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Growth and characterization of indium oxide films

P. Prathap [a], G. Gowri Devi [a], Y.P.V. Subbaiah [b], K.T. Ramakrishna Reddy [a,*], V. Ganesan [c]

[a] *Thin Films Laboratory, Department of Physics, Sri Venkateswara University, Tirupati 517 502, India*
[b] *School of Information & Communication Engineering, Sungkyunkwan University, Suwon, Republic of Korea*
[c] *UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore, India*

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Abstract

In_2O_3 films have been deposited using chemical spray pyrolysis technique at different substrate temperatures that varied in the range, 250–450 °C. The structural and morphological properties of the as-deposited films were studied using X-ray diffractometer and scanning electron microscope as well as atomic force microscope, respectively. The films formed at a temperature of 400 °C showed body-centered cubic structure with a strong (222) orientation. The structural parameters such as the crystallite size, lattice strain and texture coefficient of the films were also calculated. The films deposited at a temperature of 400 °C showed an optical transmittance of >85% in the visible region. The change of resistivity, mobility, carrier concentration and activation energies with the deposition temperature was studied. The highest figure of merit for the layers grown at 400 °C was $1.09 \times 10^{-3}\ \Omega^{-1}$.

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Keywords: In_2O_3 films; Spray pyrolysis; Physical properties

1. Introductions

Transparent conducting oxides (TCO) have been widely used in different areas due to their high optical transparency, low resistivity and wide energy band gap and hence there has been great deal of work on investigating their preparation processes and optimizing their properties. Various ternary and compound oxide materials have been developed and obtained good opto-electronic transport properties. But the preparation of doped ternary and multi-component oxide layers with suitable composition and understanding of the chemistry involved are rather difficult when compared to undoped TCO materials. In general, undoped binary oxide films are insulators in its stoichiometric condition. But the conductivity of pure oxide layers can be improved to the level of doped layers by suitably controlling the density of oxygen vacancies, each of which donate two electrons to the conduction band. This deficiency determines the conductivity of undoped oxide layers. Moreover, the ultimate attainable properties are material dependent. Among the various TCO materials available, In_2O_3 is one of the potential candidates for solar cell and sensor applications. It is an n-type semiconductor that has high electrical conductivity. This material has an optical energy band gap of 3.6 eV with good adherence to the substrate surface and high chemical inertness [1–4].

In_2O_3 films have been prepared using various physical methods such as vacuum evaporation [5], pulsed laser evaporation [6], sputtering [7], sol–gel process [8] and a variety of chemical methods [9–11]. Among the different techniques used, spray pyrolysis offers many advantages such as low cost of the apparatus and raw materials, flexibility for doping the layers and easy control over the

* Corresponding author. Tel.: +91 877 2249666x272; fax: +91 877 2248485.
E-mail address: ktrkreddy@hotmail.com (K.T. Ramakrishna Reddy).

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deposition parameters compared to other process. As the electro-optical properties of thin films are highly sensitive to microstructure as well as the level of residual stress/strain caused during the deposition process [12] and orientational changes which in turn depend on the deposition conditions [13], it is essential to study the influence of each process parameter in order to get good control over the physical properties of the films. A few reports are available on the influence of pyrolysis temperature on surface morphology [30], which predominantly affects the light transmission to absorber layer in photovoltaic devices causes to photocurrent losses. Hence, the present investigation involves the study of the effect of substrate temperature on physical properties such as structure changes, surface morphology and other opto-electronic properties of In_2O_3 layers grown by chemical spray pyrolysis at different substrate temperatures.

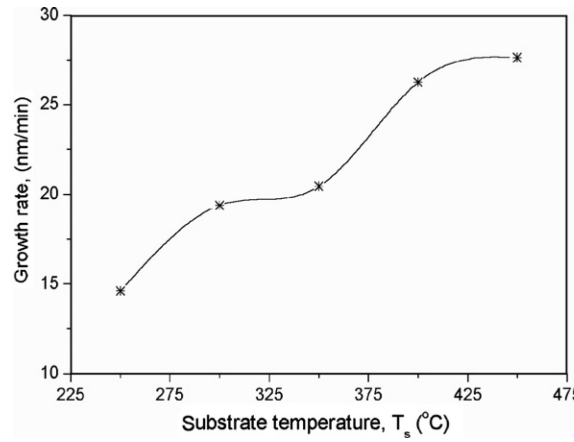

Fig. 1. Growth rate of In_2O_3 layers as a function of the substrate temperature.

2. Experimental details

In_2O_3 thin films were deposited by spray pyrolysis technique. 5 N pure $InCl_3$ and deionized water were used as the solute and solvent to prepare the solution with a concentration of 0.1 M. The layers were grown at different substrate temperatures that varied between 250 °C and 450 °C. The substrate temperature was maintained at predetermined temperature using Eurotherm temperature controller with an accuracy of ±5 °C. Chromel–Alumel thermocouple placed on glass substrate surface was used as a temperature sensor for the controller. Ultrasonically cleaned corning 7059 glass was used as the substrate. Compressed purified air was used as the carrier gas with a flow rate of 8 l/min and the solution was sprayed at a flow rate of 6 ml/min. The substrate to nozzle distance was maintained at 25 cm. The spray head was moved in the *X*–*Y* plane using a microprocessor controlled stepper motor system in order to get uniform films on the substrate. The precursor solution was sprayed onto the substrates at an interval of 1–2 min i.e., the spray process lasts for 8 s and was paused for 1–2 min, in order to overcome cooling effect caused by the mist of the precursor. After completion of the deposition process, the films were allowed to cool slowly to room temperature. The structural properties of as-grown layers were studied using Siemens X-ray diffractometer. The surface topological and morphological studies were carried out using Hitachi scanning electron microscope and Vecco atomic force microscope, respectively. The optical properties of as-grown layers were studied using Hitachi U:3400 UV–Vis–NIR spectrophotometer. The standard spectroscopic ellipsometry was used to determine the thickness of the films, which varied in the range, 162–347 nm. The electrical properties of the films were carried out using Hall effect measurements, employing van der Pauw method with silver as contacts.

3. Results and discussion

The visual appearance of all layers grown in the substrate temperature range, 250–450 °C, were pinhole free, strongly adherent to the substrate surface and whitish in appearance. Fig. 1 shows the growth rate of In_2O_3 thin films as a function of the deposition temperature. The monotonous increase in the growth rate at deposition temperatures below 400 °C probably indicates an activation energy limited process [14]. The lower thermal energy available for the molecules in the solution at lower deposition temperatures can also cause incomplete reactions between the precursors on the surface of the substrate. At higher deposition temperatures, growth rate was non-linear which may be due to the re-evaporation of indium atoms during the deposition. Ayouchi et al. [15] reported that when the substrate temperature is very high, the precursor solvent vaporizes away from the substrate and the precursor chemical reaction is carried out in the vapor phase that results in the decrease of growth rate or thickness of the sprayed ZnO films. Krunks and Mellikov [16] explained the decrease in the growth rate of spray deposited ZnO films at higher growth temperatures that mass transport to the substrate is diminished due to gas convection from the bath pushes the droplets of the precursor away. Moreover, the increase of rate of re-evaporation is also responsible for the decreased in the growth rate at higher deposition temperatures.

Fig. 2 shows the X-ray diffraction profiles of In_2O_3 films formed at different substrate temperatures in the scan range, 20°–70°. All the diffraction peaks, as indexed in the spectrum, originated from the cubic structure of In_2O_3 with the lattice constant, $a = 1.118$ nm. The layers formed at a temperature 250 °C had (400) predominant orientation with more amorphous background and those deposited between 300 °C and 350 °C consisted of randomly oriented crystallites with different peaks that correspond to (211), (222), (400), (411), (440) and (622) orientations of In_2O_3. The films grown at 300 °C showed (400) as the preferential orientation and it was slowly changed to (222) with the increase of substrate temperature. Kosltin et al. [17] reported a similar change in the preferred orientation from (400) to (222) with the increase of substrate temperature in sprayed In_2O_3 films. Korotcenkov