Effect of substrate temperature on the properties of AZO thin film deposited by using facing targets sputtering system

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ABSTRACT

Al doped ZnO (AZO) thin film was deposited by using Facing Target Sputtering (FTS) system. This work examined the properties of AZO thin film as a function of the substrate temperature. The sputtering targets were 4 inch diameter disks of AZO (ZnO: $Al_2O_3 = 98:2$ wt.%). The properties of electrical, structural and optical were investigated by 4-point probe, Hall effect measurement, x-ray diffractometer (XRD), field-emitting scanning electron microscopy (FE-SEM), and UV/VIS spectrometer. The lowest resistivity of films was $5.67 \times 10^{-4} \,\Omega$ cm and the average optical transmittance of the films was above 85% in the visible range.

Key Words: AZO, Substrate temperature, Facing targets sputtering

1. Introduction

Transparent conductive oxide (TCO) film is oxide material that is highly conductive transparent in visible light and conductive. Indium tin oxide (ITO) has been used for transparent conductive electrodes in many photo- electronic devices such as solar cells and flat panel displays [1-3]. Indium is rare metal that limits many applications. Impurity-doped ZnO semiconductor materials have attractive characteristics owing to its wide direct band gap (3.37 eV), the abundant raw materials, environmental friendliness, and high radiation resistance. It has the exciton binding energy of 60meV, that is larger than the thermal energy at room temperature [4-6]. Therefore, such properties make them well suited for the realization of many optoelectronic applications. Recent researches exhibited that Group III elements-doped ZnO films have low resistivity and high transmittance in the visible range. Based on many reported investigations of the effect of doping ZnO with impurities, Al seems to be the successful and promising element due to its advantages. Al is the dopant which resulting in high-quality, low-resistivity AZO thin films [7,8]. Generally sputtered thin films properties are influenced by substrate temperature, power, and pressure. The growth temperature plays a major role for thin film properties determination, the present study is focused on the influence of substrate temperature on structural, optical and electrical properties of Al doped ZnO films.

2. Experimental

2.1. Facing targets sputtering

Fig. 1 shows a schematic diagram of facing targets sputtering (FTS) system. The FTS system is an array

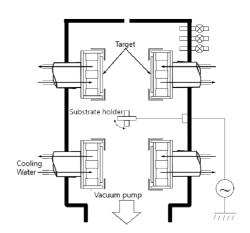


Fig. 1. Schematic diagram of FTS system.

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target of two sheets that forms high density plasma between the targets. The process is called plasma-free because the substrate is remote from the plasma, which means that bombardment from high energy particles, such as electrons and negative ions, is minimized [9].

2.2. Deposition of AZO thin film

In this work, we used the pair of AZO targets. Before film deposition, the glass substrate was cleaned by ultrasonic cleansing with distilled water and isopropyl alcohol (IPA) for 30 min. The substrate was then dried in a stream of N_2 gas. The chamber was evacuated to 2×10^{-6} Torr before the film deposition. During deposition, the pressure was maintained at 1mTorr. The AZO films on the glass substrate were deposited at different substrate temperatures (Room temperature, 100° C, 150° C, 200° C, 250° C and 300° C). The DC power fixed 90W and the thickness of films fixed 300 nm. Table 1 listed the sputtering conditions.

Table 1. Sputtering conditions

Deposition parameter	Sputtering Conditions
Targets	AZO (ZnO: Al ₂ O ₃ = 98 wt.% :2wt.%)
Substrate	Glass
Background pressure	2×10^{-6} Torr
Working gas pressure	1mTorr
Film thickness	300 nm
Substrate temperature	R.T, 100°C, 150°C, 200°C, 250°C, 300°C
DC power	90 W

2.3 Measurements

The electrical properties of the AZO thin films were examined using four-point probe (Chang-min, CMT-1000), and Hall effect measurement (ECOPIA, HMS-3000). The structural properties of the AZO thin films were analyzed by X-ray diffraction (XRD, Rigaku, D/MAX-2200). The surface morphology of AZO thin films was observed by Field Emission Scanning Electron Microscopy (FE-SEM, Hitachi). The optical transmittance AZO thin films were measured by UV-VIS spectroscopy (HP).

3. Results and Discussions

The AZO thin films exhibited n-type conductivity. The conductivity of the AZO thin films are dominated primarily by electrons generated from Al^{3+} ions on the substitutional sites of Zn^{2+} ions and Al interstitial atoms [10].

Fig. 2 shows the resistivity, mobility and carrier concentration of AZO thin films as a function of substrate temperatures. The minimum value of the resistivity of $5.67 \times 10^{-4} \,\Omega$.cm was obtained with substrate temperature of 250° C. Increasing substrate temperature caused the decrease of resistivity. The Hall mobility sharply increased from $11.07 \, \mathrm{cm}^2/\mathrm{V.s}$ to $21.45 \, \mathrm{cm}^2/\mathrm{V.s}$ and the carrier concentration slowly increased from $2.69 \times 10^{20} \, \mathrm{cm}^{-3}$ to $4.23 \times 10^{20} \, \mathrm{cm}^{-3}$ when substrate temperature increased from R.T to 250° C. Increased Hall mobility is attributed to improved crystallinity and increased crystallite sizes that weak inter-crystallite boundary scattering and increases carrier lifetime [11].

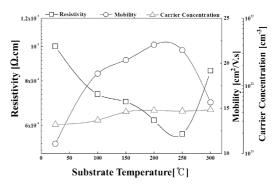


Fig. 2. Resistivity, mobility and carrier concentration of AZO thin films as a function of substrate temperatures.

Deposited AZO thin films showed a c-axis preferred orientation of the (002) (102) plane, as shown in fig.3. Deposited AZO films had a strong (002) XRD peak at value of 34.16° - 34.24° at 2θ , and a very weak (102) peak at 44.46° - 44.66° at 2θ . This suggests that the crystallographic c-axis of the AZO thin films is perpendicular to the substrate, and all AZO thin films had a polycrystalline structure. From fig.3, the preferred orientation for films is increased with sub-

strate temperature up to 250°C. The decrease of the AZO (002) peak for the films deposited at high substrate temperatures to the transition region of the Thornton structure zone model [12] where the columnar grains change their structure. In addition, a small deviation in (002) peak from the regular position was also found when the substrate was unheated, indicating some residual stress inside the film may exist.

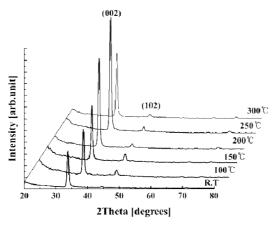


Fig. 3. XRD patterns of AZO thin films as a function of substrate temperature.

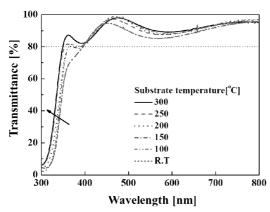


Fig. 4. Transmittance of the AZO thin films as a function of substrate temperature.

Optical transmittance of the AZO thin films was determined by a UV/Vis. spectrometer within the wavelength from 300 nm to 800 nm. Fig. 4 showed the transmittance of the AZO thin films as a function of substrate temperature. The average optical transmittance independent substrate temperature, was over

80% in the visible range (380-770 nm) indicating a high optical transparency. The insert figure showed blue shifts of the optical absorption edge as substrate temperature increased from R.T to 300°C. The optical absorption coefficient á near the optical absorption edge can be extrapolated by using

$$\alpha = \frac{t}{t} \times \frac{1}{T} \tag{1}$$

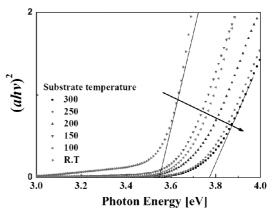


Fig. 5. Optical bandgap energy of the deposited AZO

where T is the transmittance of the AZO films near the optical absorption edge and t is the thickness of film.

For AZO thin film, a typical direct band gap semi-conductor, α obeys the following relationship with optical band gap $E_{\rm g}$,

$$\alpha h v = C(hv - E_g)^{\frac{1}{2}} \tag{2}$$

where C is a constant, h is Planck's constant, and v is the frequency of the incident photon. Fig. 5 showed the optical bandgap energy of deposited AZO thin film. The E_g value can be obtained by extrapolating the linear segments of the curves towards the x-axis. The E_g value increases from 3.54 eV to 3.77 eV from R.T to 300°C. This means that the substrate temperature affects the optical band gap of the film.

The grain size of the AZO films sharply increased above a substrate temperature of 100°C, and showed significant temperature sensitivity. The grain size can be estimated using Scherrer's formula [13].

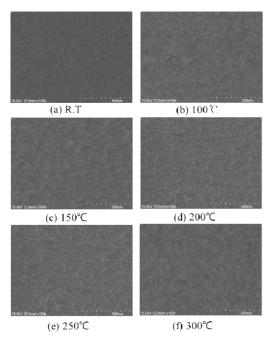


Fig. 6. The SEM images of thin films as a function of substrate temperatures.

$$D = \frac{0.94\lambda}{\beta\cos\theta} \tag{3}$$

where $\lambda = 0.154$ nm and β is the FWHM. As the films thickness increased from R.T to 250°C, the FWHM decreased from 0.69° to 0.43°, and the grain size increased from 11nm to 18 nm.

4. Conclusions

The AZO thin films were deposited on a glass substrate by using FTS system. The AZO thin films require low resistivity, high transmittance and good crystallity with a lower film thickness. The lowest resistivity of $5.67 \times 10^{-4} \Omega cm$ was obtained for a substrate temperature of $250^{\circ}C$.

The AZO thin films grew with the crystallographic *c*-axis perpendicular to the substrate, and all AZO thin films exhibited a polycrystalline structure. The best intensity of the (002) planes exhibited substrate temperature of 250°C. The optical transmittance of all AZO thin films was above 80% in the visible range. The deposited AZO films showed good transparency. The AZO thin films showed good properties, making it a suitable thin film thin film solar cells application.

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