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Original Article

Influence of oxygen flow rate on properties of indium tin oxide thin films prepared by ion-assisted electron beam evaporation

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Abstract

Indium tin oxide (ITO) thin films with various oxygen flow rates were deposited onto glass substrates by ion-assisted electron beam evaporation. All other deposition parameters were kept constant. The electrical and optical properties of the ITO thin films have been investigated as a function of oxygen flow rate. Optical transmittance and optical band gap energy were measured by spectrophotometer. Sheet resistance was measured by four-point probe method. It has been found that an oxygen flow rate at 12 sccm was suitable for improving the properties of ITO thin films. The resistivity and optical transmittance of ITO thin films were 7.2×10^{-4} Ω-cm and 84%, respectively. The optical band gap was 4.19 eV.

Keywords: indium tin oxide, evaporation, ion-assisted deposition, optical band gap

1. Introduction

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

ITO thin films are widely used in various applications such as flat panel displays (FPD), gas sensors, antireflection coatings, energy-efficient windows, heat reflecting mirrors and solar cells (Qiao *et al.*, 2004; Kim *et al.*, 1999). Many fabrication techniques have been applied to produce ITO thin films. However, different processing usually produces ITO films with remarkably different properties, such as dc and rf sputtering (Shin *et al.*, 1999), electron beam evaporation (Liu *et al.*, 2003; Pokaipisit *et al.*, 2008), spray pyrolysis (Hichou *et al.*, 2004), pulsed laser deposition (Kim *et al.*, 1999), and sol-gel process (Alam and Cameron, 2002). It is well known that ion-assisted deposition (IAD) has the advantage to produce smooth films due to the beam-enhanced migration (Liu *et al.*, 2003).

The oxygen partial pressure is defined as the ratio of oxygen flow flux to the inert gas flux. It is necessary and important to add the oxygen for obtaining films prepared from metallic targets (Shigesato *et al.*, 1992). A higher O_2

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concentration tended to yield transparent films but of high sheet resistance. A lower O_2 concentration, on the contrary, give films with a low sheet resistance and a slightly metallic appearance. The film is considered to be more oxygen deficient or metallic (Karasawa and Miyata, 1993).

In this study, thin films of ITO were fabricated using ion-assisted electron beam evaporation, and the effects of oxygen flow rate on the surface morphology, electrical and optical properties of deposited ITO thin films were investigated.

2. Experimental details

ITO thin films were deposited onto glass substrates by ion-assisted electron beam evaporation (Denton DVB SJ-26C). The evaporation source material was ITO composed of indium oxide 90 wt% and tin oxide 10 wt% with the purity of 99.99%. The base pressure of the deposition chamber was 6×10^{-6} mbar and pure oxygen gas (99.99%) was introduced during deposition. The pressure during the evaporating process was about 6×10^{-5} mbar. The thickness of the ITO thin films was measured using a quartz crystal thickness monitor during the deposition. The thickness of the deposited ITO thin films and the evaporation rate were maintained at 120 nm and 2 Å/s, respectively, for all the films. The substrate temperature during the deposition process was maintained at 150°C by quartz lamp irradiation. The distance between the electron beam evaporation source and the rotating substrate holder was 60 cm. The End-Hall ion source (Hanil Vacuum) was used to produce low-energy oxygen ions. The oxygen flow rate was regarded as a variable parameter, and hence configured at 8, 10, 12, and 14 sccm flow rates via the End-Hall ion source, monitored by a mass flow controller (MKS). The schematic diagram of the deposition system used in our experiment is shown in Figure 1.

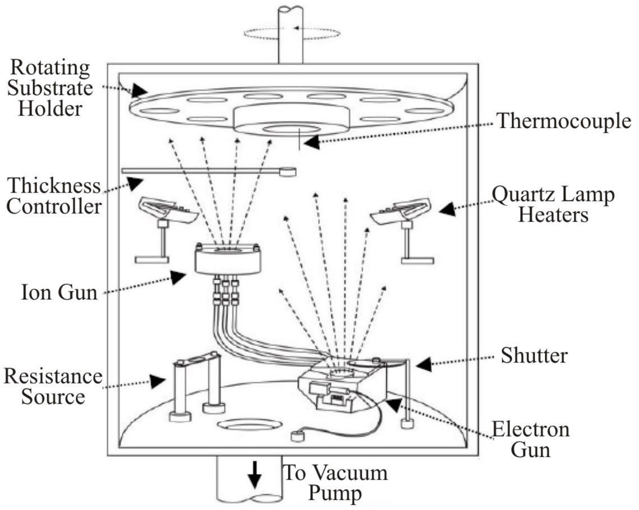


Figure 1. Schematic diagram of the deposition system.

The surface morphology and the root-mean-square surface roughness (RMSSR) of the ITO films were investigated using a NanoScope III atomic force microscope (AFM) with tapping mode. The sheet resistance of the ITO thin films were measured by a four-point probe method (Jandel). The transmittance spectra measurement was made using a UV-NIR spectrophotometer (Perkin-Elmer Lambda 900) in a double-beam configuration. Before loading the glass substrates, they were prepared by ultrasonic washer with acetone, alcohol, and de-ionized water to remove organic contaminants and blew in dry nitrogen gas.

3. Results and Discussion

ITO thin films with oxygen flow rates between 8 and 14 sccm were prepared onto glass substrates. Sheet resistance of the ITO thin films deposited by varying the oxygen flow rates at the conditions described above are shown in Table 1. The sheet resistance showed a minimum value at 12 sccm oxygen flow rate. The increase of oxygen flow rate, that is, oxygen ion beam flux, caused the increase of oxygen incorporation into the oxygen-deficient ITO films, which reduced the sheet resistance of the films by compensating oxygen vacancies in the evaporated ITO thin films. However, the increase of oxygen flow rate higher than 12 sccm increased the sheet resistance of the film.

The sheet resistance (R_s) measurements were performed using a four-point probe method. By assuming that the thickness of the films was uniform, the film resistivity (ρ) was determined using the simple relation $\rho = R_s \cdot d$, where d is the film thickness (Hamberg and Granqvist, 1986). All sheet resistance and resistivity values were determined as the average of three measurements for each film. From Table 1,

Table 1. Properties of ITO thin films at different oxygen flow rates.

Oxygen flow rate (sccm)	Sheet resistance (Ω/sq)	Resistivity (10^{-4} Ω-cm)	Transmittance (%)	RMSSR (nm)	Band gap (eV)	Figure of merit (10^{-4} Ω^{-1})
8	130	15.6	75	0.52	4.15	4.33
10	74	8.9	82	0.74	4.17	18.57
12	60	7.2	84	0.93	4.19	29.15
14	84	10.1	80	0.38	4.16	12.78

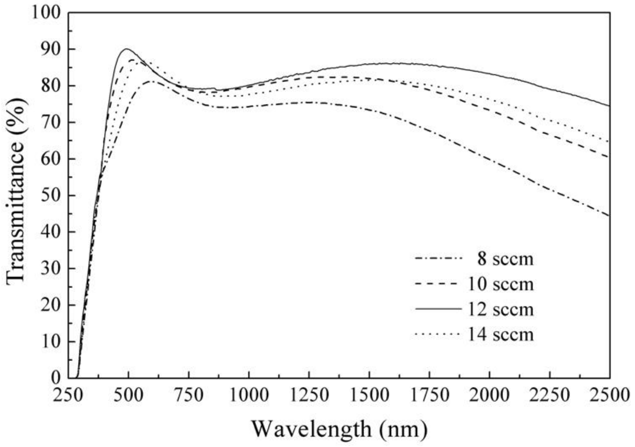


Figure 2. Transmittance spectra of ITO thin films at different oxygen flow rates.

the resistivity of ITO thin films decreases from 15.6×10^{-4} to 7.2×10^{-4} Ω-cm because the carrier concentration increases. The oxygen flow rate can change the carrier concentration due to oxygen vacancies.

Figure 2 shows the variation of transmittance spectra of the ITO thin films deposited with the same conditions described in Table 1. The transmittance increased with increasing oxygen flow rate up to 12 sccm and the further increase of oxygen flow rate decreased the transmittance. Table 1 shows the average optical transmittance (in visible range 400-700 nm) of ITO thin films at different oxygen flow rates, which have an obvious influence on the transmittance. Below 12 sccm oxygen flow rate, an increase of the optical transmittance is observed. The optical transmittance can be increased with a reasonable increase in the oxygen flow rate. When the oxygen flow rate is up to 12 sccm the optical transmittance of ITO thin films with 120 nm thickness shows the highest value. If the oxygen flow rate is above 12 sccm, the optical transmittance will begin to decrease. The results can be explained as following: when the oxygen flow rate is lower, the particles evaporated from the target cannot be oxidized enough so the prepared ITO thin films are anoxic and sub-oxides such as InO_x and SnO_x. The transmittance of ITO thin films were higher because sub-oxides can be oxidated with an increasing oxygen flow rate. However, when the oxygen flow rate is over a maximum, the redundant oxygen can be absorbed in the defects such as grain boundaries and microcracks, which is affirmed by Hamberg and Granqvist (1986). The redundant oxygen can cause optical absorption and scattering. Furthermore, we calculated the optical band gap values of ITO thin films from transmittance spectra. In the strong absorption region, the absorption coefficient (α) can be calculated from Lambert's formula (Ray *et al.*, 1983; Wang *et al.*, 2004),

$$\alpha = d^{-1} \ln (1/T) \tag{1}$$

where T and d are transmittance and film thickness, respectively.

The absorption has its minimum at low energy and increases with optical energy in a manner similar to the absorption edge of the semiconductors. The absorption coefficient for directly allowed transition for simple parabolic scheme can be ascribed as a function of incident photon energy as

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

where $h\nu$ is the photon energy. The optical band gap of ITO thin films can be determined by plotting $(\alpha h\nu)^2$ versus $h\nu$, and extrapolation method. Figure 3 indicates the variation of $(\alpha h\nu)^2$ versus $h\nu$ for ITO thin films prepared in the present study. It is observed that the optical band gap increased from 4.15 to 4.19 eV corresponding to the increase of oxygen flow rates from 8 to 12 sccm.

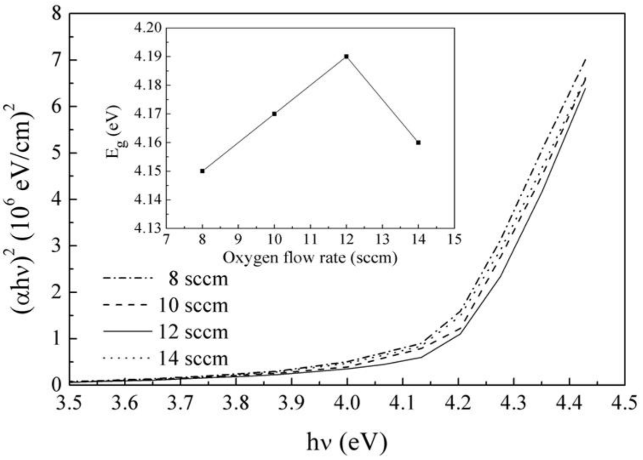
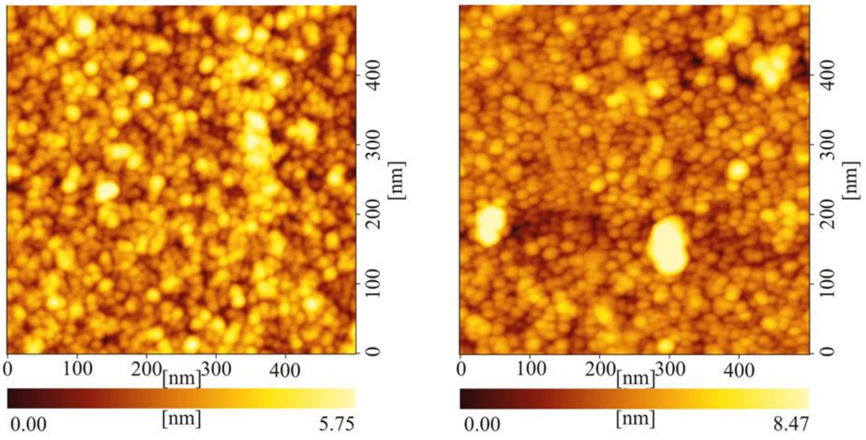


Figure 3. $(\alpha h\nu)^2$ versus $(h\nu)$ plots for ITO thin films at different oxygen flow rates.

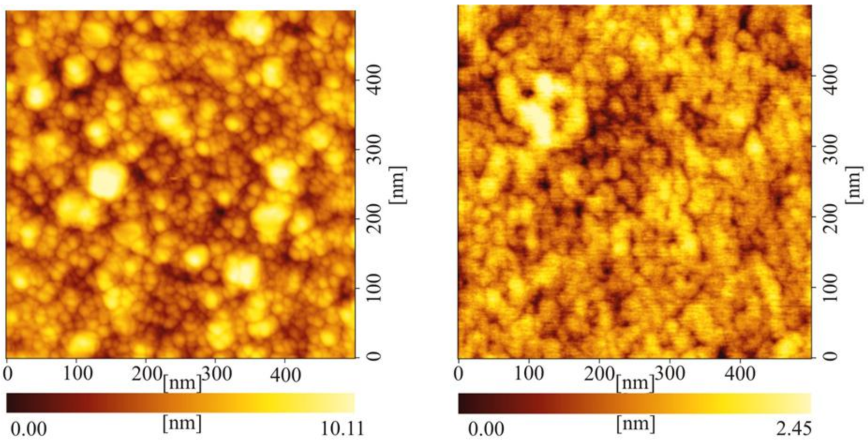
The surface morphologies of ITO thin films were investigated by atomic force microscopy. In this technique we look at surface roughness in terms of the root mean square roughness. Generally, the RMSSR parameter is a useful estimators of the average heights or depths of discrete profiles deviation. Figure 4 shows the surface morphologies of 120 nm ITO thin films deposited at a substrate temperature of 150°C as a function of oxygen flow rate. It is observed from Table 1 that the RMSSR increases from 0.52 to 0.93 nm with an increasing oxygen flow rate from 8 to 12 sccm and decrease to 0.38 nm with an increasing oxygen flow rate up to 14 sccm.

AFM image analysis was used to reveal the particle size and surface morphology of ITO thin films at different oxygen flow rates. It was found that the films consisted of nanosize crystallite with a dimension of about 14-26 nm, which is increased up to 14 sccm. AFM image analysis also shows the depth of about 4-13 nm for ITO thin films at different oxygen flow rates.



8 sccm

10 sccm



12 sccm

14 sccm

Figure 4. 2D AFM images of ITO thin films at different oxygen flow rates.

The simultaneous achievement of maximum optical transmission and electrical conductivity is not possible since these two properties are inversely related. Therefore, a figure of merit has been developed to compare transparent conducting oxide films like ITO. There have been several reports on the definition of a suitable figure of merit. The figure of merit (Φ), defined by Haacke (1976), compares the performance of ITO thin films as

$$\Phi = T^{10}/R_s \tag{3}$$

where T is the optical transmittance and R_s is the sheet resistance. The highest value of the figure of merit was observed for the ITO thin film deposited at a oxygen flow rate of 12 sccm as shown in Table 1. The good figure of merit obtained by the electron beam evaporation technique in comparison to the RF magnetron sputtering (Li *et al*., 2006) with an oxygen flow rate of 9 sccm was $2.33\times10^{-4}\ \Omega^{-1}$.

4. Conclusions

ITO thin films with a wide range of oxygen flow rate have been prepared on glass substrate using ion-assisted electron beam evaporation. The effects of oxygen flow rate on the electrical and the optical properties of ITO thin films were investigated. It was found that the resistivity of ITO thin films decreased with increasing oxygen flow rate up to 12 sccm and then it increased at an oxygen flow rate of 14 sccm. It could be concluded that an optimum oxygen flow rate for obtaining lower resistivity ITO thin films was 12 sccm. The maximum optical transmittance of the ITO thin films was 84% and it was also obtained at an oxygen flow rate of 12 sccm. The morphology of ITO thin films increased with varying oxygen flow rate. Consequently, the ITO thin films deposited at 12 sccm exhibit good electrical and optical properties. Additionally, the figure of merit demonstrated that the ITO thin film with an oxygen flow rate of 12 sccm was the best film in this study, with $\Phi = 29.15\times10^{-4}\ \Omega^{-1}$.

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