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Effects of target density on structure and properties of sputtered indium tin oxide films

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Abstract

Indium Tin Oxide (ITO) thin films were deposited from various target densities (98.7%~99.6%) using RF magnetron sputtering. Effect of the sputtering target densities on the structural, electrical and optical properties of deposited ITO thin films was investigated. The preferable (400) crystalline orientation peak was observed on the films deposited from > 99.0% target density. Higher target density produced films with higher roughness but lower resistivity. All of the deposited films showed optical transmittance more than 85% in the visible wavelength region. It is necessary to use the highest target density for sputtering deposition of ITO thin films.

Keywords: ITO thin films, transparent semiconductor, sputtering, target density, optical properties
PACS: 78.20.-e, 78.66.-w

1. Introduction

ITO thin film is an essential part in the technological field requiring both large area electrical contact and optical access in the visible portion of the light spectrum. ITO has been used as the transparent conductive oxide (TCO) layer for flat panel displays, optical diodes, window layer for solar cells and anti reflection coating for structural glasses and mirrors [1,2]. High transmittance combined with its ~3.7eV band gap, electrical conductivity, and stability has been the key properties of ITO thin films compare to the other transparent conductive oxide films such as zinc or tin oxide. A variety of methods has been applied to deposit high conductive ITO. Sputtering is the most popular method to produce device quality ITO films with low resistivity and good reproducibility [3]. Deposition and post-deposition conditions are found to be critical in controlling the structure and properties of the sputtered films. Variation in the deposition parameters may result in different structure and properties [4,5].

Target preparation is one of the important aspects for producing high quality films using sputtering deposition. During production some variation in target density may occur

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often unintentionally. In addition to target preparation, other factors which will influence structure and composition are physical sputtering parameters like, sputtering power, total pressure, partial pressure of oxygen, In this paper, we report the effect of target density (TD) on the structure and properties of ITO thin films deposited by RF magnetron sputtering.

2. Experimental Details

ITO thin films were deposited on corning glass 7059 substrates using RF magnetron sputtering from various sputtering target densities, identified as 99.6%, 99.2%, 99.0% and 98.7% (compared to ITO theoretical density is 7.19 g/cm^3). The targets were solid ITO of the same compositions, 70 mm in diameter, 5 mm in thickness and placed 50 mm from the substrate for deposition. Prior to deposition, glass substrates were cleaned in ultrasonic and organic cleaner and the chamber was evacuated down until 10^{-6} Torr using a turbo molecular pump. In order to obtain the same condition for comparison, the deposition parameters were kept constant and are given in the table 1 below.

Table 1: Sputtering deposition parameters.

Parameters	**Conditions**
Sputtering source	*Radio frequency (13.56 MHz)*
Sputtering and purging gas	*Ar (1.3 sccm)*
Working pressure	*25 mTorr*
RF power	*100 Watt*
Deposition time	*5 minutes*
Deposition temperature	*200^{o}C*

Characterization was started by X-ray diffractometer (XRD, RINT-Rigaku) with a CuK$_\alpha$ radiation for phase identification and crystallographic structure. The films microstructure and thickness were studied by Scanning Electron Microscope (SEM Hitachi S-4100). The electrical resistance of the thin films was measured using a four-point probe (CMT-SR1000N, Changmin Co.) while the optical transmittance of the thin films was measured with a UV-Vis-NIR Spectrophotometer (CARY 5G, Varian) in the wavelength range of 300–2500nm. The surface roughness (root mean square-RMS value) was measured by atomic force microscope (AFM).

3. Results and Discussion

Using the above deposition parameters, ITO thin films could be deposited at the rate of 4.5 Å/s. The thickness of the films was measured to be in the range of 130 $\pm$ 1 nm and did not vary systematically with the TDs. This result indicates that the deposition rates do not vary significantly by using different TDs.

Structure of the deposited films was observed by X-Ray diffractometer and the normalized spectra are given in Figure 1. All of the deposited films shows strong (222) peaks. The other peaks are (440) and (622) which all confirm the ITO cubic structure. It can also be concluded from the spectra that deposition from higher target densities promotes (400) peaks. This (400) peak was not found in the film deposited from 98.7% TD. The (400) peak is preferable in ITO thin films for better film properties [4,5].

As TD increases, lattice parameter (*a*) calculated from the strongest peaks (222) shows a slight decrease from 10.139Å (for 98.7% TD) to 10.126Å (for 99.6% TD). The value is generally higher than the bulk value of In_2O_3 at 10.118Å, indicating that the films are strained resulted from sputtering growth. The distortion of lattice parameters can also be attributed to the variation of oxygen defects in the In_2O_3, where more oxygen presence during sputtering deposition promotes bigger lattice parameters [6-7], as shown by films deposited from lower target densities.

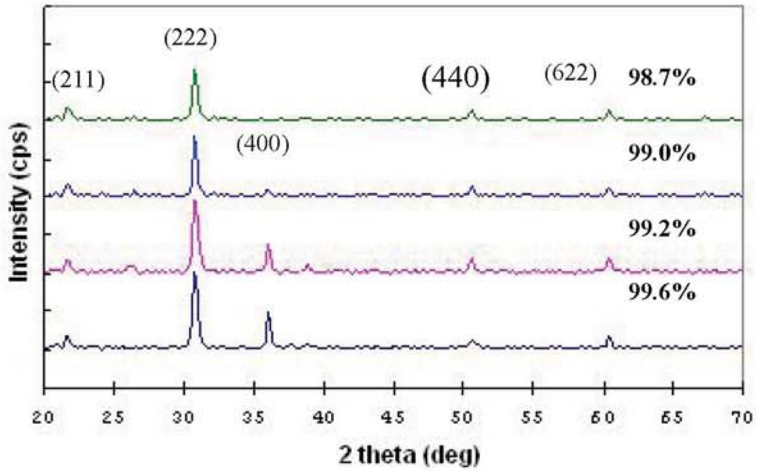
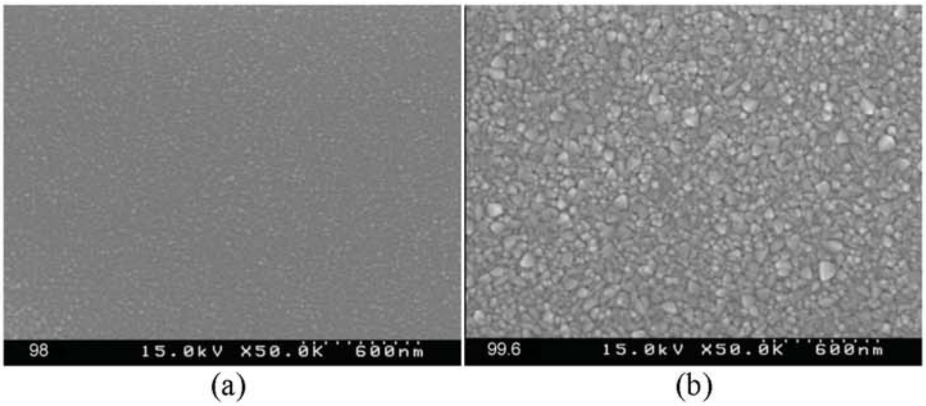


Fig. 1: XRD spectra of the deposited ITO thin films. Preferable stronger (400) peak was observed on film from the highest TD (99.6%).

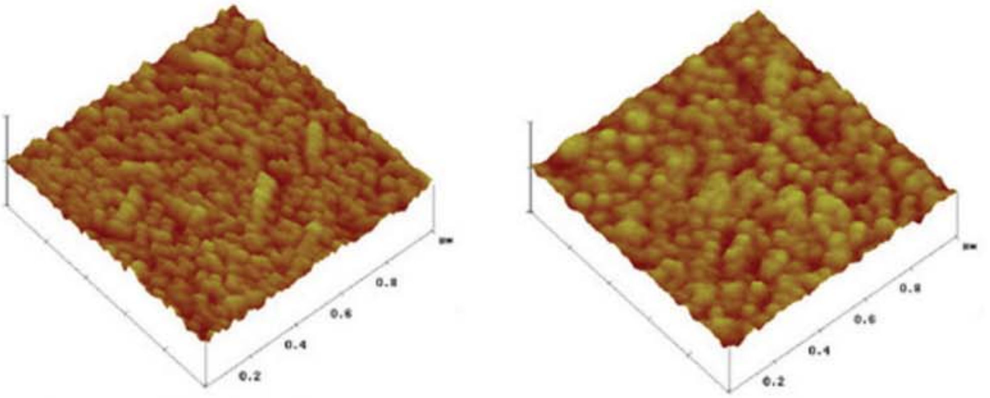
Figure 2 shows the SEM micrograph of films deposited from 98.7% and 99.6% TD. There is a significant difference in grain size observed. At 99.6% the grains depicted from surface facets look bigger compare to the 98.7% sample. As reported previously by Thilakan, et.al., the (400) crystal orientation is believed to promote larger grains as films grown exactly oriented at normal direction to the substrate and therefore having better grain uniformity, while the (222) crystal orientation prefers to have more random growth behavior in every direction and results in smaller grains [8].



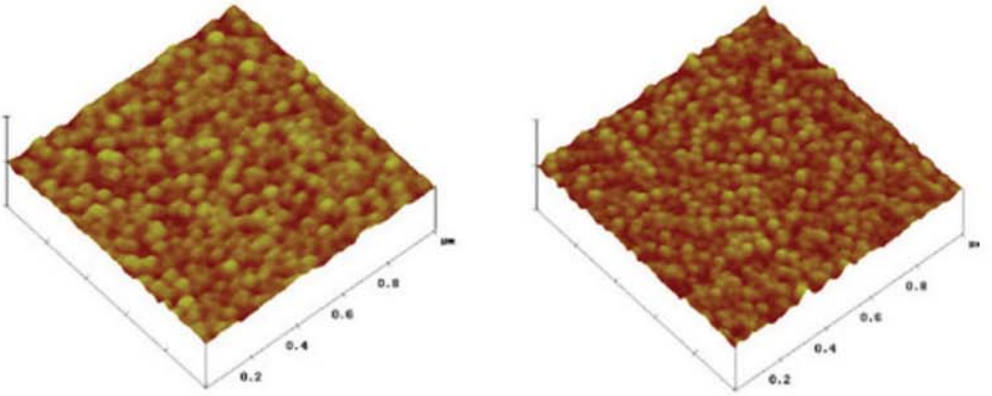
(a) (b)

Fig. 2: SEM micrograph of films deposited from 98.7% TD (a) and 99.6% TD (b).

The films surface morphology and RMS-roughness value from AFM observation is shown in the Figure 3. The highest RMS value was 5.310nm obtained from TD 99.6% while the lowest was 3.365nm from TD 98.7%. The result shows that higher TDs produced films with higher roughness. The difference was less significant for films obtained from lower TDs. While maintaining high transparency, it is also desirable in ITO thin films to have a high roughness, especially in solar cell application in order to improve the light trapping efficiency of the absorber layer. The results also correspond to the intensity of the (400) crystalline peak as shown by the XRD spectra, in which, stronger (400) peak and decreased lattice parameter yield a bigger grain size and higher surface roughness [6].



(a) TD= 99.6% RMS= 5.310nm (b) TD= 99.2% RMS= 4.725nm



(c) TD= 99.0% RMS= 3.747nm (d) TD= 98.7% RMS= 3.365nm

Fig. 3: Surface morphology and roughness measurement using AFM.RMS value decreases for films deposited from lower TDs.

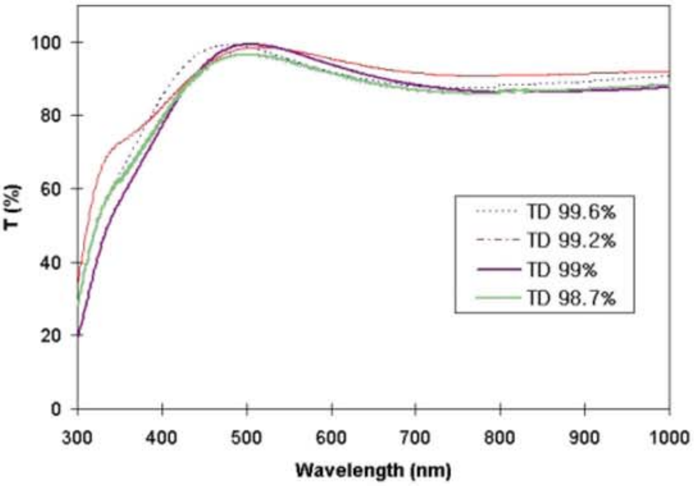


Fig. 4: Transmittance spectra of ITO thin films from various TDs

Figure 4 shows the transmittance spectra of the deposited films. All of the films possess over 85% average transmittance for visible light wavelength (400–700nm). It can be seen from the spectra that films obtained from TD $\geq$99.2% have slightly better transmittance especially in the shorter wavelength region (400-500 nm). Films deposited from TD 99.6% have the highest average transmittance.

Resistivity of the deposited thin films is presented in Figure 5, calculated from sheet resistance (measured by a 4-point probe) and film thickness (measured by SEM). The lowest thin film resistivity value of 8.9×10^{-4} $\Omega\cdot$cm was obtained from TD 99.6%. The lowest resistivity obtained in this report is higher than what has been reported earlier. This is obvious for film with lower thickness produced in this experiment. Resistivity of the films increases for the lower TDs. The presence of preferable (400) orientation peak is presumed to contribute in obtaining a lower resistivity as shown on the film from TD 99.6%. On the contrary, the resistivity was increased in the absence of (400). As discussed earlier from XRD and SEM observations stronger (400) peak yields bigger grain size and reduced the total grain boundaries which in turn increase electron mobility and conductivity of the films.

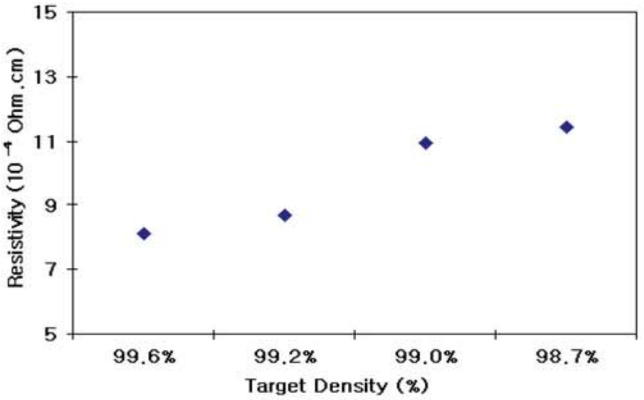


Fig. 5: Resistivity of the ITO thin films from various TDs.

This result confirms earlier report, which showed that geometry of the deposition produces films with different densities resulted in non-homogeneity of its electrical properties [4]. The resistivity of the films decreased when the density of the thin films increased.

Although the density of the film is not measured, the results seem to be in the same line as we expect the lower target densities produce less dense films, therefore the resistivity increases. Any post-deposition treatment may be employed in order to improve the properties of films deposited from lower density. Annealing may improve the structure and crystallinity of ITO films.

4. Conclusion

ITO thin films were deposited from different target densities using RF magnetron sputtering with the same deposition parameters. Structural, optical and electrical properties were investigated. Variation in target density results in properties alterations of the deposited thin films. Structure and electronic properties vary significantly when target density used is between 98.7-99.6%, while the good optical property (over 85% transmittance) is almost unaffected. The best result was obtained using the highest target density. Proved to be an important factor in producing high quality ITO thin films, it is recommended to always use the highest theoretical density for sputtering targets.

5. Acknowledgement

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