Characteristics of flexible indium tin oxide electrode grown by continuous roll-to-roll sputtering process for flexible displays

Kwang-Hyuk Choi, Sung-Woo Cho, Jin-A Jeong and Han-Ki Kim*
Department of Information & Nano Materials Engineering,
Kumoh National Institute of Technology (KIT), Yangho-dong, Gumi 730-701 , Gyeongbok, South Korea
Phone: +82-54-478-7746, E-mail: hkkim@kumoh.ac.kr

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Abstract

The preparation and characteristics of flexible indium tin oxide electrodes grown on polyethylene terephthalate (PET) substrates using a specially designed roll-to-roll sputtering system for use in flexible optoelectronics In spite of low a PET substrate temperature, we can obtain the flexible electrode with a sheet resistance of 47.4 ohm/square and an average optical transmittance of 83.46 % in the green region of 500~550 nm wavelength. Both x-ray diffraction (XRD) and field emission scanning electron microscopy (FESEM) analysis results showed that all flexible ITO electrodes grown on the PET substrate were an amorphous structure with a very smooth and featureless surface, regardless of the Ar/O_2 flow ratio due to the low substrate temperature, which is maintained by a cooling drum. In addition, the flexible ITO electrode grown on the Ar ion beam treated PET substrates showed more stable mechanical properties than the flexible ITO electrode grown on the wet cleaned PET substrate, due to an increased adhesion between the flexible ITO and the PET substrates

1. Introduction

Flexible electrodes are of considerable importance for their potential as a new generation of flexible displays due to their light weight, robust profile, ability to flex, curve, roll and fold for portability, as well as, their easy fabrication using roll-to-roll based thin film technology. To fabricate high performance and low cost flexible devices, it is necessary to prepare transparent conducting oxide with low resistance, high transmittance, superior flexibility and a smooth surface on polymer substrates, such as polyethylene terephthalate (PET), polycarbonate (PC), polyimide polyethersulfone (PES), or polyethylene naphthalate (PEN) for effective hole injection to the active layer.

In this work, we investigated electrical, optical, structural, mechanical and the surface properties of flexible ITO electrodes grown on a PET substrate using a specially designed roll-to-roll sputtering system for flexible displays. It was found that the resistivity and transmittance of flexible ITO electrodes are critically influenced by the Ar/O_2 flow ratio during continuous roll-to-roll sputtering. At an optimized Ar/O_2 flow ratio, DC power, working pressure, and rolling speed, we can obtain an flexible ITO electrode with a resistivity of 9.54×10^{-4} Ohm-cm, average transmittance of ~83.46 %, despite its preparation at room temperature. In addition, the effective surface treatment of the PET surface using an Ar ion beam enables us to make mechanically stable ITO electrode with good robustness, due to an improved adhesion between the ITO and the PET substrate.

2. Experimental

The 200 mm width polyethylene terephthalate (PET) substrate with a thickness of 188 μm was passed repeatedly over the cooling drum by motion of unwinder and rewinder for the deposition of ITO electrodes during the roll-to-roll sputtering process. The rolling speed of the PET substrate can be exactly controlled by the motor speed for unwinder and rewinder. Before ITO electrode sputtering on the PET substrate, the surface of the PET substrate was treated by the irradiation of an Ar ion beam at a DC pulsed power of 200 W, to remove surface contaminations and improve adhesion between the PET substrate and ITO anode material. Subsequently, the ITO film was sputtered on the rolling PET substrate, which is mechanically attached on cooling drum to maintain the substrate temperature below phase transformation temperature(<50℃).

Fig. 1. Picture of a specially designed roll-to-roll(R2R) sputtering system in Kumoh National Institute of Technology

The thickness of the flexible ITO electrode was measured by a profilometer. The electrical properties of flexible ITO were measured by means of a four point probe and hall measurement at room temperature. The optical transmittance was measured in the wavelength range of 250 to 800 nm by a UV/visible spectrometer. The surface morphology of the flexible ITO electrode was analyzed using a field emission scanning electron microscope (FESEM) (not shown here) as a function of the Ar/O_2 ratio.

To investigate the structural properties of flexible the ITO electrode, both x-ray diffraction (XRD) and high resolution electron microscope (HREM) examinations (not shown here) were performed. In addition, the flexibility of the flexible ITO electrode was analyzed using a laboratory made bending test system. The bending radius was approximated to 8 mm and the bending frequency was 1 Hz. During the bending test, the resistance of the flexible ITO/PET samples was measured using a multi-meter.

3. Results and discussion

Figure 2 shows the electrical properties of flexible ITO electrodes grown on a PET substrate at constant DC power of 800 W, working pressure of 3 mTorr and rolling speed of 0.2 cm/sec as a function of the Ar/O_2 flow ratio. Both the sheet resistance and resistivity of flexible the ITO electrode grown at the Ar/O_2 flow ratio of 30/1 sccm are lower than those of flexible ITO electrodes grown at the Ar/O_2 flow ratio of 20/1 as shown in Fig. 3(a).

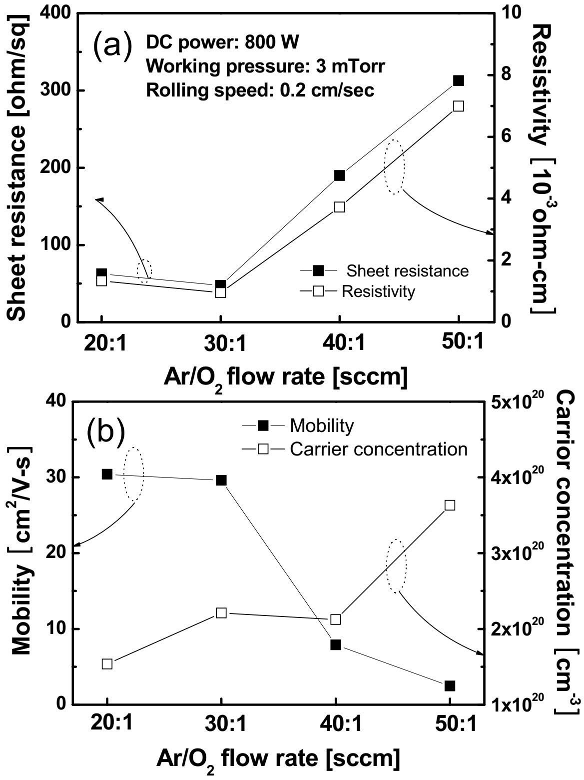

Fig. 2. (a) Sheet resistance, resistivity, (b) mobility, and carrier concentration of flexible ITO electrode grown by R2R sputtering as a function of Ar/O_2 flow ratio.

The minimum sheet resistance of 47.4 ohm/square and resistivity of 9.54×10^{-4} Ω-cm was obtained at the Ar/O_2 flow ratio of 30/1 sccm. However, the increase of Ar flow rate in the mixture of the Ar/O_2 gas above 30 sccm resulted in an abrupt increase of sheet resistance and resistivity. Tahar *et al.*, reported that increasing oxygen partial pressure resulted in a decreased in resistivity and carrier concentration and enhanced Hall mobility due to the dissipation of oxygen vacancies [1]. Therefore, the low resistivity of flexible ITO films grown at the Ar/O_2 flow ratio of 30/1 sccm can be explained by enhanced carrier mobility resulting from the dissipation of oxygen vacancy at high oxygen partial pressure.

Figure 3 shows optical transmittance spectra for the flexible ITO films grown on PET substrate as a function of the Ar/O_2 flow ratio. All flexible ITO films show similar optical transparency regardless of the Ar/O_2 flow ratio. It was found that the transparency of flexible ITO electrodes is not sensitive to the Ar/O_2

flow rate in the range between 20/1 and 50/1 sccm. It is noteworthy that the transmittance of flexible ITO films in the green region (500~550 nm) is higher than the UV region (400~450 nm). The sharp absorption edges in the transmittance spectra is caused by an extrinsic bandgap of ITO film in the range of 3.8~4.0 eV [2].

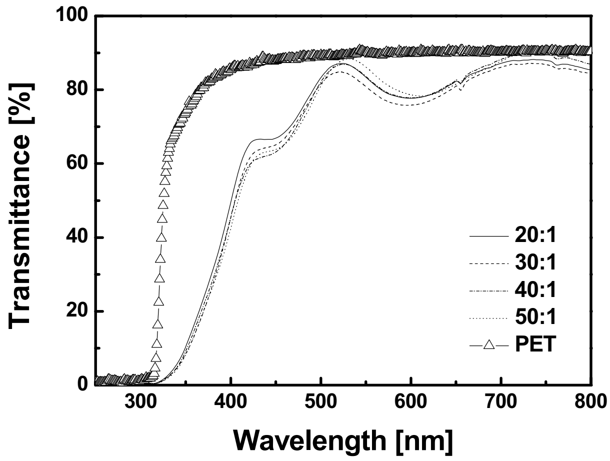

Fig. 3. Optical transmittance of the flexible ITO electrodes grown on a PET substrate using roll-to-roll sputtering system at narrowAr/O_2 flow ratio range from 20/1 to 50/1 sccm.

Figure 4 shows the XRD plots obtained from the flexible ITO electrode as a function of Ar/O_2 flow ratio. All XRD plots of the flexible ITO electrodes show only the intense PET substrate peak at the region of ~25.84 °. Due to the low substrate temperature during the roll-to-roll sputtering process, all flexible ITO electrodes show amorphous structure regardless of the Ar/O_2 flow ratio. This indicates that the substrate temperature was effectively maintained at low temperature below 50 °C by a cooling drum. To investigate the microstructure of the flexible ITO electrode grown at an optimized condition in detail, the HREM examination was employed. Figure 5 shows a cross sectional HREM image obtained from the ITO electrode grown on a Si substrate. Figure 4 shows the XRD plots obtained from the flexible ITO electrode as a function of Ar/O_2 flow ratio. All XRD plots of the flexible ITO electrodes show only the intense PET substrate peak at the region of ~25.84 °. Due to the low substrate temperature during the roll-to-roll sputtering process, all flexible ITO electrodes show amorphous structure regardless of the Ar/O_2 flow ratio. This indicates that the substrate temperature was effectively maintained at low temperature below 50 °C by a cooling drum.

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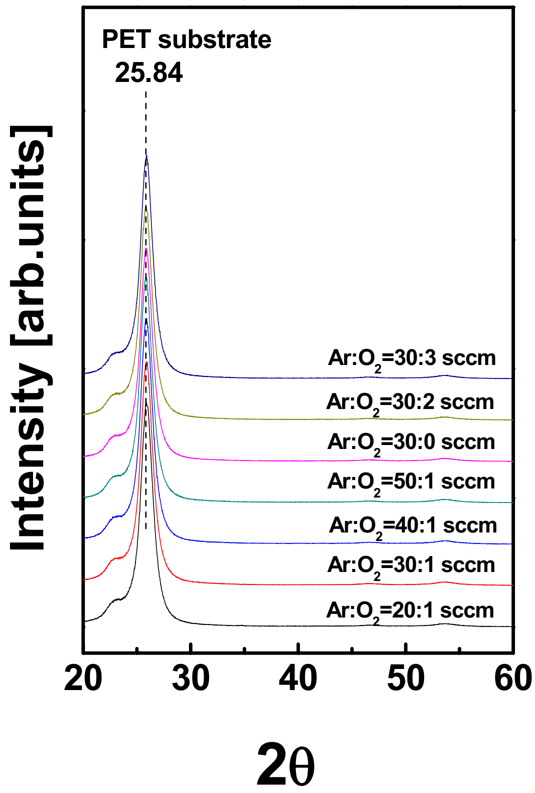

Fig. 4. XRD plots obtained from the flexible ITO electrodes grown on PET substrates as a function of Ar/O_2 flow ratio at constant DC power of 800 W, working pressure of 3 mTorr and rolling speed of 0.2 cm/sec.

Due to difficulty in the HREM sample preparation of flexible ITO electrodes grown on a PET substrate, the ITO electrode was intentionally grown on a Si substrate at identical conditions with an optimized flexible ITO growth condition. The uniform contrast of the ITO film in Fig. 5 indicates that the structure of the ITO film on a Si substrate is amorphous, as expected from the XRD results. However, some nanocrystallines, as indicated by arrows embedded in the amorphous ITO matrix, due to the low amorphous /polycrystalline transformation temperature. Even if an ITO electrode is prepared at room temperature, nanocrystallines could be easily formed at the amorphous ITO matrix at a low homogeneous temperatures (T/T_m< 0.19 ~ 150 °C) [3].

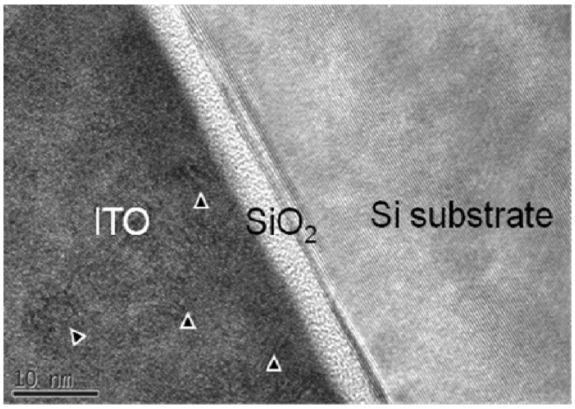

Fig. 5. Cross-sectional HREM image obtained from the flexible ITO electrode grown on a Si substrate at optimized roll-to-roll sputtering condition.

Figure 6 shows changes in the resistance of the flexible ITO electrode grown on untreated and the Ar ion treated PET substrate at a power of 200 W. It was noteworthy that the $\Delta R/R_0$ value of the flexible ITO electrode grown on untreated PET substrates increased remarkably at initial bending cycles, due to the generation and propagation of cracks.

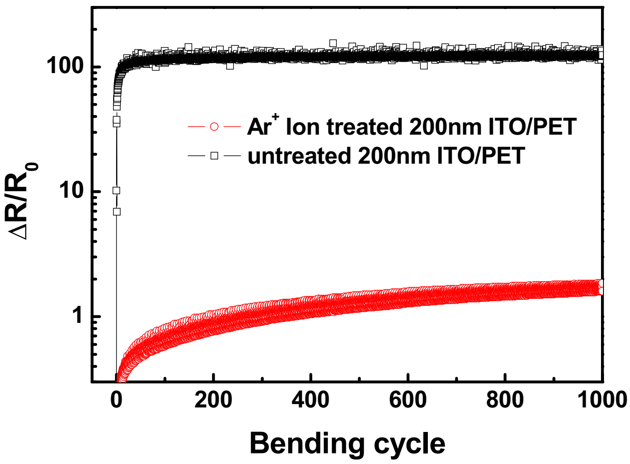

Fig. 6. Normalized resistance change after repeated bending as a function of the number of cycles for flexible ITO electrode grown on untreated and Ar ion treated PET substrate before ITO sputtering.

However, the flexible ITO electrode grown on the Ar ion treated PET substrates showed a fairly constant $\Delta R/R_0$ value throughout the bending test. The robustness of the flexible ITO electrode grown on the Ar ion treated PET substrate is attributed to the improvement of adhesion between flexible ITO films and the PET substrate. Ratchev *et al.*, reported that the ion beam treatment of PC substrates caused substantial improvement in adhesion strength between Al film and PC substrates [4]. In addition, Zaporojtchenko *et al.*, suggested that the improvement in Cu film-PC substrate adhesion, by low energy ion irradiation, is attributed to the creation of a large density of new adsorption sites [5].

4. Summary

This study has demonstrated the applicability of a roll-to-roll sputtering technique as an alternative to conventional RF or DC sputtering for continuous sputtering of flexible ITO electrode on a PET substrate. It was found that both the electrical and optical properties of the flexible ITO electrode were critically dependent on the Ar/O_2 flow ratio during continuous roll-to-roll sputtering. In addition, all flexible ITO electrode shows amorphous structure and a very smooth surface area regardless the of Ar/O_2 flow ratio, due to the low substrate temperature which is maintained by the cooling drum. Even though the flexible ITO electrode was prepared at room temperature, we can obtain the flexible ITO electrode with a sheet resistance of 47.4 ohm/square and average optical transmittance of 83.46 % in the green region between 500~550 nm wavelengths. In addition, it was found that the Ar plasma treatment of the PET substrate could improve the flexibility of the ITO electrode due to effective remove of surface contamination and increase in adhesion of the ITO film with the PET substrate. This indicates that the roll-to-roll sputtering technique is a promising continuous sputtering process for a continuous production of low cost flexible display

5. References

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