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The dielectric properties and optical propagation loss of *c*-axis oriented ZnO thin films deposited by sol–gel process

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Abstract

The *c*-axis oriented ZnO thin films were prepared on various substrates by sol–gel processes. The stability of solution was examined through solvent and stabilizer. The *c*-axis orientation and grain size of films were increased with increasing of heat treatment temperature. The optical propogation losses of ZnO films deposited SiO_2/Si(111) substrates were measured using end-coupling method. The losses result in the scattering of the interface of ZnO/SiO_2, and the ZnO grain. Dielectric constant and resistivity of thin films deposited on Pt/SiO_2/Si(111) substrates are, respectively, in the range of 7–13 and $1.7\times10^4\sim9.8\times10^5\Omega$ cm.

Keywords: A. Sol–gel processes; C. Dielectric properties; C. Optical properties; D. ZnO thin film; Orientation

1. Introduction

The physical properties, the low cost and the various fabrication techniques available make zinc oxide a promising material for optoelectronic devices. ZnO is transparent in the 0.4∼2 μm optical wavelength range, it has a fairly high refractive index, it has a fairly piezoelectric, elasto-optic, electro-optic, and nonlinear optical coefficients. ZnO films have been deposited by various techniques such as chemical vapor deposition, spray pyrolysis [1], evaporation [2] and r.f. magnetron sputtering [3,4]. There are many papers on use of doped ZnO films as transparent conductors and optical propagation loss under CO_2 laser treatment [4] while the dielectric properties and optical propagation loss of the insulating films are rarely reported.

A sol–gel process is an attractive technique for obtaining thin films and has the advantages of easy control of the film composition and easy fabrication of a large-area thin film with low cost. Sol–gel derived ZnO thin films have been reported [5–7]. Most of these studies focused on semiconducting ZnO films doped with Al or In.

In the present paper, results concerning orientation of ZnO, optical propagation loss and techniques processes, deposited on SiO_2/S(111), Pt/ SiO_2/S(111) and fused quartz substrates, are reported and discussed.

2. Experimental procedures

The starting material used in the present study was zinc acetate 2-hydrate (99.5% purity, ZnAc). The solvent was 2-propanol. Sol stabilizers was diethanolamine ($HN(CH_2CH_2OH)_2$, DEA). The ZnAc was dispersed into 2-propanol and then the DEA was added. The molar ratios were as following: DEA:ZnAc = 1:1mol. The solution of mixture was heated to 60°C for 10 min, then the water was added (ZnAc:water = 1:2 mol). The final concentrations of the solution were 0.4 mol/l. The adding of DEA and water is to keep the solutions stable and clear for a long period.

Films were coated at a spinning speed 2500 rpm for 30 s. The temperature and humidity of spinning room were kept at 20°C and below 30% RH, respectively. Gel layers were heat-treated at 200°C for 10 min in an oxygen atmosphere. The processes of coating cycle were repeated until obtaining desired thickness. Final films were treated at different temperatures for 30 min in O_2.

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The crystalline phase, crystal orientation, and crystallite size of ZnO films were determined by X-ray diffractometry (Rigaku D/Max-2400). The dielectric constant coated on $Pt/SiO_2/S(111)$ were measured with HP4275.

3. Results and discussion

3.1. Structure and orientation of thin films

Fig. 1 shows X-ray diffraction patterns of the ZnO thin films coated eight times on the $SiO_2/Si(111)$ substrates. The thin film exhibited a strong orientation of the *c*-axis perpendicular to the surface of a substrate. The *c*-axis orientation represented by the (002) reflection. The grain size of ZnO is deduced from X-ray diffraction line broadening using the Scherrer formula. Where any contributions to broadening due to nonuniform stress are neglected. The instrumental linewidth in the XRD apparatus was subtracted. The results are shown in Table 1.

With increasing of heat treatment temperatures, the grain size of ZnO was increased. Fig. 2 is the X-ray diffraction patterns of thin films coated on fused-quartz, $SiO_2/Si(111)$, and the powder of ZnO. The ZnO thin film coated on $Pt/SiO_2/Si(111)$ has also a strong *c*-axis orientation. Fig. 3 is the relations of $I_{002}/(I_{100}+I_{101})$ and heat treatment temperatures. It can be seen that the orientation of thin films is highest at 500°C. The ZnO films grow with a (002) orientation because such growth is kinetically preferred, which in turn likely reflects the fact the highest density of Zn atoms is found along the (002) plane [8].

Fig. 4 shows the change in thickness with the heat treatment temperatures. The film was coated four times. Below 500°C, the thickness continually decreased largely because of the organics reducing and film crystallization. Above 500°C, the thickness continually decreased but the rate reduced.

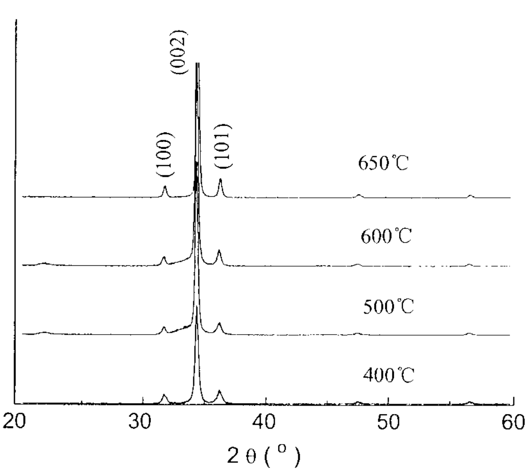

Fig. 1. XRD patterns of ZnO films deposited on $SiO_2/Si(111)$ substrates.

Table 1
Grain size of films vs. heat treatment temperatures

Heat treatment temperatures (°C)	300	400	500	600
Grain size (nm)	16	28	53	86

3.2. Optical propagation losses

The optical propogation losses of ZnO films deposited $SiO_2/Si(111)$ substrates were measured using end-coupling method. The optical propagation loss was verified by measuring the scattered light from the transmitted light beam as a function of the propagation distance [9]. The results of optical propagation losses in films coated 4 times are shown in Table 2. Table 2

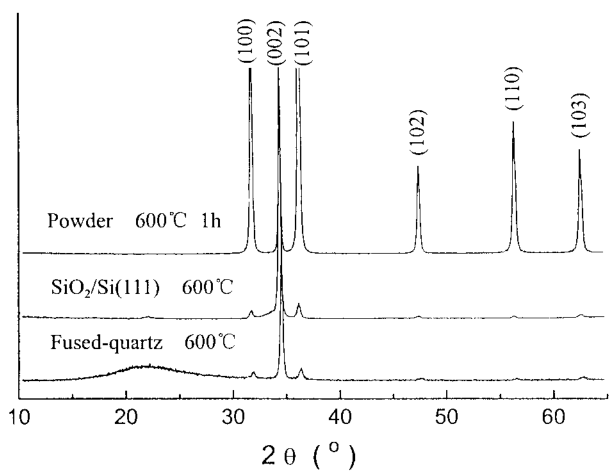

Fig. 2. XRD patterns of ZnO films and powder.

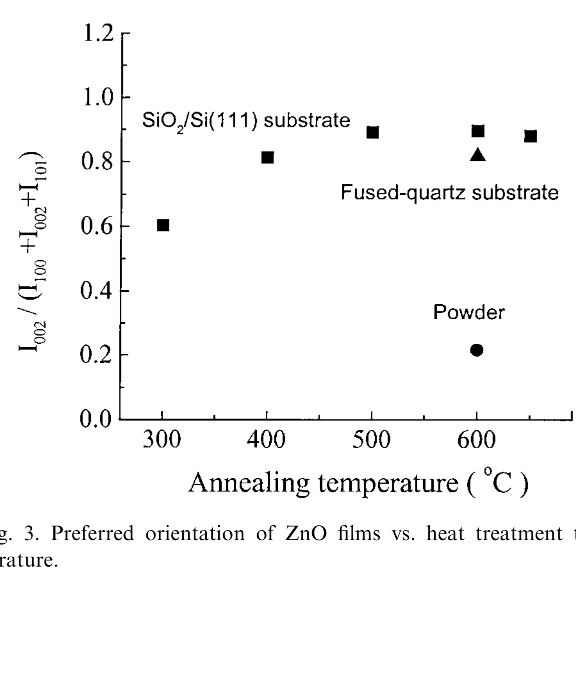

Fig. 3. Preferred orientation of ZnO films vs. heat treatment temperature.

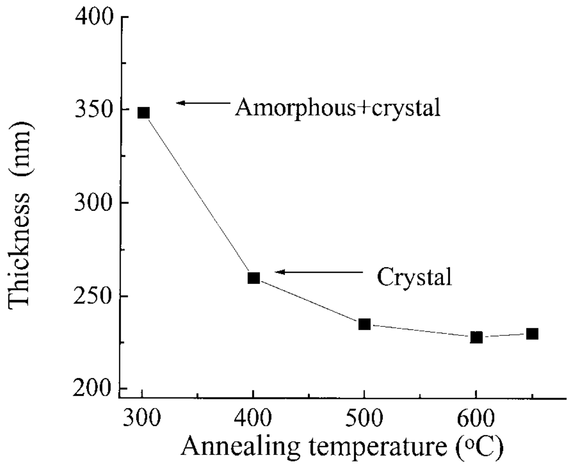

Fig. 4. Change in thickness of films with increasing heat treatment temperature.

Table 2
Optical waveguide losses vs. heat treatment temperature

Heat treatment temperatures (°C)	300	400	500	600
Optical loss (dB/cm)	2.3	3.2	6.8	9.4

shows the optical losses were increased with increasing of heat treatment temperature. Above 500°C, the losses increased rapidly with increasing heat treatment temperature.

The grain caused light scattering can be neglected at lower heat treatment temperature. For instance, about 16 nm at 300°C for 30 min is only 1/40 of the wavelength of light of 632 nm. The grain size of ZnO is about 86 nm at 600°C for 30 min. It is 1/7 of wavelength 632 nm. Therefore, when the treatment temperature is higher, the optical propogration loss is mainly from the grain scattering. Laser annealing of ZnO thin films deposited on SiO_2/Si(111) substrates was carried out by Subhadra Dutta et al. [10]. Both before and after the annealing treatment, showing a significant reduction of the optical propogration losses. The reduction in losses was mainly attributed to an improvement in the quality of the film at the ZnO/SiO_2 interface. At lower heat treatment temperature the propogration losses of sol–gel derived ZnO film was mainly attributed to the interface of ZnO/SiO_2. At higher heat treatment temperature the light scatterings caused by grain is mainly to the losses.

3.3. Dielectric properties

Fig. 5 shows the curves of dielectric constant, resistivity of thin films coated on Pt/SiO_2/Si(111) substrates with the heat treatment temperatures.

The dielectric constant of thin film was increased from 6.4 to 11.8 with heat treatment temperatures. The resistivity of thin films was also increased from 1.7×10^4 to

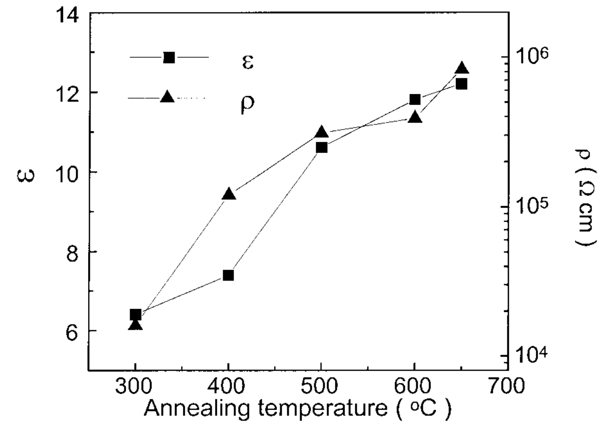

Fig. 5. Dielectric constant and resistivity as a function of heat treatment temperature.

$9.8\times10^5\Omega$ cm. The bigger grain and compact in thin film was attributed to the dielectric constant. ZnO films are known to the *n*-type semiconductors and under more oxidizing conditions, the *n*-type semiconductivity is suppressed due to elimination of resisivity values, leading to an increase of resistivity.

4. Conclusion

ZnO thin films that have a strong *c*-axis orientation were prepared on various substrates by sol–gel process. The *c*-axis orientation and grain size of ZnO films were increased with increasing of heat treatment temperature. The optical propogration losses were increased with increasing of heat treatment temperature. The losses result in the scatterings of the interface of ZnO/SiO_2, and the ZnO grain. Dielectric constant and resistivity of thin films deposited on Pt/SiO_2/Si(111) substrates are, respectively, in the range of 7–13 and 1.7×10^4 to 9.8×10^5 Ω cm.

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