Effects of Nitridation Time on Top-emission Inverted Organic Light Emitting Diode

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Abstract — A top-emission inverted organic light-emitting diode (TEIOLED) was fabricated by using Al/AlNx layer as the cathode in the device structure of glass/Al/AlNx/AlQ3/NPB/MTDATA/Au/Ag, where AlNx ultra thin layer was obtained from Al layer under 90 W microwave plasma treatments in Ar and N\textsubscript{2} mixed gas environment. The N\textsubscript{2}/Ar ratio and plasma treatment time were adjusted to obtain the maximum luminance and efficiency of 1206 cd/m\textsuperscript{2} and 0.51 cd/A, respectively, both at 17 volts. The AlNx layer surface after plasma treatment was examined by atomic force microscope to study the effects of surface roughness on the electroluminescent (EL) characteristics. The AlNx layer thickness also affected the EL results apparently.

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1. INTRODUCTION

One of the trends in the OLED (organic light emitting diode) display industry in recent years is to develop large-screen display. Large-size panel must be driven by thin film transistor (TFT) to enhance its brightness uniformity, resolution and life time. The conventional OLED have predominantly bottom-emission structure where emitted light is partially blocked by bottom-layer TFT and data line. To improve aperture ratio and minimize the effect of TFT on active-matrix panel, the research and development of top-emission OLED is inevitable [1, 2]. To integrate the process for OLED with other active components in the fabrication of active-matrix panel, including amorphous silicon (α-Si), poly-silicon (poly-Si) or complementary metal oxide semiconductor (CMOS), the whole process may be simplified if the electrode (Al pad) prepared on TFT substrate in the final step of the process is also used as the cathode of OLED. Such OLED device must employ top-emission inverted organic light emitting diodes (TEIOLEDs) structure. But when Al electrode is deposited on the substrate using thermal evaporation, spikes tend to form, which are prone to lead to organic carbonization and puncture the organic thin film after prolonged operation of OLED, causing dark spots on the emitting surfaces of OLED [3]. Many researchers have pro-posed inverted top-emission light-emitting diode by inserting an ultra thin insulating layer between the organic layer and metal electrode to increase electron injection from cathode, thereby enhancing the luminance efficiency. For example, Li et al., [4], Kurosaka et al., [5] and Kho et al., [6] respectively inserted a proper buffer layer of Al\textsubscript{2}O\textsubscript{3}/AlNx between the organic light-emitting layer and Al cathode to improve the electroluminescence of OLED. This paper utilized the plasma of Ar+N\textsubscript{2} gas mixture to treat Al surface to modify the surface roughness and form an ultra-thin AlNx insulating layer (or buffer layer) to improve the efficiency of cathode electron injection.

2. EXPERIMENT

Our experiment comprised the following steps: First ultra-sonically wash the glass substrate with in sequence acetone, methyl alcohol and DI water. Deposit cathode metal Al on the glass substrate by evaporation under 3 × 10\textsuperscript{-6} torr and then move the substrate to a microwave plasma chamber to undergo plasma treatment of Al surface with the mixture of Ar (70 sccm) and N\textsubscript{2} (10 sccm) under 0.35 torr and 90 W to form AlNx as buffer layer. Remove the AlNx-treated substrate from plasma chamber and place it in organic vacuum evaporator to deposit tris (8-quinolinolato) aluminum (AlQ\textsubscript{3}) as green-light emitting layer, NPB as hole transport layer, and m-MTDATA as hole inject layer (HIL). Finally move the substrate to another chamber to deposit Au/Ag as metal anode by deposition. The resulting device has a structure of glass/Al (80 nm)/AlNx/AlQ\textsubscript{3} (100 nm)/NPB (60 nm) /m-MTDATA (50 nm)/Au (5 nm)/Ag (10 nm) as shown in Fig. 1. The light emitting area of the device was 36 mm\textsuperscript{2} as defined by shadow mask. The experiment used KEITHLEY 2400 and SpectraScan PR650 to measure the L-J-V curve of device, atomic force microscope (AFM) and contact angle to observe its surface roughness, and four-point probe to measure the sheet resistance.
3. RESULTS AND DISCUSSION

Atomic Force Microscope (AFM) was used to observe the surface roughness of Al cathode layer after microwave plasma treatment over different durations. As shown in Fig. 2, it is found that untreated Al electrode surface has many spikes that would cause non-uniform distribution of electric field in the organic layer during the operation of OLED and render the OLED prone to damage under higher current density. Thus prior to the deposition of organic film, Al electrode should undergo surface planarization with microwave plasma, which improves surface roughness significantly. Based on the measurement of contact angle, it is also clear that there is better adhesion between the plasma-treated Al electrode and the organic layer.

Figure 2: Plasma-treated surface of Al cathode under AFM, (a) untreated Ra = 5.249 nm, (b) 40 sec Ra = 4.112 nm, (c) 60 sec Ra = 3.747 nm, (d) 80 sec Ra = 2.551 nm.

Figure 3, Fig. 4 anf Fig. 5 show respectively the J-V curve (current density vs. voltage), L-V curve (luminance vs. voltage), and η-V curve (yield vs. voltage) of the same device under plasma treatment of different durations. As shown by the J-V curve, the resistance of plasma-treated Al surface increased as treatment time increased, which led to decrease in current density as shown in Table 1. Based on the L-V curve in Fig. 4, it is found increase in the time of nitrogen treatment led to increase in threshold voltage. Untreated Al cathode caused organic layer to produce joule...
heat under higher current and same voltage and burn more easily. As shown in Fig. 4 and Fig. 5, under optimum plasma treatment time of 40 sec, the device obtained maximum luminance and yield of 1206 cd/m$^2$ and 0.51 cd/A respectively under 17 V. Device made of untreated Al electrode had extremely short life-time, while treated device with an AlNx buffer layer had significantly longer life time. Our TEIOLED has lower turn-on voltage when compared with Ref. [6].

![Figure 5](image5.png)

Figure 5: Luminance yield of TEIOLED vs. voltage under different plasma treatment times.

<table>
<thead>
<tr>
<th>Treatment time</th>
<th>Roughness (Ra, nm)</th>
<th>Contact Angle</th>
<th>Resistivity (Ω-cm)</th>
<th>Max. Luminance (cd/m$^2$)</th>
<th>Yield (cd/A)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Untreated</td>
<td>5.249</td>
<td>22.30°</td>
<td>6.55 × 10$^{-8}$</td>
<td>418@14 V</td>
<td>0.17@14 V</td>
</tr>
<tr>
<td>40 s</td>
<td>4.112</td>
<td>9.88°</td>
<td>7.40 × 10$^{-8}$</td>
<td>1206@17 V</td>
<td>0.51@17 V</td>
</tr>
<tr>
<td>60 s</td>
<td>3.747</td>
<td>8.63°</td>
<td>7.51 × 10$^{-8}$</td>
<td>474@17 V</td>
<td>0.29@16 V</td>
</tr>
<tr>
<td>80 s</td>
<td>2.551</td>
<td>7.32°</td>
<td>7.54 × 10$^{-8}$</td>
<td>256@17 V</td>
<td>0.20@17 V</td>
</tr>
</tbody>
</table>

When plasma treatment time increased to more than 40 sec, the luminance and yield of device decreased gradually instead. It might be attributed to the fact that prolonged treatment time resulted in thicker AlNx and hence high resistivity, which makes injection of electron into AlQ$_3$

![Figure 6](image6.png)

Figure 6: Shift of emission peak from 508 nm to 558 nm when thickness of organic layer increases.

![Figure 7](image7.png)

Figure 7: EL intensity of TEIOLED under different angles EL spectrum.

more difficult. The functions of AlNx may be summed up as follows: 1. Proper AlNx thickness aids the tunneling of electrons [8], for in the absence of AlNx as buffer layer, there exits a larger
potential barrier at AlQ3/Al interface to block the injection of cathode electrons into AlQ3; 2. the mobility of surface electron in organic material is lower than that of hole, and AlNx can block the circulation of holes to prevent quenching of exciton at AlQ3/Al interface.

In the TEIOLED structure discussed in this paper, Al metal with high reflectivity was used for cathode, and Au (5 nm)/Ag (10 nm) was used for anode where Au was for improving hole injection from anode into organic layer and Ag with higher conductivity and transmission was for reducing sheet resistance and enhancing light output. The penetration rate of such double-layer anode was approximately 47%. In addition, change in the thickness of organic or metal electrode would cause displacement of emission peak in microcavity. In the example of changing the total thickness of organic layer as shown in Fig. 6, when the length of microcavity increased from 120 nm to 210 nm, emission peak shifted from 508 nm to 552 nm. The change in resonance wavelength is not just a result of change in microcavity length, but also due to difference in the reflection phase of top and bottom surface electrodes. Fig. 7 shows the electroluminescence (EL) spectrum obtained at different angles, which are the angles intersecting the measuring direction of PR650 relative to the normal line of light-emitting surface of device. The measurement took place at the interval of 15° between 0° ∼ 75°. It is clear that as the measuring angle increased, the peak of EL spectrum would drop from 576 nm to 524 nm to produce blue displacement phenomenon, mainly due to the intense microcavity effect [9].

4. CONCLUSION

We have demonstrated that using the microwave plasma of Ar+N₂ mixture to treat the surface of Al electrode over the optimum treatment time of 40 sec can form an ultra thin AlNx layer as the buffer layer for injection of electrons from cathode in TEIOLED. This surface treatment effectively improves the efficiency of electron injection, enable the number of electrons and holes injected to reach a balance and prevent the phenomenon of exciton quenching at the cathode, thereby significantly enhancing the luminance, efficiency and life-time of the device. The N₂/Ar ratio and plasma treatment time were adjusted to obtain the maximum luminance and efficiency of 1206 cd/m² and 0.51 cd/A, respectively, both at 17 volts.

REFERENCES