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Effect of Films Thickness on the Properties of ITO Thin Films Prepared by Electron Beam Evaporation

Artorn Pokaipisit*, Mati Horprathum and Pichet Limsuwan

ABSTRACT

Indium tin oxide (ITO) thin films were prepared on glass slide substrates at various thicknesses by electron beam evaporation (e-beam) from a tablet of In_2O_3 and SnO_2 (9:1). The ITO thin films were fabricated at substrate temperatures of 150 °C, deposition rate of 2 Å/s and oxygen flow rate of 12 sccm. The structure, surface morphology, electrical and optical properties of ITO thin films were investigated. The structure and surface morphology of films were monitored by using X–ray diffraction (XRD) and atomic force microscopy (AFM), respectively. The optical transmittance and sheet resistance of ITO thin films were measured by spectrophotometer and four-point probe. It was found that transmittance and sheet resistance of ITO thin films on glass slide decreased as film thickness increased from 200 nm to 700 nm. The transmittance spectra in the visible region (400-700 nm) and the sheet resistance decreased from 76.63% to 57.89% and from 16 Ω/sq. to 4 Ω/sq., respectively, with increasing the films thickness.

Key words: indium tin oxide, atomic force microscopy, X–ray diffraction, evaporation

INTRODUCTION

The applications of indium tin oxide (ITO) thin films in optoelectronics were enormous. It can provide manufacturers with a tool that allows them to produce a variety of devices from semiconductor to liquid crystals. New designs of liquid crystal based devices for specific applications are under way by using this kind of substrates as a polymer matrix associated with liquid crystalline materials. Indium tin oxide is a well-known metal oxide degenerate semiconductor. It is largely utilized as transparent conducting electrode in the fabrication of various device structures due to its high conductivity. Many new materials and various manufacturing techniques have been developed. The most widely used materials for transparent conductive thin films are semiconducting which compared to the other oxides, ITO has lowest electrical resistivity and the highest optical transparency (George and Menon, 2000; Pokaipisit *et al*., 2006). Due to these unique properties, ITO thin films are becoming increasingly important in the field of electronic devices. The ITO thin films with an even lower resistivity are required to satisfy commercial demands for flat panel displays with a large size and higher image quality. Moreover, ITO has been used in a wide range of other applications including solar cells, heat-reflecting mirrors, antireflection coatings, gas sensors, antistatic window coatings, touch panel contacts and flat panel displays (Kim *et al*.,1999; Pokaipisit *et al*., 2006).

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Numerous deposition techniques have been utilized to prepare ITO thin films such as DC/RF magnetron sputtering (Shin *et al*., 1999), ion beam sputtering (Kim *et al*., 2000), electron beam evaporation (Paine *et al*., 1999), pulsed laser deposition (Kim *et al*., 1999), spray pyrolysis (Benamar *et al*., 1999) and chemical vapor deposition (Maki *et al*., 2003). In the evaporation technique, the three main kinds are classified as thermal, electron beam and resistive method. In the thermal method, the crucible that holds the source material is radiatively heated by a filament that wind around it. In the electron beam method, the current that heats the crucible is boiled off a filament and is attracted to the crucible by a high voltage. Electron beam evaporation is used with the highest melting elements. Resistive evaporation is accomplished by passing a large current through a wire or foil of the material that is to be deposited. Molecular beam epitaxy is a particularly sophisticated kind of thermal evaporation (Sze *et al*., 2002)

Indium tin oxide is essentially formed by subsititutional doping of In_2O_3 with Sn which replaces the In^{3+} atoms from the cubic bixbyte structure of indium oxide. Sn thus forms an interstitial bond with oxygen and exists either as SnO or SnO_2, accordingly it has a valency of +2 or +4 respectively. This valency state has a direct bearing on the ultimate conductivity of ITO. The lower valence state results in a net reduction in carrier concentration since a hole is created which acts as a trap and reduces conductivity. On the other hand, predominance of the SnO_2 state means Sn^{4+} acts as an n-type donor releasing electrons to the conduction band. However, in ITO, both substitutional tin and oxygen vacancies contribute to the high conductivity.

In this paper, we studied the effect of films thickness on the structure, surface morphology, electrical and optical properties of ITO thin films prepared by electron beam evaporation on glass slide substrate.

MATERIALS AND METHODS

ITO thin films were deposited on glass slide substrates by electron beam evaporation (Denton DVB SJ-26C) from a tablet of indium tin oxide (90 wt.% In_2O_3 and 10 wt.% SnO_2, 99.99% purity). The base pressure of the deposition chamber was 6×10^{-6} mbar and O_2 gas was introduced during deposition. The optimum conditions of films depositing were achieved at substrate temperature of 150 °C, deposition rate of 2 Å/s and oxygen flow rate of 12 sccm. The thickness of the ITO thin films varied in the range of 200–700 nm and was controlled by using a quartz crystal thickness monitor.

The structure of ITO thin films were characterized by X-ray diffraction (Bruker D8 Advance) measurements at room temperature with 40 kV, 40 mA, Cu K_α radiation with wavelength of 1.54 Å, 2θ between 10° and 70° of incident angles. Surface morphology of ITO thin films were scanned by atomic force microscope (SPA-400). The sheet resistance (R_S) measurements were performed using a four-point probe (MDC). The transmittance spectra measurement was made using a UV–NIR spectrophotometer (280–2500 nm) (Perkin–Elmer Lambda 900) in a double-beam configuration. Before loading the glass slide substrates, they were chemically cleaned using standard methods.

RESULTS AND DISCUSSION

ITO thin films with thickness between 200 and 700 nm were prepared on glass slide substrates. Variation of the sheet resistance according to the thickness is shown in Figure 1. The sheet resistance of a film with thickness of 200 nm is as high as 16 Ω/sq. As the film thickness increases to about 700 nm, the sheet resistance rapidly decreased to 4 Ω/sq. In further increase of the thickness, the resistance slowly decreased.

Since the resistivity of ITO thin films is known to be dependent on the structure of the thin films, X-ray diffraction patterns of the films as shown in Figure 2 were compared. For the films with thickness of 200 nm, no characteristic diffraction peak of ITO was identified and only amorphous nature of glass slide substrate was observed. The peaks appear at thickness of 400 nm and the preferential orientations of (211), (222), (400), (440) and (622) become intensive above 400 nm.

Surface morphologies of ITO thin films were investigated by atomic force microscopy (AFM). In this technique we look at surface contours, measuring average roughness R_a and root-mean-squared roughness R_q of the surface of some selected ITO thin films. The average surface

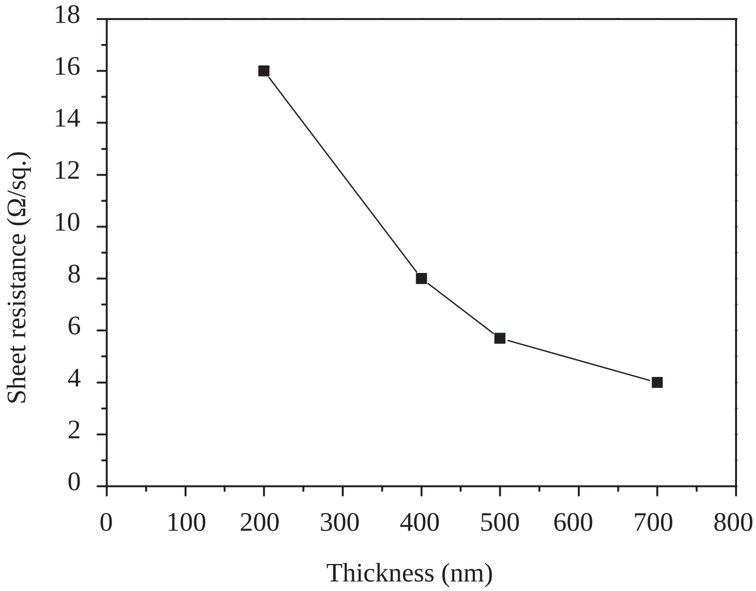

Figure 1 Sheet resistance of ITO thin films with various thicknesses.

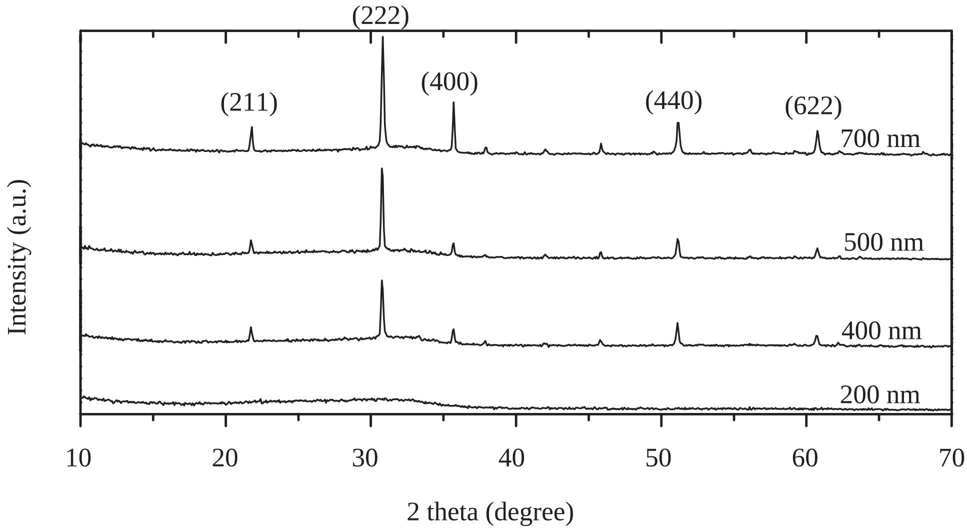

Figure 2 X-ray diffraction patterns of ITO thin films with various thicknesses.

roughness R_a, the most frequently used roughness parameter is the average surface roughness defined as

$$R_a = \frac{1}{4}\sum_{i=1}^{n}|z_i|$$

Where z_i is the height or depth of the i-th highest or lowest deviation and n is the number of discrete profile deviation. The root-mean-squared surface roughness R_q, which is defined as the root-mean-squared of the deviations in the height from the profile mean which may be defined as follows;

$$R_q = \sqrt{\frac{1}{n}\sum_{i=1}^{n} z_i^2}$$

The roughness parameters R_a and R_q are often used as quantitative parameters. Figure 3 shows the surface morphologies of the ITO thin films with films thickness of 200, 400, 500 and 700 nm. Obviously, the average surface roughness R_a and root-mean-square surface roughness R_q increase drastically with oxygen flow rate as the crystals grow larger.

It is observed from Figure 4 that the root-mean-square (RMS) surface roughness increases from 1.01 to 5.30 nm with increasing of the films thickness from 200 to 700 nm.

The transmittance of a typical ITO thin films were grown under the optimum oxygen pressure of 4 × 10^{-5} mbar. Figure 5 shows the wavelength dependent transmittance of the ITO thin films with various thickness of 200 nm (solid curve), 400 nm (dashed curve), 500 nm (dotted curve) and 700 nm (dash-dot curve), respectively, the optical transmittance decreased with thickness increased. It is also observed from Figure 6 that the average optical transmittance decreased from 76.63% to 57.89% in the visible region with

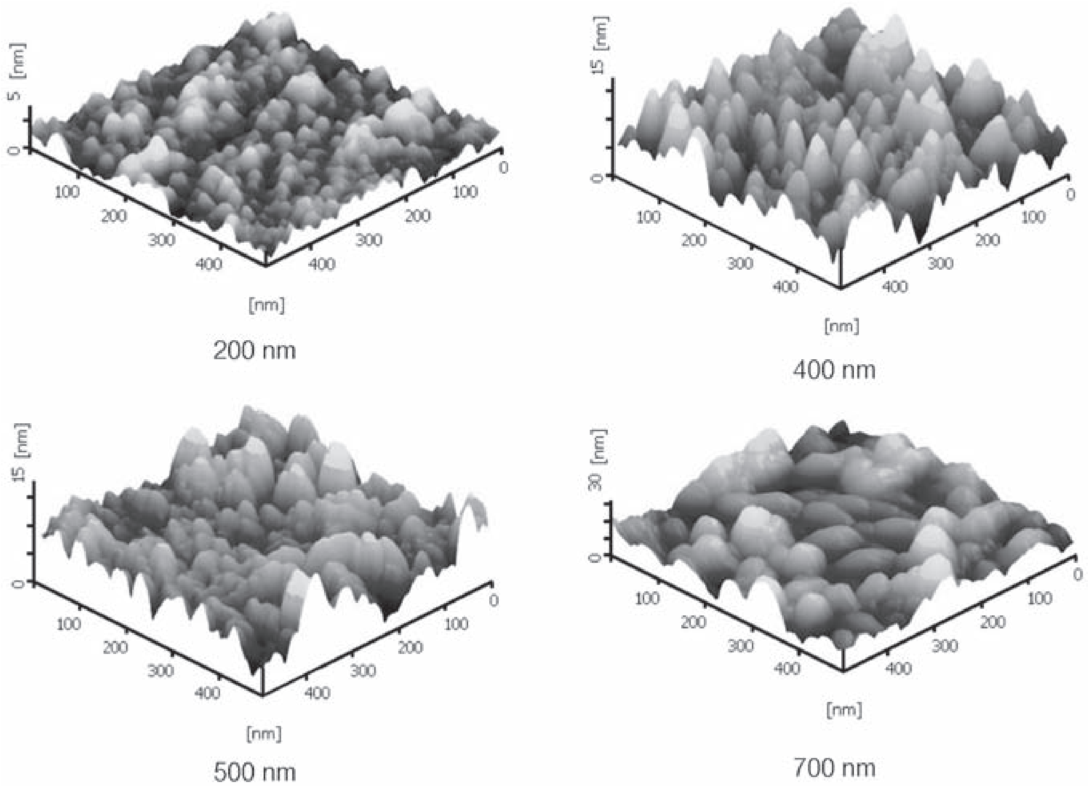

Figure 3 AFM images of ITO thin films with various thicknesses.

increasing of the thickness from 200 nm to 700 nm. The characteristics of ITO films with various thickness of 200, 400, 500 and 700 nm are shown in Table 1.

CONCLUSION

ITO thin films with various thicknesses from 200 to 700 nm have been prepared on glass

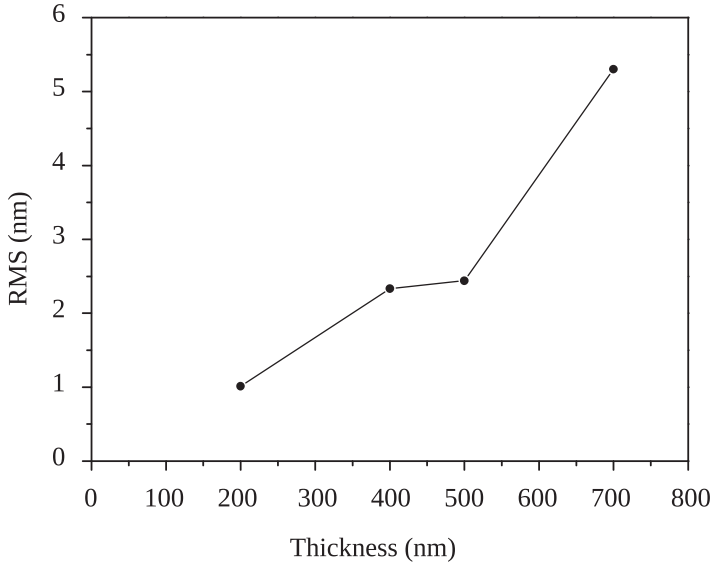

Figure 4 Surface roughness of ITO thin films with various thicknesses.

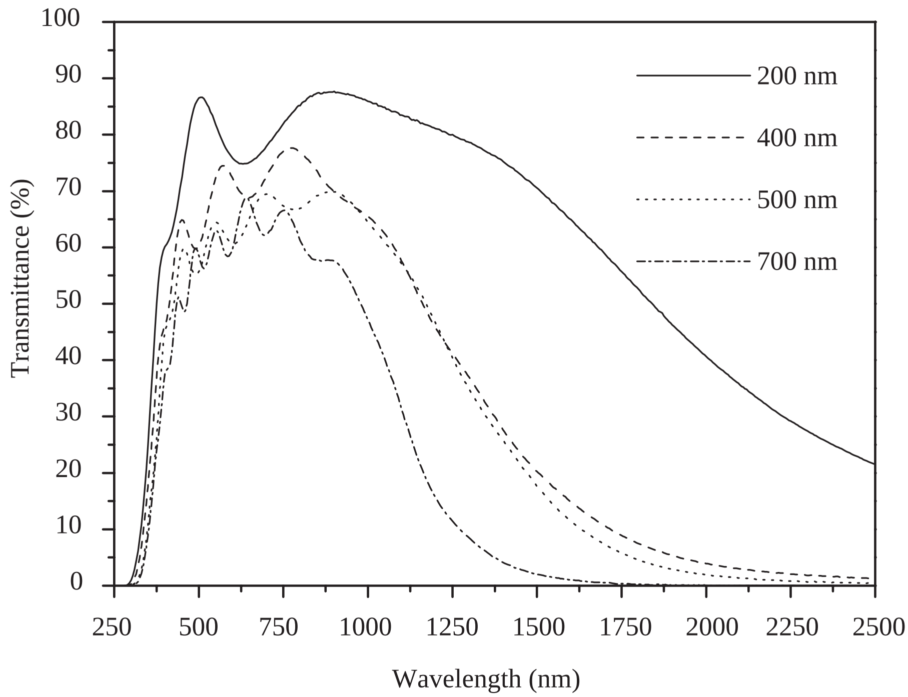

Figure 5 Transmittance spectra of ITO thin films with various thicknesses.

slide substrate by electron beam evaporation. Changes in structure and electrical properties of ITO thin films deposited on the glass slide substrate were investigated according to their thicknesses. It was observed that amorphous layer with thickness of 200 nm was formed at the interface on the glass slide substrate and polycrystalline phase evolved above the thickness and it was obviously observed that the surface roughness of ITO thin films increased as the thickness increased. The sheet resistance of ITO thin films was found to be decreased considerably along with the phase change from amorphous to polycrystalline. It is concluded that in order to prepare high quality of ITO thin films on glass slide substrates, crystallinity of the deposited films should be enhanced at the initial stage of deposition and the thickness of amorphous layer at interface be reduced using proper surface treatments.

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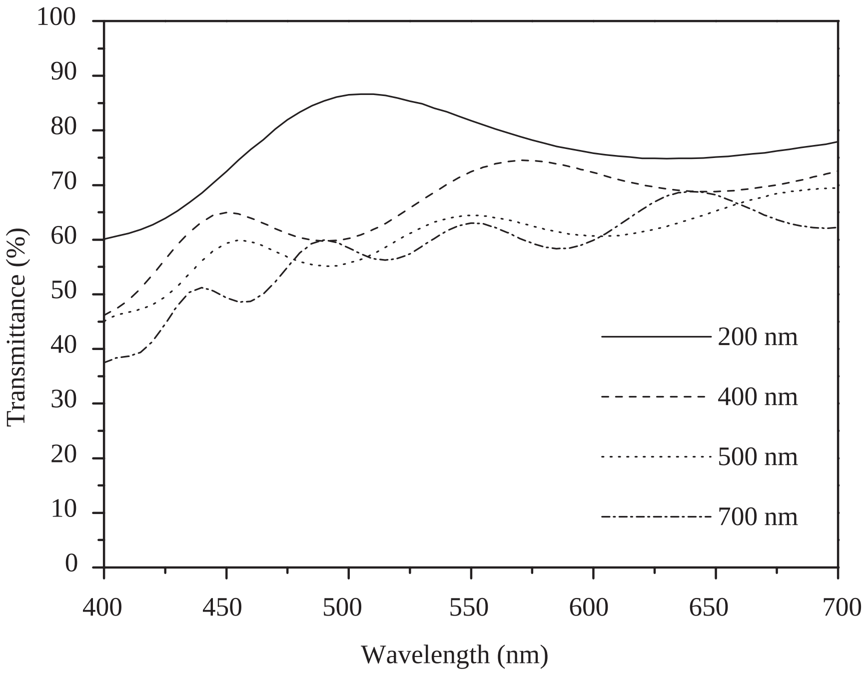

Figure 6 Transmittance spectra in visible region of ITO thin films with various thicknesses.

Table 1 Characteristic of ITO thin films with various thicknesses.

Characteristic	Sample A	Sample B	Sample C	Sample D
d (nm)	200	400	500	700
$< T >$(%)	76.63	66.17	60.23	57.89
R_q (nm)	1.01	2.33	2.44	5.30
R_S (W/sq.)	16.00	8.00	5.70	4.00

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