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Research Paper

A Study on Properties of RF-sputtered Al-doped ZnO Thin Films Prepared with Different Ar Gas Flow Rates

Seung Ik Han*, and Hong Bae Kim
Department of Semiconductor Engineering, Cheongju University, Cheongju 363-764, Korea

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Abstract This paper, Al-doped ZnO(AZO) thin films for application as transparent conducting oxide films were deposited on the Corning glass substrate by using RF magnetron sputtering system. The effects of various Argon gas flow rates on optical and electrical characteristics of AZO films were investigate sputtering method. The Carrier Concentration is enhanced as Ar gas rate increases, and also the oxygen vacancy concentration. The figure of merit obtained in this study means that AZO films which deposited Ar gas rate of 75 sccm have the highest Carrier concentration and Hall mobility, which have the highest photoelectrical performance that it could be used as transparent electrodes.

Keywords: Al-doped ZnO, Ar gas flow rate, Hall mobility, Oxygen vacancy

I. Introduction

Transparent conducting oxide (TCO) Thin films have been widely used for optoelectronic devices, such as PDP(Plasma display panel), LCD(Liquid Crystal display), OLED(Organic light-emitting diode), Solar Cell [1-4]. Until now, Indium tin oxide(ITO) layers have been popular amongst TCO materials because of low resistivity and high visible transmittance [5-7]. However, Because of the toxicity, high cost and the rareness of the indium in the ITO films, the Al doped ZnO(AZO), Ga doped ZnO(GZO) thin films have been considered an ideal alternative material due to its nontoxicity, lower mater cost, high transmittance [8-10]. When Al-doped ZnO thin films are used as an Transparent electrodes, they should be required to have high transmission capabilities in a low resistivity ($\rho \le 10^{-3}\,\Omega cm$) and wavelength range of 400-800 nm (T ≥80%) [11]. In this study, we investigated the structural, optical and electrical properties of Al-doped ZnO films deposited under different Ar gas flow rate change by RF Sputtering method and discussed the Oxygen Vacancy dependence of the Hall mobility of AZO films with varying Ar gas flow rate change.

II. Experiments

Al doped ZnO(AZO) films were deposited on a corning 1737 glass substrate by an RF magnetron sputtering with a 2inch diameter target consisting of ZnO:Al (98:2 wt%). Before the operating plasma, the substrates were cleaned with acetone, methanol and D.I. water for 5 minutes respectively. The RF power applied to the target was fixed at 50 W. The base pressure in the chamber was kept below 3.0×10^{-6} Torr. Film deposition was carried out under a fixed working pressure of 2.0×10^{-3} Torr.

The substrate temperature during the deposition was maintained at room temperature. The flow rate of the argon (99.999%) was ranged from 25 to 100 sccm. The films were deposited with a constant thickness of 100 nm. After deposition, AZO films were annealed in a vacuum state at 400°C for 30 min.

The thickness and cross sectional images of the AZO thin films were checked by field emission scanning electron microscopy (FE-SEM), and the crystallinity of the

Table 1. Processing conditions of AZO films.

Target	2 wt%, Al-doped ZnO
Substrate	Glass (Corning #1737)
RF Power [W]	13.56 MHz, 50 W
Ar flow rate[sccm]	25, 50, 75, 100
Base pressure [Torr]	3.0×10^{-6}
Working pressure [Torr]	2.0×10^{-3}
Deposition Temperature [°C]	Room temperature
Pre-sputtering Time [min]	10
Deposition Time [min]	35, 25, 18, 10
Film Thickness [nm]	±100
Annealing Temperature [°C]	400
Annealing Time [min]	30

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*Corresponding author
E-mail: gkstmddlr1@naver.com

film was analyzed with X-ray diffraction (XRD). The roughness was observed by atomic force microscopy (AFM). The transmittance of the AZO films was investigated using UV-vis-NIR spectrometer. The electrical properties were investigated using a Hall measurement and a four-point probe. Research the oxygen vacancy contents of the AZO films was studied with X-ray photoelectron spectroscopy (XPS).

III. Results and Discussion

Fig. 1 shows the XRD patterns of Al-doped ZnO thin films with various Ar gas flow rate. Regardless of the Ar gas flow rate, the same peaks of the (002) plane were observed around 2theta=34.45°, The (002) peak position of AZO films is found bigger than that of bulk ZnO material (34.42°). The strong peak of the (002) plane indicated that the structure of AZO thin films was the polycrystalline with a hexagonal structure and the c-axis of the AZO thin film grew perpendicularly from the substrate. respectively, As decrease Ar gas flow rate, AZO films show the increment of the preferred direction of ZnO (002) diffraction peak in the XRD pattern. The intensity of the diffraction peaks from (002) was increased as Ar gas flow rate decreased, indicating that preferential c-axis direction becomes more obvious.

Fig. 2 shows The full width at half maximum (FWHM) and grain size of Al-doped ZnO thin films with various Ar gas flow rate. As decrease Ar gas flow rate, the FWHM value of (002) peaks was decreased from 0.28 to 0.17. These result indicate that the crystal quality of AZO thin films is improved with decreasing the Ar gas flow rate. Ar gas flow rate shows evident influence on the crystal quality of AZO thin films

Fig. 3 shows FE-SEM images of Al-doped ZnO thin films with various Ar gas flow rate. The thickness and cross-sectional images of the AZO films were checked by field emission scanning electron microscopy (FE-SEM). all of AZO thin films were deposited in 100 nm (±10%). AZO thin films are shows grown into columnar structures. This result shows that c-axis growth is consistent like XRD result.

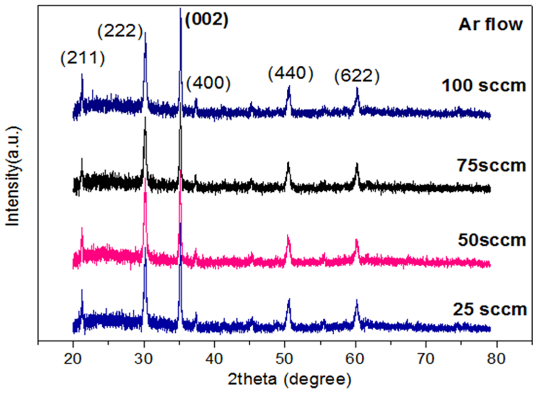


Figure 1. XRD patterns of Al-doped ZnO thin films with various Ar gas flow rate.

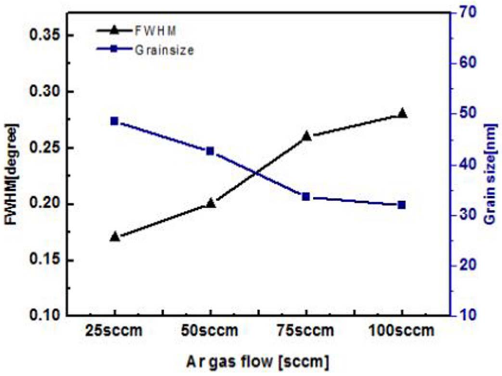
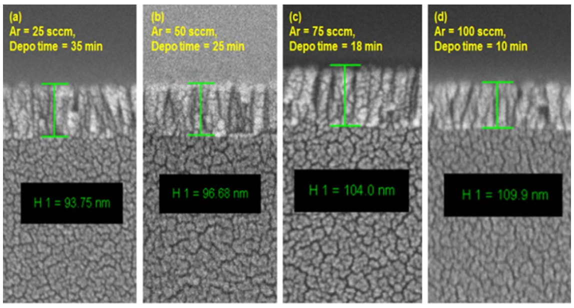


Figure 2. FWHM and grain size of Al-doped ZnO thin films with various Ar gas flow rate.



(a) 25sccm (b) 50sccm (c) 75sccm (d) 100sccm

Figure 3. FE-SEM images of Al-doped ZnO thin films with various Ar gas flow rate. (a) 25 sccm, (b) 50 sccm, (c) 75 sccm, (d) 100 sccm.

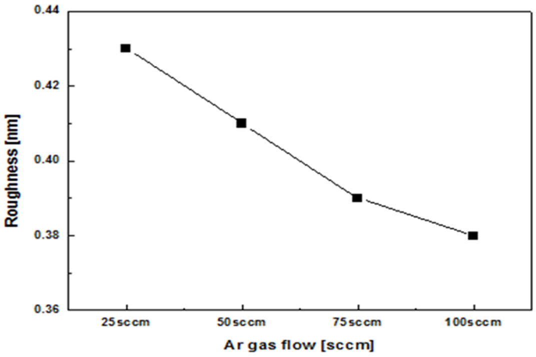


Figure 4. Roughness of Al-doped ZnO thin films with various Ar gas flow rate.

Fig. 4 shows Roughness of Al-doped ZnO thin films with various Ar gas flow rate. The root mean square (RMS) surface roughness value increased from 0.38 to 0.43 nm as the Ar gas flow rate was elongated from 100 to 25 sccm.

It is found that the AZO films surface roughness is dependent on Ar gas flow rate and grain size.

Fig. 5 shows Transmittance spectra of Al-doped ZnO thin films with various Ar gas flow rate in the wavelength range of 300~1000 nm.

The average transmittance of AZO thin films in the visible range (400~700 nm) was higher than 85%, which is

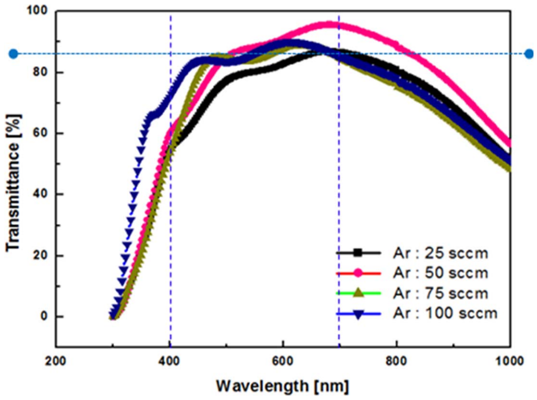


Figure 5. Transmittance spectra of Al doped ZnO thin films with various Ar gas flow rate.

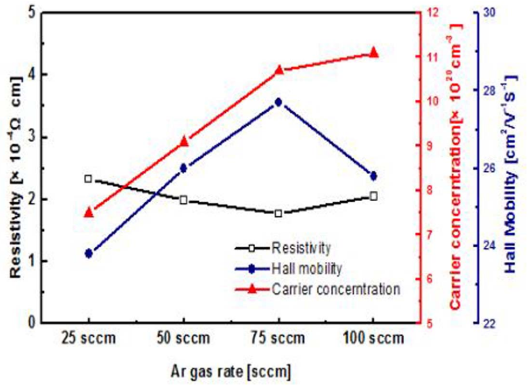


Figure 6. Resistivity, Carrier concentration, Hall mobility of Al-doped ZnO thin films with various Ar gas flow rates.

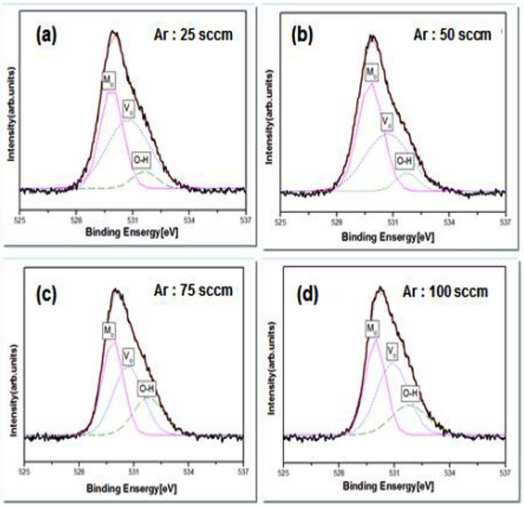


Figure 7. XPS spectra of O1s of Al-doped ZnO thin films with various Ar gas flow rate (a) 25 sccm, (b) 50 sccm, (c) 75 sccm, (d) 100 sccm.

plenum value of transparent electrodes

Fig. 6 shows Resistivities, Carrier concentration, Hall mobility of Al doped ZnO thin films with various Ar gas flow rate. As the Ar gas flow rate increased from 25 sccm to 75 sccm, the sheet resistance decreased from $2.33 \times 10^{-4}\Omega - cm$ to $1.77 \times 10^{-4}\Omega - cm$, whereas carrier concentration increased from $7.5 \times 10^{20}cm^{-3}$ to $10.7 \times 10^{20}cm^{-3}$. Also Hall mobility increased from $23.8cn^{-3}/V-s$ to $27.7cn^{2}/V-s$. The increase in sheet Hall mobility at higher Ar gas flow rate is due to the increase in Oxygen Vacancy (V_o) with increasing the number of electrons. In our case, the crystal quality in AZO thin film is degraded with increasing the Ar gas flow rate due to the number of Oxygen Vacancies. The lowest resistivity ($1.77 \times 10^{-4}\Omega - cm$) and Resistivity ($1.77 \times 10^{-4}\Omega - cm$) are obtained in the AZO thin films deposited at Ar 75 sccm. Therefore the AZO thin film deposited on 75 sccm is the best electrical properties.

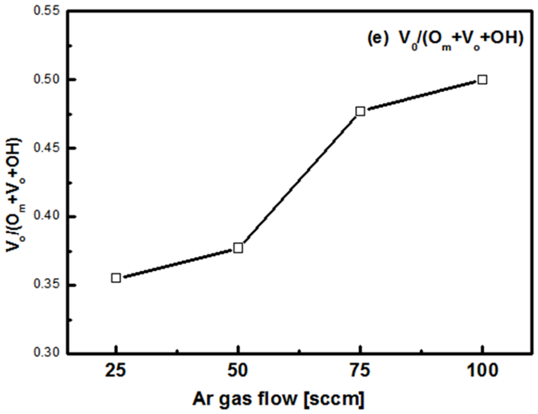


Figure 8. Oxygen Vacancy (V_o) of Al-doped ZnO thin films with various Ar gas flow rate.

Fig. 7 shows XPS spectra of O1s of Al doped ZnO thin films with various Ar gas flow rate. Each peak represents a peak associated with O_m (Metal oxygen), V_o (Oxygen vacancy), OH.

Fig. 8. Shows oxygen vacancy (V_o) of Al doped ZnO thin films with various Ar gas flow rate. As the Ar gas flow rate increased from 25 sccm to 100 sccm, the sheet Carrier concentration and Hall mobility increased from $7.5 \times 10^{20}cm^{-3}$ to $10.7 \times 10^{20}cm^{-3}$ and $23.8cm^{2}/V-s$ to $27.7cm^{2}/V-s$ whereas Ar gas flow rate increased from 75 sccm to 100 sccm, the sheet Carrier concentration increased from $10.7 \times 10^{20}cm^{-3}$ to $11.1 \times 10^{20}cm^{-3}$, but Hall mobility were decreased from $27.7cm^{2}/V-s$ to $25.8cm^{2}/V-s$. The observed result means that Carrier concentration and Hall mobility of the AZO films were influenced with Oxygen Vacancy. The carrier concentration increased as the number of electrons increased as the oxygen vacancy increased. But Oxygen vacancy becomes excessively large, which interferes with scattering of electrons and disturbs electron mobility. As a result, The electrical characteristic of the AZO films were dependent on the Oxygen Vacancy.

IV. Conclusions

Al doped ZnO thin films have been deposited on glass by RF magnetron sputtering at various Ar gas flow rate. The effect of Ar gas flow rate on the morphology, structured, optical, electrical properties of the AZO films was studied. The AZO films are oriented along the c-axis of the hexagonal structure whatever the Ar gas flow rate. FWHM was found to decrease as the Ar gas flow rate decreases. AZO thin films were transparent up to 85% in the visible range. The increase in sheet Hall mobility and Carrier Concentration at higher Ar gas flow rate is due to the increase in Oxygen Vacancy (V_o) with increasing the number of electrons. The Ar gas flow rate is a major factor to determine the structural, optical and electrical of AZO films

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