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Effect of Substrate Temperature on Structural, Optical and Electrical Properties of AZO Thin Films by RF Magnetron Sputtering for Optoelectronic Devices

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Abstract: Aluminium Zinc Oxide (AZO) thin films have been deposited on glass substrates by RF magnetron sputtering method at different substrate temperatures from RT to 400°C. The structural, optical, electrical properties of AZO thin films deposited with various substrate temperatures were investigated by X-ray diffractometer (XRD), Scanning electron microscope (SEM), UV-Vis spectrophotometer and Hall effect measurement system. X-Ray diffraction pattern of AZO films shows (0 0 2) with a hexagonal wurtzite structure preferentially oriented along C-axis. The grain size increases from 0.6 to 1.44µm with increase of substrate temperature. The average transmittance of AZO films about 84% in the UV-Visible region and sharp edge occurs at 380 nm. The band gap increases with increase of substrate temperature from 3.34 to 3.46 eV and this may be attributed to Burstein-Moss shift. The lowest resistivity of 5.37 X 10^{-4} Ω.cm, is obtained for the thin films deposited at 400°C, these results are suitable for optoelectronic applications such as transparent electrodes for solar cells, flat panel displays and organic light-emitting diodes.
Keywords: AZO thin films, RF sputtering, Structural, optical and electrical properties.

I. INTRODUCTION

Transparent conducting oxide (TCO) films have been widely used in optoelectronic devices, such as solar cells, and liquid crystal displays, heat mirrors and multiple layer photothermal conversion systems [1–3]. Zinc Oxide (ZnO) thin films have been appeared to be a promising material for applications in optoelectronic devices because of their high transparency for visible light, high electron mobility, low resistivity, low cost, non toxicity, high exciton binding energy of 60 meV, wide band gap energy of 3.3 eV, relatively low deposition temperature and high stability and easy to fabricate [4,5].The n-type dopants used in ZnO are mainly In^{3+}, Al^{3+}, B^{3+}, Ga^{3+} [6-9]. Among all these dopants Al is relatively a cheaper, abundant and non-toxic material hence Al-doped ZnO films can be prominent, low cost substitute for high cost Tin doped In_2O^3 (ITO) films in all TCO applications. Aluminium doped Zinc Oxide (AZO) thin films can be produced by various deposition techniques including chemical vapor deposition [10], sol-gel process [11], spray pyrolysis [12], evaporation [13], and RF magnetron sputtering [14]. Among all these techniques, sputtering can be considered as one of most promising techniques, as this technique allows uniform films with good adhesion, high deposition rate and large-area deposition, and low cost of equipment for commercialization of all TCO films. In this paper, we studied the effect of substrate temperature on the structural, optical and electrical properties of AZO thin films deposited on glass substrate by Rf magnetron sputtering.

II. EXPERIMENTAL DETAILS

PLASSYS 2000 deposition system was used to deposit AZO thin films on glass substrate by Rf magnetron sputtering. High purity of ZnO (99.99%) ceramic and metallic Al (99.99%) target with 2 inch diameter and 4mm thickness are used. Gas mixture of Ar (99.99%) and O_2(99.99%) with Ar/O_2 ratio of 8:3 Sccm was introduced into chamber by mass flow controllers. Glass substrate was placed parallel to target surface with a distance about 60 mm with controlled substrate rotation speed set 30 rotations per minute (rpm). The glass substrates were ultrasonically cleaned in detergent diluted in deionized water finally ultrasonic bath in isopropyl alcohol and dried in hot air flow then mounted on the sample holder. Before thin film deposition, pre sputtering was done for 10

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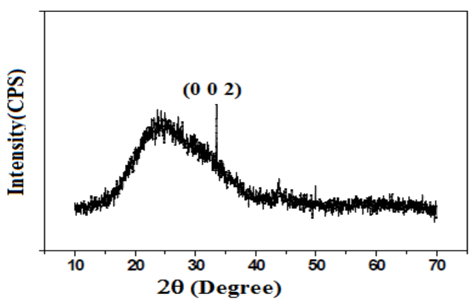
minutes to avoid the contamination on the surface of the target. The chamber was evacuated with the help of turbo molecular pump in the vacuum of $4x10^{-6}$ mbar and the sputtering pressure was $7.4x10^{-3}$mTorr.

The depositions were carried out from room temperature (RT) to 400°C and deposition time is 30mins, while radio frequency was 13.56MHz. The resulting thickness of all samples was about 250-300nm and was determined by using Stylus profile meter. The crystal structure of AZO films was characterized by X-ray diffraction (XRD), CuKα radiation. The surface morphologies were observed by field emission electron microscope (FESEM). The optical transmittance measurements were recorded as a function of wavelength in the range of 300–1200nm using UV-VIS spectrophotometer (JASCO,V-670).The electrical properties of AZO films were investigated from Hall Effect measurements system (Ecopia, HMS7000) using vander pauw geometry, at room temperature.

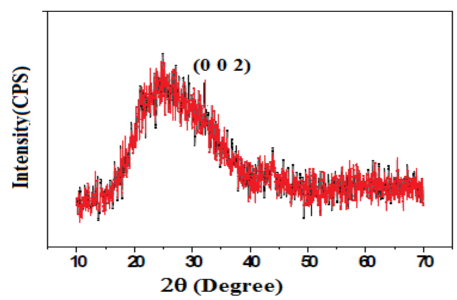
III. RESULTS AND DISCUSSIONS

A. Structural Properties

XRD spectra results that the deposited films exhibit preferential growth along C-axis of hexagonal structure. The (0 0 2) preferred orientation is due to the lowest surface free energy, which corresponds to the densest packed plane [15].The internal stresses were primarily compressive in the present case of AZO films deposited on glass substrate. It is thought that the "atomistic peening" effect caused by the bombardment of energetic species during deposition is the cause of the internal compressive stresses in the films produced using the sputtering method [16].It is observed the peak intensities are nearly identical and extremely weak. The amorphous nature of the glass substrate results in poor crystallinity in the AZO films that are produced on it. As a result, the atomistic peening effect is considered to occur weakly, which means the stress difference caused by the variation in pressure is very small.



(a)



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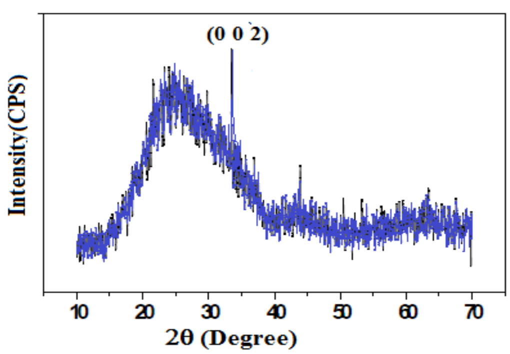

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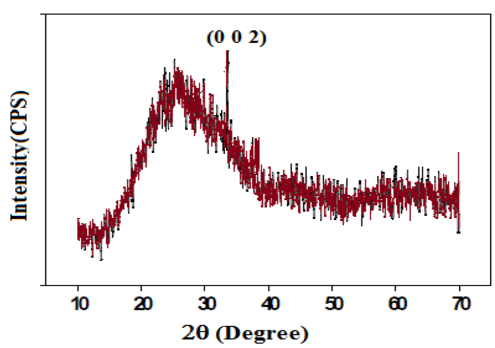
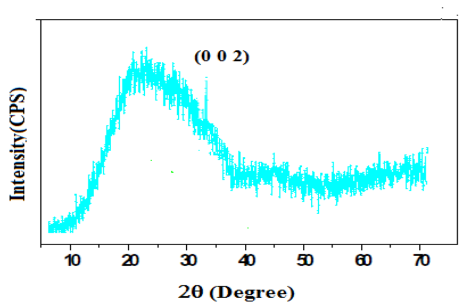


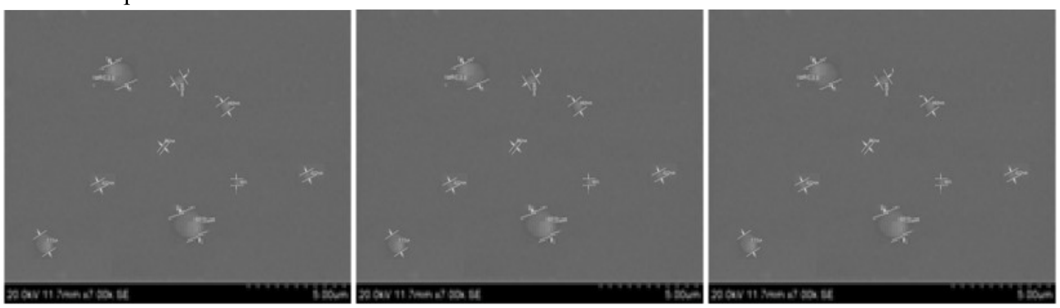
(d)



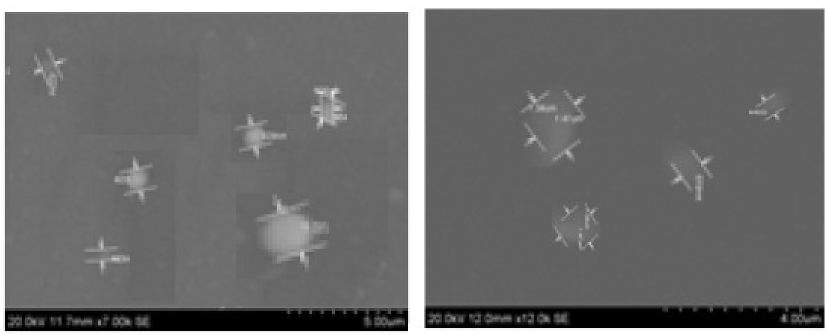
(e)

Fig.1. XRD Patterns of AZO films at different substrate temperatures (a) Room temperature (RT) (b) 100ºC (c) 200ºC (d) 300ºC (e) 400ºC

Figure 2(a)-(e) shows the SEM images of AZO films under various substrate temperatures from RT to 400°C. It is observed that SEM images of AZO films have spherical grains and grain size increases from 0.6 to 1.44μm with increase of substrate temperature. Higher substrate temperature gives the energy for surface atoms to increase mobility, which can improve the crystallinity of films. Although mobility was improved by higher substrate temperatures, they also led to the re-evaporation of poorly combined structures that made the structures irregular. As a result of this was irregular in surface morphology of films and larger grains were produced with increased substrate temperature.



(a) (b) (c)



(d) (e)

Fig.2. SEM images of AZO thin films deposited at the substrate temperatures of (a) Room temperature (RT) (b) 100ºC (c) 200ºC (d) 300ºC and (e) 350ºC.

B. Optical Properties

The optical transmittance spectra of Aluminum doped Zinc Oxide (AZO) films deposited on glass substrates from room temperature (RT) to 400ºC is as shown in figure 3(a)&3(b). The optical transmittance is increased with increasing of substrate temperature due to the weakening of scattering and absorption of light because of the increase in grain size. It was observed that the all films exhibit highly transparent about 84% in the UV-Visible region wavelength between 365nm-1200nm.The sharp UV absorption edge occurs near 380nm.

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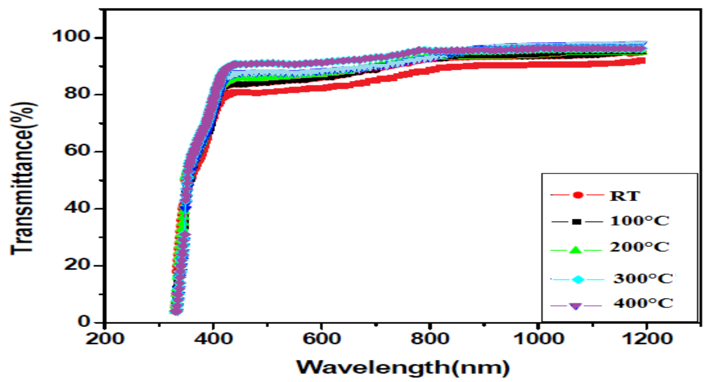

Fig.3(a). Optical transmittance spectra of AZO as a function of wavelength at different substrate temperatures

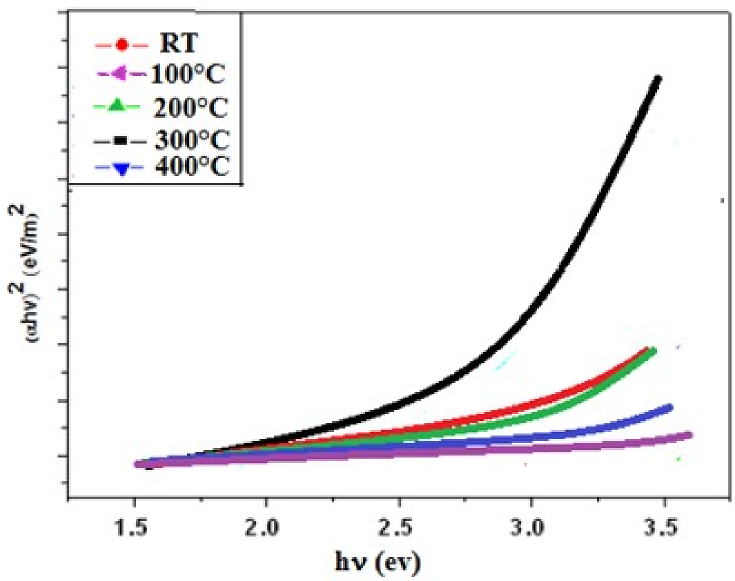

Fig.3(b). Plots of $(\alpha h\nu)^2$ versus photon energy hν of ZAO thin films with various substrate temperatures

The optical parameters such as absorption coefficient (α), extinction coefficient (K), refractive index (n) can be determined from the transmission data. The refractive index (n), extinction coefficient (k) and absorption coefficient (α) values were calculated from the following equations [17].

$$n = \frac{1+R}{1-R} + \sqrt{\left(\frac{4R}{(1-R)^2}\right) - K^2} \qquad (1)$$

Where R is reflectance, K is extinction coefficient and can be calculated from equation,

$$K = \frac{\alpha\lambda}{4\pi} \qquad (2)$$

Where α is the absorption coefficient and is obtained from relation

$$\alpha = \frac{1}{d}\ln\left(\frac{1}{T}\right) \qquad (3)$$

Where'd' is thickness of film and T is the transmittance.

The absorption coefficient (α) is found to be in order of 10^6 cm^{-1} at shorter wavelength regions. For higher values of photon energy, the energy dependence of absorption coefficient ($\alpha > 10^4$ cm^{-1}) suggests the occurrence of direct electron transitions. The optical band gap of ZAO films was evaluated from the plots of $(\alpha h\nu)^2$ versus hν as shown in Fig 3(b). The band gap increases with increase of substrate temperature from 3.34 to 3.46 eV. The observed widening of band gap was caused by increase in carrier concentration after doping with Al^{3+} ions, which is attributed to Burstein –Moss effect [17].

C. Electrical Properties

The electrical resistivity of AZO films was investigated by Hall effect measurement system at room temperature. The resistivity of AZO films deposited at different substrate temperatures is shown in figure 4. The resistivity decreases with increase of substrate temperature. The obtained resistivity values are varied from 2.2 X 10^{-3} Ω.cm to 7.93 X 10^{-4} Ω.cm. The thin films deposited at 300°C have the lowest resistivity, 5.37 X 10^{-4} Ω.cm, and at 400°C, it slightly increases to 7.93 X 10^{-4} Ω.cm. Because Al^{3+} ions are present at substitutional sites $Zn^{2+,}$ the conductivity of AZO thin films is superior to that of pure ZnO films. As substrate temperature increases, resistivity decreases because film crystallinity, carrier concentration, and carrier mobility all increase. The resistivity will decrease as the crystal orientation increases. This is because there is less carrier dispersion at grain boundaries and crystal imperfections. Low resistivity of AZO films with high transparency can be used as transparent electrodes in optoelectronic displays [17].

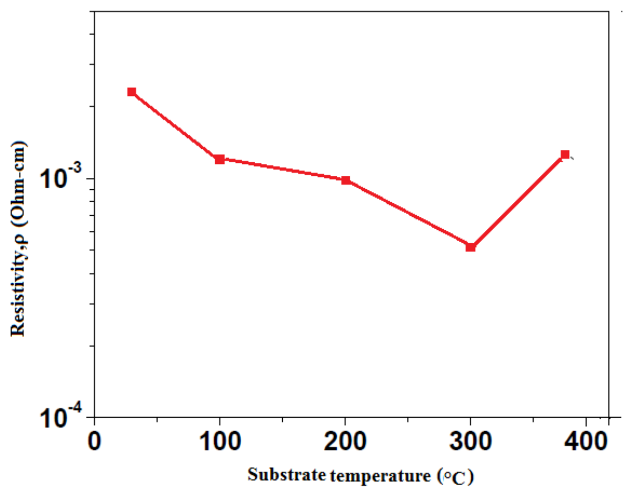

Fig.4. Variation of electrical resistivity as a function of substrate temperature

IV. CONCLUSION

AZO thin films have been deposited on glass substrates by RF magnetron sputtering technique under various substrate temperatures from room temperature to 400ºC. All deposited films are oriented along the c-axis of the hexagonal structure. . It was observed that the all films exhibit highly transparent about 84% in the UV-Visible region wavelength between 365nm-1200nm.The sharp UV absorption edge occurs near 400nm. The film resistivity decreases with increase of substrate temperature upto 300ºC and then slightly increases at 400ºC. The minimum resistivity of 5.37 X 10^{-4} Ω.cm was obtained 300ºC .From these results we conclude that AZO films are promising for optoelectronic applications such as transparent electrodes for solar cells, flat panel displays and organic light-emitting diodes.

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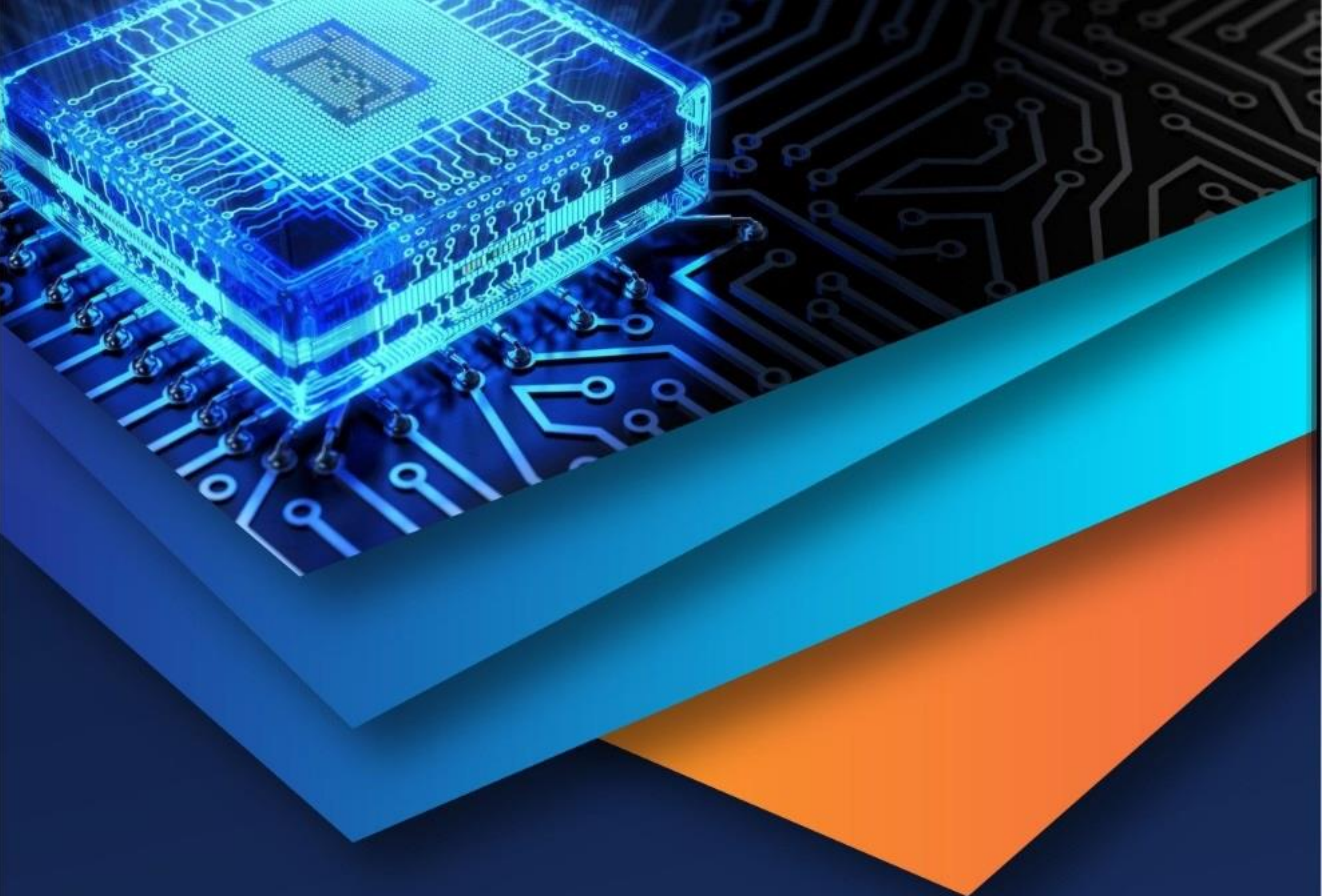

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