Transparent Organic Light-Emitting Diodes with Transparent Oxide Semiconductor of IZO for Cathode <u>Takayuki Amemiya</u>, Shigeki Naka, and Hiroyuki Okada

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1. Introduction

Organic light-emitting diodes (OLEDs) have excellent properties of a low driving voltage and a bright emission. One of interesting research topics is transparent OLEDs (TOLEDs) that emit on both sides and this type of matrix panel has been demonstrated.^{1,2)} One of the subjects in this type of OLED is an higher electron injection from the transparent cathode and stacking layer of CuPc/ITO or ITO/Li/BCP cathodes are reported. 1,2) For research topic in transparent oxide semiconductor (TOS) thin-film-transistors (TFTs), SnO₂,³⁾ ZnO₂,⁴⁾ In-Ga-Zn-O (IGZO)⁵⁾ and In-Zn-O (IZO)^{6,7)}-based TFTs are actively studied. The TOS is act as the high mobility n-type semiconductor. Especially, the IZO under low temperature annealing at 300 °C to decrease the carrier density due to a control of oxide vacancy act as the n-type TOS.⁸⁾ In this study, we have studied transparent OLEDs with relatively low temperature annealed IZO for cathode.

2. Experimental

For a bottom cathode layer, we used bi-layer of ITO/IZO for reducing a sheet resistance and electron injection. Process steps were as follows: The ITO glass substrate was thoroughly cleaned using the process of scrubbing, ultrasonic cleaning, and an UV-ozone treatment. Upper cathode layer used was IZO (Idemitsu Kosan), where, the weight ratio of In₂O₃ to ZnO was 90:10. The IZO film was deposited at room temperature using a conventional radio-frequency (RF) magnetronsputtering apparatus. Bi-layer electrode of ITO/IZO was patterned using photolithography and wet etching. The IZO etchant used was oxalic acid (HOOCCOOH, 2.5 wt%, 32 °C). And the etchant used for ITO was aqua regia (48 °C). After cleaning process, IZO film was annealed at 300 °C for 1h in air. The 2, 9- dimethyl-4,7diphenyl-1,10-phenanthroline (BCP) as electron

transport layer (ETL), tris (8-quinolinolato) aluminum (III) (Alq₃) as emission layer (EML), bis [N-(1naphthyl)-N-phenyl] benzidine (α -NPD) as hole transport layer (HTL) were used. Hole injection layer (HIL) of molybdenum trioxide (MoO₃), oxide semiconductor, was also used. Semi-transparent Au electrode was used for ultra thin anode. The MoO₃ was effective for a buffer layer in order to prevent penetration of Au atom into organic layers during Au evaporation. These layers were sequentially deposited by conventional thermal evaporation technique without vacuum breaking. The devices with ITO (200 nm), IZO (200 nm, with and without annealing) cathodes were fabricated for a comparison. Device area was $2 \times 2 \text{ mm}^2$.



Fig. 1 Current density vs voltage characteristics.



Fig. 2 Luminance vs current density characteristics.



Fig. 3 Current density vs voltage characteristics.



Fig. 4 Luminance vs current density characteristics.

3. Results and discussion

Figure 1 shows current density versus applied voltage (J-V) characteristics. In the ITO/IZO (5 nm) cathode device with annealing, operating voltage was lower than other devices. This result suggests that electron injection from cathode to organic layer is improved by IZO annealing. On the other hands, in the IZO (200 nm) cathode device with annealing, operating voltage was shifted to higher voltage. Sheet carrier concentration and resistivity of IZO was changed from 2×10^{15} to 5×10^{13} cm⁻² and from 2.5×10^{-2} to 1.4×10^{-1} Ω cm, by annealing, respectively. Figure 2 shows luminance versus current density (L-J) characteristics. In the ITO/IZO (5 nm) and IZO (200 nm) cathode device with annealing, brighter luminance was observed. This result suggests that electron injection from cathode to device was enhanced by annealing and injected hole and electron from anode and cathode, respectively, were well balanced. In addition, luminance was in proportion in overall current range. This result suggests that efficiency of exciton generation is constant because identical structures of Alq₃/ α -NPD interface. The luminance from bottom and from top emission ($J = 100 \text{ mA/cm}^2$) was 622 and 495



Fig. 5 Transmission spectra of devices. cd/m^2 , respectively. This discrepancy was due to light absorption between transparent bottom ITO/IZO and top Au. Figures 3 and 4 show *J-V* and *L-J* characteristics varied with the IZO thickness for ITO/IZO cathode devices. Within these ranges, device characteristic did not change. In thinner IZO thickness, tunneling current will be dominant.

Figure 5 show the transmission spectrum of the ITO/IZO (20nm) device. At peak wavelength of Alq_3 emission, transmittance of 65 % was observed. In addition, transmittance of ITO/IZO (20 nm) and Au is 75 and 65%, respectively. A difference of double-faced luminance, as discussed in Fig. 3, can be explained by difference of transmittance.

4. Conclusions

We had demonstrated the transparent organic light emitting diodes with IZO cathode layer with annealing and obtained brightness was over 1,000 cd/m². Higher performance TOLEDs will be expected by further optimization of device process and layer structures.

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