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Study of Electrical, Optical and Structural Properties of Al- Doped ZnO Thin Films on PEN Substrates

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Aluminum-doped zinc oxide (AZO), as one of the most promising transparent conducting oxide (TCO) material, has now been widely used in thin film solar cells. Most of the study of AZO films till date has been done on glass substrates but nowadays there is a growing interest in replacing glass with polymer substrate for the thin-film solar cell technology and many other flexible optoelectronic devices. In this study, AZO thin films were deposited at room temperature by RF magnetron sputtering on flexible substrates from a 3 inch diameter target of 2 wt % Al_2O_3 in zinc oxide. The effect of RF power on the structural, optical and electrical properties of AZO films was investigated by X-ray Diffraction (XRD), Hall measurement, and UV-visible spectrophotometery. The XRD data indicates a preferential c-axis orientation for all the films. All films exhibit high transmittance (> 85%) in visible region. Films deposited at 60 W power exhibit lowest resistivity of 1.2×10^{-3} Ω·cm.

Keywords: Al doped ZnO, RF magnetron sputtering.

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1. INTRODUCTION

Materials such as tin oxide (SnO_2), zinc oxide (ZnO) films are well-known transparent conductive films [1-3]. The advantages of ZnO are its material cost, non-toxicity, high crystallinity and stability in hydrogen plasma, compared with SnO_2. However, undoped ZnO thin films usually have high resistivity due to the low carrier concentration. Various dopants have been used so far to increase the free charge carrier concentration and hence the conductivity. Among the various dopants like Al, In and Ga, Al is commonly used for *n*-type ZnO thin films [4]. Al-doped ZnO films show a wider band gap, higher visible transmittance and lower resistivity as compared to ITO and other doped ZnO materials.

Recently extensive study of AZO thin films has been done on glass substrates and films with high visible transparency and electrical conductivity have been obtained. However, the glass substrate is heavy, brittle and not foldable so one cannot use the glass substrate where flexibility and lightweight is of concern such as smart card, electronic map flat panel display, light emitting diodes, flexible amorphous silicon solar cells, position sensitive detectors, gas sensors, surface acoustic wave devices and other optoelectronic devices [5-12]. Therefore the deposition of AZO thin films on flexible substrates is currently of great commercial and scientific importance. To our knowledge very few papers on AZO thin films deposited on flexible substrates have been published so far. The main challenge to get high transparency and high conductivity on flexible polymer substrates is that one cannot deposit the AZO thin films at high power and at high substrate temperature since organic material has poor thermal and mechanical stability [13-16]. There are several deposition techniques to deposit AZO films such as chemical vapor deposition, spray pyrolysis, pulsed laser deposition, magnetron sputtering and sol-gel [6, 9, 17]. However, most of these techniques need moderate temperature to achieve highly conducting AZO films. Among all, radio-frequency (RF) magnetron sputtering is considered as a favorable deposition technique where low substrate temperature is required [18]. Most of the study of AZO thin film has done on Polyethylene Terephthalate (PET) and Polyether Sulfone (PES) substrates. The aim of this work was to deposit highly conducting and transparent AZO thin films on flexible substrates at room temperature and at very low power. Considering the fact that for achieving AZO films with good electrical and optical properties high substrate temperatures are required, the deposition of this material at such a low temperature on the flexible substrate becomes a big challenge. In this study the deposition of AZO thin films has been done on Polyethylene Naphthalate (PEN) since PEN has high thermal and mechanical stability compared to PET substrates and other flexible substrates. We have characterized the structural, electrical and optical properties of the AZO films on PEN substrates deposited using an RF magnetron sputtering system. Systematic study was conducted to investigate the dependence of RF power on the physical properties of AZO thin films on PEN polymer substrates.

2. EXPERIMENTAL DETAILS

Fig. 1 shows the schematic diagram of the RF magnetron sputtering chamber. AZO thin films were prepared by RF magnetron sputtering from a 3 inch diameter target consisting of 98 wt % ZnO and 2 wt % Al_2O_3. The PEN polymer was used as substrate which was first cleaned in organic solution, rinsed in deionized water and dry blown with N_2 gas before loading into the chamber. The base pressure in the chamber was maintained at 10^{-6} torr. High purity Ar flow of 6 sccm was introduced through mass flow controller.

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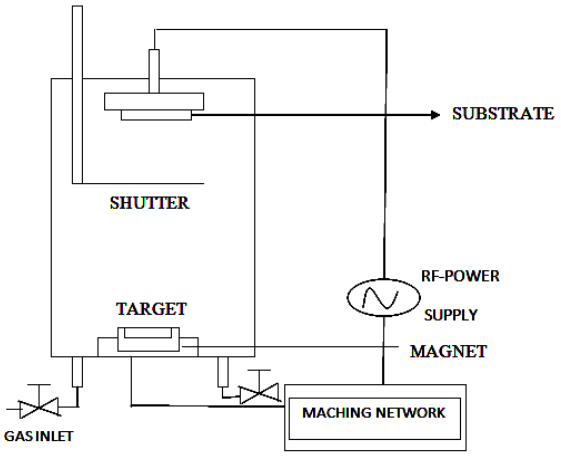

Fig. 1 – Schematic diagram of RF magnetron sputtering chamber

RF power was varied from 40 W to 100 W at 5 mtorr chamber pressure. The distance between the target and the substrate was kept constant at 50 mm.

Table 1 shows the process parameters for the AZO film deposition. All films have approximately same thickness of 200 nm as measured by Dektek Profilometer. Structural, electronic and optical properties were studied by XRD, Hall Effect and UV-VIS spectroscopy respectively. To identify the crystalline structure in the films an X-ray diffractometer with CuK_α radiation ($\lambda = 0.154$ nm) was used. The electrical resistivity, mobility and charge carrier concentration were measured by Hall Measurement. The optical transmittance of the films was measured with an ultraviolet-visible spectrometer.

Table 1 – Various deposition parameters for the AZO films

Process parameters	Value
Target specification	98 wt % ZnO and 2 wt % Al_2O_3
Substrate used	Polyethylene Naphthalate (PEN)
Target to substrate distance	50 mm
Substrate temperature	Room temperature
Power	40-100 W
Gas flow rate (Ar)	6 sccm
Deposition pressure	5 mtorr

3. RESULTS AND DISCUSSION

AZO films presented in this study were physically stable and showed good adherence to the polymeric substrate. No cracks and peel-offs of layers were observed after the deposition. However, the layers deposited at greater than 60 W power show cracks in the films when characterized by Scanning Electron Microscopy.

The film thicknesses were estimated in the range of 200 nm in all the analysed samples measured by Dektek profilometer. Fig. 2 shows the XRD data for AZO thin films deposited on PEN substrate at different powers. All AZO thin films show strong diffraction peak of (002) at 2θ location of ~ 34.2° independent of the RF power. No Al_2O_3 phase was detected in the

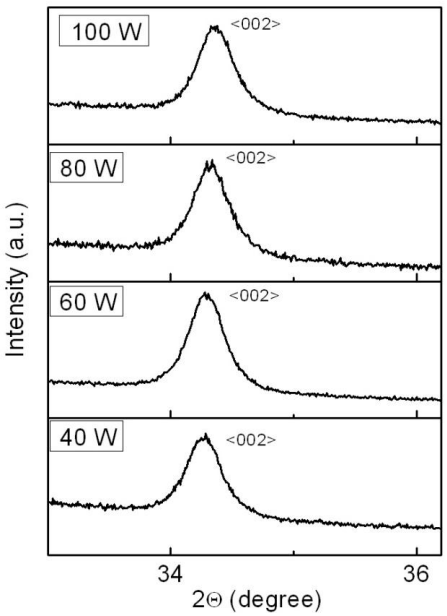

Fig. 2 – XRD curves for AZO thin films deposited on PEN substrate at different powers

XRD patterns. This may be due to aluminum replacing zinc substitutionally in the hexagonal lattice. The average grain size was calculated using Scherrer formula.

$$D = \frac{0.9\lambda}{B\cos\theta}$$

where λ is the wavelength of X-ray, B is FWHM of the peak and θ is Bragg diffraction angle. The corresponding full width at half maximum (FWHM) of the (002) diffraction peak ranged from 0.271 to 0.361. According to the results obtained from FWHM, the estimated grain size ranged from 22 to 20 nm when the RF power increased from 40 to 100 W. Hence, in spite of the low substrate temperature used in the deposition on PEN, the structural quality of the AZO films achieved was close to the one obtained in films deposited on glass at higher substrate temperature.

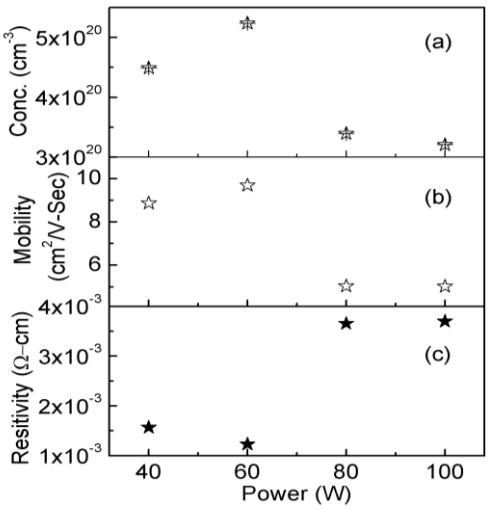

Fig. 3 – Dependence of carrier concentration (a); Hall mobility (b); and resistivity of AZO thin films on RF power (c)

Fig. 3 shows the electrical properties as a function of RF power. Initially, when the RF power increases from 40 to 60 W the resistivity decreases and reaches a value of $1.2 \times 10^{-3}\,\Omega$·cm. Resistivity increases sharply when we increases the power further. The rapid rise in the electrical resistivity of AZO films could be due to the crack formation in the AZO films as shown in Fig. 4, which terminates the film continuity and hence the conduction path for free carrier. As expected, the charge carrier concentration ($5.24 \times 10^{20}\,\text{cm}^{-3}$) and mobility ($9.69\,\text{cm}^2/\text{Vs}$) show high values at the same RF power.

Fig. 4 shows the SEM micrographs of AZO thin films deposited on PEN substrates at different power. It is clearly seen in these pictures that as the RF power increases the cracks in the film start appearing. The main reason for the crack formation in the film could be the stresses in AZO film on flexible substrates. As the RF power increases, more stresses are generated in the film that leads to the cracks. These cracks increased the resistivity of the film significantly. Also, the mobility of charge carriers reduced for the films that are deposited at greater than 60 W. Due to the crack formation the film is not continuous and the film continuity is terminated and hence the conduction path for the charge carrier is also broken. This reduces the mobility of charge carriers. The transmission spectra of all the films are shown in the Fig. 5. It is clearly seen from the figure that as the power increases the transmission in the visible range decreases. This could be due to the large scattering taking place at the cracks in the film.

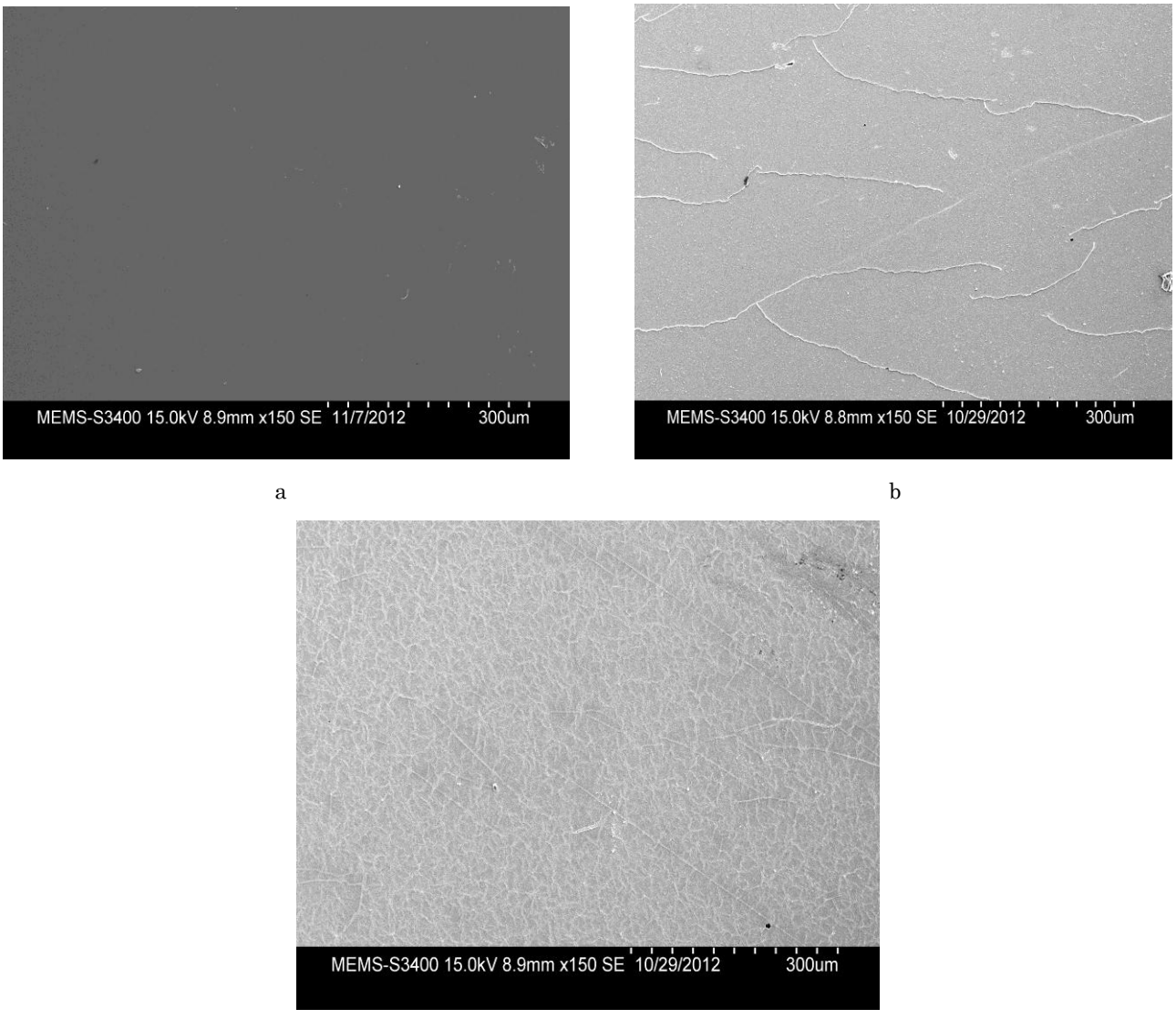

Fig. 4 – SEM micrograph of AZO films grown on PEN substrates at different power: 60 W (a); 80 W (b) and 100 W (c)

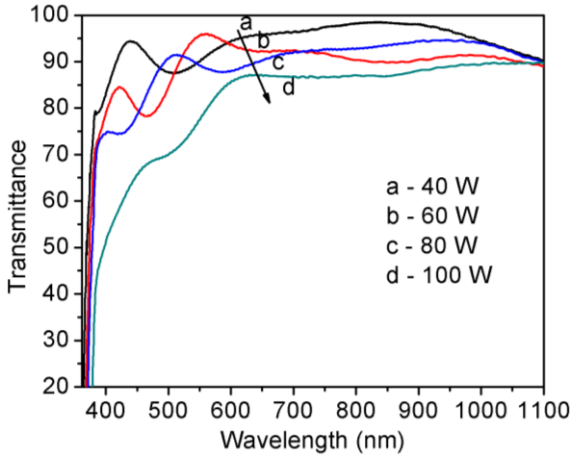

Fig. 5 – Optical transmission for AZO films as a function of RF power

4. CONCLUSION

Transparent conducting Al-doped zinc oxide films were fabricated on PEN substrates using RF magnetron sputtering at room temperature. The RF power dependence of the structural, morphological, electrical and optical properties of the films was studied. All the AZO films deposited on the PEN substrates show strong c-axis (002) preferred orientation. The minimum electrical resistivity of $1.2 \times 10^{-3}\,\Omega$·cm was obtained for the film deposited at 60 W RF power at room temperature.

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