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Effect of Substrate on Structural and Optical Properties of In_2O_3 Thin Films Prepared by Electron Beam Evaporation

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ABSTRACT

High purity Indium oxide pellets were used to prepare In_2O_3 thin films using e-beam evaporation. Ultrasonically cleaned substrates were used for the deposition of In_2O_3 thin films. The thin films were deposited at a substrate temperature of 673 K under 3×10^{-5} torr vacuum. The crystallinity of the In_2O_3 thin films was investigated using GIXRD. The films were found to be polycrystalline in nature and crystallizes in a cubic structure with preferred (2 2 2) orientation. The surface morphology, thickness and compositional analysis were investigated for these films using Scanning Electron Microscope (SEM) and Energy Dispersive X-Ray Spectroscopy (EDS). Thickness of the films was found to be 369 nm. Optical transmission measurements were carried out on In_2O_3 thin films using UV-VIS spectrophotometer in the wavelength range 300-1000 nm and it was confirmed that these thin films exhibits good transparency with an average transparency (90%) in visible region. The optical band gap was estimated from the optical transmittance data. The optical constants like optical band gap and other parameters were calculated from the optical absorption data. These films were found to be promising materials for TCO applications.

Key words: In_2O_3 thin film, quartz, GIXRD, e-beam evaporation, transmittance

INTRODUCTION

The investigation for Transparent Conductive Oxides (TCOs) (Coutts *et al*., 1999) is motivated by technical desires in many devices like automobile windows, heat-reflecting mirrors (Gillaspie *et al*., 2010) and incandescent bulbs, gas sensors (Liu *et al*., 2005), thermal detectors (Bean *et al*., 2009) ferroelectric storage and display devices (Alamria and Joraid, 2011). 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 counter parts 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 (Gupta *et al*., 2007), Direct Current (DC) magnetron sputtering (Hotovy *et al*., 2010), spray pyrolysis (Parthiban *et al*., 2010), sol-gel (Zhang *et al*., 2004), thermal evaporation (Rao *et al*., 2010) and electron beam evaporation (Shan *et al*., 2007). Surprisingly there was no report was found on substrate material effect on In_2O_3 films prepared using e-beam technique. In view of this the present study focuses on the effect of substrate material on micro structural and optical 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 and quartz substrates were cleaned by submerge in double distilled water and chromic acid and were then cleaned in a detergent solution with ultra sonicator for 15 mts. 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 quartz and 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 (Kumar *et al*., 2011a). The source material was pelletized by taking fine powder of Indium oxide (99.99%) from Sigma-Aldrich chemicals. Substrates were top mounted at 16 cm from the target material, with a miniature heater to maintain constant substrate temperature at 673 K. Initially 10^{-6} torr 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 3×10^{-5} torr inside the vacuum chamber. By operating e-gun In_2O_3 thin films are deposited on quartz and glass substrates. The films were characterized structurally using GI-XRD, surface morphological features were studied using SEM and optical properties using UV-VISIBLE spectro photometer. Throughout the deposition the power level of the e-gun was maintained constant.

Structural and morphological characterization: GIXRD of the thin films was investigated by using CuK_α radiation on a STOE high resolution X-ray powder diffractometer with parallel beam optics set to θ-2θ geometry. For all the measurements, the angle of incidence of the X-rays were kept at 0.5° and diffraction space was scanned over an angle 2θ range of 15-90°. To investigate the structural and crystallographic phases present in the films using nickel-filtered CuK_α radiation (λ = 0.15418 nm) a voltage of 40 kV and a current of 30 mA. was applied The average size of Crystallites (D) of In_2O_3 films was estimated using Debye Sherrer formula (Beena *et al*., 2010):

$$D = \frac{k\lambda}{\beta \cos\theta} \tag{1}$$

where, D-crystallite size, λ-wavelength of CuK_α radiation (0.15418 nm), k-Constant (0.9), β-FWHM, θ-Bragg's diffraction angle.

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: Optical transmittance was carried out in the wavelength range 300-1000 nm 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 (Kumar *et al*., 2011b):

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

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 (Beena *et al.*, 2007):

$$ah\upsilon = A(h\upsilon - E_g)^n \tag{3}$$

where, A is constant, υ is transition frequency and the exponent n characterizes the nature of band transition. n = 1/2 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 (Goswami, 1996). It is observed that the best straight line is observed for n = 1/2 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. $h\upsilon$.plot.

The extinction coefficient (k) was calculated by using the relation:

$$k = \frac{\alpha\lambda}{4\pi} \tag{4}$$

Where:

λ = Wavelength of incident radiation

α = Absorption coefficient

RESULTS AND DISCUSSION

X-Ray diffraction spectra recorded for In_2O_3 thin films (different substrate materials) at constant substrate temperature (673 K) using e-beam evaporation technique were shown in Fig. 1a-b. From the figure it can be concluded that the In_2O_3 thin film was found to be polycrystalline in nature and crystallizes in a cubic structure and all the peaks were well matched with the JCPDS file (card No.: 71-2195). Different peaks in GIXRD pattern which appeared at 2θ values -30.6°, 35.5° and 51.0° were due to reflections from 222, 400 and 440, respectively. The peak intensity of (222) orientation was found to be very high compared with other planes and the second strongest peak is (400). From Fig. 1c the variation of intensity with 2θ of the (222) peak differ for different substrates, this may be due to the influence of the substrate material. From the GIXRD data the average crystallite size (D) was calculated for (222) diffraction peak using Debye Scherer's formula. The average crystallite size was found to be 25 and 28 nm for quartz and glass substrates, respectively. This demonstrates the nano metric grained nature of the In_2O_3 thin films. Figure 2a-b shows SEM images at a substrate temperature of 673 K for glass and quartz substrates, respectively. From these images, it can be confirmed that the film formed on quartz substrate was highly homogeneous with unique structure compared with glass substrate. The observed SEM images are found to be crack free, glass substrate has a random molecular structure unlike quartz which have a symmetrical structure below 1100°C. As the films are prepared at 673 K the fibrous grains formed on quartz substrate are highly uniform and found to be symmetrical compared with non uniform fibrous grains observed on glass substrate. This can be attributed to symmetrical structure similarly the quartz substrate used in the present investigation. The average grain size

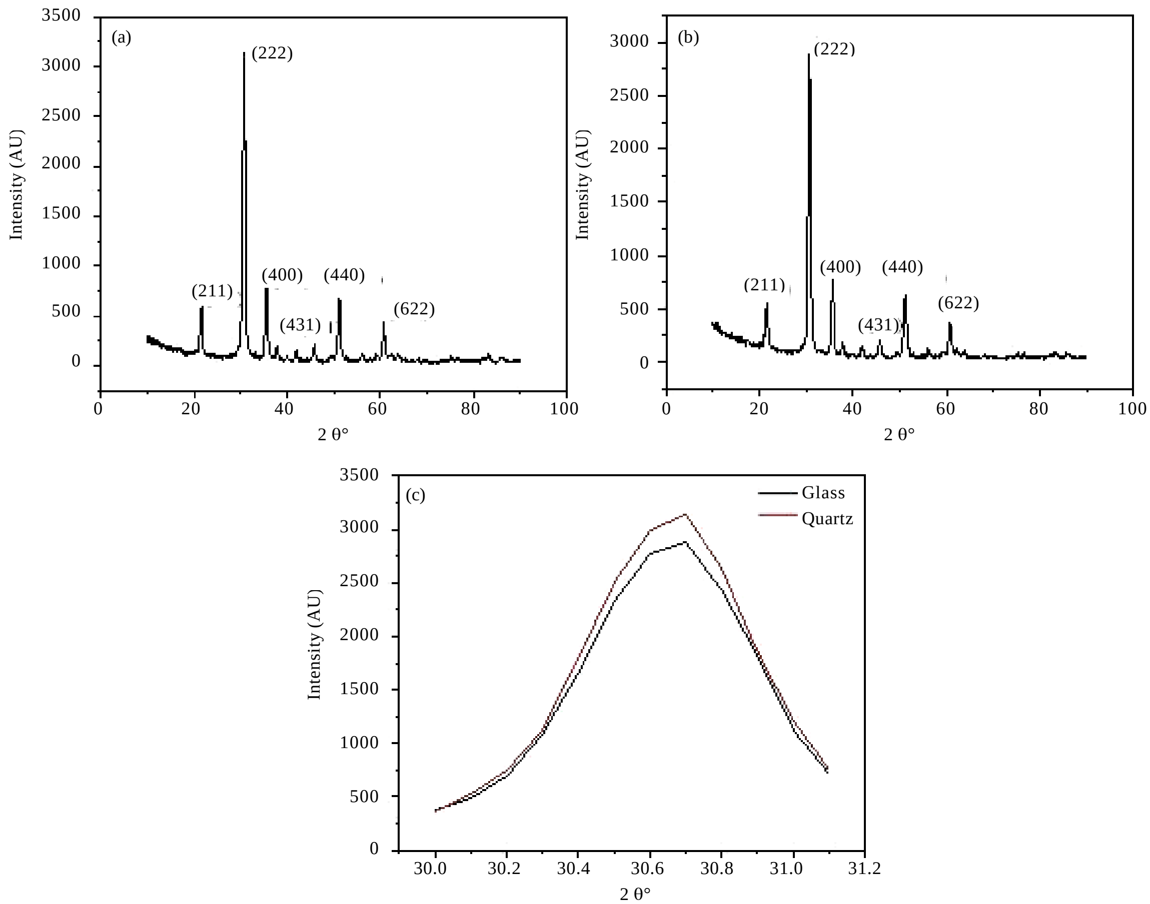

Fig. 1(a-c): GIXRD of In_2O_3 thin film on (a) Quartz substrate, (b) Glass substrate and (c) 222 GIXRD peak distribution for quartz and glass substrates

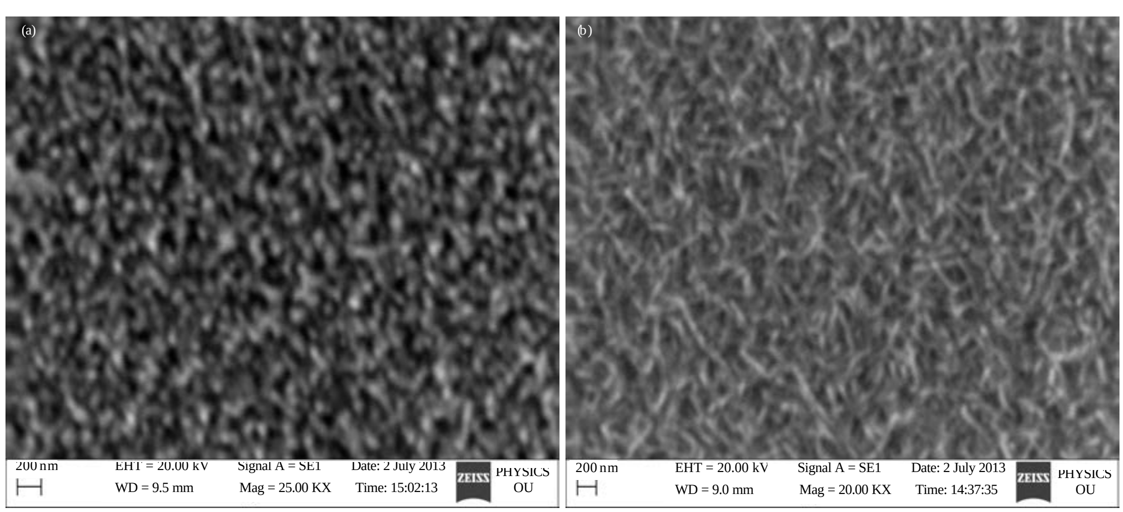

Fig. 2(a-b): SEM image of In_2O_3 thin film on (a) Glass and (b) Quartz substrate

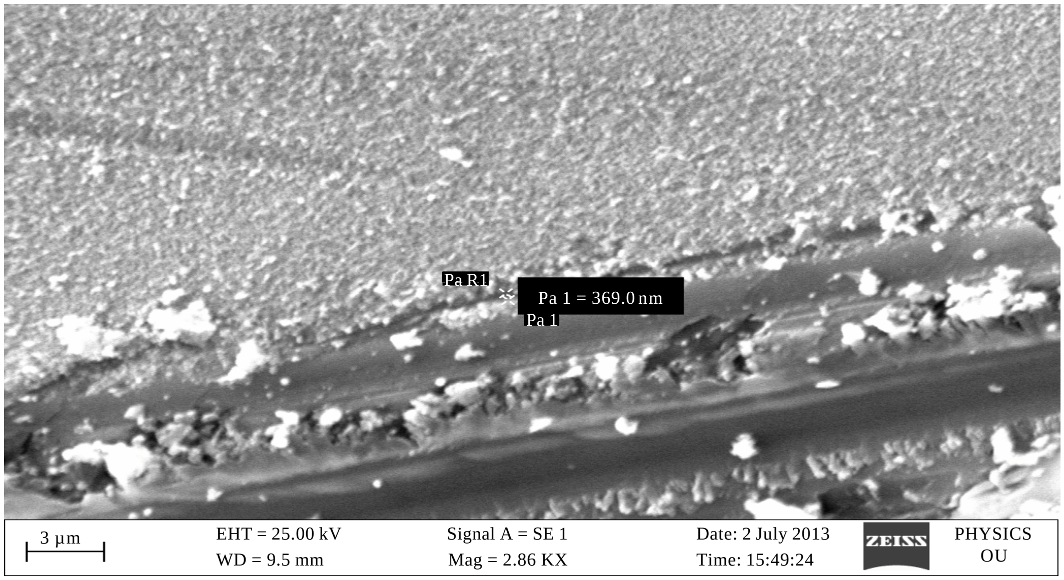

Fig. 3: Thickness of In_2O_3 thin film from SEM cross- sectional view

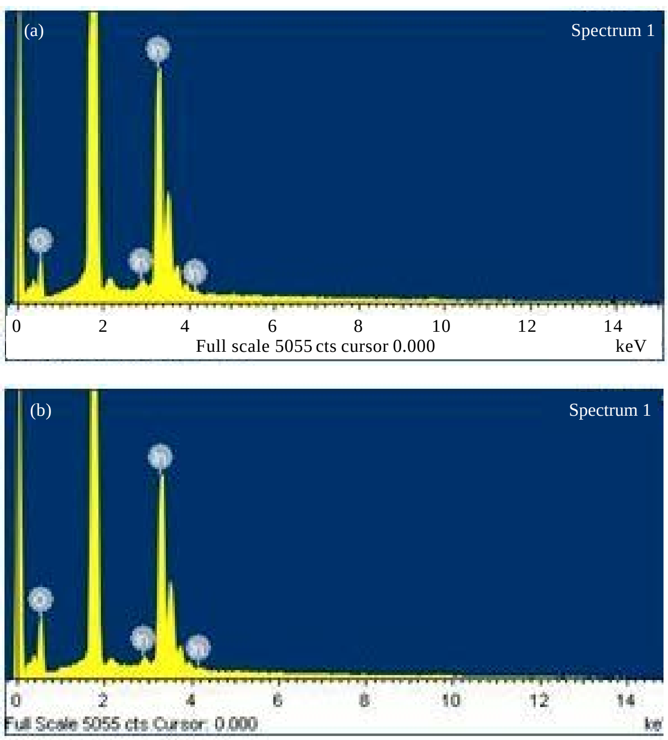

Fig. 4(a-b): EDS of In_2O_3 thin films on (a) Quartz and (b) Glass substrate

was found to be 24 nm on quartz substrate where as on glass substrate it is little higher. The thickness of the film was estimated from cross-sectional view of SEM picture was found to be 369 nm (Fig. 3). Figure 4a-b shows the elemental analysis of the deposited film using EDS. The

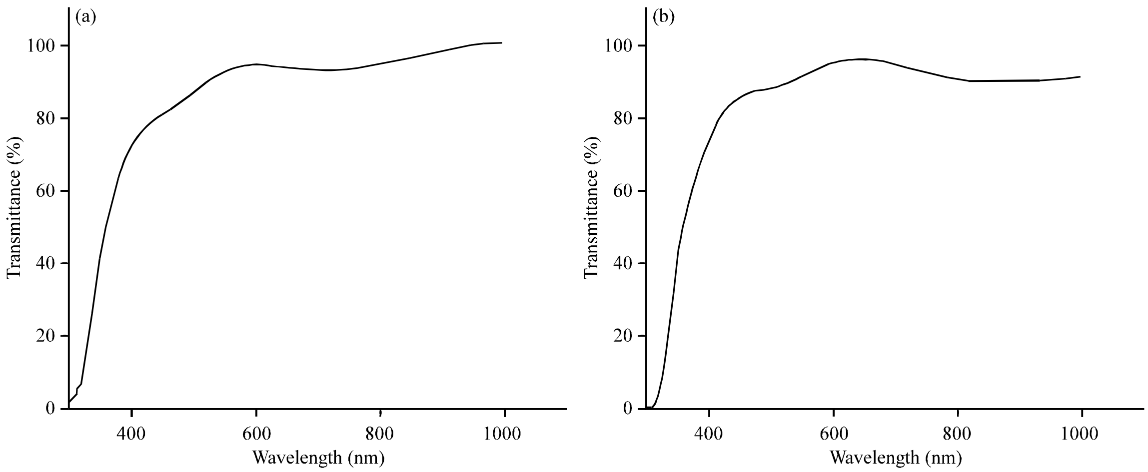

Fig. 5(a-b): Transmittance vs. wavelength of In_2O_3 film for (a) Quartz and (b) Glass substrate

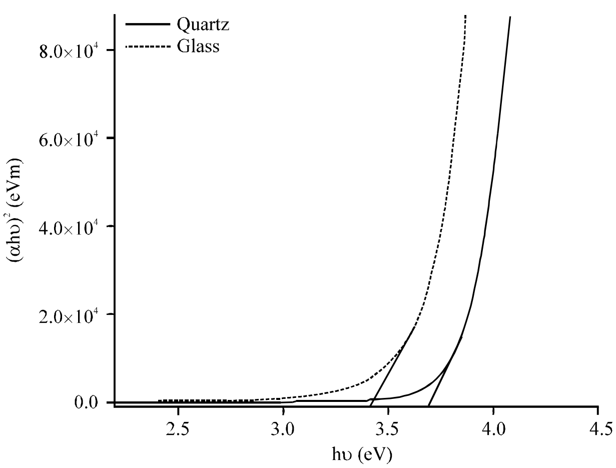

Fig. 6: $(\alpha h\upsilon)^2$ vs. $h\upsilon$ of In_2O_3 thin film on quartz and glass substrate

EDS spectrum confirms the formation of good stoichiometric films. Figure 5a-b shows optical transmittance spectra of In_2O_3 thin film recorded in the wavelength region 300-1000 nm. The transmittance of the thin films exhibited a sharp rise in the NIR (near infrared) region and the sharp rise is high on glass substrate. But in visible region both films shows the average transmittance around 90%. 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 effect. From the Fig. 6 the optical band gap was found to be 3.67 and 3.4 eV for quartz and glass substrates, respectively. The higher value of the band gap on quartz substrate may be due to smaller crystallite size. This is due to crystalline nature of quartz substrate. The variation of extinction coefficient (k) with wavelength was shown in Fig. 7a-b. The value of extinction coefficient had a maximum at fundamental

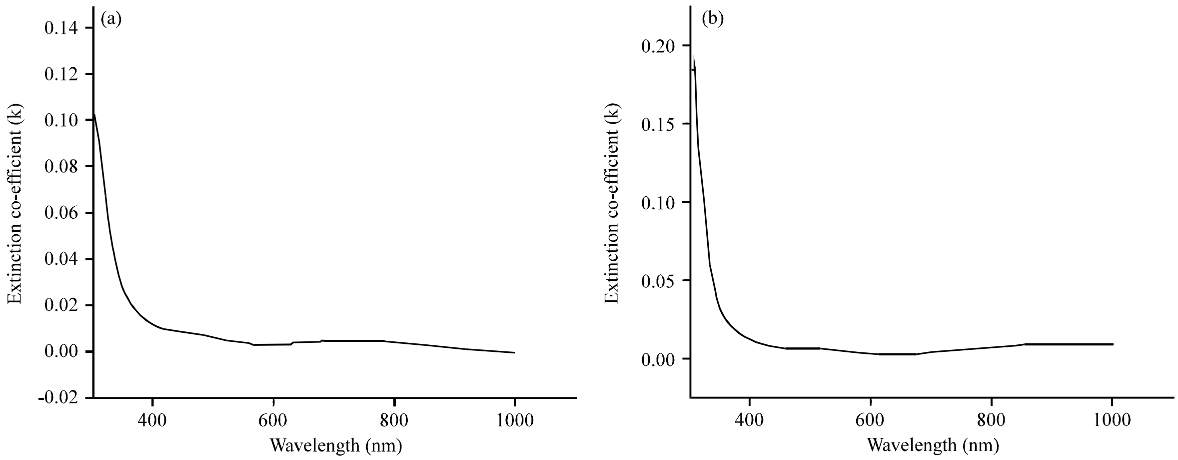

Fig. 7(a-b): Extinction co-efficient (k) vs. wavelength of In_2O_3 thin film on (a) Quartz and (b) Glass substrate

absorption edge and with increase in λ, it decreases and remains constant in visible region. The value of extinction coefficient remains un affected due to different substrates.

CONCLUSION

In_2O_3 thin films were prepared by electron beam evaporation on glass and quartz substrates at a substrate temperature of 673 K. The film deposited on quartz substrate gives low crystallite size and high intensity. From SEM images it can be concluded that the film formed on quartz substrate was highly uniform with unique structure compared with glass substrate. 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 for quartz substrate was higher than the film deposited on glass substrate. The effect of substrate on structural and optical properties confirmed that quartz substrate showing improved structural and optical properties compared with glass substrate.

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