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Study of the Aluminium Oxide Doped Zinc Oxide Thin Films Prepared by Thermal Evaporation Technique

Sowmya Palimar, Kasturi V. Banger and G.K. Shivakumar
Department of Physics, Thin Film Laboratory, National Institute of Technology, Karnataka, India

Abstract: The present study reports the observations made on investigations carried out to study structural, optical and electrical properties of aluminium oxide doped ZnO thin films obtained by thermal evaporation technique. Films obtained are found to be amorphous in nature with smooth and continuous surface. Room temperature conductivity of the film is found to be 5×10^{3} U^{-1} cm^{-1} with visible region transmittance of above 95%. The optical energy gap of the film is found to be 3.25 eV. From the calculations of activation energy, it is observed that the doped ZnO film has two donor levels, one at 142 meV and other at 43 meV. A detailed analysis of the result is reported.

Key words: ZnO thin film, thermal evaporation, optical property, electrical conductivity

INTRODUCTION

Zinc oxide (ZnO) thin film has attracted much attention in the research community due to its coexistence of conductivity and transmittance in the visible region of electromagnetic radiation spectrum. Although, verities of techniques (Jeong *et al*., 2003; Craciun *et al*., 1994; Prasada Rao and Santhoshkumar, 2009) have been adopted to obtain ZnO thin films, not much attention has been given towards its preparation by thermal evaporation technique. Thermal evaporation method is found to be one of the low cost and simple procedures to obtain ZnO thin films. This method does not demands any catalyst or high temperature growth and even the annealing condition required for the further oxidation of ZnO films obtained by this method is moderate enough to apply easily in thin film technology.

In the present study an attempt is made to obtain transparent and conducting aluminium oxide doped zinc oxide ($ZnO:Al_2O_3$) thin films by thermal evaporation technique. The structural, optical and electrical properties of these Al_2O_3 doped ZnO films are studied and reported.

MATERIALS AND METHODS

Aluminum oxide doped ZnO ($ZnO:Al_2O_3$) thin films were obtained by thermally evaporating charge, containing a mixture of 95% of ZnO [99.99% Alfa Aesar] and 5% of Al_2O_3 [99.97% Alfa Aesar] powders, on clean glass slides using tungsten boat as the evaporating source.

The glass slide is maintained at room temperature and the vacuum maintained during the evaporation is of the order of 10^{-5} torr. Gravimetric method is used to measure the thickness of the film and it is found to be 200 nm. The films obtained in the as deposited condition were dark brown in color and opaque in nature. During the process of evaporation, ZnO decomposes in to zinc and atomic oxygen according to the kinetics of vaporization of ZnO (Lvov *et al*., 2004) and the film is expected to be rich in zinc.

Structural characterization of the obtained $ZnO:Al_2O_3$ films was done using JEOL X-ray diffractometer with a scanning rate of 1° min^{-1}. The transmittance and absorbance measurements were carried out relative to the uncovered substrates at normal incidence at a spectral range of 250-850 nm using Ocean Optics Inc SD 2000 UV-Vis spectrometer. The electrical characterizations of $ZnO:Al_2O_3$ thin films were found out using Keithley source meter (2400) setup and thermally evaporated silver strips separated by a gap of 1 mm were used as the electrodes.

RESULTS AND DISCUSSION

X-ray diffractograms of $ZnO:Al_2O_3$ films did not show any peak in the θ range of 20-80°. This confirms the amorphous nature of the films.

The scanning electron micrographs of the surface of $ZnO:Al_2O_3$ film, is shown in Fig. 1. By analyzing the micrographs, it is found that film has smooth, continuous and pinhole free microstructure. This amorphous nature

Corresponding Author: Sowmya Palimar, Department of Physics, Thin Film Laboratory, National Institute of Technology, Karnataka, India

with flat and smooth surface of films will lead to the low internal stress and such films are well suited for the application of flat panel displays (Shigesato, 2010).

Figure 2 shows the transmittance spectra and optical energy gap measurements of $ZnO:Al_2O_3$ films. It is observed that the films have an excellent transmittance of above 95% in the visible region of the electromagnetic spectrum. Since annealing leads to oxidation, the increase in the transmittance may be due to the oxidation of the films. Thus the film, which was rich in zinc in the as deposited condition, might have attained stoichiometry after annealing and showed a good transmittance in the visible region of the electromagnetic spectrum. There are reports, which prove the improvement in the stoichiometry of the ZnO film after annealing (Aida *et al*., 2006; Aly *et al*., 2001).

Since ZnO is a direct band gap semiconductor allowed direct transitions can be assumed and the spectral dependence of the absorption coefficient, α can be described using the following Eq. 1 (Khairnar *et al*., 2003):

$$\alpha h\upsilon = B(h\upsilon - Eg)^{1/2} \quad (1)$$

where, B is a constant.

The variation of $(\alpha h\upsilon)^2$ with photon energy $h\upsilon$, is plotted and the linear portion of the plot is extrapolated to zero to find the optical band gap of the film (inset of Fig. 2). It is shown that the optical band gap of the $ZnO:Al_2O_3$ film is 3.25 eV.

To study the electrical property, the ohmic contact was made using metallic silver. At room temperature, $ZnO:Al_2O_3$ films showed an excellent conductivity of $5\times10^3\ \Omega^{-1}\,cm^{-1}$.

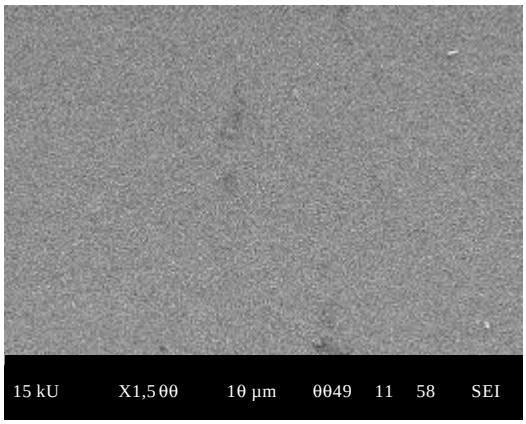

Fig. 1: SEM image showing the surface topography of ZnO thin film after doping with 5% Al_2O_3

This high conductivity obtained due to the doping of III group oxides can be explained by considering the Eq. 2 (Elmer, 2010).

According to Eq. 2 on adding group III oxides like Al_2O_3, Ga_2O_3 or In_2O_3 to ZnO, third group dopant metal atoms (In, Al, Ga) will get released. Since these atoms have one valance electron more than zinc they act as donors and built in to zinc lattice site. The additional electrons which are released during the process according to Eq. 2 and not used for the bonding are transferred to the conduction band and will contribute in improving the conductivity of the film.

$$Al_2O_3 \rightarrow 2Al_{Zn} + 2e + (\tfrac{1}{2})O_2 \quad (2)$$

To calculate the activation energy the variation in the resistance of the films with the change in temperature is recorded. The study was carried out by increasing the temperature of the film up to 300°C from room temperature. In this low temperature, extrinsic conduction predominates since ZnO has a wide band gap of the order of 3.3 eV.

The semiconducting nature of the films follows the Eq. 3 (Sze, 2006):

$$R = R_0 \exp(E_0/k_bT) \quad (3)$$

where, R is the resistance of the film at temperature T, R_0 is a constant, k_b is Boltzmann constant (1.38×10^{-23} J K^{-1}) and E_0 is the activation energy required for conduction.

A representative graph of variation of Log R with the reciprocal of temperature (1/T) is shown (Fig. 3). From the Log R vs. 1/T plots, the thermal activation energy is calculated using the Eq. 4.

$$Ea = [2.303\ k_b\ (slope)]/e \quad (4)$$

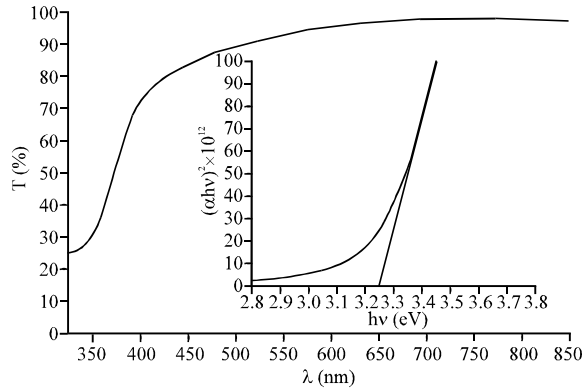

Fig. 2: Transmittance spectra of $ZnO:Al_2O_3$ thin films

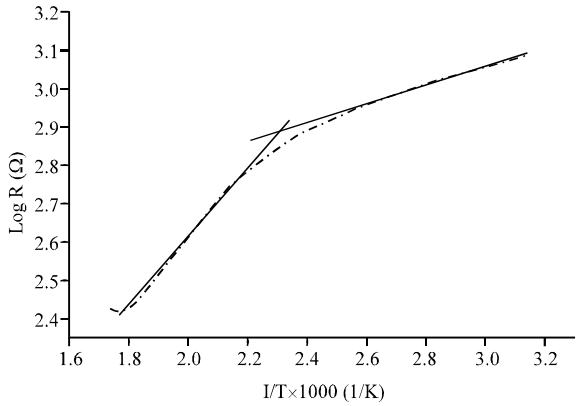

Fig. 3: Logarithmic resistance profile with respect to reciprocal of temperature for Al_2O_3 doped ZnO thin film

Where, e = magnitude of charge on electron (1.6×10^{-19} C). In the graph Log R verses 1000/T (Fig. 3), for ZnO:Al_2O_3 thin film, curve shows a linear behaviour in two regions. One at 1.7 -2.1 (1/K) and the other at 2.3-3.2 (1/K). In these two ranges the activation energy is proportional to the slope. Aluminium oxide doped ZnO thin film shows first activation energy of 43 meV in the range of (2.3-3.2 [1/K]) and second activation energy of 142 meV in the range of (1.7-2 [1/K]). The electron concentration in ZnO thin film is increased by aluminium oxide doping and the trapping levels with activation energy lower than 55 meV will support the electrical conductivity of the film with very low energy expense. This condition will support the dark conductivity of the film (Jimenez-Gonzalez *et al*., 1998).

CONCLUSION

Aluminium oxide doped zinc oxide thin films were prepared by thermally evaporating a mixture of charge containing 95% of zinc oxide powder and 5% of aluminium oxide powder. It is shown that the films obtained have a very good visible region transmittance of above 95% with a significantly high room temperature conductivity of $5\times10^{3}U^{-1}$ cm^{-1}. From the structural characterizations the amorphous nature and the smooth and continuous surface of the film is confirmed. Thus, aluminium oxide doped ZnO thin films with an excellent combination of conductivity and transmittance, well suited for the application of transparent electrodes are obtained by thermal evaporation method.

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