

Thickness dependence of the properties of indium tin oxide (ITO) FILMS prepared by activated reactive evaporation

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Tin doped indium oxide thin films were prepared on glass substrates kept at room temperature, by activated reactive evaporation (ARE). Structural, electrical and optical properties were studied for films having different thickness. The resulting films are polycrystalline and show $\approx 90\%$ transmission in the visible region. Hall effect measurements at room temperature for a film with a nominal thickness of ≈ 350 nm shows a relatively high carrier concentration $\approx 6.3 \times 10^{20} \text{ cm}^{-3}$, mobility $\approx 16 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, with a low resistivity $\approx 1.01 \times 10^{-3} \Omega \text{ cm}$.

Keywords: Thin films, ITO, ARE

1. INTRODUCTION

The interest in transparent conductors has tremendously increased, since the first report of a transparent conducting cadmium oxide films by Badeker(1907) [1]. These materials find applications in liquid crystal displays, transparent electrodes for solar cells, gas sensors, photodetectors and laser damage resistant coatings in high power laser technology [2]. The simultaneous occurrence of high optical transparency ($\approx 90\%$) in the visible region together with high electrical conductivity require the creation of electron degeneracy in wide band gap ($\geq 3 \text{ eV}$) oxides by introducing non-stoichiometry or appropriate dopants. These conditions can be achieved in various oxides of indium, tin, cadmium, zinc and in combinations thereof.

In view of the technological importance, a great deal of basic research and development has been carried out on the electrical and optical properties of indium oxide based materials. Rupprecht [3] carried out one of the first investigations on the indium oxide (In_2O_3). Since then different techniques have been used for the preparation of these oxide films [4-6]. The conductivity of these films can be increased by introducing various type of dopants such as tin, cadmium, antimony and fluorine. Considerable attention has been given to the study of tin doped indium oxide (ITO) films, because of their enhanced electrical and optical quality, compared to other transparent conductors. ITO is an In_2O_3 based material that has been doped with Sn to improve the electrical conductivity. Here tin acts as a cationic dopant in the In_2O_3 lattice and substitute on the indium sites to bind with the interstitial oxygen. The ITO films, with a band gap greater than 3 eV is highly transmitting in the visible region and hence is preferred in most of the applications.

In the present paper we report on the thickness dependence of electrical and optical properties of ITO films, prepared by activated reactive evaporation.

2. EXPERIMENTAL

Indium tin oxide films were prepared on substrates kept at room temperature by activated reactive evaporation [7]. The starting materials were In and 10Wt % Sn. The purity of each metal was 99.999 %. A resistively heated molybdenum boat was used to evaporate the material, in a conventional vacuum system. The evaporation was carried out in the presence of oxygen plasma. The vacuum chamber was first evacuated to a pressure of 10^{-5} m.bar. Then industrial grade oxygen was admitted into the chamber, through a needle valve, to a pressure of 5×10^{-3} m.bar. The anode supply was turned on, initiating the glow discharge. A bluish glow filled the whole chamber, and a steady discharge current was maintained. The indium and tin placed in the molybdenum boat was evaporated in the oxygen plasma. A slight adjustment of the needle valve was necessary to maintain the oxygen pressure at 5×10^{-3} m.bar. Optically flat glass slides were used as substrates. Substrates were cleaned with an industrial detergent, followed by running water and ten minutes ultrasonic agitation in distilled water. The substrates were dried in hot air and loaded into the chamber. The details of the deposition parameters have been reported in our previous paper [8].

The crystallinity of the films were identified using X-ray diffraction. The thickness of the films were measured using Tolansky's multiple beam interferometric method [9]. Conductivity and Hall effect measurements were carried out using van der Pauw method. Samples used for Hall measurements were $1 \text{ cm} \times 1 \text{ cm}$ in size and the ohmic contacts were made using silver paste. The transmission spectra were recorded from 2600 nm to cut off using Hitachi U-3410 UV-Vis - NIR spectrophotometer.

3. RESULTS AND DISCUSSIONS

Figure 1 shows the X-ray diffraction pattern of ITO films having various thicknesses. Figure 1(a) shows the XRD pattern of film with thickness ≈ 80 nm. It gives reflections only

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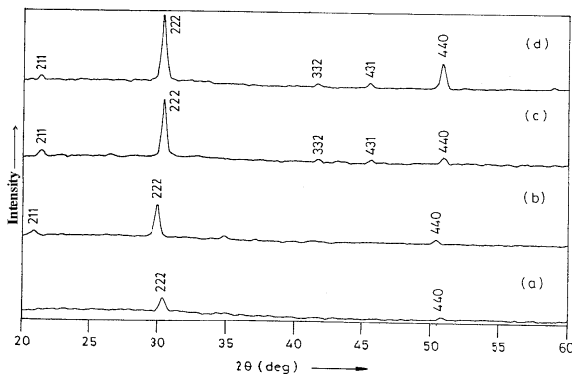


FIG. 1: X-ray diffraction pattern of ITO films of different thickness: (a) 80 nm; (b) 150 nm; (c) 270 nm; (d) 350 nm

TABLE 1: X-ray diffraction data of ITO films of different thickness with the standard data

Standard data			Film thickness							
JCPDS File No.6-416			80 nm		150 nm		270 nm		350 nm	
(hkl)	d A°	I/I ₀	d A°	I (cps)	d A°	I (cps)	d A°	I (cps)	d A°	I (cps)
(211)	4.13	14	----	----	4.125	185	4.125	349	4.125	200
(222)	2.92	100	2.91	336	2.91	849	2.91	1539	2.91	2023
(332)	2.157	6	----	----	----	----	2.155	173	2.155	100
(431)	1.984	10	----	----	----	----	1.981	120	1.981	140
(440)	1.788	35	1.786	195	1.786	106	1.786	275	1.786	856

from (222) and (440) planes, which are having relative intensity 100 and 35 respectively in the standard XRD data (JCPDS File No. 6-416). Figure 1(b) shows the XRD pattern for a film with thickness ≈ 150 nm, which gives reflection from (211) plane also, which is having a relative intensity 14 in the standard data, in addition to the (222) and (440) planes. Figure 1 (c and d) shows the XRD pattern of films having thickness 270 and 350 nm respectively. Here reflections from (332) and (431) planes with relative intensity 6 and 10 get resolved. It shows that as the film thickness increases the XRD pattern agrees very well with the standard data. In the case of film with smaller thickness, only those planes having a higher relative intensity is resolved.

It is a well-known fact that the reflected X-ray intensity depends on the penetration depth of X-ray. For films with thickness ≈ 80 nm, we obtained only the reflections corresponding to the high relative intensity in the standard data. Still the total count was very small compared to films with higher thickness. Results of XRD analysis were summarised in table 1. From this it is very clear that the films formed at room temperature are polycrystalline in nature.

The absence of reflections from (211), (332) and (431) planes of the films, with smaller thickness are not due to orientation, but due to the lack of sample thickness. This is evident by the comparative studies given in table 1. The lattice parameter is calculated to be 10.17 \AA , which is very close to that of In_2O_3 .

The grain size was calculated from the XRD pattern using Debye-Scherrer formula [10]. Figure 2 shows the variation of grain size with thickness. It can be seen that the grain size was independent of thickness, for films with thickness greater than 150 nm. Here the average grain size is ≈ 16 nm. Figure 3 shows the variation of the sheet resistance with the

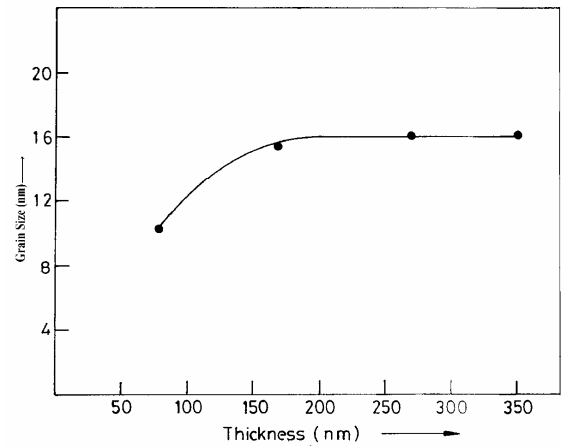


FIG. 2: Variation of the grain size with film thickness (d).

film thickness.

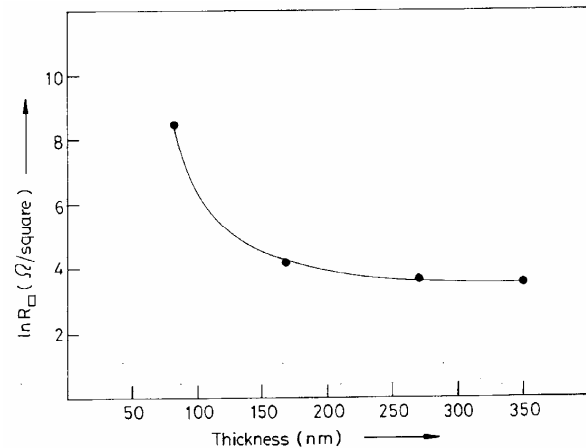


FIG. 3: Variation of sheet resistance (R) with the film thickness.

It was observed that the decrease in sheet resistance is rapid when the film thickness increases from 80 nm to 150 nm, whereas this decrease is very slow when the thickness is greater than 150 nm. A similar variation in sheet resistance with thickness has been reported for ITO and SnO_2 films [11,12]. The observed increase in sheet resistance for films having thickness less than 150 nm is probably due to the discontinuous nature of the film. The observed dependence of sheet resistance on thickness is in good agreement with Fuchs-Sondheimer theory [13].

Figure 4 shows the transmission spectra of ITO films with different thickness. The transmittance in the visible region is found to be almost independent of film thicknesses. But the transmission in the NIR region decreases with increase in film thickness.

Figure 5 shows the reflection spectra of the same set of films. It shows that there is no appreciable reflection in the NIR region. But in case of samples having greater thickness, the reflectance increases slightly for wavelengths above 2000

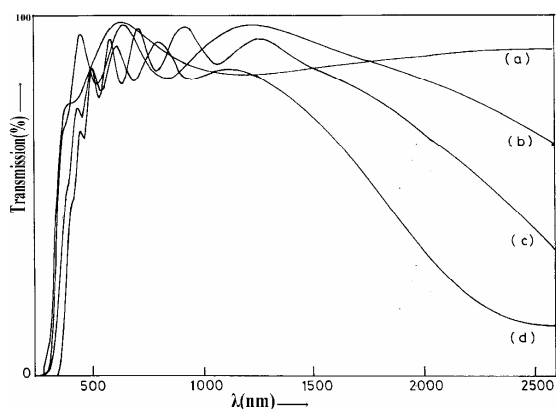


FIG. 4: Transmission spectra of ITO films of different thickness; (a) 80 nm; (b) 150 nm; (c) 270 nm; (d) 350 nm.

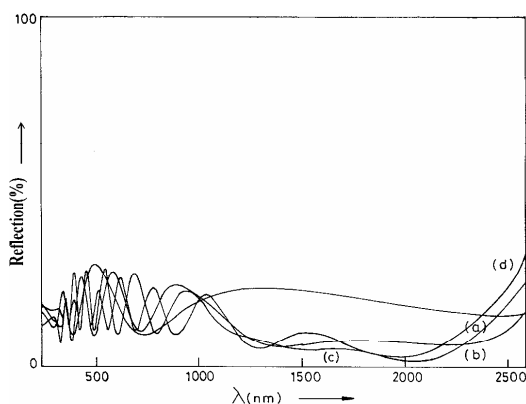


FIG. 5: Reflection spectra of ITO films of different thickness; (a) 80 nm; (b) 150 nm; (c) 270 nm; (d) 350 nm.

TABLE 2: Data obtained from Hall effect measurement of ITO films at room temperature.

d (nm)	n (cm ⁻³)	ρ (Ω-cm)	μ (cm ² V ⁻¹ s ⁻¹)
80	3.87×10 ¹⁸	3.3×10 ⁻²	48
150	1.12×10 ²⁰	1.4×10 ⁻³	34
270	3.8×10 ²⁰	1.05×10 ⁻³	28
350	6.3×10 ²⁰	1.01×10 ⁻³	16

nm. But this increase in reflectance is not consistent with the decrease in transmittance in the same region. So the decrease in transmittance with increase in thickness in the NIR region is due to the free carrier absorption, a phenomenon that is common in all transparent conductors having high carrier concentration [14].

It is also supported by the Hall effect studies. Table 2 show the results obtained from the Hall effect measurements. It shows that the carrier concentration increases with increase in film thickness. A similar variation in the carrier density with film thickness has been reported in the case of Sb doped SnO₂ films by Kaneko and Miyake [15]. The decrease in transmittance in the NIR region with increase in thickness for ITO films has been observed by other authors also[11,13,16].

4. CONCLUSION:

Highly conducting and transparent ITO films were prepared on glass substrates at room temperature by activated reactive evaporation. Crystallinity and grain size of the films were found to increase with film thickness. The Hall effect measurement shows that for a given set of deposition conditions, the thicker films have better electrical properties. The visible transmittance was found to be independent of thickness, whereas the transmittance in the NIR region decreases with film thickness.

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