Int.J.HIT.TRANSC:ECCN.Vol.7: Issue 2A (2021) Page 18-21 ISSN: 0973-6875

Available Online at www.hithaldia.in/locate/ECCN

ORIGINAL CONTRIBUTION

SOL-GEL PREPARATION, CHARACTERIZATION AND THERMOELECTRICAL PROPERTIES OF ALUMINUM DOPED ZINC OXIDE FILMS

G.K.Paul

Department of Physics, Hooghly Mohsin College, Hooghly – 712101

ABSTRACT

ZnO films doped aluminum has been prepared using sol-gel technique. The films are polycrystalline in nature having wurtzite structure and a tendency of growth of (002) reflection with doping. The crystallite size becomes increase with doping. The thermoelectric power (TEP) measurements curves show a peak around 215 K for all the samples, which might be due to interaction between charge carrier and anisotropic lattice vibrations.

Keywords: Sol-gel; Zinc oxide; Thermoelectric power; Thin film;

1. INTRODUCTION

ZnO thin films are technologically important due to their range of electrical and optical properties, which make them suitable for a variety of applications, such as solar cell electronics, gas sensors and optical wave-guide devices etc. A wide variety of coating techniques for the transparent thin film are known such as thermal evaporation, chemical vapor deposition [1], RF-magnetron sputtering [2], spray pyrolysis [3] etc. Sol-gel [4-6] method is another attractive technique for obtain thin films and has the advantages of easy control of the film composition and easy fabrication of a large area film with low cost. In this article we report the preparation, structural and thermoelectrical properties of Al doped films prepared by sol-gel techniques.

2. EXPERIMENTAL

The samples are prepared on silica-glass substrates. Thoroughly cleaned and heat treated at 550^0 C. The precursors used are zinc acetate di-hydrate (extra pure), dehydrated isopropyl alcohol and diethanolamine (DEA) as sol-stabilizer. Zinc acetate has been added to alcohol so as to prepare concentration of 0.6 mole/liter. The resultant solution was mixed thoroughly on a magnetic stirrer. Then equimolar quantity of diethanolamine (DEA) has been introduced drop by drop into the solution until the solution becomes transparent. For aluminum doping, we have mixed the reagent $Al(NO_3),9H_2O$. Al/Zn ratio varied from 0 to 2.0 at%. The solution after proper mixing was filtered and kept for few days. The films were prepared from this solution by drain coating method having withdrawal rates of about 6 cm/minute. The films after each dip coating were kept in a furnace in air at 120^0 C for 30 minutes and then quickly put into another furnace at 550^0 C and heated for 30 minutes. The process was repeated ten times for each sample. We have carefully chosen those which have similar thicknesses and uniformity. The average thicknesses of film chosen are about 250 nm.

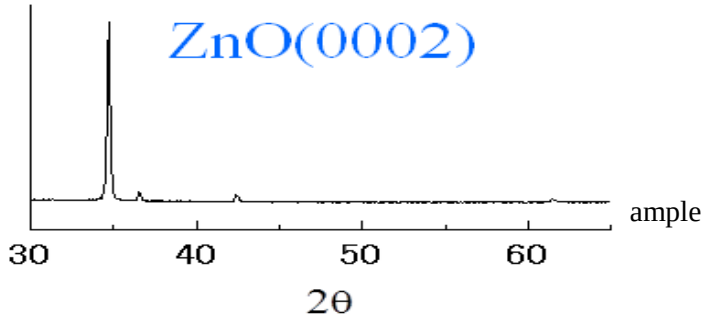
We recorded the X-ray diffraction pattern (XRD) in an X-ray diffractometer (Philips model PW1730) using CuK_α as a source of radiation.

*Corresponding Address: gkpaul76@yahoo.in

The scanning electron micrographs of two samples have been taken from HITACHI (Model S-2300). The thermoelectric power (TEP) of the samples were carried out at high vacuum using lead as electrode and varying the temperature difference between hot and cold end up to maximum of 15K. Copper-constantan thermocouples were used to measure the temperature difference. TEP was calculated from the slope of a Seebeck voltage vs. mean temperature difference graph. The measurements of TEP which were done from room temperature to about 190 K.

3. RESULTS AND DISCUSSION

The doping of Al was performed at 2 at %. Samples were prepared and annealed 550^0 C for 30 minutes and in reduced atmosphere for 30 minutes at 400^0 C. Our samples were polycrystalline in nature having wurtzite structure. The X-ray diffraction spectra for one (2 at%) Al doped ZnO thin films prepared at the temperatures (550^0C) shown in Fig.1. indicate that the films are of polycrystalline nature. These films have a better orientation along (002) direction. A very important result is that the preferential orientation of the crystallites is not lost with annealing, as occurs in ZnO films prepared by other techniques.



The observed microstructures of the films are also different for two films. The two microstructures obtained from SEM have been shown in Fig.2. For undoped sample (Fig.2a) the crystallite size is smaller than the 2 at % doped (Fig.2b) sample where the crystallite sizes are larger. The grain size is around 40 nm for the film. The grains are regular in size and shape and densely packed with little porosity.

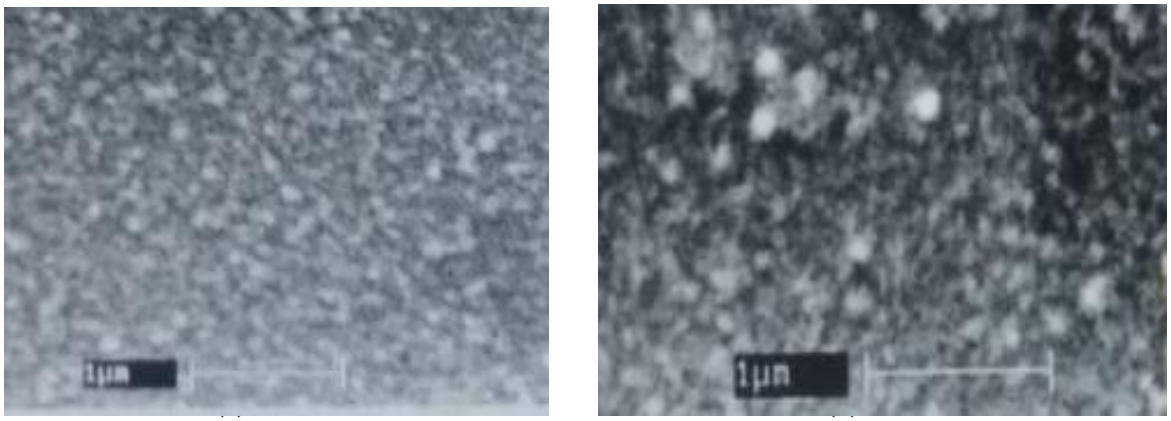


Fig.2. SEM micrographs of (a) Undoped and (b) 2 at % Al doped ZnO samples.

Thermoelectric power (TEP) measurement

We measure the TEP of doped ZnO films especially at low temperature upto 190K. In Fig.3. the plot of TEP against temperature has been shown. The Seebeck coefficients are negative within the whole temperature range examined indicating n-type of conduction. There is a steady decrease in magnitude of TEP with lowering of temperature but a sudden increase at a temperature near about 215K. This cannot be related to sudden change in effective mass (m^*) for otherwise it will be reflected in the mobility curve. Such changes have earlier been noticed in germanium by Geballe and Hall [7] below 50 K, which was explained on the basis of the theory, proposed by Herring [8].

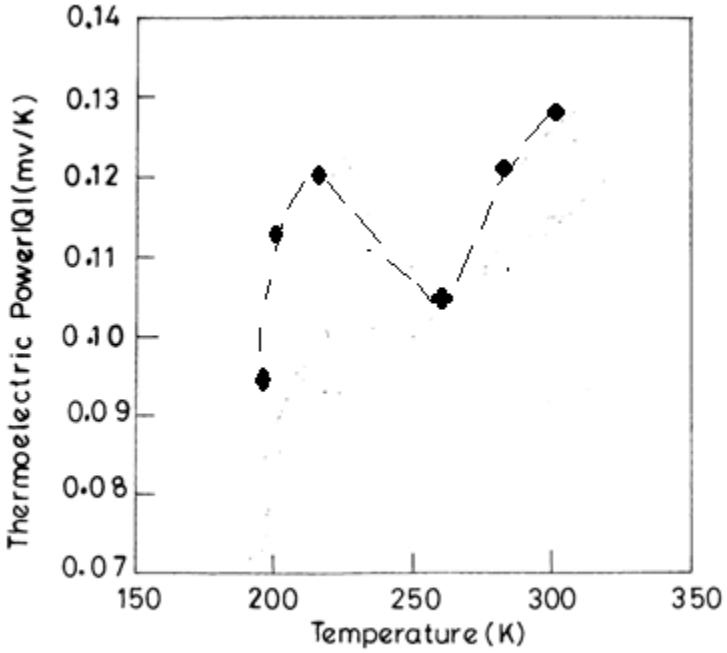


Fig.3

4. CONCLUSIONS

Several samples of aluminum doped zinc oxide films have been prepared by sol-gel metho]. An doping concentration of 2 at % aluminum and annealing temperature 550^0 C found to be the good quality films. The thermoelectric power A hump in thermoelectric power has been noticed around 215 K, the origin of which might be due to interaction between phonon and mobile charge carrier.

ACKNOWLEDGEMENTS

The author gratefully acknowledge with thanks the cooperation received from Dr. S.Bera of Tamralipta Mahavidyalaya, Tamluk, Purba Medinipur, West Bengal, India in carrying out this study.

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