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Influence of Deposition Temperature on Indium Tin Oxide Thin Films for Solar Cell Applications

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Abstract. The transparent conductive oxide (TCO) layer is an important layer to improve the efficiency of solar cells. The key requirements are low sheet resistance and high transparency in visible range. In order to optimize the deposition parameters to get high quality TCO, Indium tin oxide (ITO) thin films were deposited on Corning 1737 glass substrate at deposition temperature ranging from 50 to 200 °C by using RF Sputtering technique. It is observed from X-ray diffraction (XRD) measurements that the films deposited at 50°C were nanocrystalline in nature. The film crystallinity was improved with increase in deposition temperature and saturated at 150 °C. The average crystallite size was estimated by Williamson-Hall method, which are in the range of 21 to 27 nm for the films deposited at 100 to 200 °C respectively. The film deposited at 150 °C showed 91-98% of transmission in the wavelength range of 400-1500 nm, low sheet resistance of 7.5 Ω/\square and surface roughness of 6.1 nm. These highly crystallized, low sheet resistance and low temperature ITO films are very useful for improving efficiency of solar cells.

INTRODUCTION

Indium tin oxide (ITO) thin films are used in many electronic devices such as solar cells [1,2], liquid crystal displays (LCDs) [3], light emitting diodes (LEDs) [4,5] and also in biology to determine glucose concentration due to electroluminescent properties of ITO films [6]. The ITO thin films have significance importance in solar cells as transparent conducting oxide and antireflection layer [1,7–9]. In order to improve performance of solar cells, the key requirements of ITO films are high transparency, high conductivity and low sheet resistance [10–13]. However, these properties are sensitive to deposition condition like substrate temperature, process pressure, Ar gas flow rate, RF power and annealing temperature etc [14,15].

This paper presents the influence of deposition temperature on morphology, structural, optical, and electrical properties of ITO thin films deposited by RF sputtering technique. The films were studied using XRD, UV-Vis-NIR and I-V measurements. The morphology of ITO films has been studied by FESEM and AFM measurements.

EXPERIMENTAL DETAILS

The Indium Tin Oxide (ITO) thin films were deposited on Corning 1737 glass substrate using radio frequency sputtering (RF Sputtering) technique. The deposition parameters used for deposition of ITO films were: Argon flow rate (AFR) of 7 SCCM, RF power of 80 W, process pressure (PP) of 0.062mbar and substrate temperature varied from 50 to 200 °C.

The X-ray diffraction (XRD)(Rigaku) was used to study the structure, crystallite size and micro strain of the ITO films. Transmission spectra of the ITO thin films were measured with UV-Vis-NIR spectrometer (Shimadzu

3101PC) in the wavelength range of 200 to 1500 nm and used to estimate band gap of the films. Atomic Force Microscope (Agilent 5500) and Field Emission Scanning Electron Microscope (FESEM) (ZEISS, SIGMA) measurements were used to study the surface morphology of the films. The sheet resistance of ITO films was measured using four probe current-voltage (I-V) measurements.

RESULTS AND DISCUSSION

Fig.1(a, b) show the XRD pattern of *SP385-SP388* ITO films and peak fitting of *SP388* film respectively. It is observed that the films deposited at 50°C were nanocrystalline in nature. The film crystallinity was improved as deposition temperature was increased and completely crystallized as well as saturated at 150 °C. The intensity of the diffraction peaks was also increased with increasing deposition temperature. This indicates that films deposited at higher temperature are highly crystalline and oriented with reduced crystalline defects [16–18]. The preferential direction of growth for films deposited at 50 and 100 °C is [222]. Most intense peak was observed corresponding to (400) plane for ITO films deposited at 150 and 200 °C. Here 2θ represents the angle between the direct X-ray beam and scattered beam.

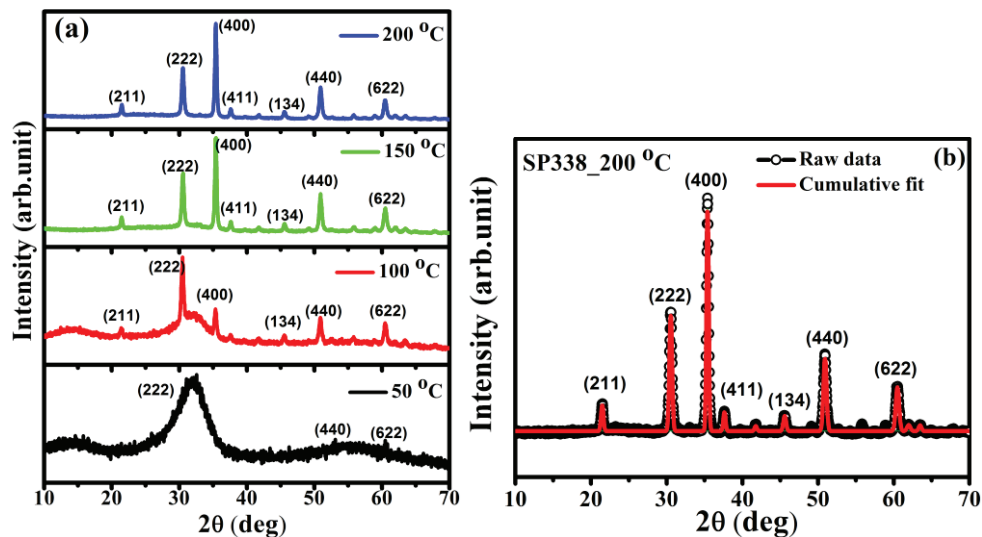


FIGURE 1. (a) XRD pattern of *SP385-SP388* ITO films, (b) Fitted peaks in *SP388* film.

The Williamson-Hall (W-H) method has been used to calculate accurate crystallite size and micro strain in the films [19,20]. Fig. 2 shows the W-H plot of *SP388* film deposited at 200 °C and the estimated crystallite size and micro strain of the ITO films are listed in Table 1. It was observed that the crystallite size of films has increased and micro strain in the films has decreased as increase in deposition temperature of 50 to 200 °C. The ITO films have been completely crystallized at 150 °C and then saturated at deposition temperature of 200 °C. The *SP337*(150 °C) and *SP338*(200 °C) films have crystallite size of 26.86 and 27.50 nm and very less micro strain of 0.1% in the films. The *SP336* (100 °C) showed crystallite size of 21.12 nm and also slightly higher micro strain than other films. The crystallite size of 1.57 nm of *SP335*(50 °C) film was calculated from Debye-Sherrer formula [21,22].

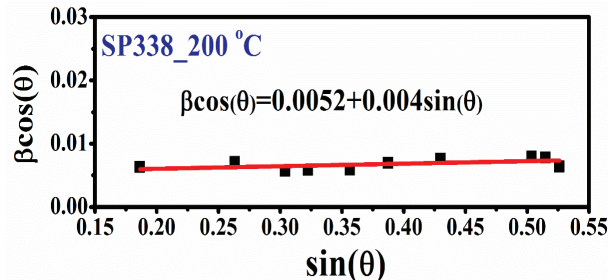
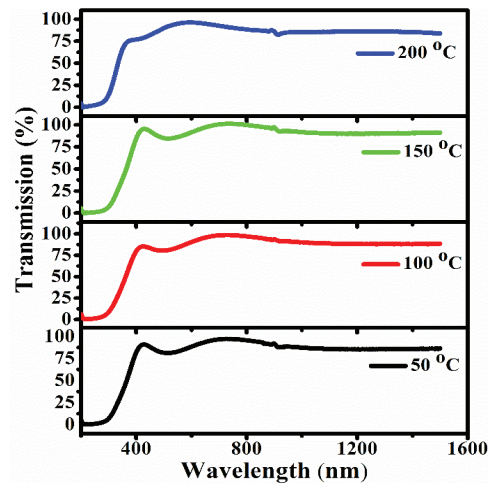


FIGURE 2. Williamson-Hall plot of *SP388* ITO thin film.

TABLE 1. Calculated crystallite size(*d*) and micro strain in the films.

| Sample | Dep. temp (o C) | <i>d</i> (nm) | Strain (%) |
|--------|-----------------|---------------|------------|
| SP338 | 200 | 27.50 | 0.1 |
| SP337 | 150 | 26.86 | 0.1 |
| SP386 | 100 | 21.72 | 0.2 |
| SP385 | 50 | 1.57 | - |

Fig. 3 shows the UV-Vis-NIR transmission spectra of SP385-SP388 films. All the films shown high transmission of 90-98% in the wavelength range of 400-1500 nm. The optical bandgap of films has been estimated using Tauc's plot [23] and values are given in Table 2. It is clearly observed that absorption edge of the films shifted to lower wavelength side with increase in deposition temperature. The optical bandgap of the ITO films has increased and thickness has slightly decreased as deposition temperature is increased.

**FIGURE 3.** UV-Vis-NIR Transmission spectra of ITO films.**TABLE 2.** Estimated thickness, optical bandgap and surface roughness of films.

| Sample | Thickness (nm) | E_g (eV) | Surface roughness (nm) |
|--------|----------------|------------|------------------------|
| SP338 | 86±7 | 3.98±0.02 | 10.7±0.4 |
| SP337 | 90±8 | 3.85±0.01 | 6.1±0.5 |
| SP386 | 100±7 | 3.74±0.01 | 13.8±0.4 |
| SP385 | 110±6 | 3.55±0.02 | 14.3±0.6 |

Fig. 4(a-d) show the FESEM surface morphology of the SP385-SP388 (50-200 °C) ITO films respectively. It is clearly observed that the growth of the films changes with change in deposition temperature. The films are uniform and the grain size of the films has increased with increase in deposition temperature.

Fig. 5(a-d) show the AFM surface morphology of the ITO films correspond to 50-200 °C respectively. It was observed that at lower deposition temperature (50 and 100 °C), films are not uniform, with voids in the films and high surface roughness. With further increase in deposition temperature, uniform films with very less surface roughness were deposited. Measured RMS roughness values are 14.38, 13.81, 6.11 and 10.77 nm corresponding to 50-200°C respectively (Table 2). At lower temperature, atoms have less energy to occupy stable position, which create path to formation of voids and rough films. At higher deposition temperature of 150 °C and above, atoms have sufficient energy to diffuse on surface as well as bulk of the films. This results in strong bonds with neighbouring atoms and very uniform films. It is observed that surface roughness of SP338 film is higher than SP337 film, this could be due to increased grain size at deposition temperature of 200 °C.

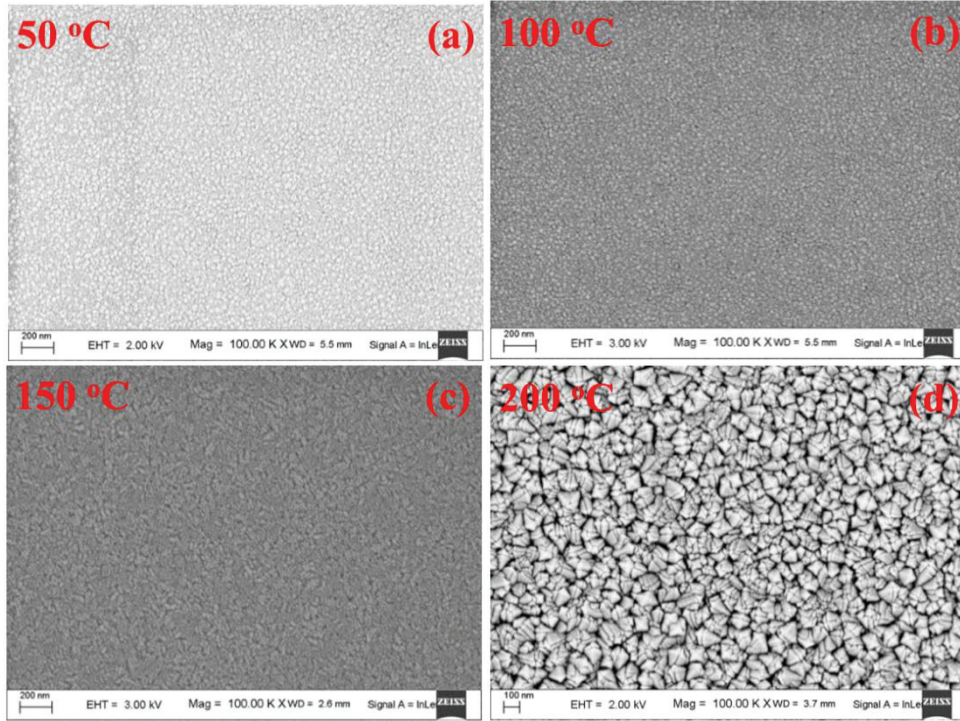


FIGURE 4(a-d).FESEM surface morphology of the *SP385-SP388* (50-200 °C) ITO films.

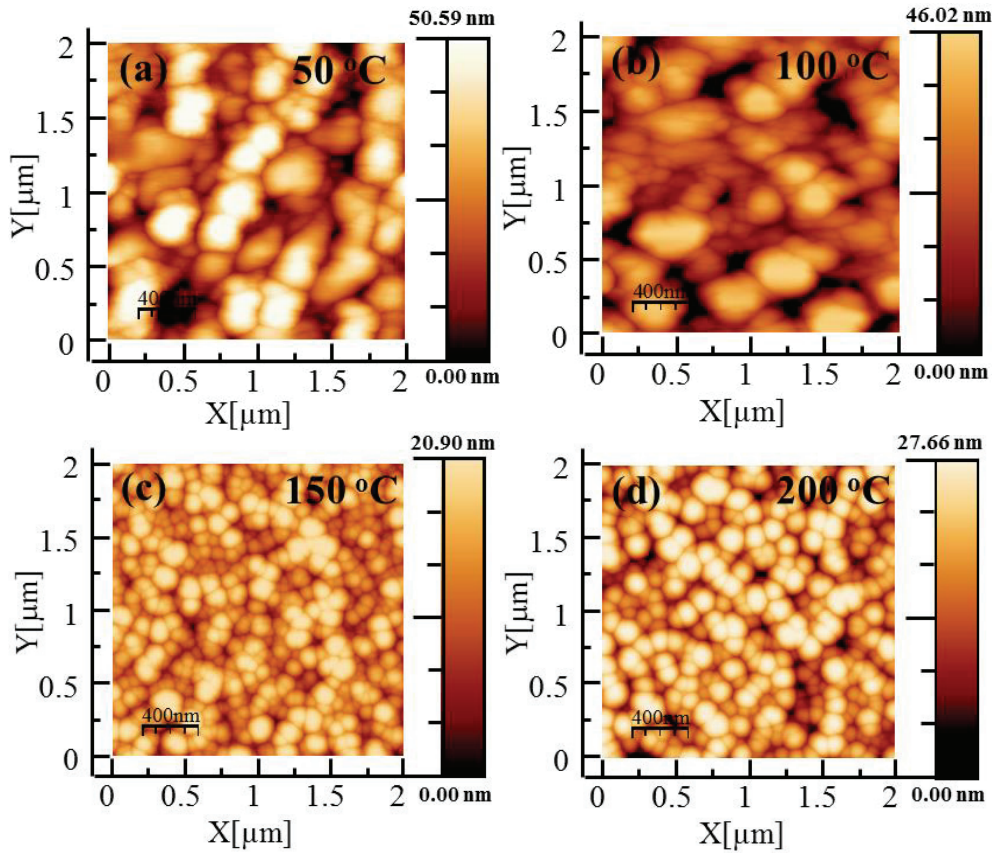


FIGURE 5(a-d).AFM morphology of ITO films.

Sheet resistance and resistivity of ITO films were estimated from four probe I-V measurements and values are given in Table 3. The separation between each probe is 1 mm. It is found that sheet resistance and resistivity have decreased with increase in deposition temperature. Low sheet resistance of 7.5 and 7.4 Ω/\square and resistivity of 6.7×10^{-5} and $6.4 \times 10^{-5} \Omega\text{cm}$ is obtained for films deposited at 150 and 200 °C respectively. These values are one order lower than films deposited at 50 and 100 °C.

TABLE 3. Estimated resistivity and sheet resistance of films.

| Sample | Resistivity (Ωcm)($\times 10^{-5}$) | Sheet Resistance (Ω/\square) |
|--------|--|--|
| SP338 | 6.4 \pm 0.2 | 7.4 \pm 0.3 |
| SP337 | 6.7 \pm 0.1 | 7.5 \pm 0.2 |
| SP386 | 25.0 \pm 0.4 | 25.0 \pm 0.3 |
| SP385 | 37.3 \pm 0.2 | 33.9 \pm 0.5 |

CONCLUSIONS

Indium tin oxide (ITO) thin films were deposited by RF Sputtering technique with varying deposition temperature of 50 to 200 °C. It is observed that the films deposited at 50°C were nanocrystalline in nature. The film crystallinity was enhanced with increases in deposition temperature and saturated at 150 °C. Williamson-Hall plot has been used to estimate crystallite size of the ITO films, which are 21.7, 26.8 and 27.5 nm corresponding to films deposited at 100, 150 and 200 °C respectively. It is found that films deposited at 150 °C have high transmission of 91-98% in the wavelength range of 400-1500 nm, low sheet resistance of 7.5 Ω/\square and surface roughness of 6.1 nm. These highly crystallized, transparent, low sheet resistance and temperature ITO films are very useful for improving the interface properties and efficiency of solar cells.

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