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Improvement of mechanical reliability by patterned silver/Indium-Tin-Oxide structure for flexible electronic devices

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ABSTRACT

We report the effect of silver (Ag)-buffer layer Indium-Tin-Oxide (ITO) film on a polyethylene terephthalate substrate on the electrical, optical and reliable properties for transparent–flexible displays. The electrical and optical characteristics of an ITO-only film and an Ag-layer-inserted ITO film are measured and compared to assess the applicability of the triple layered structure in flexible displays. The sheet resistance, the resistivity and the light transmittance of the ITO-only film were 400 Ω/sq, 1.33×10^{-3} Ω-cm and 99.2%, while those of the ITO film inserted with a 10 nm thick Ag layer were 165 Ω/sq, 4.78×10^{-4} Ω-cm and about 97%, respectively. To evaluate the mechanical reliability of the different ITO films, bending tests were carried out. After the dynamic bending test of 900 cycles, the sheet resistance of the ITO film inserted with the Ag layer changed from 154 Ω/sq to 475 Ω/sq, about a 3-time increase but that of the ITO-only film changed from 400 Ω/sq to 61,986 Ω/sq, about 150-time increase. When the radius is changed from 25 mm to 20 mm in the static bending test, the sheet resistance of the ITO-only film changed from 400 to 678.3 linearly whereas that of the Ag-layer inserted ITO film changed a little from 154.4 to 154.9. These results show that Ag-layer inserted ITO film had better mechanical characteristics than the ITO-only film.

1. Introduction

Indium-Tin-Oxide (ITO) thin films are widely used as transparent electrodes in optoelectronic and electro-optic devices such as solar cells and flat panel displays [1–3]. This is due to their unique properties of both low electrical resistivity and high transmittance in the visible spectral region [4,5]. Recently, demands for the flexible display panels have increased because they are inexpensive and lightweight. However, the mechanical characteristics of ITO thin films have limited the use in flexible display panels. Flexible display panels are made using a polymer substrate instead of glass, which is used in non-flexible display panels. The properties of ITO films are very different from the physical properties of a polymer substrate. When the display is produced by a traditional fabrication process, cracks can be induced in the oxide films during the production and/or during their use.

Typically, the Young's modulus of the typically used polyethylene terephthalate (PET) substrate is 5.3 G N/m^2 while the Young's modulus of the widely used transparent conductor ITO is 100 G N/m^2 [6,7]. ITO films can be easily damaged by externally applied bending stress resulting from the difference in the Young's modulus between the ITO film and the PET substrate. To increase the mechanical stability of ITO films against bending stress, triple layered structures utilizing a buffer layer such as Ag sandwiched between two ITO layers have been suggested, and the change in the conductance of the ITO film with the bending of the substrate was studied [8,9]. When a triple layer structure was used, the transmittance of the ITO film decreased due to the metal layer covering the ITO layer.

In this paper, we aim to enhance the mechanical stability of the ITO film by patterning it and depositing Ag along the pattern, i.e., by changing the ITO structure horizontally instead of vertically as is done in triple layer structured ITO films. The transmittances and electrical conductivities of an ITO-only film and an Ag-layer-inserted ITO film on PET were studied and compared. The mechanical stability of the different ITO films against bending stress was also examined by static and dynamic bending tests.

2. Experiment

Fig. 1 shows the process flow of fabricating an ITO-film inserted with an Ag layer on a PET substrate. A thin layer ITO film of about 30 nm in thickness was deposited on a PET substrate using the roll-to-roll sputter-coating method [10]. For patterning, the ITO surface was covered with a layer of photo-resist (HPR504), which was deposited at rotation speed of 5000 rpm for 1 min. It was then exposed to ultra-violet light through a patterned mask. After the exposure, the photo-resist

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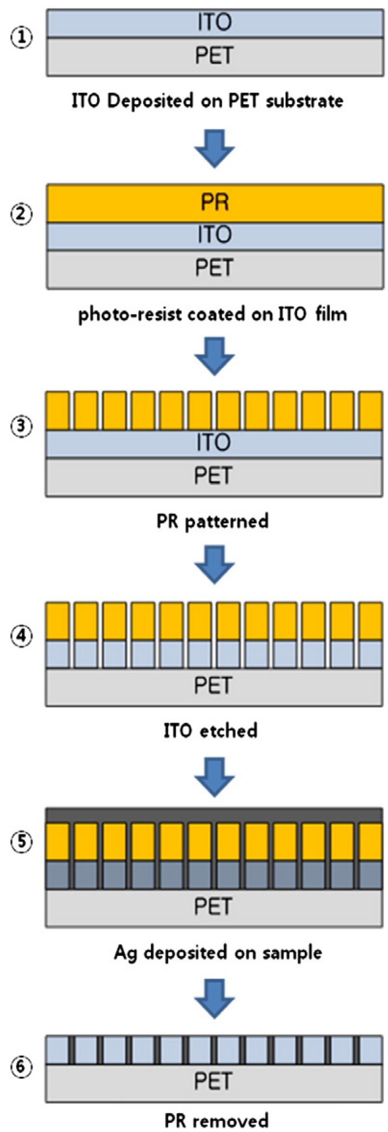

Fig. 1. Schematic of the patterned ITO film inserted with Ag layer on PET substrate.

was developed and cured at 125 °C. The ITO was etched using 5% oxalic acid ($C_2H_2O_4$) at 45 °C at etch rate of 40 nm/min [10]. Ag layers with different thicknesses (10, 20 and 30 nm) were deposited by thermal evaporation in vacuum of around 4×10^{-4} Pa. The lift-off procedure was carried out ultrasonically using acetone to completely remove the residual Ag and/or the photo-resist layer. The patterned width of Ag was 4 μm spaced 90 μm apart. For reference, another sample was made by depositing only an ITO layer on the PET.

The sheet resistances of the ITO-coated PET specimens were measured using a Four-Point Probe method (Quatek QT-50), and the carrier concentration and resistivity were determined by a Hall-effect measurement system under a magnetic field of 0.55 T (Ecopia HMS-3000). Optical transmittances of the ITO-coated PET specimens were measured using a UV–visible spectrophotometer (Shimadzu UV-1601). The dynamic bending test was performed by repeatedly bending the sample at a constant R value of 16 mm. The electrical resistances of the ITO-only coated and Ag-layer-inserted ITO specimens were recorded after 0–900 bending cycles. The static bending test was carried out by changing the radii of the ITO films from 25 to 20 mm. The sheet resistances of the specimens were measured. The SEM (JSM 7000F, JEOL, 15 kV) images of the surfaces before and after the dynamic 900-cycle bending test were obtained.

3. Results and discussion

Fig. 2 shows the electrical characteristics of ITO-only film and Ag-layer-inserted ITO film samples. As the thickness of the Ag layer increased, the sheet resistance decreased from 400 Ω/sq to 135.4 Ω/sq; the resistivity decreased from 1.33×10^{-3} Ω-cm to 4.467×10^{-4} Ω-cm; and the carrier concentration increased from 2.8×10^{20} cm^{-3} to 1.1×10^{21} cm^{-3}. It is believed that the high metallic conductivity of the inserted Ag layer is the reason for the reduced resistivity and increased carrier concentration of the Ag-layer-inserted ITO electrodes.

The change in resistivity of the Ag inserted ITO films with increasing Ag thickness can be explained by using the following basic relation:

$$\rho = \frac{1}{ne\mu}$$

where ρ is the resistivity, n is the number of charge carriers, e is the charge of the carrier, and μ is the mobility of the charge carriers [11]. From this equation we see that changes in charge carrier mobility or carrier concentration or both can affect the resistivity. The decrease in resistivity with the increasing Ag thickness can be attributed to the increases in carrier concentration and Hall mobility. The increase in the carrier concentration with the increase in the thickness of the Ag layer can be understood based on the Schottky theory.

Fig. 3 shows the schematic diagram of the energy bands of the ITO and Ag before (Fig. 3(a)) and after (Fig. 3(b)) their contact. Because the work function of silver (Φ = 4.4 eV) is smaller than that of ITO (Φ = 4.5–5.1 eV), electrons from Ag transfer to ITO to align the Fermi levels at equilibrium [12–14], causing band bending at the ITO-Ag contact, an accumulation-type contact. The electrons are easily injected from Ag into ITO because there is essentially no barrier for electron flow; as a result, the overall carrier concentration increases and the resistivity decreases.

Fig. 4 shows the transmittances of the ITO film-only and Ag-layer-inserted ITO film samples over a wide range of wavelengths. As the thickness of the Ag increases, the average optical transmittance of the Ag-layer-inserted ITO samples decreases from 99.2% to 92.9% at 550 nm wavelength because of the high optical absorption in the Ag layers. When the thickness of the Ag layer is below 20 nm, transmittance is over 90% even in the high wavelength range. However, when the thickness of the Ag layer is 30 nm, the transmittance of the ITO film drops to 87%. To see the possibility of using the ITO films with inserted Ag for display applications, a logo is placed under the Ag inserted ITO films and photographs were taken. Since the transmittance of Ag was less than that of ITO, it was thought that grid lines could appear when a picture is taken. Fig. 4 shows a photo of the ITO film with the 10 nm thick Ag layer. The 10 nm-thick Ag layer is transparent enough and grids are not present. Although not shown, the 30 nm-thick Ag layer inserted film showed very pale grids, which could be seen with bare eyes but not in a photograph.

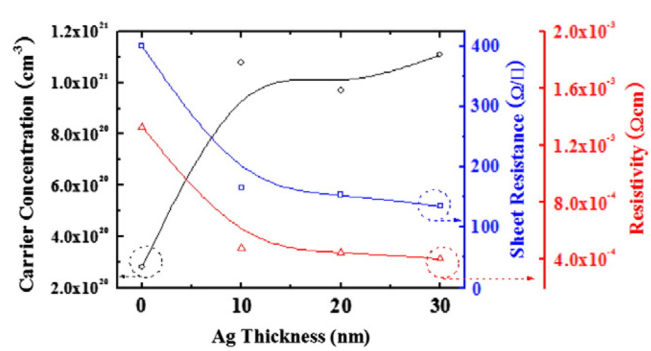

Fig. 2. Carrier concentration, electrical resistivity and sheet resistance variation with different Ag-layer thicknesses.