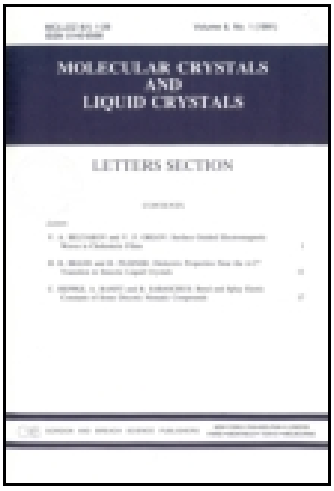

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Characteristics of Ion Beam Assisted ITO Thin Films Deposited by RF Magnetron Sputtering

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Indium tin oxide (ITO) thin films have been deposited onto glass substrates at low temperature (100 °C) by the ion beam assisted RF magnetron sputter technique at different ion beam energy. The structural, surface morphology and electrical characteristics of the ITO thin films were investigated as a function of ion beam assisted energy. With increasing ion beam assisted energy from 0 eV to 100 eV, electrical resistivity of the ITO films reduced from 1.13 × 10^{-3} Ω·cm to 5.5 × 10^{-4} Ω·cm. Hall mobility and carrier concentration slightly increased, which attribute to slightly the crystal growth and harden ITO film. The preferential crystalline orientation of the sputtered films is observed gradually a change from the (222) to the (400) crystalline orientation with increasing ion beam assisted energy.

Keywords Indium tin oxide; resistivity; ion beam; x-ray diffraction; electro-optic property; transmittance

Introduction

Indium tin oxide (ITO) is one of the most attractive materials for application as transparent conductive electrode in optoelectronic and electronic devices such as liquid crystal displays (LCDs), organic light emitting diodes (OLEDs)· flat panel displays (FPD)· solar cells, and surface layers in electroluminescent applications, owing to its high transmittance in the visible spectral region, low electrical resistivity and thermal stability. [1–5] ITO thin films have been deposited by various techniques such as various sputtering, evaporation, plasma-assisted electron beam evaporation, and chemical vapor deposition (CVD) [6–8]. In order to optimize electrical and optical characteristics such as high transmittance and low resistivity, ITO thin films deposition is carried out at relatively high temperature (≥300°C) and it was annealed above ITO crystallization temperature (≥200°C). However,

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some ITO-based devices, such as flexible electro-optical devices and photovoltaic devices based on amorphous silicon, need the deposition of ITO at the low substrate temperature. With conventional techniques, low temperature deposition is difficult. Although there are some techniques for the low temperature deposition with low electrical resistivity such as sputtering and reactive evaporation technique, the oxygen gas flow during the ITO film deposition is required to be controlled accurately owing to quite different ITO films properties with small variations on oxygen gas flow. Whereas, ion beam assisted sputtering technique is suitable for good electrical conductivity with low temperature deposition of ITO films [9–10]. Furthermore, ion beam assisted sputtering technique has merits such that it can be deposited dense and smooth films without almost exposure damage.

In this research, the effects of ion beam assistant energy under radio frequency (RF) magnetron power on electrical and optical properties of transparent indium tin oxide (ITO) film were investigated.

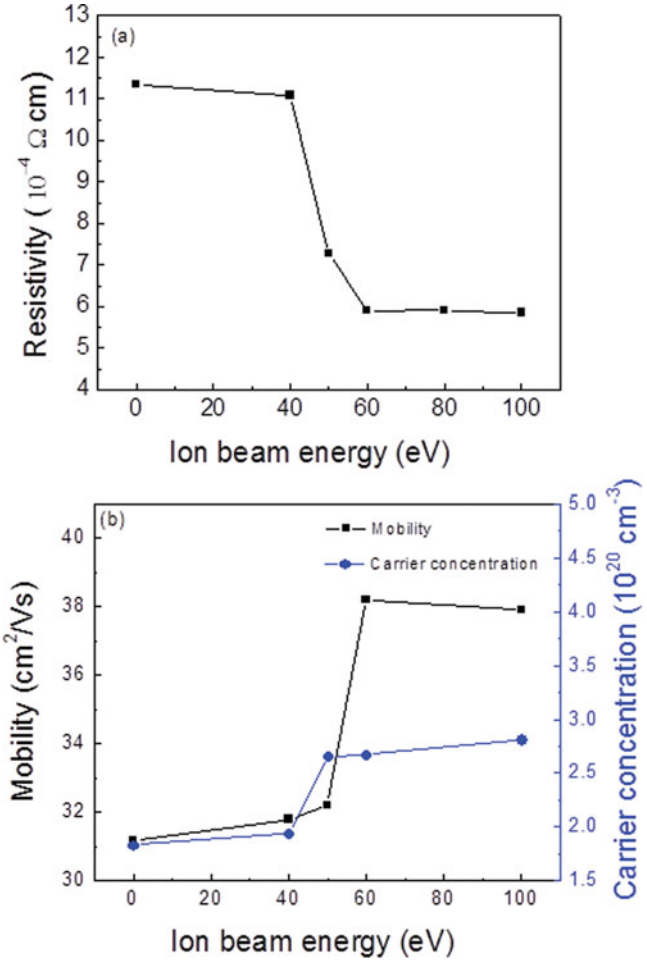

Figure 1. (a) Variation of electrical resistivity and (b) dependence of Hall mobility and carrier concentration of ITO films deposited at different ion beam assistant energy with 50 W of RF power.

Experimental

The ITO films were deposited by the ion beam assisted RF magnetron sputtering on the glass substrates. The ITO target used was in 90 wt.% In_2O_3 and 10 wt.% SnO_2. All the substrates were ultrasonically cleaned in acetone and distilled water, successively, and then dried on a hot plate at 100°C for 5 minutes. The deposition was carried out at 20 sccm of Ar gas that was controlled by a mass flow-meter. The base pressure of the sputtering system was 5×10^{-6} Torr and the working pressure was 8×10^{-4} Torr. The applied RF power was 50 W and this was kept constant during all processes. The depositions were carried out at 100°C for 40 minutes. The ion beam energy is varied from 0 to 100 eV.

The crystalline structure of the deposited ITO films was analyzed by X-ray diffraction pattern observed from X-ray diffractometer (the X'Pert PRO MPD, PANalytical). Electrical resistivity, carrier mobility and concentration values were measured from Hall effects by using a Nanometrics HL5500 Hall System. Spectral reflectance as a function of applied voltage was measured using a cary 5000 UV-VIS-NIR spectrophotometer (Agilent-Korea).

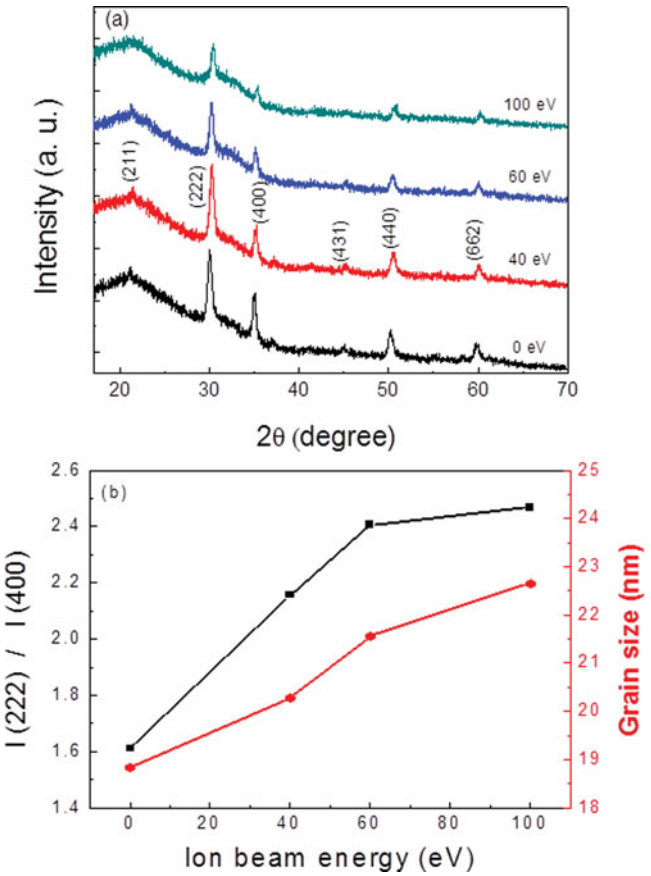

Figure 2. Characteristics of (a) the XRD patterns, (b) I(222) / I(400) and grain size of the ITO films deposited at various ion beam assisted energy.

Results and Discussion

We investigated the electrical and optical characteristics of the ITO films deposited with several ion beam assistant energy conditions. Figure 1 shows the electrical properties of the ITO films deposited as a function of ion beam assisted energy with 50 W of RF magnetron sputtering power. As shown in Figure 1 (a), the electrical resistivity gradually decreased as increasing ion beam exposure energy. With increasing ion beam assistant energy from 0 eV to 100 eV, their resistivity varied from 1.13×10^{-3} Ω·cm to 5.6×10^{-4} Ω·cm. Especially, in the range from 40 eV to 60 eV, the electrical resistivity decreased rapidly. It is explained that the ITO films with increasing with ion beam assisted energy may be ascribed to decrease of grain boundary scattering due to larger grain size, densification, and so on. Furthermore, the minimum resistivity obtained at 100°C of substrate temperature with 100 eV of our experiment is 5.6×10^{-4} Ω·cm, which is similar compared with 3.4×10^{-4} Ω·cm achieved at substrate temperature of 350°C reported by Boonyopakorn [11]. The dependence of Hall mobility and carrier concentration of ITO films deposited at different ion beam assistant energy with 50 W of RF power is shown in Figure 1 (b). As increasing ion beam assisted energy, the mobility and carrier concentration was founded to be in the range 31.2 $cm^2/V{\cdot}s$ $\sim$ 37.9 $cm^2/V{\cdot}s$ and 1.84×10^{20} cm^{-3} $\sim$ 2.81×10^{20} cm^{-3}, respectively. These may be due to decrease grain boundary and harden ITO film due to ion beam collision and current with increasing ion beam assisted energy.

Figure 2(a) shows the XRD patterns of the ITO films deposited at various ion beam assisted energy. The X-ray diffraction peaks of the ITO films at about 21å, 30å, 35å, 45å,

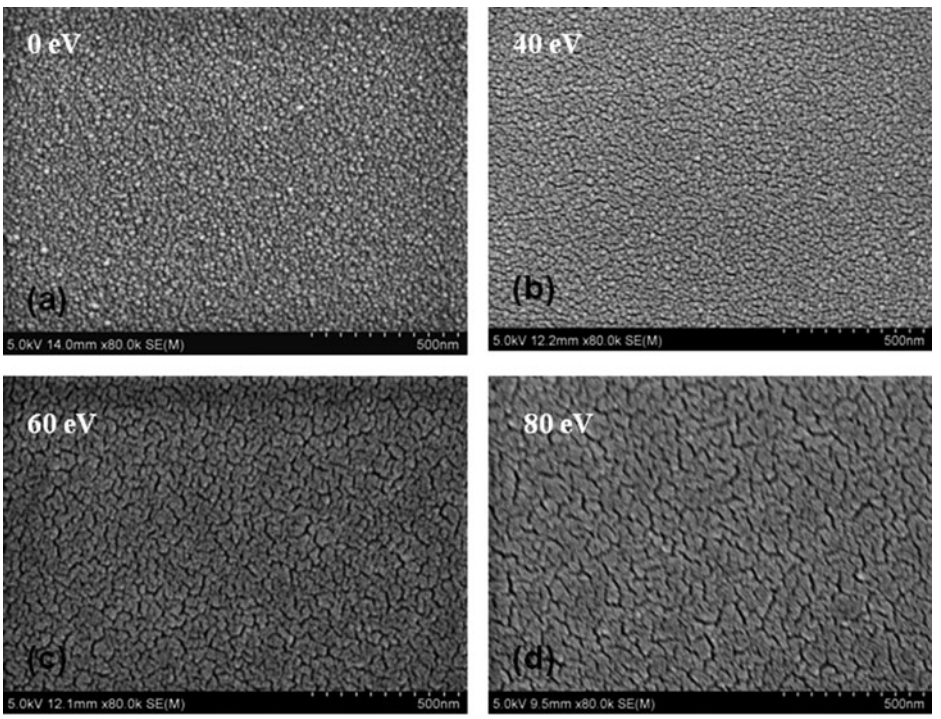

Figure 3. SEM images of the surface of the ITO films deposited with various ion beam assistant energy.

51å, and 60å coincide with reference peaks of the (211), (222), (400), (430), (440), and (622) planes of the bixbite cubic structure of In_2O_3. All the deposited films regardless of ion beam assisted energy showed polycrystalline structure and a strong (222) and (400) preferred orientation. The ITO film texture changes from (222) to (400) as ion beam assisted energy is increased. According to Chopra et al. [12], electrical resistivity of an ITO thin film mainly depends on a carrier concentration. The carrier concentration of ITO is directly related to the deviation from nonstoichiometry of indium oxide which is due to oxygen vacancies and substitute Sn affect. The XRD peak intensity ratio, I(222) / I(440) is directly related to the carrier concentration.

This change in the preferential orientation is more obviously showed in Fig. 2 (b). As shown in Fig. 2 (b), the gain sizes, D, are calculated from the full width at half maximum (FWHM) by using the Scherrer's formula as follows [13]:

$$D = \frac{0.9\lambda}{B cos\theta_B}$$

where $\lambda = 0.154186$nm, θ is the Bragg diffraction angle and B is the FWHM in radian of the diffraction line peak at an angle of the Bragg diffraction. With increasing ion beam assisted energy from 0 eV to 100 eV, grain size of the ITO films increased slightly from about 19 nm to 23 nm. With the increasing ion beam assisted energy, larger grain sizes may be explained by higher adatom mobility, which enhanced the resistivity of the deposited films.

To investigate the possible correlation between electrical properties and the surface morphology of ITO films, its surface morphologies according to ion beam assisted energy using a field emission scanning electron microscope (SEM) were observed. Figure 3 shows SEM images of the surface of the ITO films deposited at various ion beam assisted energy. As increasing ion beam assisted energy, the grain size increased slightly. This can be explained that the surface morphology also play an important in the electrical properties of ITO films, as shown in Fig 1.

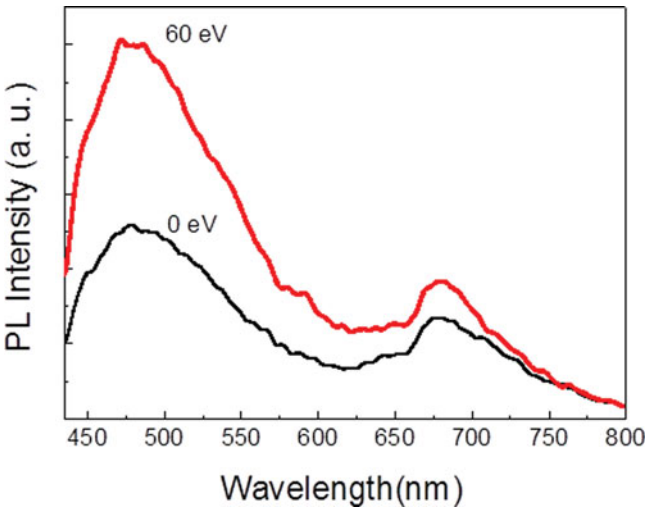

Figure 4. Room temperature PL spectra of the ITO films deposited at different ion beam assisted energy: (a) 0 eV and (b) 60 eV.

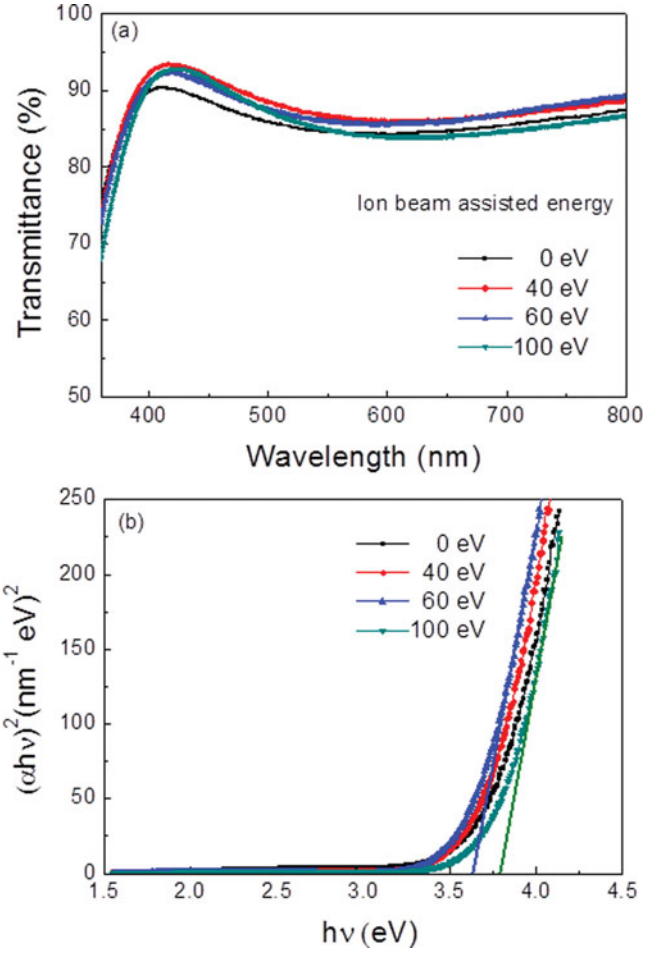

Figure 5. (a) Optical transmittance spectra and (b) Corresponding plot of $(\alpha h\nu)^2$ versus $h\nu$ for ITO thin films deposited at different ion beam assisted energy.

Figure 4 shows strong and broad PL emission spectra of the deposited ITO films at different ion beam assisted energy with 100°C of substrate temperature by 50 W of RF power. The intensity of the PL emission spectrum at 480 nm increased compared to that of unexposed ITO film (0 eV) as increasing ion beam assisted energy by 60 eV. It is known that the PL peak mainly attributed to effect of oxygen deficiencies from In_2O_3 [14]. As shown in Fig. 1, this is associated with an increase in the carrier concentration, which is related to enhance the electrical resistivity.

Figure 5 shows the optical transmittance spectra and corresponding plot of $(\alpha h\nu)^2$ versus $h\nu$ for ITO thin films deposited at different ion beam assisted energy. The average transmittance in the visible range was about 85%, as shown in Fig. 5(a). In Fig. 5(b), the optical band gap energy (E_g) can be determined for the direct allowed transition by using the relation [15]:

$$\alpha h\nu = A\left(h\nu - E_g\right)^{1/2}$$

where A is a constant, α is an absorption coefficient and $h\nu$ is the photon energy.

From the transmission data as shown in Fig. 5(b), the optical band gap energies for the ITO films deposited at different assisted energy was between 3.64 and 3.79 eV. Furthermore, as increasing ion beam assisted energy by 60 eV, band gap energy was decreased slightly. It may be considered as that the XRD peak intensity of polycrystalline structure of (222) plane due to stoichiometric oxides (In_2O_3) increase compared to (400) plan direction with the increase in ion beam assisted energy as shown in Fig. 2(b).

Conclusions

The effects of ITO thin films deposited at different ion beam assistant energy at 100°C of substrate temperature by RF magnetron sputtering were investigated. The electrical resistivity and mobility gradually decreased as increasing ion beam exposure energy. The resistivity obtained at 100 eV of ion beam assistant energy decreased to 5.6×10^{-4} Ω·cm. The XRD spectra showed preferential orientation of polycrystalline structure of (222) plane with increasing ion beam assisted energy by 60 eV. The optical transmission of ITO films is about 85% in the visible wavelength region of spectrum. Moreover, based on these results we can conclude that the electrical properties can be controlled by ion beam assisted energy. We expect that our proposed method can be used at flexible devices which require low temperature process such as flexible displays, foldable e-paper and solar cell.

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