim, wherein the channel is between 50 μ m and 100 μ m wide.
6. A method according to any preceding claim, wherein the channel is abutted by a channel wall.
15
7. A method according to claim 6, wherein the channel wall is between 5 and 50 μ m wide.
8. A method according to claim 7, wherein the channel is a 70 μ m wide channel abutted by a 20 μ m channel wall, defining a 90 μ m pitch.
9. A method according to any of claims 6 to 8, wherein the channel wall is defined by a resistive material.
20
10. A method according to claim 5, wherein the channel wall is defined by a positive resist material.
11. A method according to claim 5, wherein the channel wall is defined by a negative resist material.
25
12. A method according to any of claims 6 to 11, wherein the channel is a dual bank channel.
13. A method according to claim any preceding claim, wherein the print head comprises nozzles having a diameter of between 10 μ m and 30 100 μ m.

14. A method according to either claim 13, wherein the pitch between adjacent nozzles is between 50 μ m and 100 μ m.
15. A method according to any preceding claim wherein the print head speed is between 20m/s and 250m/s
- 5 16. A method according to claim 15, wherein the print head speed is approximately 24m/s.
17. A method according to claim 15, wherein the print head speed is approximately 240m/s.
18. A method according to any preceding claim, wherein the ink is a
10 conductive organic material.
19. A method according to claim 18, wherein the conductive organic material is chosen from the list consisting of: LEP, Interlayer, PEDOT and PEDOT:PSS.
20. A method according to claims 18 or 19, wherein the ink further
15 comprises a solvent taken from the list consisting of: cyclohexylbenzene, an alkylated benzene, toluene and xylene.
21. A method according to any preceding claim in which the organic electronic device is an OLED display.
22. A method according to claim 21, wherein the OLED display is a
20 monochrome display.
23. A method according to claim 21, wherein the OLED display is a red, blue green (RGB) display.
24. A method according to claim 21, wherein the OLED display is an RGB rectilinear anode layout display.
- 25 25. A method according to claim 21, wherein the OLED display is an active matrix display
26. A method according to claim 21, wherein the OLED display a passive matrix display.
27. An organic electronic device fabricated according to a method as
30 claimed in any one of claims 1 to 26.

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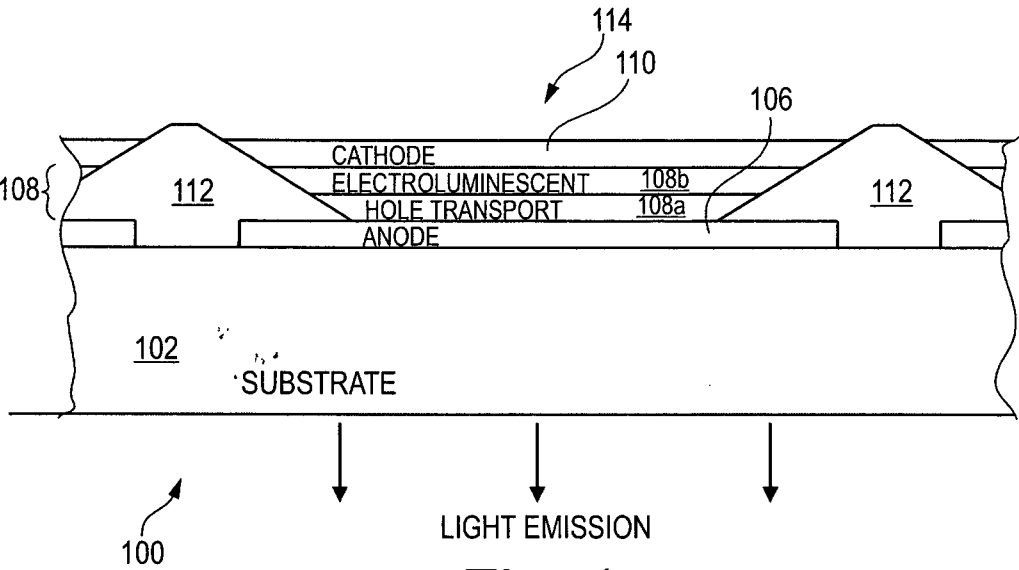


Fig. 1
(PRIOR ART)

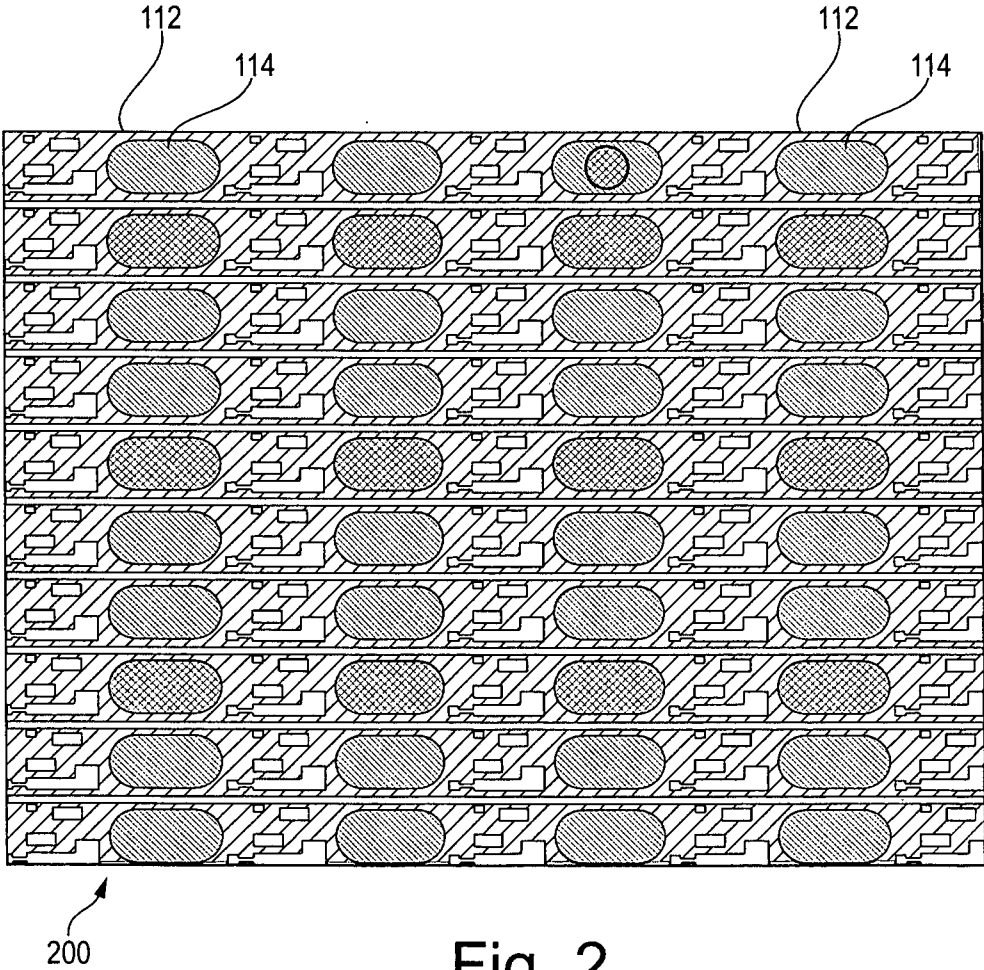


Fig. 2

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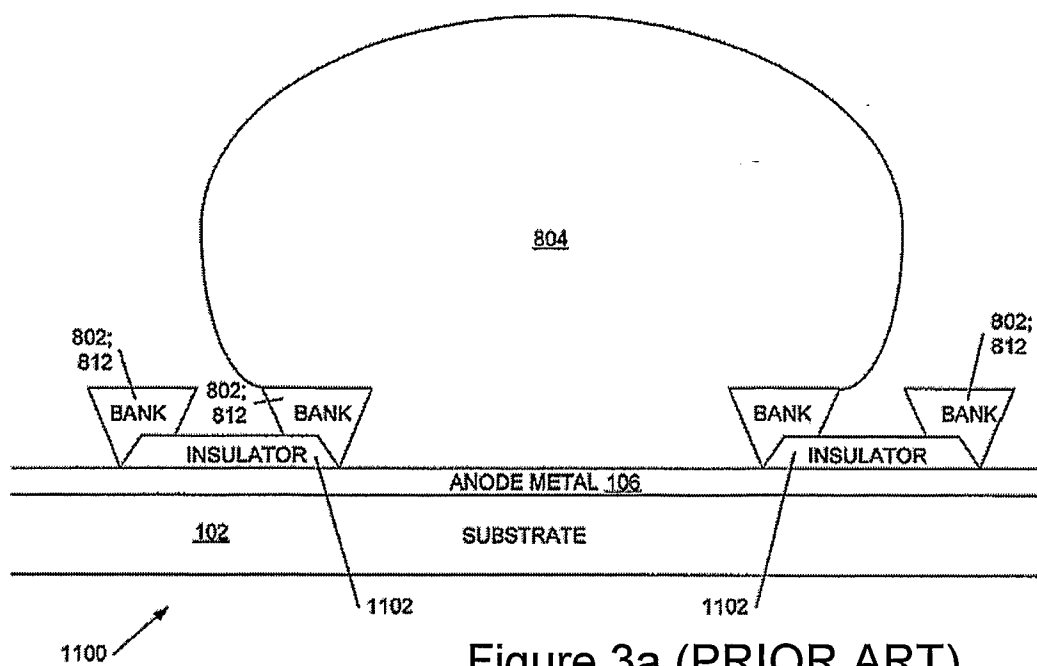


Figure 3a (PRIOR ART)

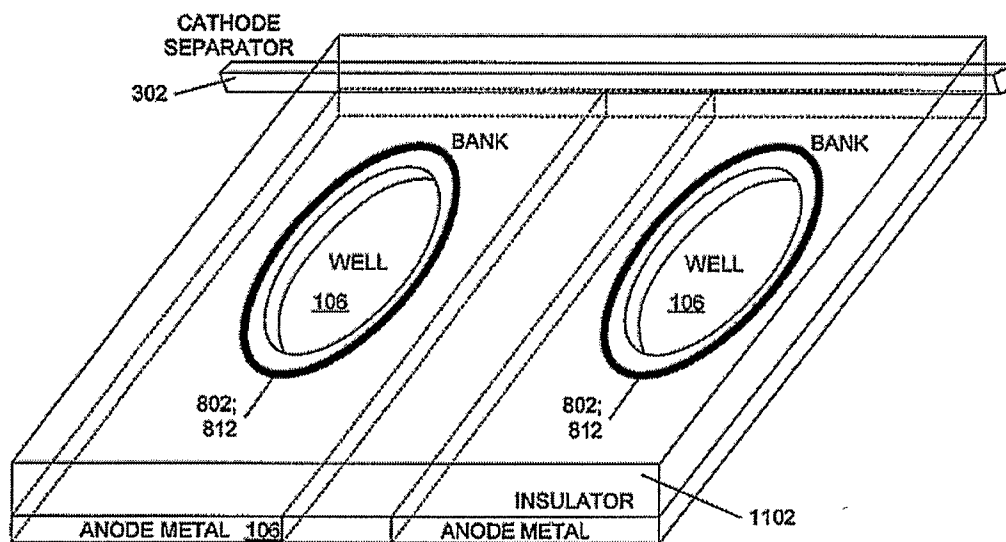


Figure 3b (PRIOR ART)

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