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Structure-composition-property dependence in reactive magnetron sputtered ZnO thin films

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Abstract

Thin films of zinc oxide (ZnO) were prepared by dc reactive magnetron sputtering on glass substrates at various oxygen partial pressures in the range 1×10^{-4} – 6×10^{-3} mbar and substrate temperatures in the range 548–723 K. The variation of cathode potential of zinc target on the oxygen partial pressure was explained in terms of target poisoning effects. The stoichiometry of the films has improved with the increase in the oxygen partial pressure. The films were polycrystalline with wurtzite structure. The films formed at higher substrate temperatures were (002) oriented. The temperature dependence of Hall mobility of the films formed at various substrate temperatures indicated that the grain boundary scattering of charge carriers was predominant electrical conduction mechanism in these films. The optical band gap of the films increased with the increase of substrate temperature. The ZnO films formed under optimized oxygen partial pressure of 1×10^{-3} mbar and substrate temperature of 663 K exhibited low electrical resistivity of $6.9 \times 10^{-2} \Omega \text{cm}$, high visible optical transmittance of 83%, optical band gap of 3.28 eV and a figure of merit of $78 \Omega^{-1} \text{cm}^{-1}$. © 2002 Published by Elsevier Science B.V.

1. Introduction

Thin films of zinc oxide (ZnO) received much attention due to its unique piezoelectric and piezooptic properties made suitable for surface acoustic wave devices [1], low loss optical waveguides and modulators [2] and optoelectronic devices [3,4]. Because of the wide optical band gap, ZnO films have been used as window layers in copper indium diselenide based thin film hetero-

junction solar cells to enhance the short circuit current [5]. Another important advantage of ZnO is its chemical stability in the presence of hydrogen plasma which makes it to be useful in the fabrication of amorphous silicon solar cells by plasma enhanced chemical vapour deposition [6]. Recently it has been recognized as a promising alternative material to transparent conducting indium tin oxide and tin oxide coatings due to its low cost and nontoxicity [7]. Apart from the above, ZnO films have been used in many devices such as gas sensors [8] and high temperature solid lubricants in gas turbine engines [9]. Zinc oxide films have been prepared by various physical vapour deposition

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techniques such as reactive evaporation, activated reactive evaporation, rf/dc sputtering, ion beam sputtering, atomic layer epitaxy etc., as well as a plethora of chemical methods. Among these methods, dc reactive magnetron sputter deposition has gained importance due to the industrial nature of the process.

In this paper, we report a systematic investigation on the deposition of zinc oxide films by dc reactive magnetron sputtering in order to study the structure-composition-property dependence vis-a-vis the process parameters. Electrical and optical properties of zinc oxide films prepared with different oxygen partial pressures and substrate temperatures have been discussed in relation to the composition and structure of the films.

2. Experimental

Zinc oxide films have been deposited on glass substrates using a magnetron sputter deposition system developed in the laboratory. The sputtering cathode is fitted with a 100 mm diameter zinc target (99.99% purity) in sputter down configuration. The sputtering chamber was evacuated to a base pressure of 10^{-6} mbar using diffusion pump with a liquid nitrogen trap. The chamber pressure was monitored using a Pirani-Penning gauge combination. Pure oxygen and argon have been used as reactive and sputter gases respectively. Individual gas flows have been controlled using Tylan mass flow controllers (model FC 260). After reaching the base pressure, oxygen gas was first introduced into the chamber, and the flow rate was adjusted to maintain the required partial pressure. Later argon gas flow was controlled to maintain the sputtering pressure. After each deposition, initially argon gas flow was closed and the partial pressure of oxygen was checked. It was found to be the same as that of set pressure. Also prior to each deposition, the target was sputtered in pure argon environment, with the shutter covering the substrate. This would ensure the same target condition for each deposition. Our earlier studies [10] showed that films with a thickness uniformity of $\pm 2\%$ can be deposited at a target substrate distance of 65 mm. Hence all the films discussed in

Table 1

Deposition conditions of the dc magnetron sputtered ZnO films

Sputtering target	99.995% pure zinc (100 mm diameter and 3 mm thick)
Substrate	glass
Target-substrate distance	65 mm
Base pressure, p_u	2×10^{-6} mbar
Oxygen partial pressure, p_{O_2}	1×10^{-4} – 6×10^{-3} mbar
Substrate temperature, T_s	548–723 K
Sputtering pressure, p_w	6×10^{-2} mbar
Sputtering current, I	200–300 mA
Sputtering voltage, V	280–420 V

the present study have been deposited at this distance. A resistive heater has been used to heat the substrate up to about 723 K. A chromel alumel thermocouple has been used to measure the temperature of the substrate on the top surface. Table 1 shows various deposition parameters used in the preparation of zinc oxide films in the present study. It is well known that during deposition of zinc oxide films re-sputtering due to negative oxygen ions takes place. This effect would be predominant when the deposition is carried out at low pressures. In the present study, in order to reduce re-sputtering effects, the deposition has been carried out at slightly higher pressures of the order of 6×10^{-2} mbar. This would result in reducing the energy of the bombarding species and hence minimization of re-sputtering.

3. Results and discussion

During reactive sputtering an important process parameter that decides the formation of the oxide film on the substrate is the oxygen partial pressure. This depends on the rate of sputtering of metal and is related to the discharge current. Hence optimization of oxygen partial pressure is needed for the deposition of oxide films. In this section first we discuss this optimization process and later the characteristics of the films deposited at optimized parameters.

3.1. Optimization of process parameters

Glow discharge plays a major role on the reactive sputtering process. The variation of glow

discharge parameters like the cathode potential and the discharge current often gives an indication on the nature of the films being deposited. The confirmation on this is obtained by the composition and structure of the deposited films. Hence optimization of process parameters has been carried out by studying these characteristics.

3.1.1. Glow discharge characteristics

Fig. 1 shows the variation of cathode potential of the zinc target as a function of oxygen partial pressure for different discharge currents. It can be seen that for all the discharge currents, the cathode potential decreases with an increase in the oxygen partial pressure and a minimum is observed at a critical oxygen pressure. Any further increase in the oxygen partial pressure results in an increase in the cathode potential. These observations have been consistent with many studies on reactive magnetron sputtering of metals in oxygen and argon environment. These variations have been explained on the basis of target poisoning and secondary electron emission from the oxidized targets. This plot serves as an indication on the target condition, and oxide films are generally deposited at the oxygen partial pressures where the cathode potential steeply reduces reaching a minimum. For a discharge current of 250 mA, the crit-

Fig. 1. Variation of cathode potential on zinc target as a function of oxygen partial pressure.

ical oxygen partial pressure is about 1×10^{-3} mbar. Yet another effect of the oxygen partial pressure is on the deposition rate. It has been observed that the deposition rate is about 15 nm/min when the oxygen partial pressure is lower than 1×10^{-4} mbar and drops to about 10 nm/min at the critical oxygen partial pressure and later remains almost constant. This is again in conformity with the studies on reactive sputter deposition process.

3.1.2. Composition studies

X-ray photoelectron spectroscopic study was performed in the binding energy range of 125–1125 eV. The peaks corresponding to Zn 2p, O 1s and C 1s are observed corresponding to binding energies of 1030, 530 and 285 eV respectively. The appearance of carbon peak is due to surface contamination which disappeared after argon ion bombardment for 5 min [11]. X-ray photoelectron spectra of ZnO films formed at 573 K under various oxygen partial pressures are shown in Fig. 2. The narrow scan of the spectra in the energy range of 1015–1055 eV exhibited two intense peaks of zinc corresponding to Zn 2p_{3/2} and Zn 2p_{1/2}. The position of the peak of zinc as well as oxygen changed when the oxygen partial pressure was increased. The peaks of Zn 2p_{3/2} and O 2s levels were used to calculate the individual atomic concentrations as well as O/Zn ratio. The atomic concentration of zinc and oxygen was calculated using the sensitivity factors [12] as C = 1, O = 2.93 and Zn = 18.92. The atomic concentration of zinc and oxygen at different oxygen partial pressures is given in Table 2. It can be seen that the films formed at a deposition rate of 10 nm/min and an oxygen partial pressure of 1×10^{-3} mbar and above are stoichiometric. This corresponds to the critical oxygen pressure discussed in the previous section.

3.1.3. Structure

The structure of the films has been studied using X-ray diffraction. The films deposited without substrate heating are amorphous and the crystallinity sets in at a deposition temperature of 548 K. The films formed at lower oxygen partial pressures ($< 1 \times 10^{-3}$ mbar) showed the presence of unreacted zinc also along with zinc oxide. However,

Fig. 2. X-ray photoelectron spectra of (a) Zn 2p and (b) O 1s levels of ZnO films formed at different oxygen partial pressures.

Table 2

Chemical composition of ZnO films formed at different oxygen partial pressures

Oxygen partial pressure (mbar)	Atomic concentration (%)		
	Zn 3p _{3/2}	O 1s	O/Zn
5×10^{-4}	54.7	45.3	0.83
8×10^{-4}	51.5	48.5	0.94
1×10^{-3}	50.4	49.6	0.98
5×10^{-3}	50.2	49.8	0.99

higher oxygen partial pressures resulted in single phase zinc oxide. This also confirms that this oxygen partial pressure is critical in depositing single phase zinc oxide films [13]. Hence all further depositions have been carried out at this oxygen partial pressure.

Fig. 3. Electrical resistivity of ZnO films formed at different oxygen partial pressures.

3.1.4. Electrical resistivity

Fig. 3 shows the dependence of electrical resistivity of the films deposited with a discharge current of 250 mA and substrate temperatures of 573 K and at different oxygen partial pressures. The electrical resistivity of the films deposited at an oxygen partial pressure of 3×10^{-4} mbar is $4 \times 10^1 \Omega \text{ cm}$ and reduces to a minimum of $8.2 \Omega \text{ cm}$ at an oxygen partial pressure of 1×10^{-3} mbar and a further increase in the oxygen partial pressure causes an increase in the electrical resistivity. The initial decrease in the electrical resistivity is due to the increase in the oxidation of the growing film and the minimum observed at 1×10^{-3} mbar oxygen partial pressure is due to the formation of stoichiometric zinc oxide film as discussed earlier with reference to composition and structure. However, at higher oxygen partial pressures, the excess oxygen is chemisorbed on the film surface and bound to the defects, thereby causing an increase in the electrical resistivity. Similar phenomena were observed in rf sputtered ZnO films [14]. Thus it can be seen that films with low resistivity can be deposited in a narrow region of oxygen partial pressures. Such a narrow electrical conducting window was also observed by Sato et al. [15] and Ellmer et al. [16], in dc magnetron

sputtered aluminum doped zinc oxide films. Detailed studies on the electrical and optical properties of zinc oxide films prepared at different oxygen partial pressures were reported in an earlier publication [17].

The glow discharge characteristics, structure, composition, and electrical resistivity studies showed that there exists a critical oxygen partial pressure at which stoichiometric oxide films can be deposited and this pressure is dependent on the discharge current. Based on these studies, all further films discussed in the present study have been deposited at an oxygen partial pressure of 1×10^{-3} mbar and a discharge current of 250 mA, corresponding to a deposition rate of about 10 nm/min and various substrate temperatures in the range 548–723 K.

3.2. Substrate temperature effect

The oxygen partial pressure, as shown above plays a major role in depositing stoichiometric oxide films. However, as mentioned in the earlier discussion, the films deposited at ambient temperature are amorphous and hence in order to improve the quality of the films, substrate heating has been resorted to. Under the optimum oxygen partial pressure and deposition rate, zinc oxide films have been deposited at different substrate temperature in the range of 548–723 K. The effect of substrate temperature on the physical properties of the deposited films is discussed below.

3.2.1. Structure

The X-ray diffraction spectra of the films deposited at different substrate temperatures are shown in Fig. 4. All the films are polycrystalline with wurtzite structure. The films formed at a substrate temperature ≥ 573 K showed the presence of single phase zinc oxide with (002) orientation. The films deposited below this temperature were a mixture of metallic zinc and zinc oxide. At higher substrate temperatures the intensity of (002) phase increased. The variation of d -values for (002) plane has been shown in Fig. 5. It can be seen that the d -value increased from 0.2599 to 0.2609 nm with increasing substrate temperature. The d -values for the films deposited at 663 K is

Fig. 4. X-ray diffraction spectra of ZnO films formed at different substrate temperatures.

0.2605 nm which is close to the ASTM standard data (0.2603 nm). This increase in the lattice parameter with temperature can be due to the presence of compressive stresses in the films. Grain size of the films has been calculated using the Scherrer's relation [18]

$$L = 0.9\lambda / \beta \cos \theta \quad (1)$$

where λ is the wavelength of the X-rays, β is the half intensity width of the peak and θ is the diffraction angle. The grain size increased from 20 to

Fig. 5. Variation of d -value of ZnO films as a function of substrate temperature.

45 nm with increase in substrate temperature from 545 to 663 K due to the improvement in crystallinity of the films. It decreased to 38 nm at a substrate temperature of 723 K due to the deterioration in the structure of the films probably due to the re-evaporation of the deposited film. Such a deterioration in the crystallinity was earlier reported by Kim et al. [19] in the case of rf magnetron sputtered zinc oxide films. Zhang and Brodie [20] reported for rf magnetron sputtered films, (002) orientations at 473 K while mixed phase at lower temperatures. Such an observation of c -axis orientation at higher substrate temperatures was also reported by Labeau et al. [21]. Our results are consistent with these reported data.

3.2.2. Electrical resistivity

Fig. 6 shows the variation of electrical resistivity and Hall mobility of the films at different substrate temperatures. The electrical resistivity of the films formed at 548 K was high ($3.4 \times 10^1 \Omega \text{ cm}$) and decreased to a minimum value of $6.9 \times 10^{-2} \Omega \text{ cm}$ at a substrate temperature of 663 K and showed a marginal increase for the films deposited at 723 K. Similar behaviour was observed in the Hall mobility of the films. The Hall mobility increased from 9.2 to $16.8 \text{ cm}^2/\text{V s}$ with increase in substrate temperature from 543 to 663 K and there after decreased to $15.8 \text{ cm}^2/\text{V s}$ at 723 K. The Hall mobility measurements indicated that the films are n-type. The electron density increased from 2×10^{16} to $5.4 \times 10^{18} \text{ cm}^{-3}$ with the increase in

Fig. 6. Electrical resistivity and Hall mobility of ZnO films formed at different substrate temperature.

substrate temperature from 545 to 663 K and later decreased to $1 \times 10^{18} \text{ cm}^{-3}$ for the films deposited at 723 K [22]. At low substrate temperatures, the electrical resistivity of zinc oxide films indicated a quasi-intrinsic behaviour. It is known that the n-type conductivity in undoped nonstoichiometric zinc oxide is due to the oxygen vacancies and interstitial zinc atoms which act as donors, giving rise to higher carrier density when the substrate temperature is increased. In addition, the grain size also increased upto a substrate temperature of 663 K. Therefore, the decrease in the electrical resistivity of the films formed upto 663 K could be associated with a combination of the above two effects. The same factors are responsible for the increase in the electrical resistivity of the films formed at higher substrate temperatures. The electrical resistivity data presented here is in good agreement with that reported by Sato et al. [15] in dc magnetron sputtered zinc oxide films.

The electrical conduction in ZnO may be primarily dominated by electrons generated from O^{2-} vacancies and Zn interstitial atoms. In order to

study the electrical conduction mechanism in these films, the dependence of Hall mobility on the temperature in the range 150–303 K was studied. The temperature dependence of Hall mobility was found to obey the Sato's relation [23]

$$\mu = Lq[1/2\pi m^*kT]^{1/2} \exp[-E_b/kT] \quad (2)$$

where L is the average grain size, q is the electron charge, m^* is the effective mass of the electron (assumed to be $0.35 m_e$), T is the temperature and E_b is the grain boundary potential barrier height. This relation indicated that the plot of $\ln(\mu T^{1/2})$ vs $1/T$ would be a straight line if the grain boundary scattering of the charge carriers is operated in the films. The values of E_b could be calculated from the slope of the above plot. These plots are shown in the Fig. 7. These are linear through out the temperature range of investigation, indicating the existence of grain boundary scattering. The variation of the grain boundary potential barrier height as a function of substrate temperature is shown in the Fig. 8. The potential barrier height decreased from 0.059 to 0.036 eV with increase in the substrate temperature from 548 to 663 K and then showed a marginal increase to 0.040 eV for the films deposited at 723 K. The former case is due to the increase in the Hall mobility and the grain size and decrease of scattering of the charge carriers at the grain boundaries. It was reported that for ion beam sputtered zinc oxide films, the grain boundary potential barrier height sharply decreased from

Fig. 7. Plots of $\ln(\mu T)^{1/2}$ vs $10^3/T$ of ZnO films formed at different substrate temperatures.

Fig. 8. Variation of potential barrier height with substrate temperature of ZnO films.

0.037 to 0.012 eV in the substrate temperature range 300–473 K [24].

3.2.3. Optical properties

Fig. 9 shows the optical transmittance spectra of ZnO films formed at different substrate temperatures. The absorption edge was observed around 380 nm for all the films investigated. The transmittance of the films above the absorption edge

Fig. 9. Optical transmittance spectra of ZnO films formed at different substrate temperatures.

decreased with an increase in substrate temperature. The absorption edge shifts towards higher energy side for the films deposited at higher substrate temperatures. At higher substrate temperatures, the films became polycrystalline and the grain size increased. This results in scattering of light and the coherence between the primary light beam and reflected light between the film boundary is lost. This results in the disappearance of interference fringes and decrease in transmittance [25]. The optical absorption coefficient (α) was evaluated from the optical transmittance data where the reflection losses were taken into consideration. The absorption coefficient around 380 nm was found to increase from 2×10^4 to $3.4 \times 10^4 \text{ cm}^{-1}$ with increase of substrate temperature from 548 to 723 K. These studies indicated that the decrease of optical transmittance and increase of absorption coefficient with the increase in substrate temperature may be due to the presence of oxygen vacancies. The optical band gap evaluated from the transmittance data increased from 3.23 to 3.39 eV with the increase in substrate temperature from 548 to 723 K. The widening of the band gap is related to the increase in carrier concentration. Similar results were reported earlier for nonstoichiometric ZnO films [26,27].

The refractive index of the films was evaluated from the interference of the optical data using the Swanepoel's envelope [28] method. Fig. 10 shows the refractive index of ZnO films formed at a substrate temperature of 663 K. The refractive index of the films decreased from 2.03 to 1.87 with the increase of wavelength from 400 to 1000 nm. The refractive index was also calculated for the films deposited at different substrate temperatures and observed that there was no significant influence of substrate temperature on the refractive index of the films.

The figure of merit (F) of the films was calculated from the electrical resistivity and optical transmittance data using the relation [29]

$$F = -1/\rho \ln T \quad (3)$$

where ρ is the electrical resistivity and T is the transmittance. It was found that the figure of merit of the films increased from 3 to $78 \Omega^{-1} \text{ cm}^{-1}$ as the substrate temperature was increased from 548 to

Fig. 10. Wavelength dependence of refractive index of ZnO films.

663 K and decreased to $30 \Omega^{-1} \text{ cm}^{-1}$ for the films deposited at 723 K.

4. Conclusions

ZnO films were deposited on glass substrates by dc reactive magnetron sputtering of metallic zinc target under various oxygen partial pressures and substrate temperatures. The variation of cathode potential and deposition rate on the oxygen partial pressure was studied. The influence of oxygen partial pressure and substrate temperature on the chemical composition, crystallographic structure, electrical and optical properties of the films was systematically studied. The XPS studies showed that the stoichiometry of the film improved with the increase of the oxygen partial pressure. The XRD studies revealed that the films were polycrystalline with wurtzite structure and at substrate temperatures $\geq 663 \text{ K}$ were (002) oriented. The films formed at a substrate temperature of 573 K showed low electrical resistivity of $8.2 \Omega \text{ cm}$ at an oxygen partial pressure of $1 \times 10^{-3} \text{ mbar}$. The temperature dependence of Hall mobility in the films formed at different substrate temperatures revealed the grain boundary scattering of charge carriers was predominant electrical conduction mechanism in these films. The optical band gap of the films increased from 3.23 to 3.39 eV with the increase of the substrate temperature from 548 to 723 K. The ZnO films prepared at an optimized

oxygen partial pressure of 1×10^{-3} mbar and substrate temperature of 663 K were polycrystalline with wurtzite structure and low electrical resistivity of $6.9 \times 10^{-2} \Omega \text{cm}$, high visible optical transmittance of 83%, optical band gap of 3.28 eV and a figure of merit of $78 \Omega^{-1} \text{cm}^{-1}$.

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