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Optical and electrical properties of crystalline indium tin oxide thin film deposited by vacuum evaporation technique

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Indium tin oxide (ITO) thin film was deposited on glass substrate by means of vacuum evaporation technique and annealed at 200 °C, 300 °C and 400 °C in air for 1 h. The characterization and properties of the deposited film samples were analyzed by X-ray diffraction (XRD), scanning electron microscopy (SEM), and UV-VIS-NIR spectroscopy techniques. From the XRD patterns, it was found that the deposited thin film was of crystalline at an annealing temperature of 400 °C. The crystalline phase was indexed as cubic structure with lattice constant and crystallite size of 0.511 nm and 40 nm, respectively. The SEM images showed that the films exhibited uniform surface morphology with well-defined spherical grains. The optical transmittance of ITO thin film annealed at 400 °C was improved from 44% to 84% in the wavelength range from 250 nm to 2 100 nm and an optical band gap was measured as 3.86 eV. Hall effect measurement was used to measure the resistivity and conductivity of the prepared film.

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Smart technology is the most important factor to lead a developed and comfortable human life and transparent conductive thin film technology is one of them. After the discovery of the transparent conductive oxide films researchers have splurged to investigate their structural, optical, and electrical properties in order to make them suitable for the advanced technological devices like solar cells, light emitting diodes, photodiodes, flat panel displays, photo catalysts, optical memories, gas sensors, energy efficient windows etc[1-6]. In the field of technological and industrial applications including solar cell, liquid crystal displays, and photo detectors indium tin oxide (ITO) thin film is one of the most promising materials because of their higher optical transparency (≈90% in the visible spectrum range) and higher conductivity ($\approx 2\times10^4\ \Omega^{-1}\mathrm{cm}^{-1}$)[7] in comparison to other transparent conducting oxide films such as ZnO and SnO_2[8].

Typically, less than one-micron thick thin films can be conductive or dielectric (nonconductive) and are used in myriad applications. In stoichiometric form ITO materials behave as an insulator while the materials are conductor in their non-stoichiometric form with a wide direct band gap of 3.6 eV[9]. The deposition condition (time, temperature, atmosphere, and pressure), type and quantity of the dopant agent, and heat treatment method affect the properties of the ITO thin films. It is noteworthy to mention that annealing temperature plays a very significant role on the fabrication of ITO thin films. The demand for high technology products increased majority of the research on the ITO thin films predominated on the improvement of the various properties such as optical transparency, conductivity, and their microstructures deposited on glass substrate by a variety of techniques. The conventional vacuum evaporation technique among others is the most preferred technique because of its nonreactive nature with substrate and low cost.

In the present study, the effect of annealing temperature on the structural and morphological properties of ITO thin

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films produced by vacuum evaporation technique were examined by means of the X-ray diffraction (XRD) and scanning electron microscopy (SEM) analyses. The optical and electrical properties of ITO were evaluated by means of UV-VIS-NIR spectroscopy.

In_2O_3 (American Element, 99.9% pure) and SnO_2 (American Element, 99.9% pure) were deposited on a properly cleaned glass substrate with the help of a high vacuum coating unit (Edwards, UK; Model: E306A) under a vacuum of 10^{-5} Torr in order to form the ITO thin film. The powder materials of In_2O_3 and SnO_2 with the ratio of 9:1 were placed into a thin tantalum boat used as a source heater. The boat was heated indirectly by providing current through the electrodes. The distance between the source and substrate was maintained as 14.5 cm for each film. The substrate temperature was fixed at 373 K for the film. The substrate was heated by a spiral resistance heater and the temperature was measured by a chromel–alumel thermocouple placed on the middle of the substrate. The deposition of the film on the substrate was controlled by using a source shutter. When all the parameters (vacuum, deposition temperature etc.) were optimized, then the source shutter was displaced and deposition of the film started on the substrate. The film was annealed at different temperatures for duration of 1 h in air.

The phase identification along with structural properties of the synthesized ITO thin film was carried out using X-ray diffractometer (Philips PANalytical X'PERT-PRO) equipped with CuK_α radiation (0.15418 nm). The surface morphology was investigated using scanning electron microscopy (Model No. S-3400 N, Hitachi). The optical properties of the prepared films were analyzed by a Shimadzu, UV-3100 spectrophotometer. Finally, the sheet resistance of the samples was measured with a four-probe model and the resistivity of the film was calculated. The sheet resistance of the prepared films was calculated by using a simple relation

$$R_s=\rho/t, \tag{1}$$

where R_s is the sheet resistance, ρ is the density, and t is the thickness.

The ITO thin films prepared by vacuum evaporation technique were annealed at 200 °C, 300 °C and 400 °C and the corresponding XRD patterns are shown in Fig.1(a). The XRD patterns clearly demonstrate that crystalline thin film was obtained for the annealing temperature of 400 °C. A pure phase of ITO was evidenced to achieve from the X-ray reflectometry analysis[10]. The crystallite size of ITO was measured by Scherrer formula,

$$D=\frac{k\lambda}{\beta\cos\theta}\ , \tag{2}$$

where k=0.94 is the Scherrer constant, β is the full width at half maximum, λ is the wavelength of X-ray used, and θ is the Bragg angle.

The dislocation density for cubic thin film was estimated from Williamson and Smallman's formula,

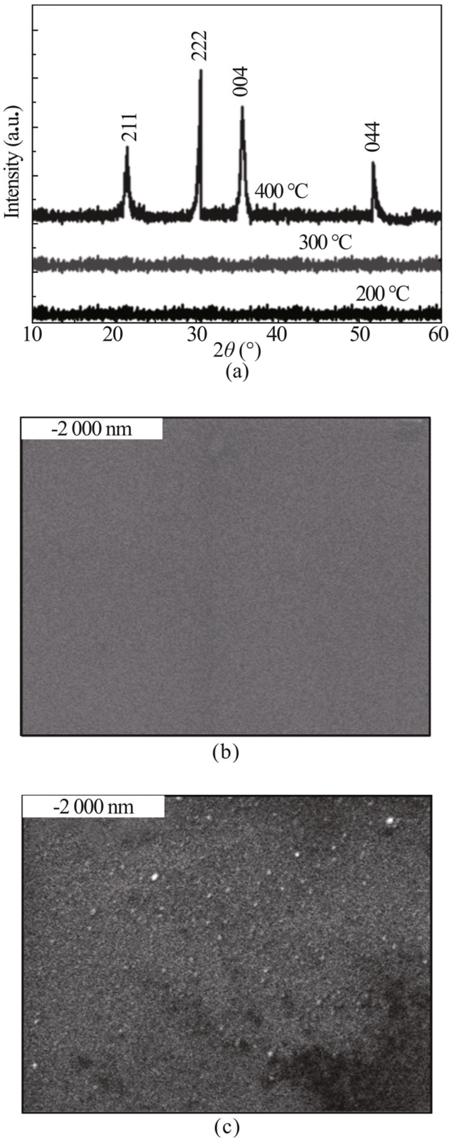
$$\rho=\frac{1}{D^2}\ , \tag{3}$$

and the micro strain of the film was estimated using the relation:

$$\varepsilon=\frac{\beta\cos\theta}{4}\ . \tag{4}$$

The average size of the crystallite was determined as 40 nm, and the number of crystallites per unit area of the film was determined as 4.77×10^{17} m^{-2}. The strain (ε) and the dislocation density (ρ) of the film were determined as 3.67×10^{-3} $lin^{-2}\cdot m^{-4}$ and 1.12×10^{16} $lines\cdot min^2$, respectively.

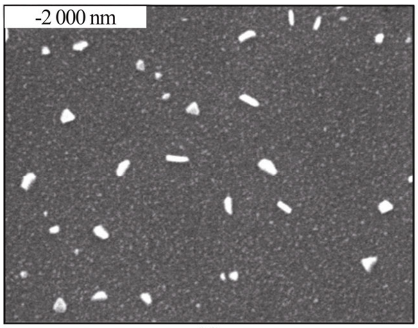
SEM images of the ITO thin films annealed at 200 °C, 300 °C and 400 °C are shown in Fig.1(b)—(d), respectively. It can be seen from Fig.1(b) that the ITO thin film annealed at 200 °C does not show obvious grain. With increasing annealing temperature, the grain becomes obvious, as shown in Fig.1(c) and (d), resulting in the increase of surface roughness[11].



(a)

(b)

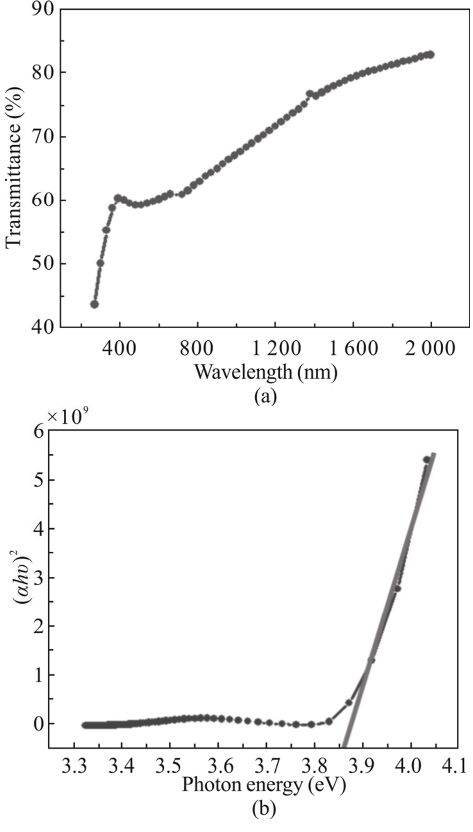
(c)



(d)

Fig.1 (a) XRD patterns of indium oxide thin film annealed at different temperatures, and their corresponding SEM images at (b) 200 °C, (c) 300 °C and (d) 400 °C

It is obvious that high transmittance of thin film indicates its low surface roughness, low refraction index, small crystallite size, high porosity, and good homogeneity[12]. The transmission spectra as a function of wavelength for the crystalline ITO thin film is shown in Fig.2(a). It is clear from the transmittance spectra that 60% to 65% transmittance occurred in the visible region. The values obtained in the present study is higher than the previously reported transmittance value of less than 50% where the ITO thin film was prepared with 50% Sn[13]. The transmittance of the transparent conductive oxide films improves with the crystalline nature reflecting that the better crystallinity decreases the scattering of light which results in excellent transparency[14].



(a)

(b)

Fig.2 (a) Transmittance versus wavelength in UV-Vis-NIR region for ITO thin film; (b) The determination of band gap from ($\alpha h\upsilon$)^2 versus ($h\upsilon$) plot for ITO thin film

The transmittance of the transparent conductive oxide films is also proportional to the concentration of oxygen[15]. The optical band gap E_g of the prepared samples for the direct allowed transitions has been calculated from the transmittance spectra by using the equation

$$\alpha h\nu = A(h\nu - E_g)^{1/2}, \tag{5}$$

where A is a constant, α is the absorption coefficient, h is the Planks constant, ν is the incident photon energy, and E_g is the optical band gap.

The absorption coefficient (α) is reckoned from the relation,

$$\alpha = \frac{1}{d}\ln\left(\frac{1}{T}\right), \tag{6}$$

where d is thickness of the films, and T is transmittance.

Fig.2(b) depicts the calculated band gap of the prepared sample and the value obtained at optimized annealing temperature for the prepared ITO thin film is 3.86 eV which fall nicely in the reported range 3.5—4.3 eV[14,16].

The measured electrical parameter of the prepared ITO thin film as a function of temperature is shown in Fig.3. It is evidenced from the graph that the resistivity of the prepared sample decreases with increasing temperature and this behavior is desirable for semiconductor type transparent conductive oxide. The obtained resistivity of the prepared ITO sample reveals that the thin film is highly degenerative n-type conductivity. The incorporation of tin dopant in indium oxide films changes the overall electrical property significantly[17]. We obtained a minimum resistivity of 4.5×10^{-2} Ω·cm for temperature 475 K. The increase of mobility is related to the reduction of grain boundary scattering. The electronic transport property for the ultrathin films is entirely related to the microstructure of the films and doping concentration[17].

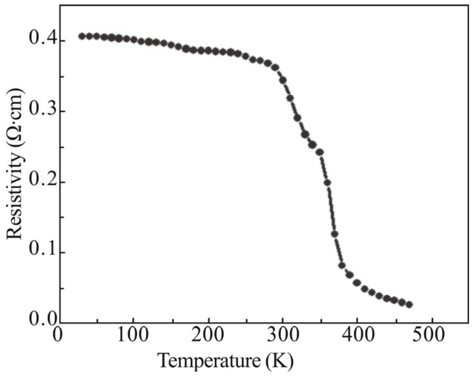


Fig.3 Temperature dependence resistivity of ITO thin film annealed at 400 °C

Transparent conductive ITO thin films were successfully prepared by the vacuum evaporation technique. The XRD spectroscopy revealed that the prepared ITO thin film annealed at 400 °C was polycrystalline in nature with cubic structure. The optical transmittance of the films at 400 °C annealing temperature has demonstrated that 60% to 65% transmittance occurred in the visible region. The observed energy band gap of the crystalline ITO thin films was measured as 3.86 eV. Hall Effect measurements showed that in the case of the prepared samples annealed at 400 °C, a rise in sample temperature effectively increases the carrier concentration and hence reduces its resistivity with an improvement in the mobility. From these findings it can be said that crystalline ITO thin films might be applicable for optoelectronic and solar cell applications and the vacuum evaporation method is an effective process for producing uniform thin films with good quality.

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