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Optical and electric properties of indium tin oxide thin films deposited by a sputtering method using a moving gun

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Transparent conducting oxide (TCO), indium-tin oxide (ITO) thin films were deposited on a polymer substrate by a RF magnetron sputtering method using a moving gun. A moving gun was used to minimize thermal damage to the substrate and underlying structure. The conditions used for depositing the ITO films were varied. A working pressure in the range of 0.67 to 2.67 Pa and an oxygen flow rate in the range of 0 to 0.3 SCCM were used. The roughness, mobility, electrical resistivity and optical transparency of the ITO films deposited on the polymer substrates were evaluated. The electrical resistivity of ITO films deposited on polyethylene terephthalate substrates at room temperature had a relatively low electrical resistivity, 9.014×10^4 Ω·cm. ITO films had a higher electrical resistivity when prepared using a, higher oxygen flow rate and working pressure. The optical transmission of the ITO films was above 85% in the wavelength range 450 to 750 nm. The band gap of the ITO films decreased with an increase in both working pressure and oxygen pressure.

Key words: ITO, RF magnetron sputtering, optical transparency, transparent conducting oxide.

Introduction

Because of their low resistivity and high optical transmission in the visible spectral region, transparent conducting oxide (TCO) thin films are widely used in a variety of applications, including optoelectronic devices, such as plasma display panels (PDPs), organic light emitting devices (OLED), and solar cells, *etc.* [1, 2]. Indium-tin-oxide (ITO) has been the subject of extensive investigations and is generally considered to be an important functional TCO material. ITO is an n-type highly degenerated semiconductor with a wide band gap (E_g is about 3.3 ~ 4.3 eV). ITO films have been fabricated by DC or RF magnetron sputtering [3-7], ion beam sputtering [8], electron beam evaporation [9], and pulsed laser deposition [10]. Among these methods, magnetron sputtering is the most attractive technique because of its high deposition rate, good quality and large area coating. The majority of studies of ITO films to date have involved the use of glass substrates. However, the deposition of high quality ITO films on polymer substrates would be highly desirable for applications of flexible display devices. Depositing a high quality ITO thin film on a flexible polymer substrates represents a critically important key technology. Because of the difference in thermal expansion coefficients between the coating material and the polymer, a thermal stress can occur, causing the substrate to bend, resulting in thin film cracking, ultimately leading to element invalidation. As a result, problems associated with high process temperatures, high thermal expansion coefficients and thermal stress problems must be addressed [11, 12]. It is necessary to minimize plasma thermal damage for good quality thin films.

Thermal damage by plasma can alter the underlying structure of an electronic device or the substrate itself. This issue has been investigated by several workers using a RF magnetron sputtering method. In this study, a moving gun was used to deposit the ITO film on the polymer substrate, using an RF sputtering method, in an attempt to minimize or eliminate thermal damage. ITO thin films were prepared on a pyrex 7740 and polymer (PET) substrates using various deposition conditions (O_2 content, working pressure) by an RF magnetron sputtering method with a moving gun. The microstructure, optical and electrical properties of the products were evaluated as a function of deposition conditions and the surface morphology of the plasma-treated polymers were examined by atomic force microscopy.

Experimental Details

Using an ITO (90% In_2O_3 + 10% SnO_2) target, the ITO films were deposited on pyrex 7740 and polyethylene terephthalate (PET) substrates by RF magnetron sputtering technique with a moving gun. The base pressure of the sputtering chamber was below

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0.00067 Pa while the pressure (P) during the sputtering process was about 0.75 ~ 4.00 Pa. Mixtures of Ar and O_2 (Ar : O_2) from 40 : 0 to 40 : 0.3 SCCM were used as the sputtering gas. The target-substrate distance was fixed at about 10 cm, where the substrate was placed parallel to the target surface. The substrate temperature was set at room temperature. The thicknesses of the ITO films were approximately 130 ~ 155 nm. The effects of the O_2 content, and working pressure on the structural, optical and electrical properties of the films were investigated by X-ray diffraction (XRD), atomic force microscopy (AFM), optical transmission and Hall effect measurements, respectively. An XRD analysis of the films was performed using a Rigaku model D/max-2400 (with Cu-K_α radiation). The surface morphology of the thin films was examined by an atomic force microscope (DI, MultiMode SPM). Spectral transmittance in the wavelength range of 200 ~ 800 nm was carried out using a Shimadzu UV-365 spectrophotometer (Shimadzu, spectral range 200 ~ 2500 nm). In addition, the electrical properties *i.e.* resistivity, Hall mobility and carrier concentration of the films were measured using a standard fourprobe Van der Pauw method in a magnetic field of 0.9 mT at room temperature.

Results and Discussion

Fig. 1 shows XRD patterns of ITO/pyrex 7740 and ITO/PET films for the deposition conditions (100 W,

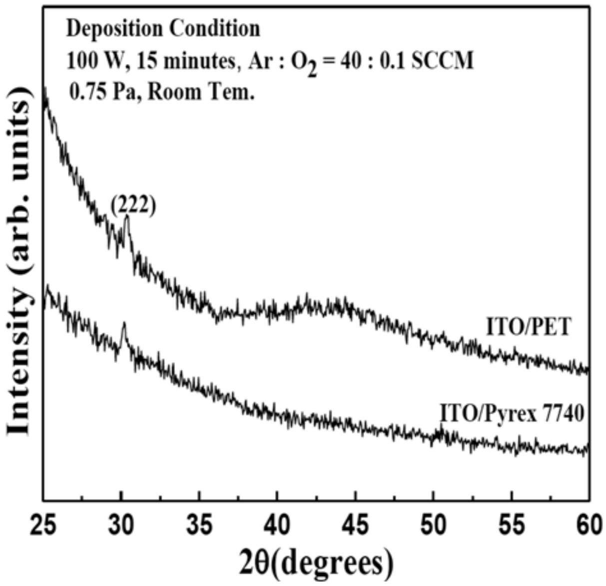

Fig. 1. XRD patterns of ITO/Pyrex 7740 and ITO/PET films deposited at room temperature.

15 minutes, Ar : O_2 = 40 : 0.1 SCCM, W. P. : 0.75 Pa) at room temperature. The XRD pattern of the thin films showed a preferred (222) orientation. The XRD pattern of films with deposition conditions indicated a similar pattern.

The structure of a polymer surface created using an atmospheric plasma can be at a nm scale and can influence the adhesion, optical and wettability

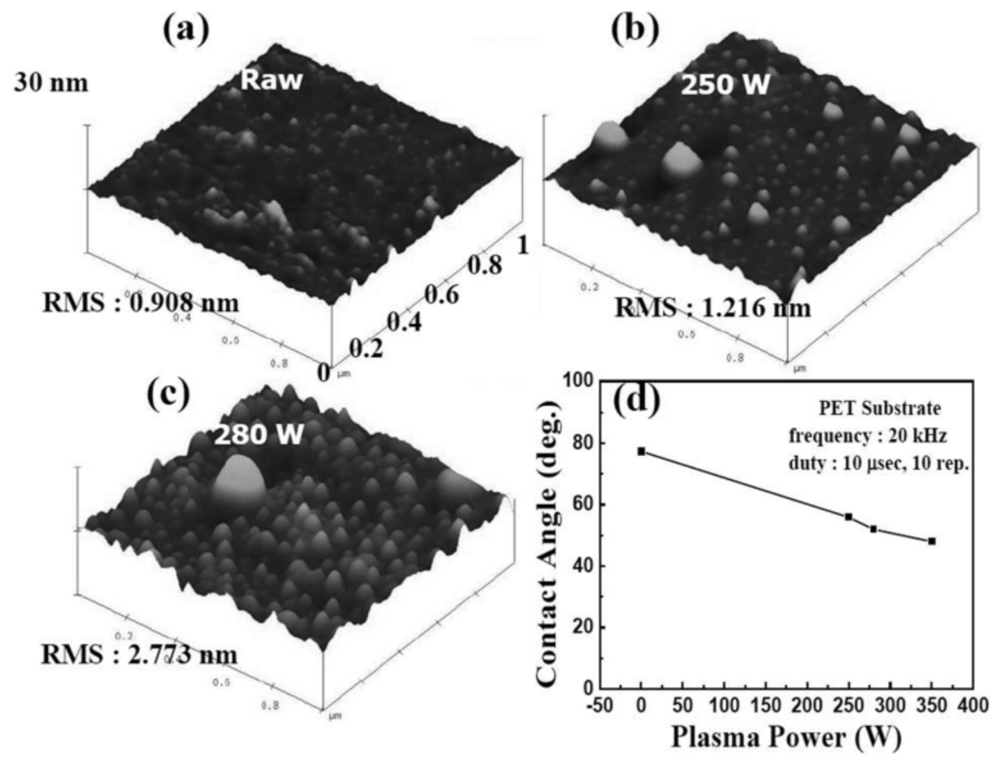

Fig. 2. AFM 3-D images of PET substrates as a function of the plasma treatment power used. (a) 0 W (Virgin), (b) 250 W and (c) 280 W.

properties of a material. Therefore, we also examined the morphology and contact angle of the PET polymer surface by plasma treatment. An atmospheric plasma was generated in a capacitive-coupled device operating at 20 kHz with various powers (250 and 280 W).

Fig. 2 shows AFM 3-D images of the PET substrate as a function of the plasma treatment power. After the plasma treatment, the root mean square (RMS) roughness, as measured by AFM, for the polymer substrate (PET) indicated that the larger the plasma power used, the higher was the roughness. The RMS values were determined to be 0.908, 1.216 and 2.773 nm for PET films at fluences of 0 W (virgin), 250 W and 280 W, respectively. This suggests that the distribution of ridges is greatly influenced by the plasma treatment used. The contact angles of water for the plasma-treated PET, were measured according to the plasma power. When the surface of the PET was exposed to air plasma, the contact angles for water on the surface decreased considerably. The values of the contact angle were 77.3, 55.9 and 51.9^{o} for PET at fluences of 0 W (virgin), 250 W and 280 W, respectively. The PET treated with a plasma power of 280 W showed the lowest relative angle (51.9^{o}). Such a decrease in contact angle shows that some modification reactions, resulting in hydrophilic behavior had occurred on the polymer surfaces by exposure to the plasma.

Fig. 3(a) shows the RMS of ITO/Pyrex 7740 and ITO/PET films as a function of the working pressure. If the pressure is too low, the average mean free sputtering particle path tends to become longer and the bombardment energy is excessively strong, thus

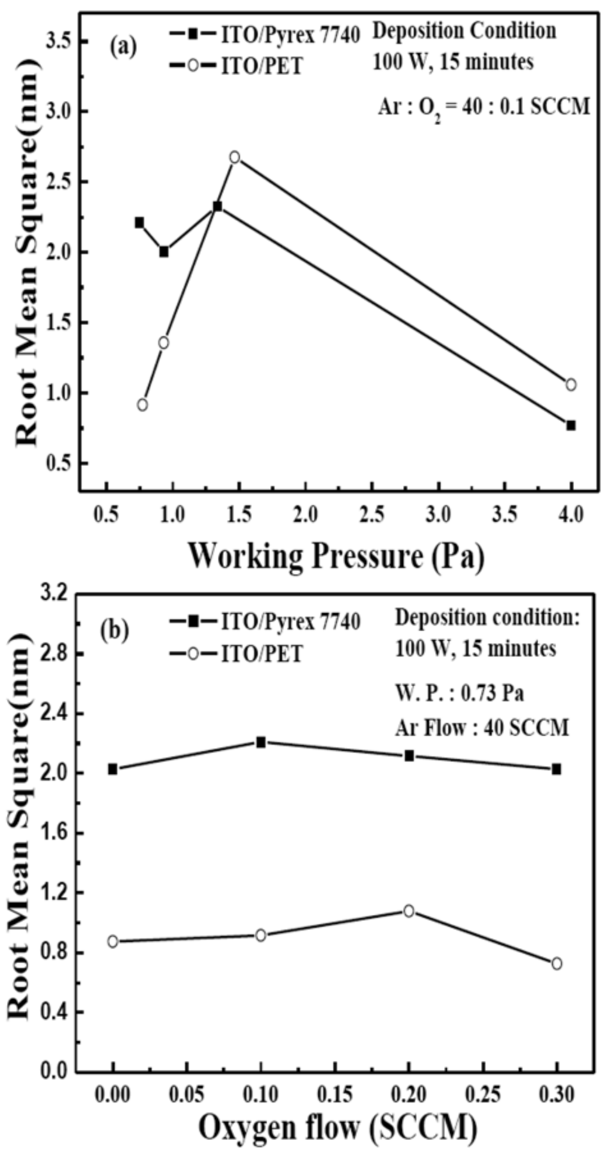

Fig. 3. Root mean square roughness of ITO/Pyrex 7740 and ITO/PET films with deposition conditions of (a) working pressure and (b) oxygen flow, respectively.

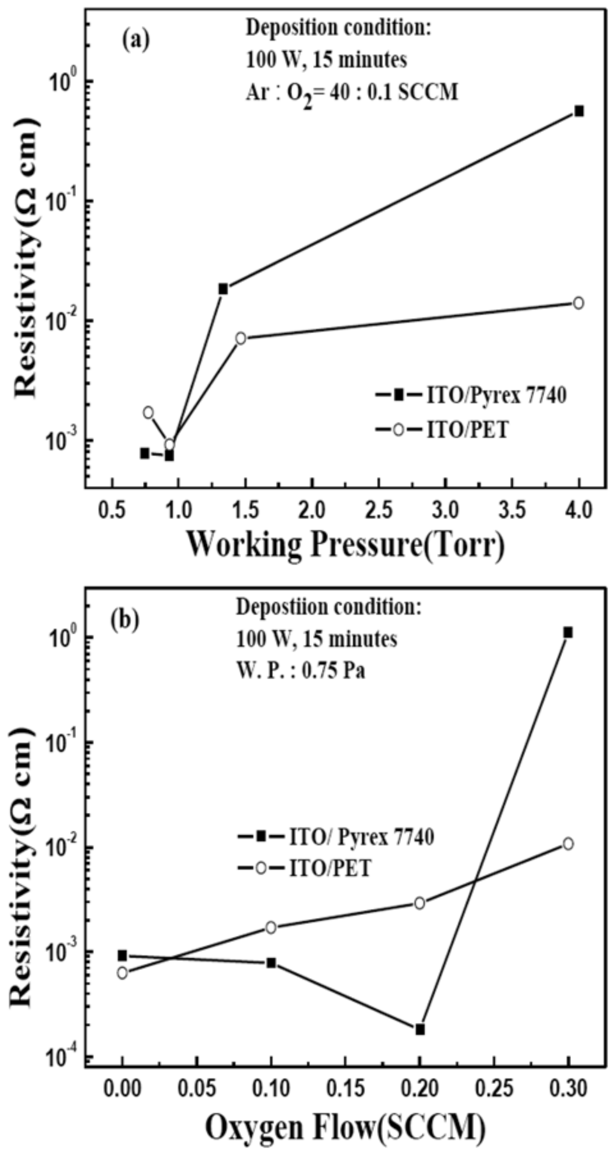

Fig. 4. Resistivity of ITO/Pyrex 7740 and ITO/PET films with deposition conditions of (a) working pressure and (b) oxygen flow, respectively.

causing damage to the substrate to increase. However, the pyrex 7740 substrate was not affected by the bombardment energy of the plasma. Therefore, as shown in Fig. 3(a), the RMS of ITO/Pyrex 7740 films tend to decrease generally for a working pressure in the range of 0.67 to 4.00 Pa. The RMS of the ITO/PET films increased in the working pressure region of 0.75 to 1.33 Pa. The RMS of the films was measured after deposition. It can be concluded that the surface roughness of the initial polymer substrate caused by plasma damage relaxed after the ITO deposition. In addition, ITO/PET films deposited at a high working pressure (4.00 Pa) had a relatively low RMS value because of the low extent of plasma damage, as the result of the use of the moving gun. Fig. 3(b) shows RMS data for ITO/Pyrex 7740 and ITO/PET films as a function of oxygen flow rate. The RMS of the ITO/ Pyrex 7740 and ITO/PET films remained unchanged for oxygen flow in the range of 0 to 0.3 SCCM.

Fig. 4 shows the resistivity of the ITO/Pyrex 7740 and ITO/PET films with deposition conditions of (a) working pressure and (b) oxygen flow, respectively. The resistivity of the ITO/Pyrex 7740 and ITO/PET films tends to generally increase with increasing deposition conditions. A lower resistivity was obtained in the case where the working pressure was reduced to 0.93 Pa. When the working pressure was increased to 4.00 Pa, the resistivity of the ITO thin films increased. Canhola *et al.*[13] used RF magnetron sputtering for depositing an ITO thin film onto a glass substrate and the resistivity was decreased with a lower working pressure.

Data on the optical transmission of the ITO films grown on the substrate PET as a function of deposition conditions was collected. The ITO thin films were deposited on the substrate using a working pressure of 100 W, 15 minutes and Ar : O_2 = 40 : 0.1 SCCM (Fig. 5(a)). At wavelengths below 800 nm, the optical transmission was similar for all films. However, the ITO thin films (PET substrate) deposited at 4.00 Pa showed about an 80% transmission in the wavelength range of 450 to 800 nm. The ITO thin films were deposited on the substrate PET with an oxygen flow at 100 W, for 15 minutes and a working pressure of 0.75 Pa (Fig. 5(b)). The optical transmission of the resulting ITO film was above 85% in the wavelength range from 450 to 750 nm. Therefore, we predict that the deposition conditions for ITO films using a moving gun is a working pressure of 0.67 to 2.70 Pa and an oxygen flow of 0 to 0.2 SCCM.

The optical transmittance data for the ITO/pyrex 7740 films with deposition conditions were analyzed based on the well- known relationships [14] :

$$\alpha h\nu = C_d(h\nu - E_{g1})^{1/2} \quad (1)$$

$$\alpha h\nu = C_i(h\nu - E_{g2})^{2} \quad (2)$$

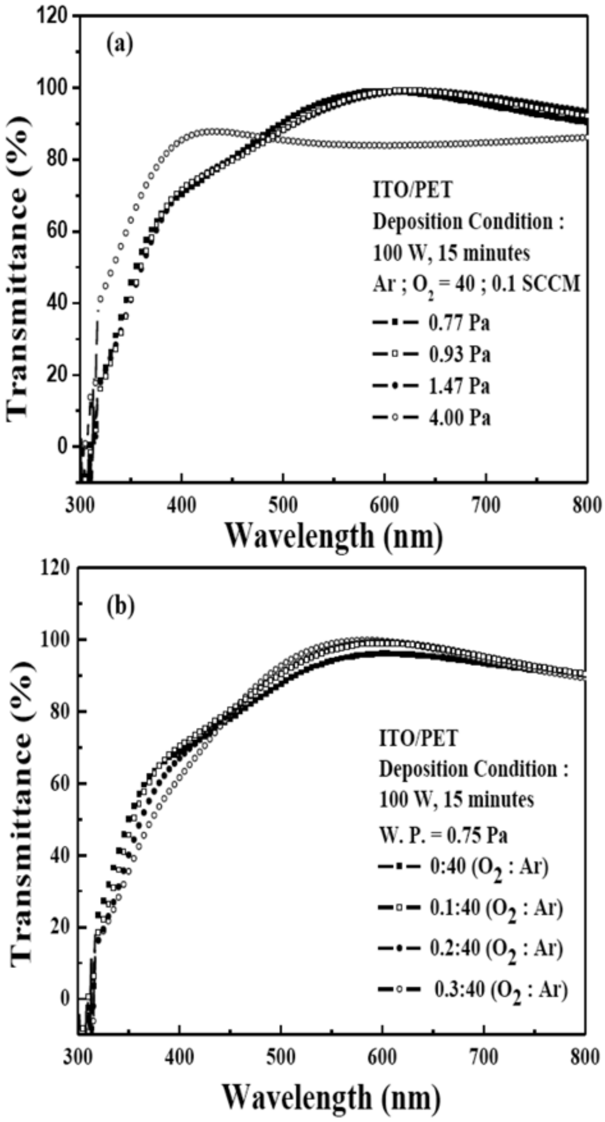

Fig. 5. Transmittance of ITO/PET films with deposition conditions of (a) working pressure and (b) oxygen flow, respectively.

for direct and indirect transitions, respectively, where C_d and C_i are constants, α is an absorption coefficient, hν is the photon energy, E_{g1} and E_{g2} are the direct and indirect band gaps, respectively. The ITO/pyrex 7740 films obeyed the directed transitions [Eq. (1)]. In the plot of $(\alpha h\nu)^2$ *vs.* hν, the band gaps were obtained by extrapolating the linear parts on the hν axis. Fig. 6 shows the extrapolation results for the transmittance measurement of the ITO/pyrex 7740 films with deposition conditions (a) working pressure and (b) oxygen flow. The E_g values were 3.56, 3.53, 3.57 and 3.45 eV for a working pressure of 0.75, 0.93, 1.33 and 4.00 Pa, respectively. In addition, the E_g values were 3.51, 3.60, 3.46 and 3.23 eV for oxygen flows of 0, 0.1, 0.2 and 0.3 SCCM, respectively. Generally, the band gap was decreased with an increase in working pressure and oxygen pressure.

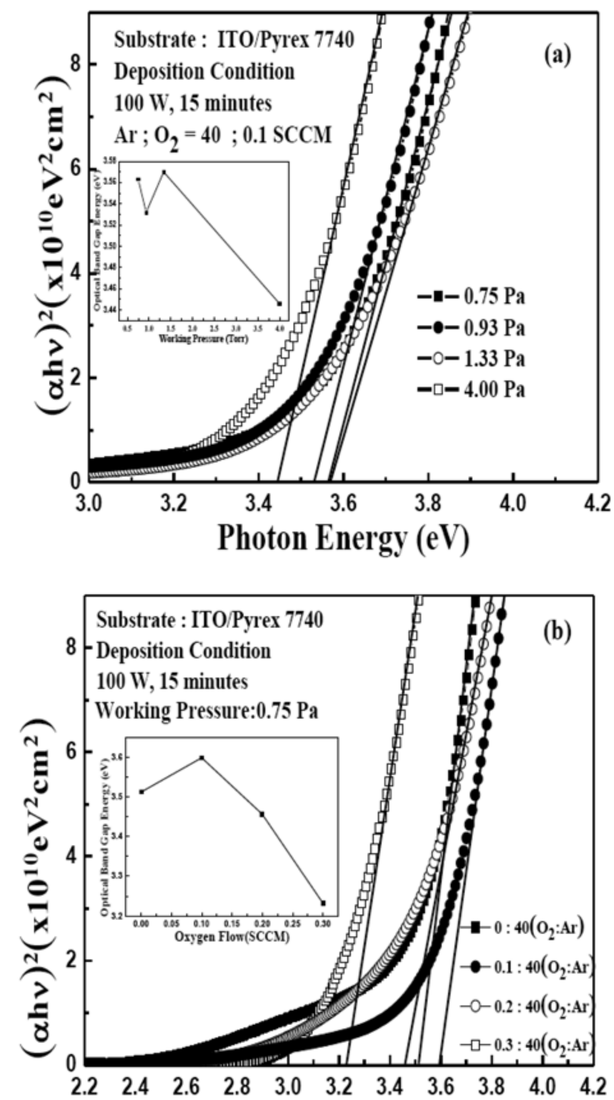

Fig. 6. Extrapolation results for the transmittance measurement of the ITO/pyrex 7740 films with deposition conditions (a) working pressure and (b) oxygen flow.

Conclusions

We deposited high quality ITO thin films on Pyrex 7740 and a polymer (PET) substrate by RF magnetron sputtering using a low damage sputter gun, a moving gun in order to minimize thermal damage to the films and the polymer substrate. The findings indicate that surface roughness increases with increasing plasma power (RMS: $1.216 \sim 2.773$ nm for PET films at fluences of 250 and 280 W). A low RF power, a high vacuum and a low oxygen flow rate are needed to produce ITO films suitable of use in TCO. The electrical resistivity and roughness of the ITO on the PET substrate were $9.014 \times 10^{-4}\,\Omega \cdot$cm and 0.88 nm at 100 W, W. P. = 0.75 Pa and 0.1 SCCM oxygen flow rate. The optical transmittance of the ITO films was above 85% in the wavelength range 450 to 750 nm. The band gap of the ITO films decreased with an increase in working pressure and oxygen pressure.

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