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Growth and Micro Structural Characterization of In_2O_3 Thin Films Prepared by Electron Beam Evaporation Technique.

Veeraswamy Y*, Vijayakumar Y, and Ramana Reddy MV.

Thin Films and Nano Materials Research Laboratory, Department of Physics, Osmania University, Hyderabad-500007, Andhra Pradesh, India.

ABSTRACT

High purity Indium oxide powder was used to prepare In_2O_3 thin films using electron beam evaporation technique. Ultrasonically cleaned substrates were used for the deposition of In_2O_3 thin films. Well-defined and good quality In_2O_3 thin films were formed by maintaining substrate temperatures between 300K - 573K. The thin films were characterized using XRD, SEM, AFM, EDS and UV-VIS spectroscopy, to study the effect of substrate temperature on the properties of In_2O_3 thin films. The XRD patterns suggest that the films deposited at room temperature are amorphous in nature. The crystalline nature of the films increases with increase in substrate temperature and it was found that the films crystallizes in a cubic structure with preferred (2 2 2) orientation. Thickness of the films was found from cross sectional view of SEM images. Optical transmission measurements were carried out using UV-VIS spectrophotometer in the wavelength range 300-1000nm and it was confirmed that these thin films exhibits good transparency and it was noticed that the transmittance was decreased with increasing substrate temperature. Surface morphology studies of the films, using AFM, reveal the formation of nanostructured indium oxide thin films. The results obtained in the present work was presented and discussed.

Keywords: In_2O_3 thin film, AFM, XRD, electron beam evaporation, transmittance.

Corresponding author

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INTRODUCTION

The investigation for transparent conductive oxides (TCOs)[1] is motivated by technical desires in many devices like automobile windows, heat-reflecting mirrors[2] and incandescent bulbs, gas sensors[3], thermal detectors[4] ferroelectric storage and display devices [5]. In opto-electronic devices, these metal oxide semiconductor thin films have been attracting innumerable attention due to their exceptional properties, which differ from those of their bulk counterparts and their potential applications in transparent and conductive oxide thin films. Among them, Indium oxide (In_2O_3) has been investigated extensively for its semiconducting properties. Degenerate Indium oxide thin films show unique shape and size dependent properties. In_2O_3 thin films have been formed by number of different deposition techniques which include PLD [6], direct current (DC) magnetron sputtering [7], spray pyrolysis [8], sol-gel [9] ,thermal evaporation [10] and electron beam evaporation[11]. Surprisingly there was very few reports were found on substrate temperature on In_2O_3 films prepared using e-beam technique. In view of this the present paper focuses on the effect of substrate temperature on micro structural characteristics of In_2O_3 thin films.

EXPERIMENTAL

In the deposition of In_2O_3 thin films first priority goes to cleaning of the substrate. In order to achieve desirable film properties, cleaning of the substrate surface prior to film deposition is very much essential. For this the glass substrates were cleaned by submerges in double distilled water and chromic acid, and was then cleaned in a detergent solution with ultra sonicator for 15mts. After washing with double distilled water, they were rinsed with acetone and dried in an oven to get moisture free substrates.In_2O_3 thin films were grown on glass substrates by e-beam deposition method. The vacuum chamber was pumped with diffusion pump and rotary pump combination. The pressure in the chamber was measured using digital pirani and penning gauge combination [12].The source material was pelletized by taking fine powder of Indium oxide (99.99%) from Sigma-Aldrich chemicals. Substrates were top mounted at 15cm from the target material, with a miniature heater to maintain substrate temperature 300-573K. Initially 10^{-7} mbar base pressure was maintained in the vacuum chamber, then oxygen gas was admitted through a needle valve for oxygen atmosphere and final pressure was maintained at $3x10^{-5}$ mbar inside the vacuum chamber. By operating e-gun In_2O_3 thin films are deposited on glass substrates for different substrate temperatures 300-573K. The films were characterized structurally using XRD; surface micro structural characterization was investigated by SEM and AFM. The optical properties were studied by using UV-VISIBLE spectro photometer. Throughout the deposition the power level of the e-gun was maintained constant.

Micro structural characterization

X'pert PRO Panalytical diffractometer was used to investigate the structural and crystallographic phases present in the films using nickel-filtered CuK_α radiation (λ= 0.15418 nm) under a voltage of 40 kV and a current of 30 mA. The average size of Crystallites (D) of In_2O_3 films was estimated using Debye Sherrer formula [13].

$$D = \frac{.098\lambda}{\beta cos\theta} \quad \text{---------- (1)}$$

Where D=crystallite size, λ=wavelength of CuK_α radiation (0.15418nm), **β**=FWHM, θ=Bragg's diffraction angle.

The surface morphology of the In_2O_3 thin films were analyzed by AFM using Park systems XE-70, with scanning area was 1µm x 1µm. The following three characteristic parameters for the analysis of the AFM measurements were used: (i) the Root Mean Square (RMS) Roughness(*Rq*), which gives the standard deviation within a given area; (ii) the Mean Roughness (*Ra*), which represents the arithmetic average of the deviations from the center plane; (iii) the difference between the highest and lowest points (peak and valley) on the surface relative to the mean plane (*Rpv*). The AFM measurements were performed in non contact mode. The film morphology was examined by Zeiss Supra 50VP 3500 SEM. In_2O_3 thin films were mounted on stubs with carbon tape and the samples were set into secondary electron detection mode. The elemental composition was recorded with Energy-dispersive X-ray spectroscopy (EDS).

Optical characterization

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Optical transmittance was carried out in the wavelength range 300nm-1000nm using a Shimadzu UV-2450 UV-Visible single beam spectrophotometer. Beam calibration with indium oxide thin film was done by a bare glass substrate during the recording of the spectra.

Determination of band gap

The absorption coefficient was calculated from optical transmittance data using Lambert's principle [14]

$$\alpha = -\frac{\ln(T)}{t} \quad \text{---------- (2)}$$

Where T is the transmittance and t is the thickness of the film.

The optical band gap (E_g) of the films can be estimated using the relation:

$$\alpha h\upsilon = A(h\upsilon - E_g)^n \quad \text{---------- (3)}$$

Where A is constant, υ is transition frequency and the exponent n characterizes the nature of band transition. n= ½ and 3/2 corresponds to direct allowed and direct forbidden transitions and n= 2 and 3 corresponds to indirect allowed and indirect forbidden transitions respectively [15]. It is observed that the best straight line is observed for n= ½, which is expected for direct allowed transition.

The optical band gap E_g was estimated by extrapolating the straight line portion of $(\alpha h\upsilon)^2$ vs hυ plot (fig.6)

RESULTS AND DISCUSSION

XRD

The XRD patterns of the films shown in fig(1) were compared for different substrate temperatures on glass substrates. The peaks appeared of substrate temperature 373K and which has been found to match with the JCPDS file (card no: 71-2195). The crystallinity increases with increasing substrate temperature. Three prominent peaks corresponding to (222), (400) and (400) planes are observed in the spectra, in addition to other peaks with low relative intensities. The intensity count of (222) plane increases with increasing substrate temperatures. The average crystallite size (D) was determined using the Scherrer formula tabulated in table (1). That the crystallite size increases with increasing substrate temperature. As the substrate temperature increases, the energy of the surface species increases and they migrate through the lattice to get deposited at appropriate lattice sites to form more stoichiometry crystalline phase or it may be due to development of compressive stress in the film, and it confirms that the lattice constant decreases from 10.18-10.1A^o with increasing of substrate temperature from 300K to 573K.Bender et al (2001) has observed similar result.

Table 1: Variation physical parameters with substrate temperatures.

Substrate temperature(K)	Crystallite size(nm)	Lattice parameter(A^o)	Transmittance (%)	Energy gap (eV)
373	27	2.94	91	3.15
473	36	2.93	80	3.53
573	45	2.92	89	3.46

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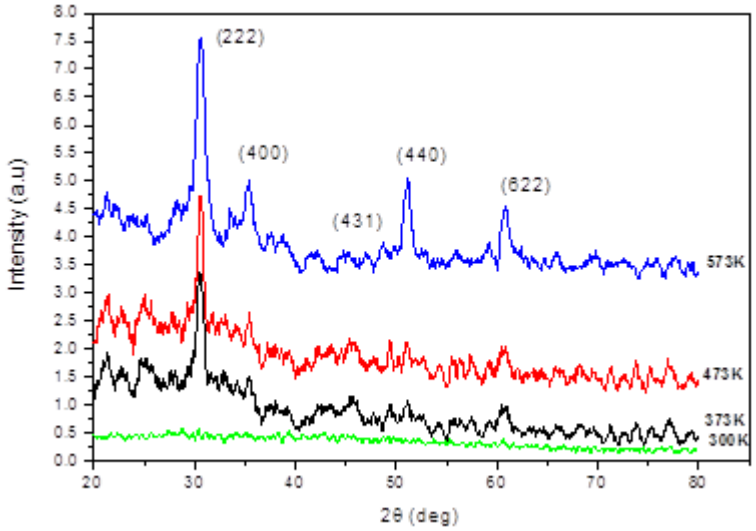


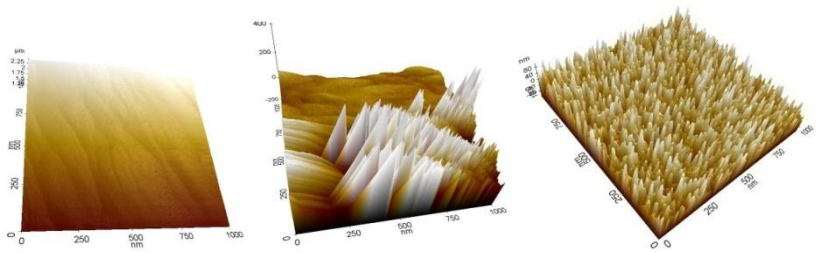
Figure 1: Comparison of XRD of In_2O_3 thin films for different substrate temperatures.

AFM

Fig (2)(a,b,c) shows the AFM micrographs of In_2O_3 thin films on glass substrates at different substrate temperatures (373K, 473K, and 573K). The surface of the film founds to be continuous without any pinholes/cracks. Surface roughnesses of all the films were tabulated in table (2). From this the film formed at substrate temperature 373K represents neither sharp peaks nor hillocks. While substrate temperature increases to 473K the micrograph changes to sharp peaks like saw tooth possessing very high roughness. At a substrate temperature of 573K the thin film composed of unique shape and size Hillocks results conical shape. These hillocks have comparatively flatter tops and their sides appear to be faceted as well as. It may be due to bulk diffusion of indium atoms, and was may be result of substrate temperature.

Table 2: Variation of Micro structural parameters with substrate temperatures

Substrate temperature(K)	RMS Roughness(Rq)(nm)	Mean Roughness(Ra)(nm)	Peak to Valley (Rpv)(nm)
373K	7	5	34
473K	126	113	426
573K	26	20	100



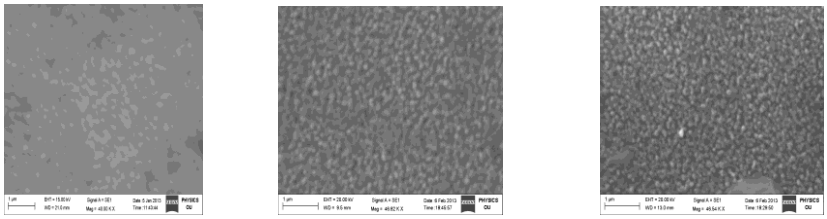
a **b** **c**

Figure 2 (a,b,c): AFM images for different substrate temperatures (373k,473K,73K).

SEM

SEM picture of In_2O_3 thin films prepared at different substrate temperatures 373-573K were shown in Fig(3)(a,b,c).It had polycrystalline or granular structure.The most distinguishable grains were observed in the film which was grown at a substrate temperature 573K. Fig (4) represents the cross sectional view of the film

and the thickness was found to be 1560nm The EDAX analysis revealed that the nearly good stoichiometric films were formed as shown in in Fig (5).



a b c

Figure 3 (a,b,c): SEM Micrographs for different substrate temperatures(373K,473K,573K).

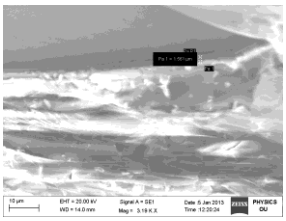


Figure 4: SEM cross sectional view of In_2O_3 thin film.

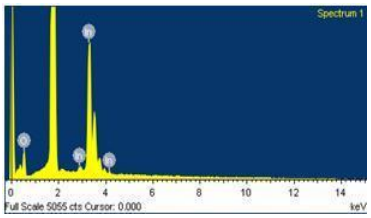


Figure 5: EDS of In2O3 thin film deposited by e- beamtechnique

UV-Vis-spectroscopy

The fig.(6) shows optical transmittance spectra of In_2O_3 thin film recorded in the wavelength region 300-1000nm.The transmittance of the thin films exhibited a sharp rise in the NIR(near infrared) region and the sharp rise high at 373K and 573K but low at substrate temperature 473K. The ripple pattern seems to arise on account of interference between light and nano structured materials. This could be explained on the basis of three dimensional quantum size effects. From the Fig.(7) the optical band gap was found to be 3.15eV,3.53eV,3.46eV for substrate temperatures 373K,473K,573K respectively. The higher value of the band gap may be due to smaller crystallite size.

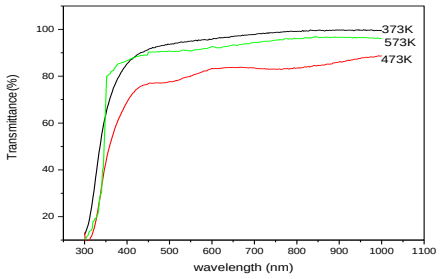


Figure 6: Transmittance spectra of In_2O_3 thin films for different substrate temperatures

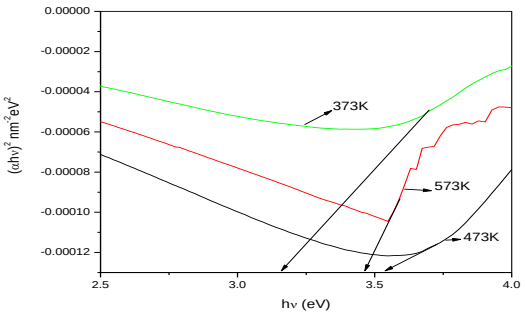


Figure 7: Optical band gap of In_2O_3 thin films for different substrate temperatures.

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CONCLUSIONS

In_2O_3 thin films were prepared by electron beam evaporation on glass substrates at different substrate temperatures 300-573K. The film deposited at 300K was amorphous in nature and low crystallite size and high intensity at higher temperatures. From AFM Hillocks with high uniformity appears at high substrate temperature. From SEM images it can be concluded that the film formed at substrate temperature 573K was highly uniform with unique granular structure compared with low substrate temperatures. Compositional analysis by EDS confirms that the sample with clear peaks of In_2O_3 is around nominal compositions. From the optical transmittance spectra the energy band gap was higher for substrate temperature 473K. The thin films of In_2O_3, deposited by Electron beam evaporation technique shows effect of substrate temperature on micro structural characterization.

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