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Influence of Annealing Temperature on the Properties of ITO Films Prepared by Electron Beam Evaporation and Ion-Assisted Deposition

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ABSTRACT

Indium tin oxide (ITO) films were deposited on glass substrates by electron beam evaporation and ion-assisted deposition. Evaporation material source was 90 wt% In_2O_3 and 10 wt% SnO_2 (purity of 99.99%). The ITO films were annealed in the air at 200, 250, 300 and 350 °C for 1 h. The structures, electrical and optical properties of ITO films were investigated. The deposited films were analyzed by X-ray diffractometer, four-point probe method and UV-NIR spectrophotometer. It was found that the lowest resistivity (2.25×10^{-4} Ω-cm) and highest optical transmittance (83%) of ITO films were obtained at the annealing temperature of 300 °C. The grain size increased from 36.69 to 46.73 nm with increasing annealing temperature.

Key words: indium tin oxide, annealing temperature, electron beam evaporation

INTRODUCTION

Indium tin oxide is widely utilized in numerous industrial applications due to its unique combined properties of transparency to visible light and electrical conductivity. ITO films are highly degenerate n-type semiconductors, low electrical resistivity (~10^{-4} Ω-cm), and high carrier concentration. Furthermore, ITO is a wide band gap semiconductor (E_g = 3.5-4.3 eV) (Kim *et al*., 1999), high transmittance (>80%) in the visible range of the electromagnetic spectrum. The optical and electrical properties of ITO films are affected strongly by the concentration of Sn^{4+}.

This unique combination of electrical and optical properties has led numerous researchers to a thorough investigation of the growth and characterization of ITO films. Due to their physical properties, it has potential applications in many devices, such as flat panel displays, solar cells, surface heaters for automobile windows, energy-efficient windows, camera lenses, gas sensors, antireflection coatings and heat reflecting mirrors (Chandrasekhar *et al*., 2001; Qiao *et al*., 2004). For these applications, the film should be a high electrical conductivity and a high visible transparency film.

Many fabrication techniques have been used to produce ITO films. In addition, different processes usually produce ITO films with a significantly different properties, such as dc and rf sputtering (Joshi *et al*., 1995; Shin *et al*., 1999), electron beam evaporation (Paine *et al*., 1999; Pokaipisit *et al*., 2006), spray pyrolysis (Manoj *et*

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al., 2007), pulsed laser deposition (Chandrasekhar *et al*., 2001) and sol-gel process (Alam *et al*., 2002). From all of these techniques, the results show that the electron beam evaporation technique is one of the effective methods to obtain good quality ITO films (Paine *et al*., 1999).

In this paper, we prepared ITO films by electron beam evaporation and ion-assisted deposition (IAD) and investigated the structural, electrical and optical properties of the ITO films as a function of annealing temperature.

MATERIALS AND METHODS

The ITO films were deposited onto glass substrates by electron beam evaporation (Denton DVB SJ-26C) and ion-assisted deposition using a starting material of ITO tablet with a composition of 90 wt% In_2O_3 and 10 wt% SnO_2 (purity of 99.99%). The base pressure of the deposition chamber was 6×10^{-6} mbar while the pressure during evaporating process was about 6×10^{-5} mbar. The pure oxygen (99.99%) was flowed through the End-Hall ion source (Hanil Vacuum Inc.) and it was kept constant at 12 sccm by mass flow controller. The angle between the incident oxygen ion beam and the normal of the substrate was fixed at 45°. The distance between the evaporating source and the rotating substrate holder was fixed at 60 cm. The substrate temperature during the deposition process was maintained at 150 °C by quartz lamp irradiation. The thickness of deposited films was controlled using a quartz crystal thickness monitor in order to obtain a film thickness of 500 nm with an evaporating rate of 2 Å/s. Before loading the glass substrates, there were chemically cleaned using standard methods. After the deposition process, ITO films were annealed in the air at 200, 250, 300 and 350 °C for 1 h.

The crystalline quality and orientations of ITO films were determined by X-ray diffractometer (XRD) analysis at room temperature (Bruker D8 Advance), using Cu K_α radiation with wavelength of 0.154 nm and incident angles in the range of 2θ between 10° and 70°. The X-ray tube voltage and the current were 40 kV and 40 mA, respectively. The electrical sheet resistance and transmittance spectra were measured by a four-point probe method and a UV–NIR spectrophotometer (Perkin–Elmer Lambda 900) in a double-beam configuration, respectively.

RESULTS AND DISCUSSION

The effect of annealing temperature on the optical property of ITO films was examined. The ITO films with thickness of 500 nm were annealed at 200, 250, 300 and 350 °C for 1 h. Figure 1 shows the transmittant spectra of ITO films for different annealing temperatures and Table 1 shows the average transmittance in the visible region (λ = 400-700 nm). It is clear that the transmittance increases with increasing the annealing temperature from 200 to 300 °C. Further annealing temperature (350 °C), the transmittance decreased. In the visible region, the film with thickness of 500 nm (as deposited film, ASD) has a transmittance higher than 60%, but it is increased up to 80% after annealing.

The annealing temperature has influences on the electrical property of the ITO films. Table 1 shows the variation of resistivity as a function of annealing temperature for the ITO films with thickness of 500 nm. As seen in Table 1, the resistivity of the ITO films decreased as the annealing temperature was increased from 200 to 300 °C and then increased as the annealing temperature was further increased. The lowest resistivity of 2.25×10^{-4} Ω-cm was obtained from the film that was annealed at 300 °C.

Figure 2 shows XRD patterns of the ITO films deposited on glass slide substrates annealed at 200, 250, 300 and 350 °C for 1 h. The XRD results suggest that the annealing temperature has the strong effects on the orientation of ITO films.

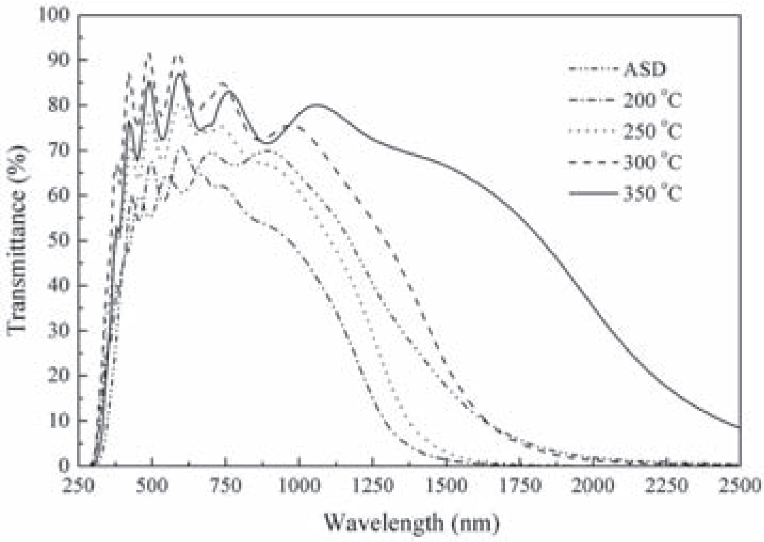

Figure 1 Transmittant spectra of ITO films annealed at different temperatures.

Table 1 Transmittance, resistivity and grain size of ITO films annealed at different temperatures.

Annealing temperature (°C)	Optical transmittance (%)	Resistivity ($\times 10^{-4}$ Ω-cm)	Grain size (nm)
RT	60	2.85	36.69
200	62	2.77	39.94
250	72	2.49	44.63
300	83	2.25	44.69
350	77	5.14	46.73

In this experiment, the annealing temperature was not found to cause a marked change in the crystal structure of the films under investigation at temperatures from 200 to 350 °C. Table 2 shows a comparison between XRD results and standard pattern for pure indium oxide, In_2O_3. The ITO films was found to showed a preferential orientation in the (222) plane. Table 1 reveals that the grain size increased from 36.69 to 46.73 nm with increasing the annealing temperature. The ITO films, grain size in the range of 40-50 nm were annealed at temperatures from 200 to 350 °C. They were determined from the full width at half maximum (FWHM) of the X-ray diffraction peak (222) using Scherrer's formula (Guozhong *et al.*, 2004),

$$D = \frac{0.94\lambda}{\beta \cos\theta} \quad (1)$$

where λ is the X-ray wavelength, β is the FWHM of a diffraction peak and θ is the diffraction angle.

CONCLUSION

The ITO films were deposited onto glass substrates by electron beam evaporation and ion-assisted deposition from an ITO tablet with 10 wt% SnO_2 and annealing at temperatures range 200-350 °C. The structural, electrical and optical properties of the films were studied as a function of annealing temperature. From the transmittant measurement using a spectrophotometer, it was found that optical transparency of ITO films were

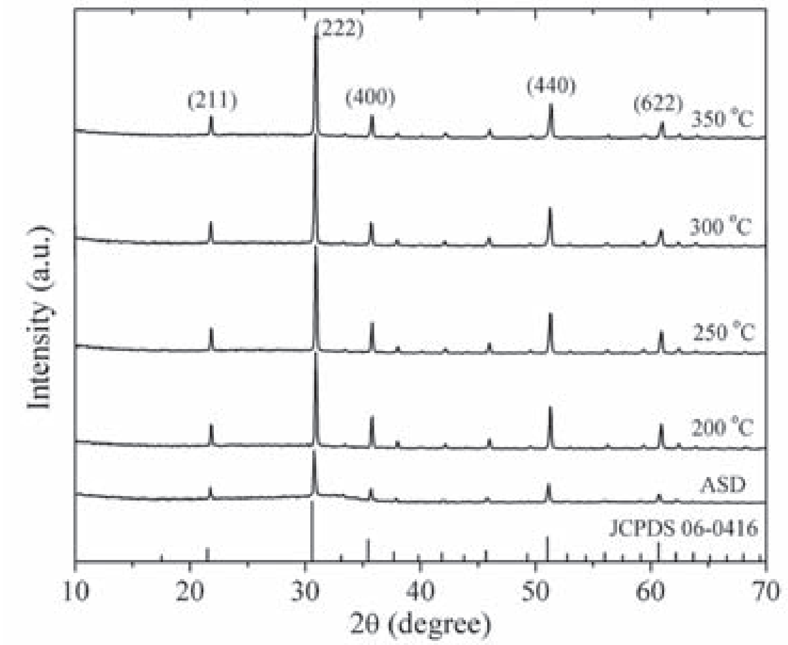

Figure 2 XRD patterns of ITO films annealed at different temperatures.

Table 2 Comparison between the obtained results of XRD peak (annealed at 300 °C for 1 h) and standard peak of indium oxide.

Standard 2θ (°)	Observed 2 (°)	hkl
21.50	21.79	211
30.58	30.89	222
35.47	35.71	400
51.04	51.27	440
60.68	60.89	622

up to 83% in the visible region as the annealing temperature increased. The resistivity of the films had a minimum value of 2.25×10^{-4} Ω-cm when annealed at 300 °C. The grain size increases from 36.69 to 46.73 nm with annealing temperature increase.

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