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# Thickness Dependence of Electrical and Optical Properties of Sol Gel ZnO Coatings

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Single and multilayer transparent conducting zinc oxide film has been prepared on boro silicate substrates by the sol gel dip coating process. Each layer was kept at 600°C in a conventional furnace for 15 min. The final coatings were then tempered under a flux of forming gas ($N_2/H_2$) at 400°C for 2 h. The coatings were characterized by surface stylus profiling, optical spectroscopy (UV, near IR) and combined Hall and van der Pauw techniques. The thickness, sheet resistance, electrical resistivity, mobility and transmittance of obtained ZnO coatings were studied as a function of dipping speeds and the number of coatings. The film resistivity decreases as the number of coatings increases while the carrier mobility increases. The transmittance of ZnO multilayers is slightly decreased with increasing the number of coatings.

**Key Words: ZnO coatings, Electrical and optical properties, Thickness.**

## INTRODUCTION

Transparent conductive oxide films (TCO) that are both transparent in the visible region and electrically conducting have been studied widely for over 20 years as a result of their extensive applications in opto-electronic devices. Zinc oxide (ZnO) film exhibits a wide range of technological applications, such as SAW devices, transparent conducting film[1–3], solar cell windows[4, 5], gas sensors[6] and surface acoustic wave devices[7]. For the wide variety of applications, numerous ZnO film preparation methods have been attempted: r.f. and magnetron sputtering[1, 8, 9], spray pyrolysis[10–13], chemical vapour deposition (CVD)[14–16] and sol gel process[17].

The sol gel method is an attractive technique for obtaining thin films and has the advantage of easy control of film composition and easy fabrication of large area thin film with low cost. The usual starting materials of the sol gel process are metal alkoxides and they are hydrolysed to form a sol.

In the present work, a suitable sol from simple salts such as acetates was obtained[18], which is more convenient to use and less costly. In this paper, the thickness dependence of different electrical properties of sol gel ZnO thin films was investigated.

## EXPERIMENTAL

The sols were prepared using methodologies already described[17, 18] in the literature. The sol was prepared by dissolving diethanolamine (DEA, Fluka puriss) in isopropanol (*i*PrOH). Then zinc acetate dihydrate ($ZnOAc{\cdot}2H_2O$, Fluka puriss) was added under stirring. The molar ratio DEA : ZnOAc were 1 : 1 mol. The solution mixture was heated to 60°C for 10 min, and then the water was added (ZnOAc : water = 1 : 2 mol). The final concentration of the solution was 0.4 mol/L. The adding of DEA and water is to keep the solutions stable and clear for a long period. ZnO thin films have been prepared by dip-coating 1.1 mm thick alkali free glass (slide dimensions $60 \times 100$ mm$^2$) at withdrawal speeds in the range 1–10 mm/s and heating the sample directly in a furnace at 600°C for 15 min. The coating process was repeated after cooling down to room temperature in order to increase the thickness of the coatings. All samples have been tempered in a furnace under forming gas flux (95N2/5H2) of 200 l/h to decrease the resistivity. The samples were heated from room temperature to 400°C at a rate of 500°C/h, held at this temperature for 2 h and cooled down to 37°C by convection under forming gas flux. The films were kept at 25°C and humidity 40% RH; all measurements carried out on these films were at the same conditions. The film thickness, t, was measured with a Tencor P10 profilometer, and sheet resistance, R, of the films was measured by linear four-point method. The electrical resistivity, ρ, was determined by the relation $\rho = Rt$. The electrical properties have been measured with the van der Pauw and Hall method (NMR Technologies) with an applied magnetic field of 1.3 T. The optical absorption spectra were measured using a double-beam spectrophotometer (Shimadzu UV-140). An absolute specular reflectance accessory (VW-Geometry) was used for near normal (7°) reflection measurements in the range 200–3000 nm.

## RESULTS AND DISCUSSION

The variation of single layer thickness of ZnO thin film with the withdrawal speeds is plotted in Fig. 1. A non-linear dependence of the film thickness on the dipping speeds is observed. Fig. 1 shows that the layer thickness increases as the speed of dipping increases. Fig.2 shows the thickness of the films as a function of the number of coatings. The thickness of the coatings increases non-linearly with the number of layers.

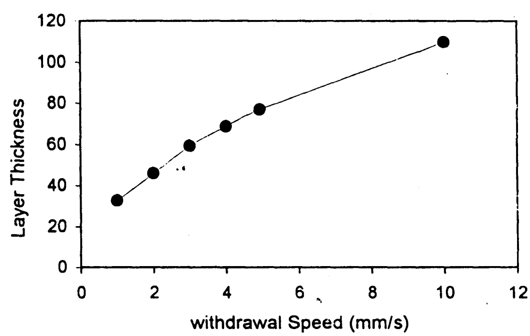


Fig. 1. Thickness dependence of single layer of ZnO thin film at different withdrawal speeds

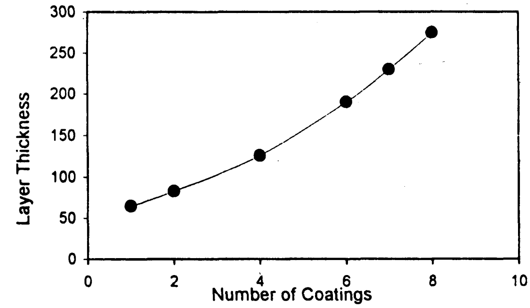


Fig. 2. Thickness dependence of multilayer system of ZnO thin film *vs.* the number of coatings

The electrical properties of single layer coatings obtained with different withdrawal speeds are presented in Fig. 3. The sheet resistance decreases with increasing the speed of dipping, until it reaches to the minimum value of 8.7 MΩ at a speed value of 10 mm/s. This decrease of sheet resistance of single layer is attributed to the presence of columnar grains[19], which enhances the conductivity of the coatings because of easier movement of carriers due to lesser grain boundaries. The electrical properties of multilayers are presented in Fig. 4. The

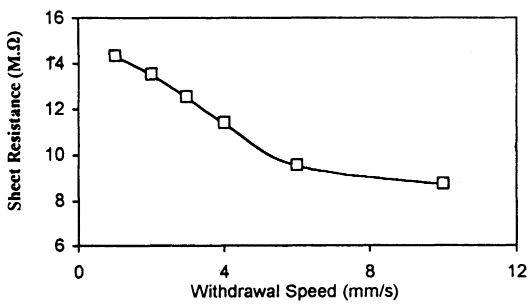


Fig. 3. Sheet resistance of single layer of ZnO thin film at different withdrawal speeds

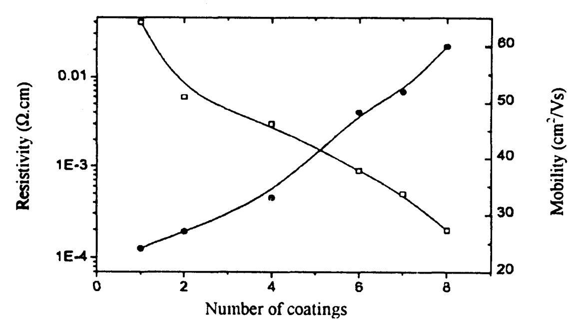


Fig. 4. Electrical properties of multilayer system of ZnO coatings *vs.* the number of coatings

resistivity of the films decreases by a factor of $10^2$ with the number of coatings. This variation is almost essentially due to the strong increase of mobility as the carrier density increases (Fig. 5). Fig. 5 shows the variation of carrier density as a function of the number of coatings. The carrier density increases as the number of coatings is increasing. The strong increase in mobility up to a value of 36

$cm^2/Vs$ as the number of deposited layers increases (Fig. 5) is attributed to higher mean free path and lower scattering of the electrons[20]. From the relation $\rho_{ee} = \frac{1}{ne\mu}$, where $\rho_{ee}$ is the electrical resistivity, n is the carrier concentration and $\mu$ is the mobility. It is evident that the electrical resistivity is decreased mainly by the increase in mobility for the columnar grown sol gel coatings. This increase in mobility is also associated with the observed increase in grain size of ZnO thin films.

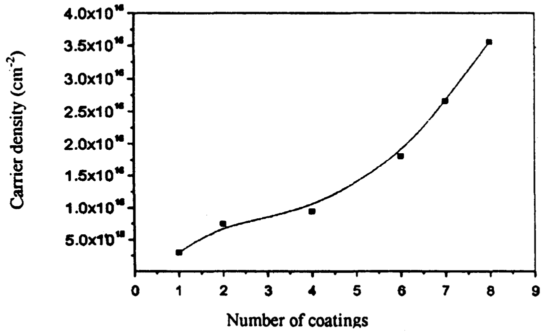


Fig. 5. Variation of carrier density of multilayer system of ZnO coatings with the number of coatings

The optical properties of single layer coatings with different dipping speeds are presented in Fig. 6. It shows the optical transmittance spectra in the wavelength ($\lambda$) range 300–3000 nm for single layer of ZnO thin films with different thicknesses from 37 nm to 109 nm. The optical spectra for all samples giving transparency in the

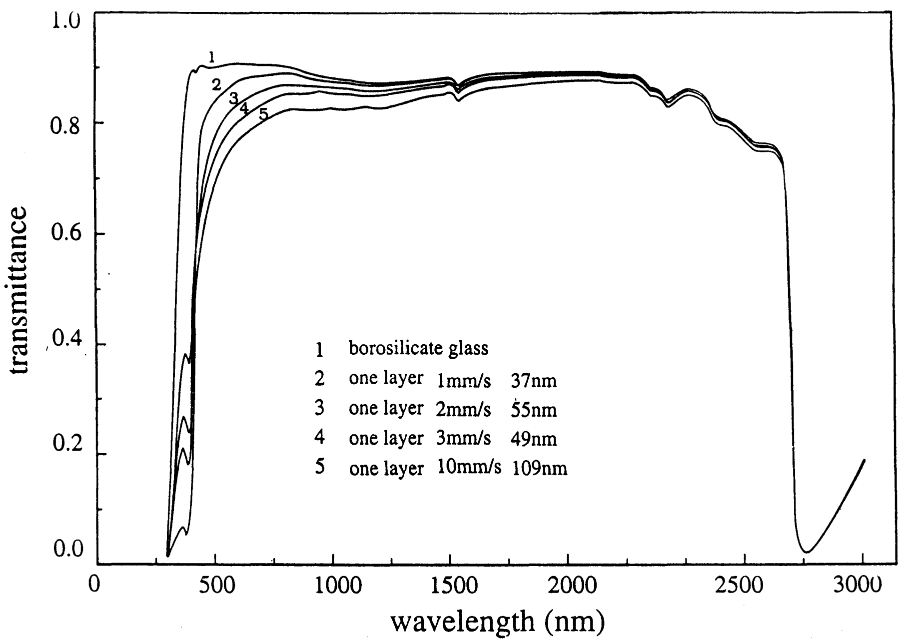


Fig. 6. Optical transmittance spectra for single layer of ZnO thin films at different dipping speeds

different thicknesses from 37 nm to 109 nm. The optical spectra for all samples giving transparency in the visible range above $\lambda = 300$ nm are observed, depending on the dipping speeds of single layer. The transmittance of ZnO single layer is decreased gradually with increasing the dipping speeds. For example, the transmittance has a value of 0.9 for the first layer of dipping speeds 1 mm/s and then decreases nearly to a value of 0.8 for the fourth layer of dipping speed 10 mm/s. Fig. 7 shows the variation of transmittance of single layer of thickness 360 nm with different withdrawal speeds. The transmittance is decreased gradually as

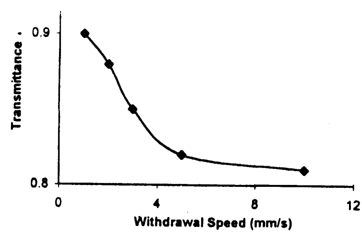


Fig. 7. Transmittance of single layer of ZnO thin film at different dipping speeds

the withdrawal speeds are increased. This decrease in the transmittance in the near IR region is due to an increase in free electron concentration in the film[21]. The optical transmittance spectra of multilayer coatings with different number of coatings are presented in Fig. 8, which shows the variation of the transmittance

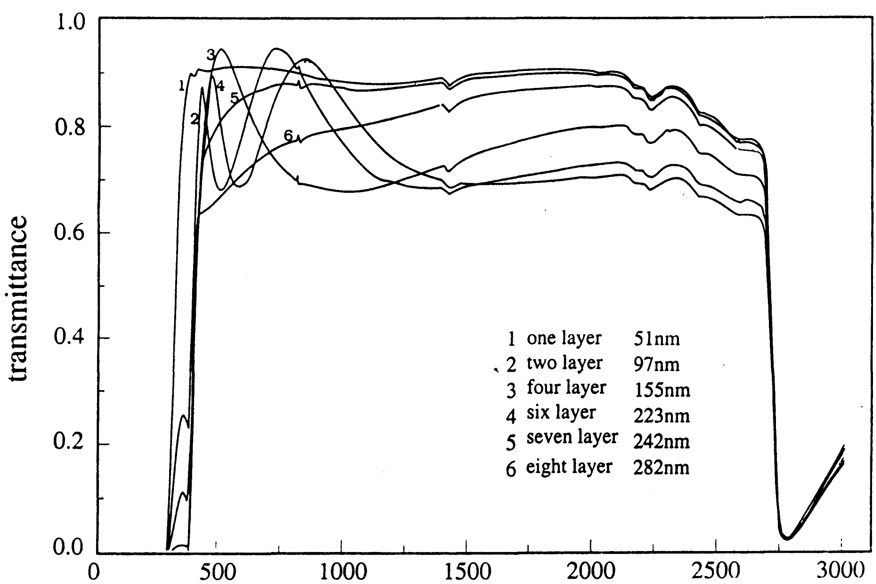


Fig. 8. Optical transmittance spectra for multilayers of ZnO coatings

of multilayer with the number of coatings from one layer of thickness 51 nm to eighth layer of thickness 282 nm. A marked interference pattern is observed for multilayer coatings but not for a single layer coating although they have almost the same thickness. This is due to differences in refractive index and possibly due to differences in surface roughness[21].

## Conclusion

ZnO films were fabricated by the sol gel dip-coating methods; electrical properties as well as its optical properties were investigated. ZnO thin films can serve as transparent conducting materials due to their good electrical and optical properties. The electrical properties of single layer and multilayer coatings have been studied as a function of dipping speeds and the number of coatings. The electrical resistivity of multilayers is decreased as the number of coatings is increased, which enhances the conductivity of thin films. This decrease of resistivity is also associated with observed increase in both mobility and carrier density. The transmittance of ZnO multilayers is slightly decreased with increasing the number of coatings.

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