Study of AZO Thin Films Under Different Ar Flow and Sputtering Power by rf Magnetron Sputtering

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Abstract—**Transparent conductive aluminum-doped zinc oxide (ZnO: Al, AZO) thin films with different Ar flow (from 30 to 60 sccm) and sputtering power (from 150 to 250 W) were prepared on the Coring glass substrate by using rf magnetron sputtering. The lowest resistivity of 6.11 × 10^{-4} Ω-cm and highest transmittance of 92.8 % was obtained at sputtering power of 250 W and Ar flow of 40 sccm. In the transmission spectra, the absorption edge was about 440 nm and optical transmittance was about 80 - 92.8 % in the visible range. The observed property of the AZO thin films is suitable for transparent conductive electrode applications.**

Keywords:—Zinc oxide, AZO, RF magnetron sputtering, transparent conductive oxides, Ar flow, Sputtering power

I. Introduction

In recent years, transparent conductive oxides (TCOs) have been extensively studied because they are essential elements in thin film optoelectronic devices applications such as thin film solar cells, flat-panel displays, and light emitting diodes [1-3]. For these films applications, the average optical transmittance up to 80% in the visible wavelength range and the resistivity of below ~10^{-3} Ω-cm are required.

At present, the commonly used TCO material is indium tin oxide (ITO), and there are growing concerns are due to its toxicity, high cost and the limited availability of the element indium. Aluminum-doped zinc oxide (AZO) thin films are attracted interest as transparent conductive oxide (TCO) films prepared materials due to the advantages of AZO films are cheap and abundant elements [4]. In addition, AZO thin films have an excellent chemical stability and specific electronic/optical properties of a wide band gap (Eg ≒ 3.4 to 3.9) semiconductor. Therefore, AZO thin films are usually used as transparent conducting electrodes in solar cells. Several studies using different deposition methods have been reported [5]-[8], such as sol-gel processes [5], pulsed laser deposition [6], sputtering [7] and molecular beam epitaxy [8]. RF (Radio-Frequency) Sputtering method is an effective technique due to its ability to produce reasonable quality thin films at a high deposition rate [9]. Based on the progress in the previous works, it is important to better understand the influences of rf magnetron sputtering at different Ar flows and sputtering power to obtain the optimum conductivity and transmittance.

In this study, the Al-doped ZnO (ZnO: Al, AZO) thin films were prepared by using the rf magnetron sputtering at different Ar flows (from 30 to 60 sccm) and sputtering power (from 150 to 250W) to examine the optical and electronic properties. In the transmission spectra, the absorption edge was about 440 nm and optical transmittance was about 80 - 92.8 % in the visible range. The lowest resistivity of 6.11 × 10^{-4} Ω-cm (61.1 Ω/sq) and highest transmittance of 92.8 % was obtained at sputtering power of 250 W and Ar flow of 40 sccm. The observed property of the AZO thin films is suitable for transparent conductive electrode applications.

II. Experimental

The AZO structure were deposited on glass substrate (corning eagle XG) by using rf magnetron sputtering of AZO using an AZO ceramic target (99.9995 purity, 20 cm diameter, 0.5 cm thickness, Al_2O_3 : ZnO = 2 : 98 wt %). The dimension of the glass substrate is 25 × 25 × 0.5 mm^3. The sputtering chamber was initially pumped down to 3.0 × 10^{-6} torr. The deposition of AZO layers was performed in an argon (purity: 99.99%) atmosphere and the deposition pressure was maintained at 1.1 × 10^{-3} torr and the rf power was controlled in the range of 150 - 250 W. The rotating speed of the substrate was 10 rpm. The temperature was controlled using a feedback controlled heater. Substrate temperature during deposition was maintained within ±5 °C. Ar flow was controlled in the range of 30 - 60 sccm. The thickness of the deposited films was measured by an Alpha-step (α-step, Kosaka Laboratory Ltd. ET-4000). The resistivity (ρ), carrier concentration (n) and Hall mobility (μ) were measured by the four point probe method (Jiehan 5000 Electrochmical workstation, SRS-400) and Hall Effect measurement (Ecopia HMS 3000). Conventional u-2u XRD studies on the films were carried out in Rigaku (BRUKER D8 ADVANCE) diffractometer using CuKα (λ = 0.154056 nm) radiation to investigate the crystallinity and crystal orientation of the films. The AZO thin films surface roughness were performed by (NT-MDT Solver P47) AFM system. Optical transmittance was measured using a UV-vis-IR spectrophotometer (JASCO V-670) in the range of 300 - 800

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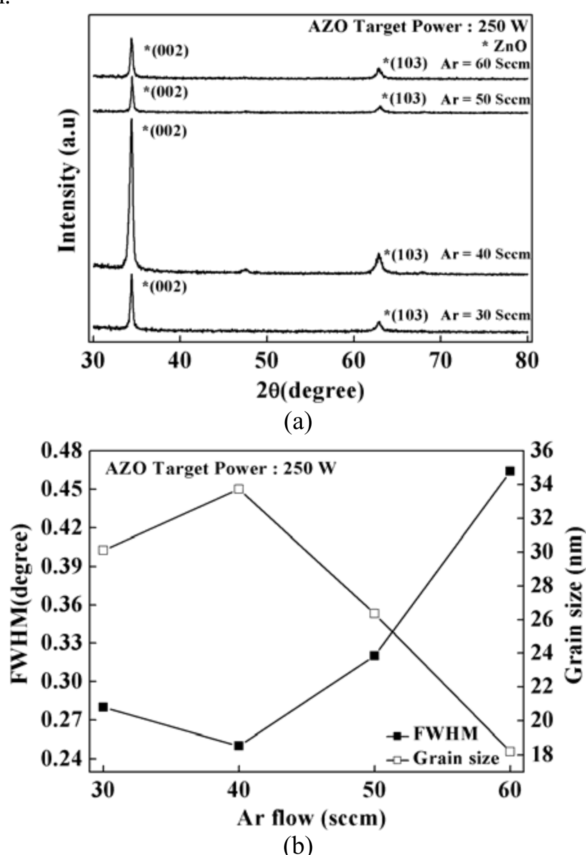

Fig. 1. (a) XRD patterns and (b) full width at half maximum (FWHM) of XRD (002) peaks and the grain size corresponding to the AZO thin films as a function of Ar flows. (sputtering power is fixed at 250W)

III. Results and Discussions

Fig. 1(a) shows the XRD patterns corresponding to the AZO thin films as a function of Ar flows. All of films show strong <002> peaks at 34.45° along with <103> were observed for AZO thin films and indicated polycrystalline nature of the thin films. The positions of the diffraction peaks were not changed. It noted that the structures of the AZO thin films are maintained with the variation of Ar flow. As the Ar flow was increased to 40 sccm, the peak intensity increased to a maximum and then decreased at 60 sccm. The results show the good crystallinity and large grain size of the Ar flow at 40 sccm due to the smallest full width at half maximum (FWHM) [10]. Fig. 1(b) shows the FWHM of XRD (002) peaks and the grain size corresponding to the AZO thin films as a function of Ar flow. The grain size of the AZO thin films as a function of Ar flows was calculated using Scherrer's formulation [11]:

$$D = \frac{0.9\lambda}{\beta\cos\theta} \tag{1}$$

where $\lambda = 1.54$ and β = B-b (B is the observed FWHM and b is the instrument function determined from the broadening of the monocrystalline silicon diffraction line). The maximum grain size is around 33.72 nm at sputtering power of 250 W with Ar flow of 40 sccm. Increasing the crystallite size of the AZO films can decrease grain boundary scattering and increase the carrier lifetime for achieving the lower resistivity of the AZO thin films. Song et al. reported that a 50% variation of the

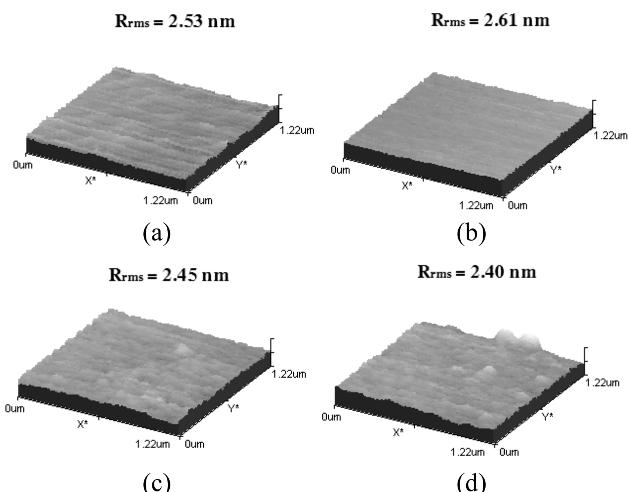

Fig. 2. AFM images of the AZO thin films with sputtering power of 250 W deposited at various Ar flows of (a) 30 sccm, (b) 40 sccm, (c) 50 sccm and (d) 60 sccm.

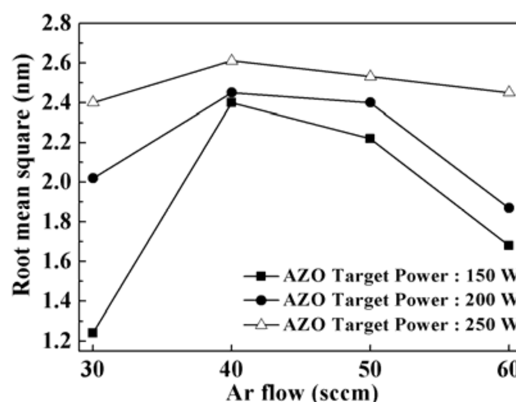

Fig. 3. Root mean square (RMS) roughness of the AZO thin films with sputtering power of 150 W, 200 W and 250 W at various Ar flow of 30 Sccm, 40 Sccm, 50 Sccm and 60 Sccm.

crystallite size was found when the deposition pressure decreased from 3.2 to 0.2 Pa in the case of ZnO: Al films [12]. Fig. 2 shows the AFM images of the AZO thin films with sputtering power of 250 W deposited at various Ar flow of 30, 40, 50 and 60 sccm. Surface roughness is one of the important properties of the AZO thin films for many opto-electronics applications, because the level of surface roughness will dominate the carrier mobility and light scattering [13]. In Fig. 2 rough and non-uniform surface structures of the AZO thin films under different Ar flow were observed. Fig. 3 shows the Root mean square (RMS) roughness of the AZO thin films with sputtering power of 150 W, 200 W and 250 W at various Ar flows of 30, 40, 50 and 60 sccm. When Ar flow increased from 40 to 60 sccm under sputtering power of 250 W, the RMS roughness decreased from 2.61 to 2.45 nm. This is because the energy of particles arriving at the surface of the thin films decreased with increasing the argon pressure, leaving less energy for surface diffuse due to collisions with more atoms [14]. However, the sputtering power increased from 150 to 250 W at Ar flow of 40 sccm. The RMS roughness of AZO films is increased due to the density of the thin films is enhanced under increasing the sputtering power. Surface roughness can more

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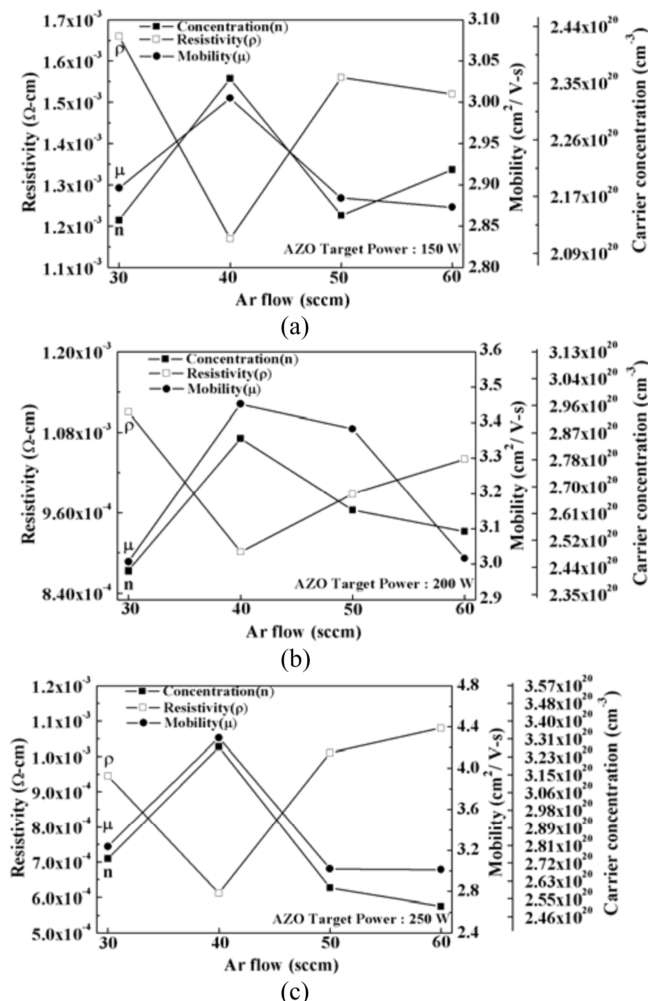

Fig. 4. Electrical resistively (ρ), Hall mobility (μ) and carrier concentration (n) of the AZO thin films with sputtering power of (a) 150 W, (b) 200 W and (c) 250 W at various Ar flows of 30 sccm, 40 sccm, 50 sccm and 60 sccm.

affect the carrier mobility [12]. The morphologies of the AZO thin films were affected by the Ar flow and sputtering power. Surface roughness is one of the important properties of the AZO thin films for many opto-electronics applications, because the smooth structure can reduce the scattering of incident light, which makes the contribution to increase the transmittance [15].

Fig. 4 shows the electrical resistively (ρ), Hall mobility (μ) and carrier concentration (n) of the AZO thin films (thickness is fixed as 100 nm) with sputtering power of 150 W, 200 W and 250 W at various Ar flows of 30, 40, 50 and 60 sccm. Resistivity of the AZO thin films increases from 1.17×10^{-3} Ω-cm (sheet resistance of 117 Ω/sq) to 6.11×10^{-4} Ω-cm (sheet resistance of 61.1 Ω/sq) at Ar flow of 40 sccm as the sputtering power was increased from 150 W to 250 W. The decrease in resistivity with increase sputtering power can be attributed to the high dense surface of the AZO thin films and reduced grain boundary scattering, thus forming the continuous and decreasing the interface scattering. In addition, the resistivity of the AZO thin films decreases from 1.16×10^{-3} Ω-cm to 7.53×10^{-4} Ω-cm at sputtering power of 250 W as the Ar flow of 30 and 40 sccm. The initial decrease in resistivity with increase in Ar flow can be attributed to an decrease in the grain size of the AZO thin films from 30.11 to 33.72 nm and high plasma dense with the increasing the growth Ar flow, therefore, reducing the grain boundary scattering and decreasing the resistivity. The increase in resistivity of the AZO thin films growth at higher Ar flow above 40 sccm may be due to contamination of the alkali ions from the glass substrate [13]. The doping effect of some Ar^+ into the thin films might caused the higher resistivity with higher deposition pressure [16]. Since increasing the grain size of the AZO thin films with increased Ar flow, the grain boundary scattering and lattice defects were decreased, and caused the enhancement of mobility from 3.01 to 4.3 cm^2/V-s at Ar flow of 40 sccm under sputtering power increased from 150 W to 250 W. Therefore, there is better way to decrease the resistivity by increasing the Ar flow due to increased mobility. The improvement of crystallinity can cause a decrease in grain boundary scattering and an increase of carrier lifetime [17, 18]. The increase in both carrier concentration and Hall mobility would consequently lead to an increase of conductivity.

Fig. 5 shows the transmission spectra of AZO thin films deposited at various Ar flows. The transmittance spectrum of the AZO thin films was measured in the wavelength from 300 to 800 nm. In the transmission spectra, the absorption edge was about 440 nm and optical transmittance was about 80 – 92.8 % in the visible range. This means that the Ar gas pressure does not have a significant effect on the transparency of the thin films over the visible light wavelength range. However, the

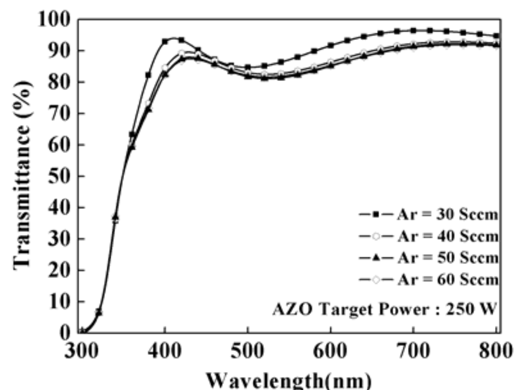

Fig. 5. Transmission spectra of AZO thin films with sputtering power of 250 W deposited at various Ar flows of 30 sccm, 40 sccm, 50 sccm and 60 sccm.

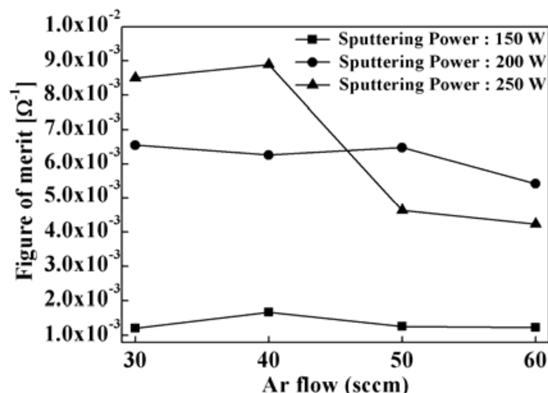

Fig. 6. A plot of the figure of merit for AZO thin films with sputtering power of 150 W, 200 W and 250 W at various Ar flows of 30 sccm, 40 sccm, 50 sccm and 60 sccm.

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short wavelength cut-off in transmittance of AZO thin films has a clear shift towards to the long wavelength range with increasing the Ar gas pressure, indicating that the optical energy gap is reduced [19]. Maximum transmittance of around 96.4 % was observed for the prepared AZO films with sputtering power of 250 W at Ar flow of 30 sccm.

Fig. 6 shows the plot of the figure of merit for AZO thin films with sputtering power of 150 W, 200 W and 250 W at various Ar flows of 30, 40, 50 and 60 sccm. The FOM (φ_{TC}) is calculated for the AZO thin films as defined by Haacke [20]

$$\varphi_{TC} = T_{av}^{10} / R_s \qquad (2)$$

where T_{av} is the optica measured transmittance of the AZO film from 300 to 800 nm and Rs is the sheet resistance. In Fig. 6, the highest peak can be observed at Ar flows of 40 sccm under the sputtering power of 250 W in the 300 – 800 nm wavelength region. Below this region, there was a steep fall in FOM due to the higher sheet resistance at Ar flows up to 50 sccm. The decrease of the resistivity is attributed to the improvement of the crystallinity of the AZO thin films. As discussed above, the AZO thin films prepared at 40 sccm Ar flows and 250 W sputtering power shows better performance with simultaneously low sheet resistance of 61.1 Ω/sq and higher transmittance of 92.8 %. The method gives a more realistic estimate of the actual merit of the AZO film for transparent electronics.

IV. Conclusion

The AZO thin films have been successfully deposited on glass substrate by rf sputtering. The prepared AZO thin films show the optical transmittance greater than 92.8 % in the visible range, the lowest electrical resistivity of 6.11×10^{-4} Ω-cm (61.1 Ω/sq) and the maximum figure of merit achieved is 8.89×10^{-3} Ω^{-1} at sputtering power of 250 W and Ar flow of 40 sccm. For the XRD analysis, the results show the good crystallinity and large grain size of the Ar flow of 40 sccm due to the smallest full width at half maximum. Such results make these AZO thin films good candidates for future transparent conductive electrode applications.

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