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The effect of film thickness on physical properties of fluorine-doped indium oxide thin films

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In this paper, fluorine-doped indium oxide films of thicknesses ranging from 115 nm to 1290 nm were prepared using the spray pyrolysis technique by varying the amount of spray solution and keeping constant the substrate temperature, doping concentration and air flow rate. The preferential growth orientation was determined using the X-ray diffraction (XRD) spectra of doped indium oxide films of various thicknesses. The sheet resistance decreases gradually with the film thickness and reaches a stable value. It has also been observed that an increase in thickness deteriorates the optical properties of the deposited films beyond some limit.

Key words: *In_2O_3:F; transparent conductive oxide; spray pyrolysis*

1. Introduction

Optically transparent and electrically conductive films of tin, indium and zinc oxide (doped and undoped) have been studied due to their increasing practical applications [1–7]. Owing to their high optical transmittance and electrical conductivity, the films are useful in photovoltaic and photothermal applications. Transparent conducting oxide (TCO) films, which can be deposited by numerous techniques, exhibit high transmittance in the visible spectral region, high reflectance in the IR region and relatively good metallic conductivity [4–10]. Their electrical as well as optical properties can be studied by controlling the deposition parameters [11].

The transmission of light in the visible region suggests a wide band gap, characteristic of transparent conducting materials. The only way to obtain a combination of good transparency in the visible range and simultaneous high electrical conductivity is to create electron degeneracy with appropriate dopants in the oxide films [1, 9]. In

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this report, we have characterized the physical properties of In_2O_3:F films, deposited using the spray pyrolysis technique, with respect to variations in film thickness.

2. Experimental

The spray pyrolysis unit consisted of a heater capable of heating the substrate up to 700 °C and a temperature controller unit to control the substrate temperature. The spray technique is one of the most commonly used techniques for preparing transparent and conducting oxides. It owes this to its simplicity, non-vacuum system of deposition and hence inexpensive method for large-area coatings.

In order to prepare the solution for depositing fluorine-doped indium oxide films, the following procedure was adapted. The starting material, indium metal, was dissolved in HCl and the solution heated in order to complete the reaction. After evaporating the excess water, remaining indium chloride ($InCl_3$) was recrystallized twice or thrice in order to obtain pure indium chloride, and finally a solution of ammonium fluoride was added to the $InCl_3$ solution for depositing fluorine-doped indium oxide films. The optical transmission was evaluated using a UV visible spectrophotometer (UV/VIS-2100 Shimadzu). X-ray diffraction (XRD, Philips-pw-1830) was used to characterize the crystal structure of the films. The thicknesses of the films were calculated from the interference pattern observed in the visible region with the formula given by Manifacier [12].

3. Results and discussion

In_2O_3:F films of various thicknesses were prepared by changing the time of deposition, which resulted in a change of the volume of solution sprayed. In_2O_3:F (F/In = 10 wt. %) thin films were deposited onto Corning 7059 glass substrate at film thicknesses ranging from 115 nm to 1290 nm. All other parameters were kept constant, i.e. substrate temperature T_s = 425 °C, flow rate of 6 dm^3 per minute (lpm), the distance between the substrate and nozzle D_{sn} = 30 cm and solution composition. Figure 1 shows the X-ray diffraction patterns of F-doped indium oxide films of various thicknesses T. It can be seen that the preferred orientation [400] becomes more predominant as the thickness increases, however above 920 nm the intensity of the (400) plane is saturated. A similar behaviour has been obtained by Agashe et al. in SnO_2:F films deposited by the spray pyrolysis method [13]. Films thinner than 300 nm exhibited an amorphous structure.

Figure 2 shows the variation of the sheet resistance of these films at room temperature as a function of thickness. The sheet resistance of thinner films is much higher due to a predominant contribution of the amorphous phase and that of thicker films is much lower due to the presence of crystalline phases. A similar behaviour of the dependence of sheet resistivity on thickness has been obtained by Chaudhuri et al. [14] and Martinez et al. [15].

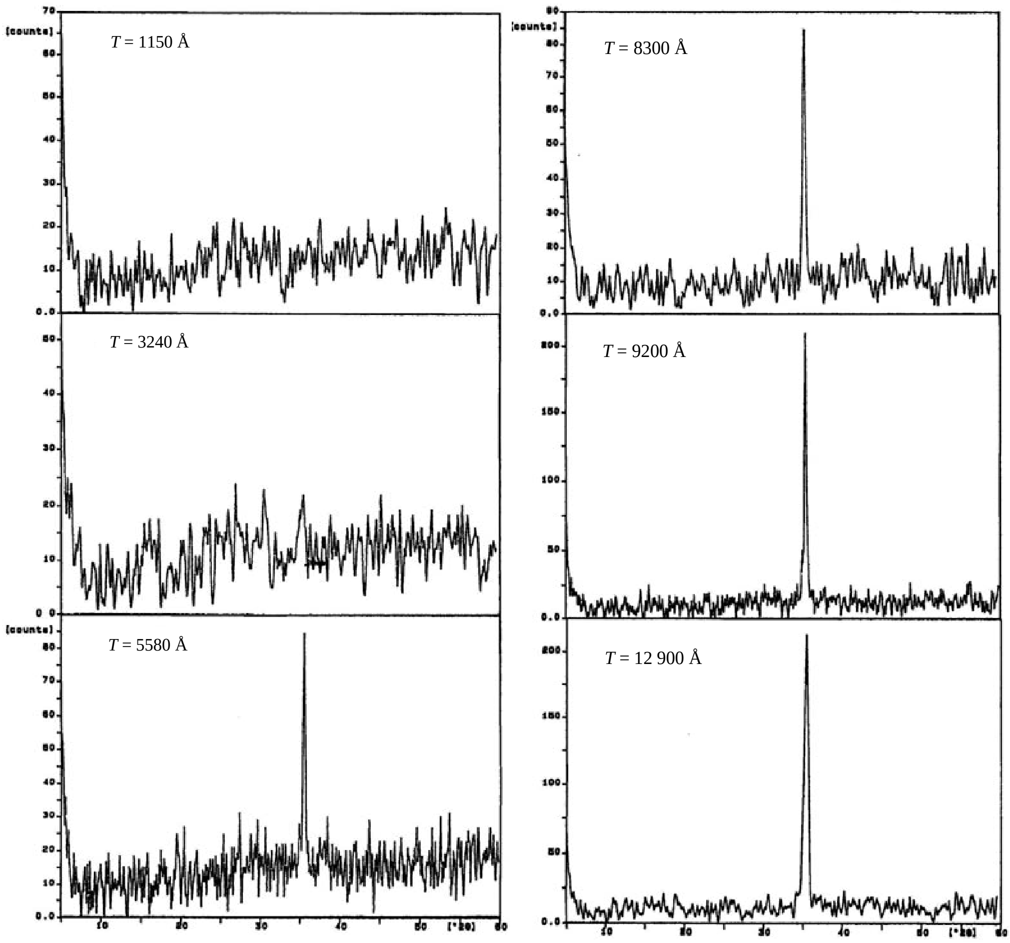

Fig. 1. Variation of X-ray diffraction patterns of F doped indium oxide thin films with thickness

Figure 3 shows the change in the average transmission (at a wavelength of 550 nm) of a series of In_2O_3:F thin films with increasing thickness. The results show that the transparency of the films decreases to a value of 70% for films having a thickness of 1300 nm.

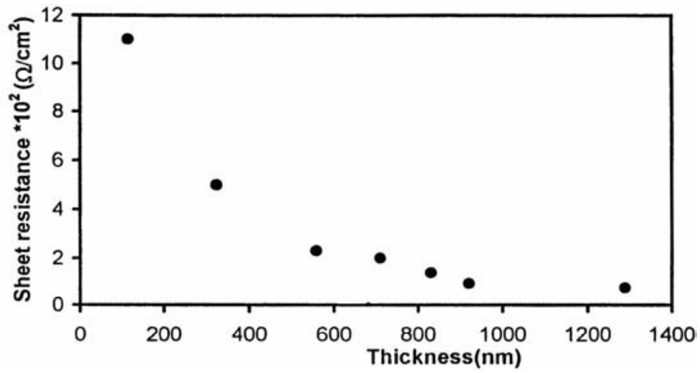

Fig. 2. Variation of room temperature sheet resistance of F-doped indium oxide thin films with thickness

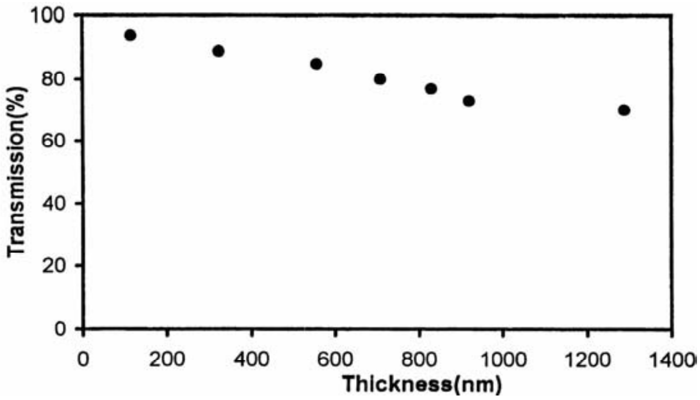

Figure 3. Variation of visible transparency of F-doped indium oxide thin films with thickness

4. Conclusions

Transparent conducting In_2O_3:F films have been prepared using the spray pyrolysis technique on glass substrate with varying the thickness. We observed that the physical properties of coated films can be improved by increasing film thickness. This results in a decrease in sheet resistance and nearly saturates at a thickness of about 600 nm. A further increase in thickness results in a decrease of the optical transmission. The structure of the films was revealed by XRD. All films exhibited a preferred orientation along the [400] direction. Films deposited with a thickness below 300 nm showed an amorphous structure.

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