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Effect of ITO/Ag/ITO Multilayer Electrode Deposited onto Glass and PET Substrate on the Performance of Organic Light-Emitting Diodes

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The transparent conductive oxide (TCO) electrode is an important part of organic light-emitting diodes (OLEDs), especially for the OLEDs based on flexible substrate. We fabricated indium tin oxide (ITO)/Ag/ITO multilayer films deposited on polyethylene terephthalate (PET) film by using sputtering technique at room temperature in order to achieve high transmittance and low sheet resistance for application to flexible OLED devices. The OLEDs made with ITO/Ag/ITO multilayer deposited on flexible film exhibited lower operating voltage and higher current efficiency than the OLEDs made with commercial single layer ITO glass substrate.

Keywords Ag thin interlayer; anode electrode; indium tin oxide (ITO); multilayer; organic light-emitting diodes (OLED); transparent conductive oxide (TCO)

Introduction

The organic light-emitting diode (OLED) has attracted much interest recently due to its potential use as a flexible display and also for solid-state lighting [1]. The transparent conductive oxide (TCO) electrode is an important part of the OLED devices, especially for the OLEDs based on flexible substrate. Indium tin oxide (ITO) is known as one of the best TCO material, however, it is difficult to satisfy the electrical requirement ($>10\,\Omega/\square \approx 1\times10^{-4}\,\Omega\cdot cm$) for OLEDs based on polymer films which require low temperature process and no additional heat treatment [2]. Recently, the ITO/Ag/ITO multilayer electrodes have been studied as a promising anode for OLED because the electrical characteristics could be improved by adopting Ag

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interlayer between ITO layers. The ITO/Ag/ITO multilayer films were found to give lower sheet resistance than the single layer ITO even though they were deposited under low temperature condition and without additional heat treatment [3–7]. The effect of ITO/Ag/ITO multilayer film used as anode of OLED on the performance of the OLED device has not been studied in detail with the specific condition of ITO/Ag/ITO multilayer deposition and consequent OLED device properties.

In this study we deposited ITO/Ag/ITO multilayer electrode on PET film and the fabricated devices and discussed the effect of ITO/Ag/ITO multilayer electrode deposition conditions on the performance of the OLED devices.

Experimental

The ITO/Ag/ITO multilayer films (total thickness $\approx$ 100 nm) were sequentially deposited both on glass (thickness = 0.7 mm) and polyethylene terephthalate (PET, thickness = 188 μm) substrates by in-line RF and DC sputter. Both the bottom and top ITO layer were deposited with RF sputter using In_2O_3 ceramic target containing 5 wt% SnO_2. The Ag interlayer was sputtered on top of the bottom ITO layer by using metallic Ag (99.999% purity) target. Subsequently, the top ITO layer was deposited on the Ag/ITO layer under the same deposition condition as the bottom ITO layer. The deposition conditions for ITO/Ag/ITO multilayer films on glass and PET substrates were described in detail elsewhere [8]. A commercial single layer ITO glass was used as a reference in the OLED device fabrication for comparison. The ITO/Ag/ITO multilayer coated film substrate was patterned by standard photolithographic method, and then cleaned and dried carefully for dehydration. The baking time and temperature of the patterned ITO/Ag/ITO multilayer anodes on glass and PET substrates were 60 min at 230°C and 60 min at 150°C, respectively. The structure of the anode part of OLED panel with insulator layer is shown in Figure 1. Figure 2 shows the schematic diagram of the fabricated OLEDs with the configuration of hole injection layer (HIL, α-NPD, 60 nm), electron transport layer/emissive layer (ETL/EML, Alq3, 40 nm), electron injection layer (EIL, LiF, 0.5 nm), and cathode (Al, 150 nm) for the analysis of the effect of ITO/Ag/ITO electrodes.

The sheet resistance of the ITO/Ag/ITO thin film was measured by a 4-point probe (Mitsubishi Chem., MCP-T610). The optical transmittance spectra of the TCO films were obtained by using UV-VIS spectrophotometer (Konica Minolta CM-3600d). Electro-optical properties of the fabricated OLED devices were measured by spectroscan PR650 (Photoresearch Inc.) with I-V source (Keithley 2400).

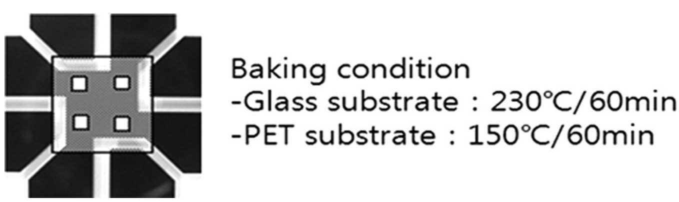


Figure 1. Schematic diagram of ITO/Ag/ITO multilayer anode electrodes patterned on PET substrate.

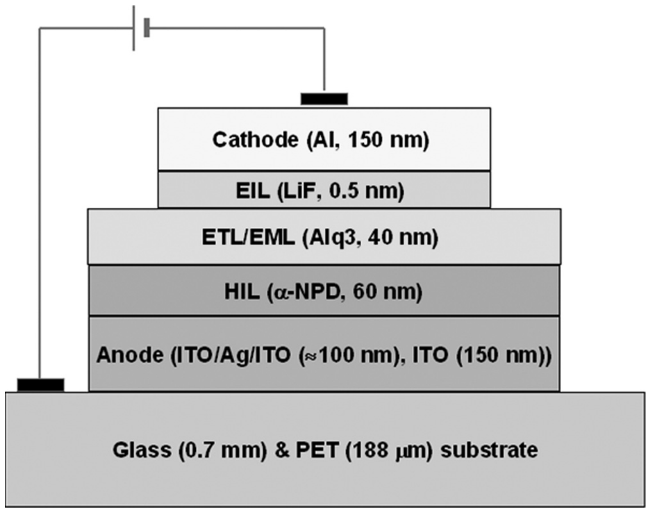


Figure 2. Configuration of the OLED device fabricated on the ITO/Ag/ITO multilayer anode electrodes.

Results and Discussion

Figure 3 shows the optical transmittance spectra of ITO/Ag/ITO multilayer films deposited on glass and PET substrates by sputtering technique at room temperature. For comparison a commercial single layer ITO deposited on glass substrate was included. The transmittance of ITO/Ag/ITO multilayer film at long wavelength region was lower than that of commercial ITO/glass sample. The decrease of the transmittance at long wavelength in ITO/Ag/ITO multilayer film could be explained by the increase of electronic absorption due to the shift of plasma resonance frequency to the long wavelength region as the sheet resistance was decreased [2,5,9]. The transmittance of ITO/Ag/ITO multilayer films on glass and PET substrate were

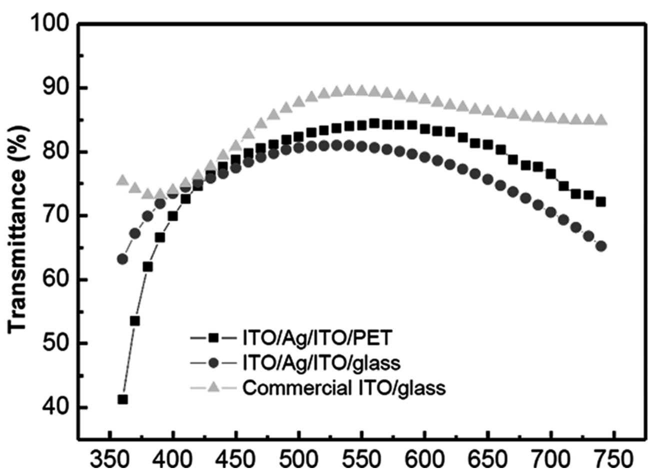


Figure 3. Optical transmittance spectra of ITO/Ag/ITO multilayer films deposited on glass and PET substrates at room temperature.

81.0 and 83.7% at 530 nm, respectively. However, the total sheet resistance of ITO/Ag/ITO multilayer films deposited on glass and PET substrate at room temperature exhibited very low values of 6.93 and 6.85 Ω/□, respectively. These values were superior to those of commercial single layer ITO glass (10 Ω/□) deposited at higher temperature. Furthermore when the size of glass and PET substrate was increased to $200 \times 200\,mm^2$, the optical transmittance and sheet resistance of ITO/Ag/ITO films showed less than 3% deviations.

It was reported that the low sheet resistance of transparent conductive electrode could result in the reduction of operating voltage and enhancement of current in the light-emitting devices [10]. The effect of ITO/Ag/ITO multilayer anode deposited

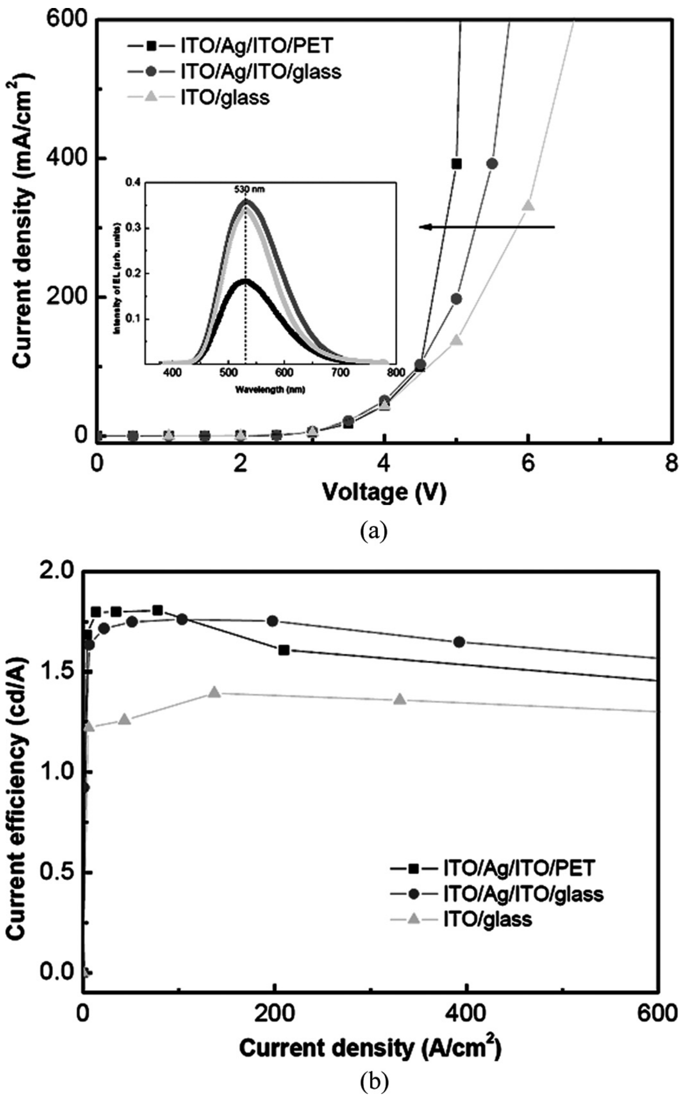
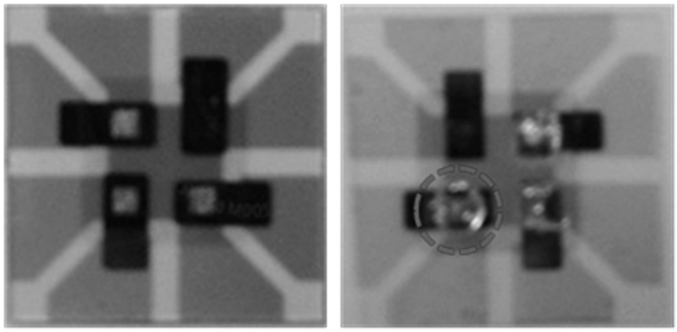


Figure 4. (a) Current density-voltage and (b) current efficiency-current density characteristics of the OLEDs fabricated with ITO/Ag/ITO multilayer anode electrodes deposited on glass and PET substrates at room temperature.



(a) (b)

Figure 5. Photographs of the OLEDs fabricated with ITO/Ag/ITO multilayer anode deposited on (a) glass and (b) PET substrate after injection of high current density (>500 mA/cm^2).

onto glass and PET substrate on the performance of the fabricated OLED is shown in Figure 4. All the OLED devices exhibited turn on voltage near 3 V and peak of EL spectra around 530 nm as shown in Figure 4(a). The operating voltages of the OLEDs at same current density were in the order of sheet resistance values as the arrow indicates in Figure 4(a). The operating voltages of the OLEDs made with the ITO/Ag/ITO multilayer anode deposited on glass and PET substrate were 5.26 and 4.86 V at the current density of 300 mA/cm^2. Figure 4(b) shows the current efficiency-current density curves of the OLEDs made with ITO/Ag/ITO multilayer anode deposited on glass and PET substrates at room temperature. The maximum current efficiency of about 1.80 cd/A was obtained in the OLED device fabricated with ITO/Ag/ITO multilayer anode deposited on glass substrate and PET substrates. It was of interest, however, that the current efficiency of the OLED (1.39 cd/A) with a commercial single layer ITO glass had slightly lower value than that of the OLED (1.81 cd/A) with ITO/Ag/ITO multilayer anode deposited on PET substrate. However, the current efficiency-current density curve showed abrupt break-down at above 5.0 V in the OLED fabricated with ITO/Ag/ITO multilayer anode deposited on PET substrate, while the OLED fabricated on ITO glass exhibited break-down at above 10 V. This phenomenon might be caused by the thermal degradation of the OLED at the junction between the inorganic electrode and organic layer of OLED under high current density. The PET substrate could not dissipate well the phonon emission (heat) to the atmosphere due to its high thermal resistivity (0.25 W/m K) compared to that of glass substrate (1.1 W/m K) [11]. These results suggest that both high conductivity and thermal energy dissipation are required for the flexible substrate in the OLED devices to avoid thermal breakdown. The OLED devices with and without thermal break-down are shown in Figure 5.

Conclusions

In summary we could deposit ITO/Ag/ITO multilayer thin film on PET film substrate by using sputtering technique at room temperature which had high transmittance and low sheet resistance suitable for fabrication of flexible OLED devices. The

OLEDs made with ITO/Ag/ITO multilayer anode electrodes exhibited lower operating voltage and higher current efficiency than the OLEDs made with commercial single layer ITO glass substrate. These results could be explained by the low sheet resistance due to the adoption of Ag interlayer in the ITO/Ag/ITO multilayer anode electrode. The thermal stability of the plastic substrate (PET), however, needs to be increased for the fabrication of stable flexible OLED devices.

Acknowledgments

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