SYNTHESIS OF INDIUM TIN OXIDE (ITO) AS A TRANSPARENT CONDUCTING LAYER FOR SOLAR CELLS BY RF SPUTTERING

ASHOK JADHAVAR, AJINKYA BHORDE, VAISHALI WAMAN, ADINATH FUNDE, AMIT PAWBAKE, RAVINDRA WAYKAR, DINKAR PATIL AND SANDESH JADKAR

Abstract: Thin films of indium doped tin oxide (ITO) were prepared using indigenously designed and locally fabricated RF sputtering unit. Electrical, optical, morphology and structural properties of these films are investigated as a function substrate temperature (T_{sub}). The UV-Vis Spectroscopy analysis showed that the deposited films have an average transmission ~ 85 % in the visible region of solar spectrum. Electrical characterization revealed that the electrical conductivity of ITO films is 2.82x10³ S/cm. The low angle XRD analysis showed that the films have preferred orientation in (400) direction with average particle size 26.4 nm. The SEM images show nice surface morphology with uniform size distribution and crack free film deposition. The obtained results indicate that the synthesized ITO can be useful as a transparent conducting electrode in a-Si:H based solar cells.

Keywords: Indium Tin Oxide (ITO), Optical Properties, RF Sputtering, Transparent conducting Oxide (TCO) etc.

Introduction: Indium doped tin oxide (ITO) thin films are widely used in opto-electronics devices, flat panel display, photo-voltaic and electro-chromic applications. (EC) The attractiveness of ITO is related to its low resistivity and high optical transmittance from visible to near infrared (NIR) light. However, these properties are strongly dependent on preparative methods and respective process parameters. Varity of deposition methods such as reactive ion plating [3], DC diode sputtering [4], RF sputtering [5], reactive evaporation [6-7], electron beam [8], chemical vapor deposition, spray pyrolysis [9] etc. have all been used to deposit ITO films. Each method has its own advantages and limitations. Among these RF sputtering has been used extensively for the synthesis of ITO films due to high deposition rate and possibility of deposition at low substrate temperature.

In the present work, an attempt have been made to synthesized ITO films using indigenously designed and locally fabricated RF sputtering unit. The influence of substrate temperature on electrical, optical, morphology and structural properties has been investigated systematically. **Film Preparation:** Fig. 1 shows schematic of indigenously designed, locally fabricated Radio Frequency (RF) sputtering system employed for the deposition of the Indium Tin Oxide (ITO) thin films.

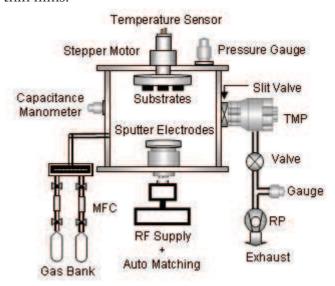


Fig. 1: Indigenously designed, locally fabricated RF sputtering system

It consists of a cylindrical stainless steel chamber

(process chamber) coupled with a turbo molecular pump (TMP) followed by a roughing pump which yields a base pressure less than 10⁻⁷ Torr. A target of 3 inch diameter (99.99%, Vin Karola Instrument, USA) was used for the deposition of ITO films and was kept facing the substrate holder approximately 5 cm away. The substrate holder can be rotated using a stepper motor with variable speed. The substrates can be placed on substrate holder which is heated by inbuilt heater using thermocouple temperature controller. The pressure during deposition was kept constant by automated throttle valve and measured with capacitance manometer. Sputtering and reaction gases can be introduced in the process chamber through a specially designed gas bank assembly which consist of mass flow controllers (MFCs) and gas mixing. The process parameters employed during the deposition of ITO thin films are listed in Table 1.

Table 1: Process parameters employed during the deposition of ITO thin films

Process parameter	Value
Deposition Pressure	5 Torr
Deposition Time	30 min
RF Power	100 W
Distance between substrate	7 cm
holder and target electrode	
Ar gas flow rate	60 sccm
Substrate temperature	100°C-250°C

Prior to each deposition, the substrate holder and deposition chamber were baked for one hour at 100 °C to remove any water vapor absorbed on the substrates and to reduce the oxygen contamination in the film. After that, the substrate temperature was brought to the desired value by appropriately setting thermocouple and temperature controller. Deposition was carried out for desired period of time, and films were allowed to cool to room temperature in vacuum.

Film Characterization: To measure the sheet resistance (R_{Sheet}) of ITO films, Al electrodes, 1 cm apart were deposited on top of the film using vacuum thermal evaporation technique with base pressure > 10⁻⁵ mTorr. After the electrodes deposition, the film was loaded to a specially designed sample holder and the contacts were made using silver paste. A voltage of 60 V DC was applied between electrodes and the current was measured using an electrometer (Keithley, Model 6514). The measurements were carried out at room temperature and atmospheric pressure. Low angle X-ray diffraction pattern were obtained by X-ray diffractometer (Bruker D8 Advance, Germany) using $CuK\alpha$ line ($\lambda =$ 1.54056 Å). The average crystallite size was estimated using the classical Scherrer's formula [29]. The scanning electron microscopy (SEM) images were recorded using JSM-6360A scanning electron microscope with operating voltage 10 kV to study the surface morphology of the films. The band gap of the films was deduced from transmittance and reflectance spectra of the films deposited on corning glass and were measured using a JASCO, V-670 UV-Visible spectrophotometer in the range 250-1100 nm by using the procedure followed by Tauc [25]. Thickness and Refractive index of the films was determined by UV-Visible spectroscopy using the method proposed by Swanepoel [28]. Hall measurements were carried out using Ecopia Hall Effect Measurement System (HMS-3000).

Results and discussion:

The variation of sheet resistance and deposition rate as a function of substrate temperatures is shown in figure 2. As seen from the figure at low temperature (< 200 °C) the sheet resistance decreases with increase in substrate temperature and at high temperature (> 200 °C) it increases again. The minimum sheet resistance (10 Ω) has been obtained for the ITO film deposited at substrate temperature 200 °C.

Thus, substrate temperature 200 °C is an optimized substrate temperature for our system

ISBN 978-93-84124-30-4 127

RF sputtering unit. At this substrate temperature the deposition rate is reasonably high (44 nm/min).

The low angle X-ray diffraction (XRD) pattern of Indium doped tin oxide (ITO) thin films prepared at different substrate temperatures is shown in figure 3. As seen from the XRD pattern the prepared films are polycrystalline having peaks at $2\theta \sim 21.6^{\circ}$, 30.6° , 35.5° , 37.8° , 48.4° , 60.7° associated with (211), (222), (400), (411), (521), (622) diffraction planes respectively. These diffraction peaks consistent with JCPDS Data

Card # 89-4598 for ITO films. These results confirm the formation of Indium doped tin oxide (ITO) thin films by RF sputtering. The dominant diffraction peak is (400) signifying that the films have preferred orientation in (400) direction. However, the film prepared at substrate temperature 250 °C, the dominant diffraction peak is (211) suggesting that the preferred orientation in ITO films prepared using RF sputtering changes from (400) to (211) direction.

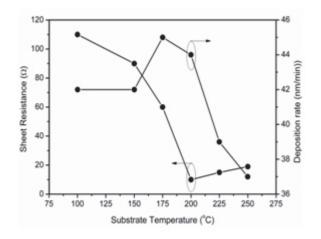


Fig. 2: Effect of substrate temperature on sheet resistance and deposition rate

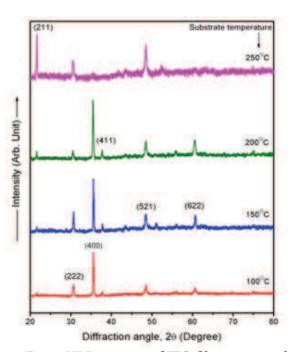
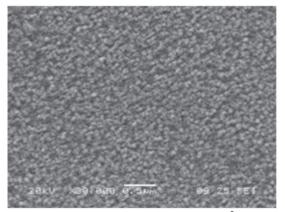
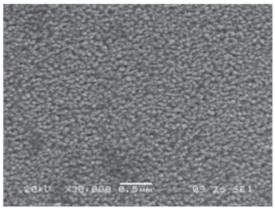


Fig. 3: XRD pattern of ITO films prepared at different substrate temperatures

Figure 4 show the scanning electron microscopy (SEM) image of ITO thin film deposited at 100 °C and 200 °C.



Substrate Temperature 100 °C



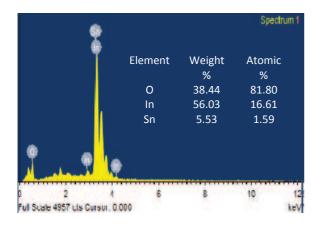
Substrate Temperature 200 °C

Fig. 4: SEM image of the ITO thin film deposited at 100 °C and 200 °C

IMRF Journals 128

It has been observed that with increase in substrate temperature the uniformity in the film structure improves and the film prepared at substrate temperature 200 °C shows nice surface morphology with uniform size distribution and crack-free deposition.

Fig. 5 shows the Energy-dispersive X-ray spectroscopy spectrum (EDS) of ITO film deposited at substrate temperature 200 °C. The inset shows the weight % and atomic % of element present in the film. These results confirm the formation of nearly stoichiometric ITO films by using RF sputtering. Fig. 6 represents the transmission spectra of ITO films deposited at different substrate temperatures.



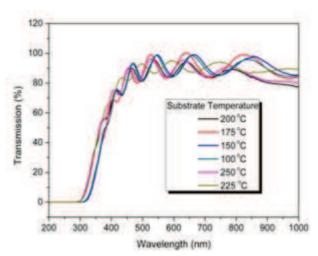


Fig. 5: EDS of ITO film deposited at substrate temperature 200 °C

Fig. 6: Transmission of films prepared at different substrate temperatures

It can be seen from the transmission spectrum that all films have the transmission above $82\,\%$ in the ultraviolet-visible region of the electromagnetic spectrum. We have estimated the optical band gap of films using the Tauc method. Fig. 7 display typical Tauc plot for the ITO film prepared at substrate temperature 200 °C.

It has been observed that with increase in substrate temperature no significant change in optical band gap. The band gap was found to be constant $\sim 3.26\pm0.2$ eV over the entire range of substrate temperature studied.

The semiconductor properties of all prepared ITO films are also analyzed by using by Hall measurement. Table 2 shows the values of the Hall parameters obtained for the film deposited at substrate temperature 200 °C.

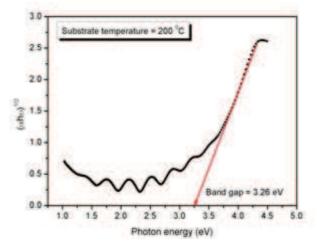


Table 2: Hall measurement parameters obtained for ITO film deposited at 200 °C

wop		
Hall parameter	Value	
Bulk concentration (cm ⁻³)	-3.69x10 ²⁰	
Charge carrier mobility	38.5	
$(cm^2/V.s)$		
Hall coefficient (cm³/C)	1.69X10 ⁻²	
Conductivity (S/cm))	2.28×10^3	
Sheet concentration (cm ⁻²)	-4.19X10 ¹⁶	

ISBN 978-93-84124-30-4 129

From the Hall measurement analysis it has been confirmed that the deposited films are n-type with significantly high conductivity.

Conclusion: In the present study thin films of indium doped tin oxide (ITO) were prepared using indigenously designed and locally fabricated RF sputtering unit. Electrical, optical, morphology and structural properties of these films are investigated as a function substrate temperature (Tsub). The UV-Vis spectroscopy analysis showed that the deposited films have an average transmission ~ 85 % in the visible region of solar spectrum. The maximum electrical conductivity obtained for films is 2.82x103 S/cm.

The low angle XRD analysis showed that the films have preferred orientation in (400) direction. The scanning electron microscopy (SEM) images show nice surface morphology with uniform size distribution and crack free film deposition. The obtained results indicate that the synthesized ITO can be use as a transparent conducting electrode in a-Si:H based solar cells.

Acknowledgement: Authors gratefully acknowledge CNQS, Savitribai Phule Pune University and MOU program between BARC, Mumbai and Savitribai Phule Pune University for their financial support.

References

- 1. L. Dawar and J. Joshi, Mater. Sci. 19 (1984) 1.
- 2. T. Vink, W. Walrave. J. Daams, P. Baarslag and J. Meerakker, Thin Solid Films 266 (1995) 145.
- 3. J. Machet, J. Guile, P. Saulnier and S. Robert, Thin Solid Films 80 (1981) 149.
- 4. S. Takaki, K. Matsumoto, and K. Suzuki, Appl. Surf. Sci. 33/34 (1988) 91.
- 5. H. Cui and S. Xi, Thin Solid Films 288(1996)325-329.
- 6. C. Nunes de Carvalho, G. Lavareda, E. Fortunato and A. Amaral, Thin Solid Films, 427 (2003) pp. 215-218.
- 7. S. Naseem, I. Rauf, K. Hussain and N. A. Malik Thin Solid Films 156 (1988) 161.
- 8. D. Paine, T. Whitson, D. Janiae, R. Beresford, and C. Yang, Appl. Phys. 85 (1999) 8445-8450.
- 9. M. Huang, Z. Hameiri and T. Muller, Energy Procedia, 33 (2013) 91–98.

* * *

1School of Energy Studies, Savitribai Phule Pune University, Pune 411 007 (India) 2Modern College of Arts, Science and Commerce, Shivajinagar, Pune 411 005 (India) 3Department of Physics, Savitribai Phule Pune University, Pune 411 007 (India) 4Department of material Science and Materrulogy, IIT, Powai, Mumbai 400 076 (India) *Corresponding Author: sandesh@physics.unipune.ac.in

IMRF Journals 130