

## The Crystallization and Physical Properties of Al-doped ZnO Films

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**Abstract.** In this paper, Al-doped ZnO (AZO) films have been deposited by RF sputtering at room temperature with thickness 300 nm from an Aluminium Zinc Oxide target in Argon 99.99% ambient and without substrate heating with RF power of 300 watt. After deposition, the samples have been annealed at temperatures ranging from 300 to 500 ° C in vacuum for 1h. The effect of flow rate of the Argon gas and annealing, on the Structural properties and grain size have been investigated by XRD using Cu(K $\alpha$ 1) radiation ( $\lambda= 0.154056$  nm) in  $\theta$ -2 $\theta$  scan mode. It is shown that the structural properties and grain size of the AZO thin films are dependent on the flow rate of the Argon gas. The experimental results show that good qualities of crystalline structure appeared when AZO film prepared by flow rate of Argon gas 10 SCCM. But the effect of annealing on the structural properties of the AZO thin films is low. A high degree of preferred orientation which depends on the various flow rate of argon gas is exhibited. The  $\theta$ -2 $\theta$  scan data of AZO films exhibit a strong 2 $\theta$  peaks at nearby 34.53°, corresponding to the (002) peaks of AZO. It is indicated that the film is a crystalline with a preferential orientation along c-axis. There is a 2% variation in 2 $\theta$  peaks of AZO films. ( i.e. 34.53°  $\pm$  2%) have been prepared at various flow rate and annealing temperature.

**Keywords:** AZO, sputtering, annealing, thin film, ZnO.

### 1. Introduction

Recently, AZO has been extensively studied because they combine attractive properties with high visible transparent and electrical conductivity and this material is excellent alternative to replace indium tin oxide (ITO) and tin oxide (SnO<sub>2</sub>) as transparent conducting electrodes in solar cells device and flat panel displays [1,2]. In addition, AZO is also potentially useful in surface acoustic devices (SAW) [3] and optical waveguides [4]. Properties and, therefore, device performance are also expected to improve drastically by using single-crystalline or high c-axis orientation film in place of no oriented polycrystalline films. Specially, AZO thin films with high c-axis orientated crystalline structure along (002) plane can reduce the electrical resistivity due to an increase in carriers mobility by reducing the probability of the scattering of the carriers at the grain boundary [5]. Generally, c-axis orientation films of AZO are fabricated by RF magnetron sputtering [6], chemical vapour deposition [7], pulsed laser deposition [8], spray pyrolysis [9] and sol-gel process [10], etc. At present RF sputtering systems are commonly used for AZO deposition in acoustic applications.

The characteristics of AZO films are generally affected by the preparation conditions, such as the deposition methods, working pressure, substrate temperature and the types of substrates. In this study, AZO thin films were deposited by RF sputtering from aluminium doped zinc oxide target at pure argon atmosphere. The influence of sputtering conditions (mainly the argon flow rate) on the properties of AZO films was studied.

AZO films were deposited by an RF sputtering system using AZO (99.99% purity) target with a diameter and a thickness of 76 and 3 mm, respectively. The sputtering was carried out in an argon (99.99% purity) gas atmosphere by supplying 300 W RF power at a frequency of 13.56 MHz. The flow rate of the argon gas was set between 5 to 30 SCCM. The distance between target and substrate was about 80 mm. The chamber was down to 8\*10<sup>-6</sup> Torr using a diffusion pump before introducing the argon sputtering gas into the chamber.

The AZO film was grown at room temperature at a pressure of  $3.3 \times 10^{-2}$  Torr. The physical structure of the films (crystalline structure or microstructure) were analysed by XRD using CuK $\alpha$ 1 radiation ( $\lambda = 0.154056$  nm) in  $\theta$ - $2\theta$  scan mode.

## 2. XRD analysing

### 2.1. Investigation of argon gas flow rate on structural properties

The crystalline structure and orientation of the AZO thin films were investigated by XRD. Fig.1 shows the XRD spectra of AZO thin films deposited at room temperature with an argon flow rate between 5 to 30 SCCM. The films exhibited a high degree of preferred orientation which depends on the various argon flow rate. The  $\theta$ - $2\theta$  scan data of AZO films exhibit a strong  $2\theta$  peaks at  $34.53^\circ$ , corresponding to the (002) peaks of AZO. C-Axis (002) diffraction peaks were distinguishably observed in the grown AZO films, we surmise that the c-axis oriented AZO film is obtained. It is noteworthy that C-Axis (002) diffraction peak of AZO thin film deposited with setting of argon gas flow rate on 10 SCCM, has highest intensity between all samples.

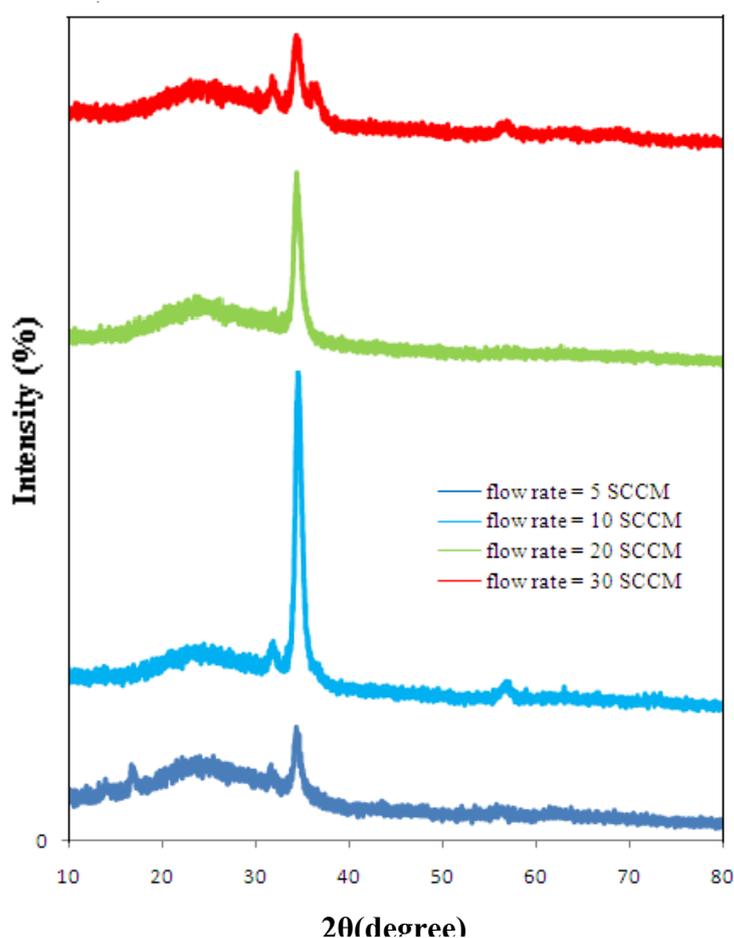


Fig. 1: XRD patterns of Al-doped ZnO thin films deposited on glass substrate with various argon gas flow rate.

### 2.2. Low shift in the position of ZnO (002) pick

A high degree of preferred orientation which depends on the various flow rate of argon gas is exhibited. The  $\theta$ - $2\theta$  scan data of AZO films exhibit a strong  $2\theta$  peaks at nearby  $34.53^\circ$ , corresponding to the (002) peaks of AZO. It is indicated that the film is a crystalline with a preferential orientation along c-axis.

But accurate investigation indicated that there is a variation in  $2\theta$  value of c-axis, resulting from change in amount aluminum in thin film. Because the ionic radii of Zn $^{+2}$  and Al $^{+3}$  are 72 pm and 53 pm, the lengths of the c-axis are expected to shorten about 1.1% and 1.8%, respectively, if all the Al atoms are substituted into Zn sites in the crystal. Thus the position of the ZnO (002) peak is expected to shift  $\sim 0.5^\circ$ .

Fig. 2 represented variation of  $2\theta$  peak as a function of the gas flow deposition and the annealing temperature in vacuum. From Fig. 2 can be seen that in various annealing temperature the respective maximum location of  $2\theta$  peak is achieved at gas flow 20 SCCM.

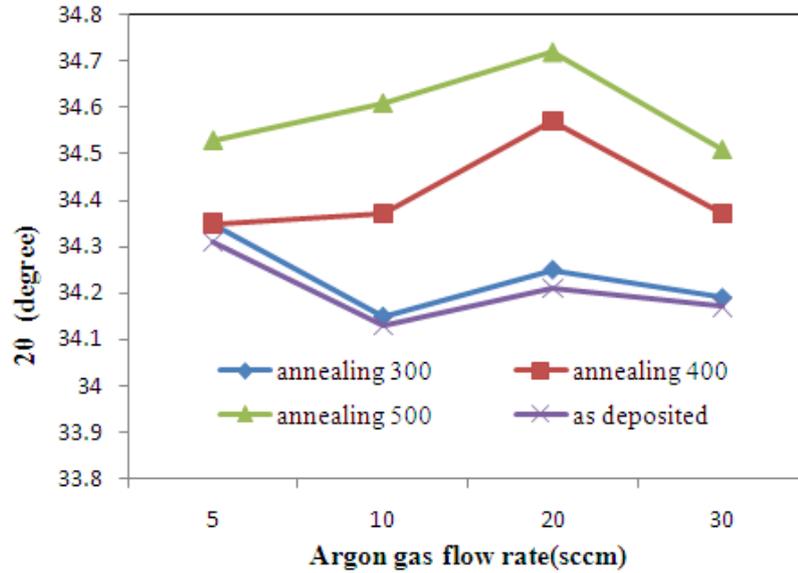


Fig 2: The location of  $2\theta$  peak for AZO thin films deposited on glass substrate in various argon gas flow rate within 5 to 30 SCCM and annealing at  $300^{\circ}\text{C}$ ,  $400^{\circ}\text{C}$  and  $500^{\circ}\text{C}$  for 1h in vacuum pressure in  $10^{-5}$  Torr

### 2.3. FWHM and grain size

XRD spectra show that the AZO films deposited by RF sputtering, have a polycrystalline hexagonal wurtzite structure and are strongly oriented with the (002) faces perpendicular to the substrate surface ( $c$ -axis orientation). We investigated the FWHM of the (002) peak of the AZO thin films, and calculated grain size of them. The grain size can be deduced from x-ray diffraction by

$$t = \frac{0.9\lambda}{\Delta\theta * \cos\theta_B}$$

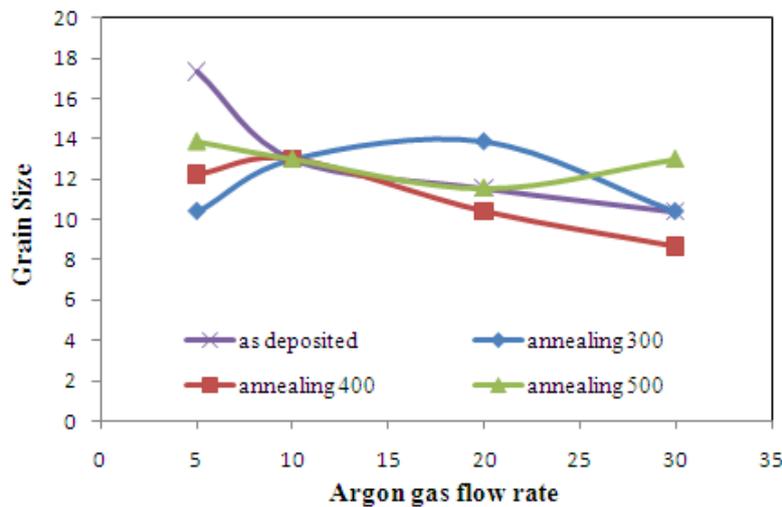


Fig. 3: Grain size of the AZO films have been prepared at different gas flow and various annealing temperature

where  $t$  is the grain size,  $\Delta\theta$  is the FWHM, and  $\theta_B$  is the Bragg angle, and  $\lambda=1.5418 \text{ \AA}$  (X-ray wave length). According to the XRD spectra, FWHM of samples deposited with different argon gas flow rate, can be calculated. The grain size has been measured for the AZO films with different gas flow and is represented in Fig. 3 as a function of the annealing temperature in vacuum. For the as-grown samples, the grain size is in

the 10-18 nm range with a tendency to decrease when the gas flow increases. When the annealing is performed in vacuum, the grain size becomes 10-12 nm at 300C, decreased to 8-12 at 400 C, and increased to 10-18 at 500C.

### 3. Conclusion

In summary, the structure of the AZO films with various argon gas flow rate and annealing temperature in vacuum on crystalline structure of AZO thin films deposited by RF sputtering system were investigated. The structural characteristics reveal that flat and well defined columnar films with C-Axis (002) diffraction peaks were formed. It is noteworthy that C-Axis (002) diffraction peak of AZO thin film deposited with setting of argon gas flow rate on 10 sccm, has highest intensity between all samples. It seems that deposition with higher flow rate of argon gas, ions have lower opportunity deposition on the c-axis oriented. Accurate investigation indicated that there is a variation in  $2\theta$  value of c-axis, resulting from change in amount aluminum in thin film. Thus the position of the ZnO (002) peak is expected to shift  $\sim 0.5^\circ$ . Annealing in vacuum also is becoming more regularity in physical structure, and intensity of peak in XRD spectra increased. Grain size of AZO thin films, were affected on conductivity and transparency of films. AZO thin films deposited with lower argon gas flow rate, could have larger grains size. Sample deposited by setting argon gas flow rate on 10 sccm, has appropriated grain size.

### 4. References

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