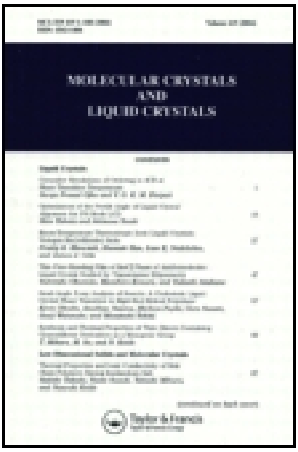

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Properties of Ga-Al Doped ZnO with Various Thicknesses Prepared by Facing Targets Sputtering Method

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Gallium-Aluminum doped Zinc Oxide (Ga-Al doped ZnO) thin films prepared on glass substrates by using facing targets sputtering methods. Electrical, structural and optical properties of Ga-Al doped ZnO thin film with various thicknesses were studied in detail. Crystal structure of the Ga-Al doped films was hexagonal wurtzite. Increased thickness of Ga-Al doped ZnO thin film, the resistivity decreased and crystallity improved. As the results, The resistivity of Ga-Al doped ZnO thin films with thickness of 500 nm exhibited 4.17×10^{-4} *Ω.cm and average optical transmittance of all thin films showed above 85% in the visible range.*

Keywords Ga-Al doped ZnO; TCO; FTS; various thicknesses

Introduction

Transparent conductive oxide (TCO) films have been widely studied transparent electrode for flat panel display, thin film solar cells and various optoelectronic devices [1–3]. One of the TCO films, ITO has been widely used various applications. However, the ITO has disadvantages, including toxicity, high price, and instability in hydrogen plasma. Accordingly, ZnO thin films have promoted the development of inexpensive material. The ZnO thin films with a low resistivity, a high transparency, a nontoxicty, chemical stability in hydrogen plasma, and low cost have been studied extensively. The ZnO thin films are n-type semiconductors with intrinsic defects, such as interstitial zinc atoms and oxygen vacancies [4,5]. Its electrical conductivity can be increased by doping with group-III elements, such as aluminum [6], boron [7], gallium [8], and indium [9]. Compared with undoped ZnO, gallium-aluminum doped ZnO (Ga-Al doped ZnO) thin films have lower resistivity and better stability [10]. The Ga-Al doped ZnO thin film may be deposited with a high transmittance above 85% in visible range and a low resistivity under 10^{-3} [Ω-cm] in various sputtering conditions. In this work, transparent conducting Ga-Al doped ZnO thin films deposited on glass substrates by using facing targets sputtering methods. The thickness

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dependence of structural, electrical and optical properties for the Ga-Al doped ZnO films are investigated in detail.

Experimental

Figure 1 showed schematic of FTS method used deposition process. The FTS method was designed to array the two targets to face each other and so form high density plasma between the targets. The FTS method was able to restrain the bombardment of the substrate by high-energy particles because the substrate's position was located outside of the plasma. Consequently, the FTS system suppresses the substrate damage caused by high-energy particles, such as electrons and partial ions [4,5,11]. In this work, chamber was evacuated to 2.6×10^{-4} Pa before the film deposition began used rotary pump and turbo molecular pump. Before each run, targets were pre-sputtered in a pure argon atmosphere for 10 min in order to remove the natural surface oxide layer of Ga doped ZnO (GZO) and Al doped ZnO (AZO) targets. Glass substrate was ultrasonically cleaned using Acetone, isopropyl alcohol (IPA) and deionize water at 30 min and it blown dry with N_2 gas. More details about the sputtering conditions were given in Table 1. Thicknesses of Ga-Al doped ZnO thin films were measured by using a surface profiler (Alpha-step, TENCOR, USA). Electrical properties of the thin films were measured by using Hall effect measurement (HMS-3000, Ecopia, Korea). Optical transmittance and structural properties were measured by using

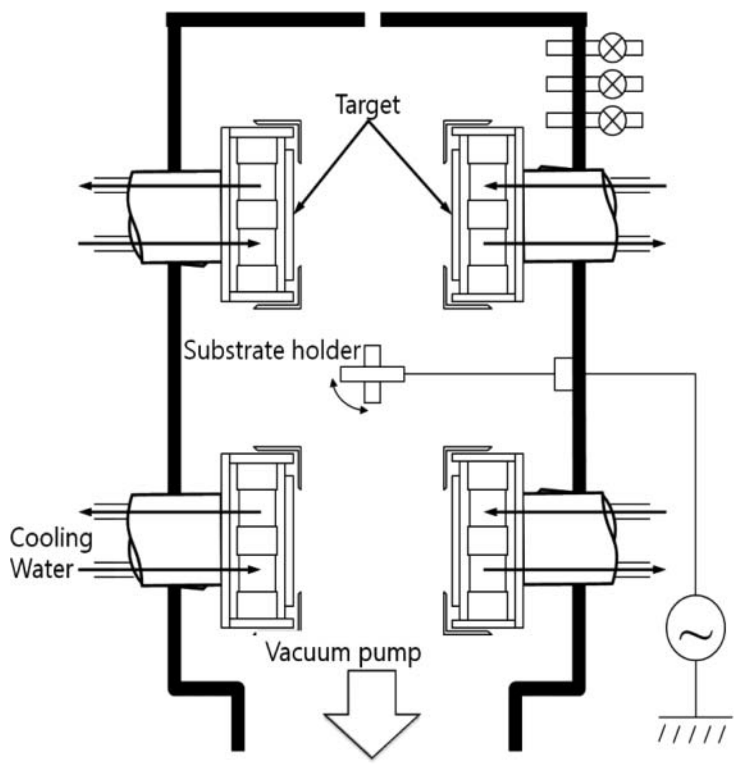

Figure 1. Schematic of FTS method using deposition process.

Table 1. Sputtering conditions.

Deposition Parameter	Sputtering Conditions
Targets	GZO (ZnO : Ga_2O_3 = 97: 3 wt.%, 4N, 4 inch) AZO (ZnO : Al_2O_3 = 98: 2 wt.%, 4N, 4 inch)
Substrate	Glass
Base pressure	2.6×10^{-4} Pa
Working gas pressure	0.13 Pa
Film thickness	100 nm – 500 nm
Substrate temperature	200°C
Input power	80 W
Working gas	Ar 12 sc cm

UV/VIS spectrometer (HP8453, Hewlett-Packard, USA) and X-ray diffractometer (RINT 2000series, Rigaku, JAPAN).

Results and Discussion

Resistivity, Hall mobility and carrier concentration of Ga-Al doped ZnO thin films with various thicknesses was shown in fig. 2. The carrier concentration increased from 4.2×10^{20} cm^{-3} to 9.5×10^{20} cm^{-3} when thickness of films increased from 100nm to 500nm. The Hall mobility slowly increased from 11.38 cm/V.s to 15.65 cm /V.s when thickness of films increased from 100 nm to 500 nm. Increased Hall mobility is attributed to improved crystallinity and increased crystallite sizes that weak inter-crystallite boundary scattering

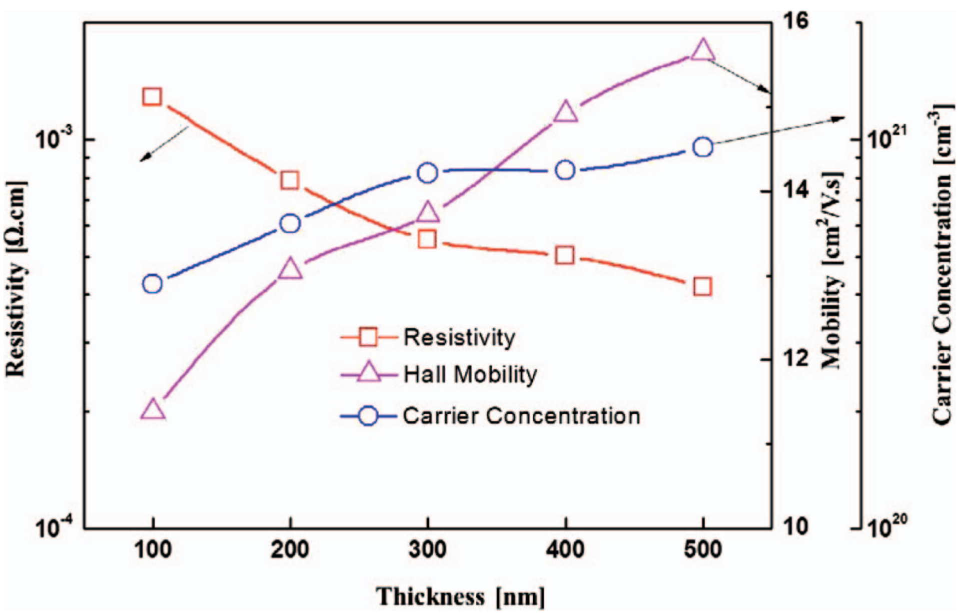

Figure 2. Resistivity, Hall mobility and carrier concentration of Ga-Al doped ZnO thin films with various thicknesses.

and increases carrier lifetime [12]. The resistivity is proportional to the reciprocal of the product of carrier concentration n and mobility μ as the following equation

$$\rho = \frac{1}{ne\mu} \tag{1}$$

From the relation, lower electrical resistivity of Ga-Al doped ZnO thin films is caused by the higher product of the carrier concentration n and mobility μ [13]. Therefore, the increased mobility and carrier concentration made the resistivity of the films decrease from $1.29 \times 10^{-3}\Omega$.cm to 4.17×10^{-4} Ω.cm when thickness of films increased from 100 nm to 500 nm.

Figure 3 showed XRD spectrum of Ga-Al doped ZnO thin films with various thicknesses from 100nm to 500nm. As shown in Fig. 3, only (002) peak is observed at 2θ $33.85^\circ \sim 34.07^\circ$ for all samples. These results indicated that the Ga-Al doped ZnO thin films were polycrystalline with the hexagonal structure and exhibited c-axis orientation perpendicular to the substrate. The c-axis orientation in the Ga-Al doped ZnO thin films can be explained by the "survival of the fastest" model proposed by Drift [14].

Al_2O_3 and Ga_2O_3 phase was not found from the XRD patterns. Gallium and aluminum atoms replace zinc in the hexagonal lattice and gallium and aluminum segregate to the non-crystalline region in grain boundary. With increasing the film thickness, the locations of the measured diffraction peaks do not change significantly but the intensities of the peaks become more intense and sharper. The crystallinity of the films improved and grain size became larger when elevating the film thickness [15].

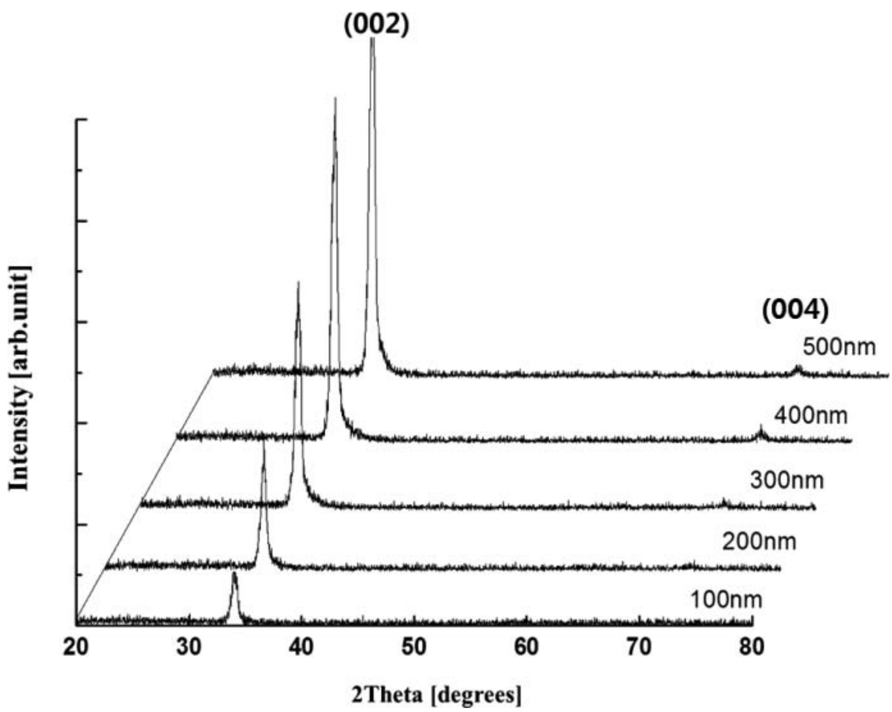

Figure 3. XRD spectrum of Ga-Al doped ZnO thin films with various thicknesses from 100 nm to 500 nm.

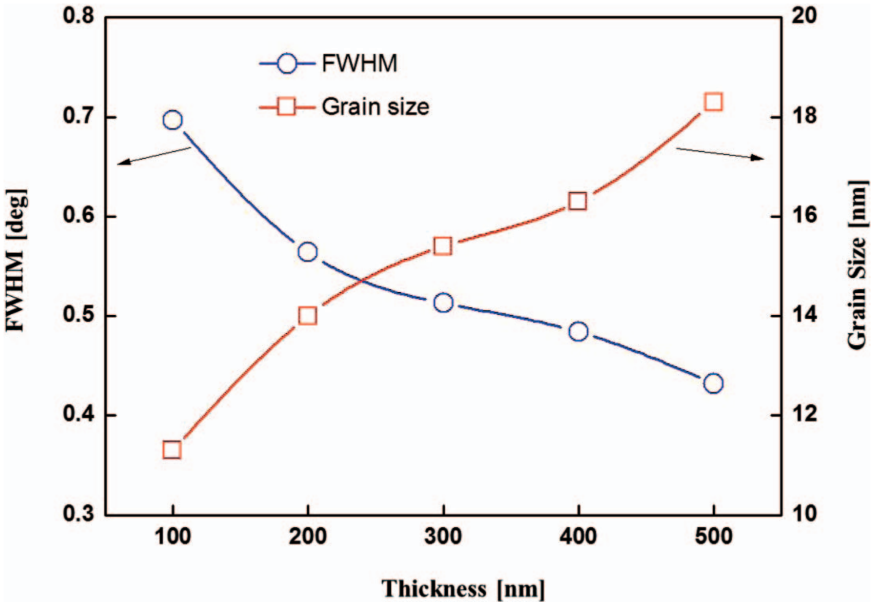

Figure 4. Full width at half-maximum (FWHM) and grain size of the Ga-Al doped ZnO thin films with various thicknesses.

Fig. 4 showed full width at half-maximum (FWHM) and grain size of the Ga-Al doped ZnO thin films with various thicknesses. The grain size can be estimated using Scherrer's formula [16].

$$D = \frac{0.94\lambda}{\beta \cos \theta} \tag{2}$$

where $\lambda = 0.154$ nm and β is the FWHM. As the films thickness increased from 100 nm to 500 nm, the FWHM decreased from 0.697° to 0.432°, and the grain size increased from 11.3nm to 18.3 nm.

Figure 5 showed the optical transmittance of Ga-Al doped ZnO thin films with various thicknesses in visible range. The average optical transmittance in visible range decreased from 91% to 86% when thickness of films increased from 100 nm to 500 nm. The insert figure showed red shifts of the optical absorption edge as thickness of films increased from 100 nm to 500 nm. The optical absorption coefficient α near optical absorption edge can be extrapolated by using

$$\alpha = \frac{l}{t} \times In\frac{1}{T} \tag{3}$$

where T is optical transmittance of film near the optical absorption edge and t is thickness of film. For Ga-Al doped ZnO thin film, a typical direct band gap semiconductor, α obeys the following relationship with optical band gap $E_{g,}$

$$\alpha h\nu = C\left(h\nu - E_g\right)^{\frac{1}{2}} \tag{4}$$

where C is a constant, h is Planck's constant, and ν is the frequency of the incident photon. Figure 6 showed the photon energy of deposited Ga-Al doped ZnO thin film. The E_g value

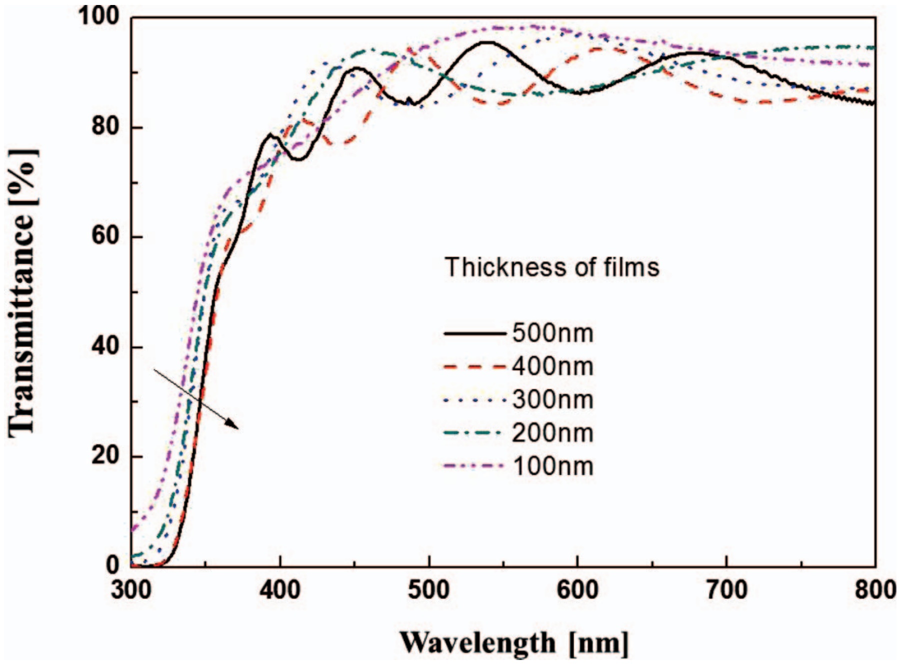

Figure 5. Optical transmittance of Ga-Al doped ZnO thin films with various thicknesses in visible range.

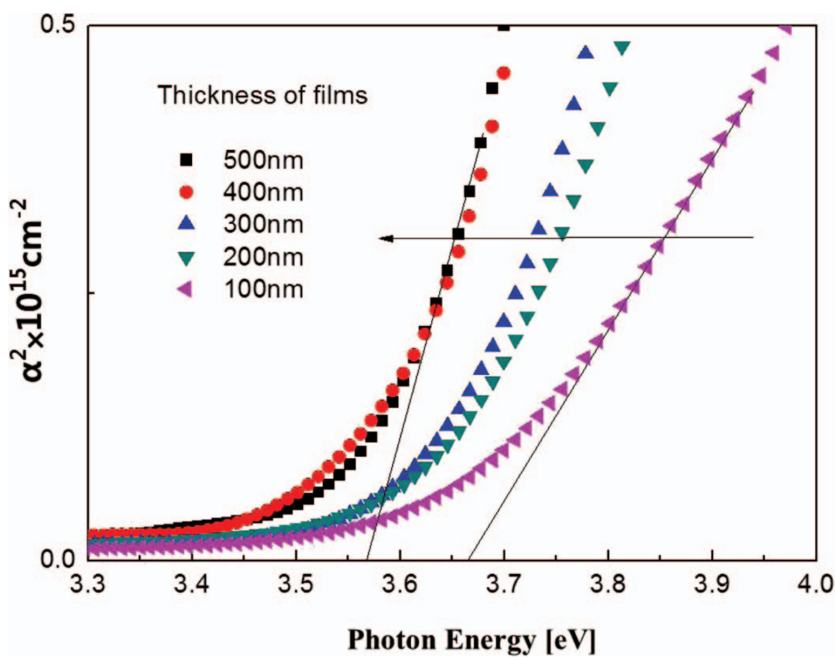

Figure 6. Photon energy of deposited Ga-Al doped ZnO thin film with various thicknesses.

can be obtained by extrapolating the linear segments of the curves towards the x-axis. The E_g value increases from 3.57eV to 3.65eV from 100 nm to 500 nm. It is clearly seen that with decreasing film thickness, the E_g of the films was broadened. This means that the thickness also affects the band gap of the film.

Conclusions

Ga-Al doped ZnO thin films were deposited on glass substrate by using facing targets sputtering with various thicknesses. The structural, electrical and optical properties of the Ga-Al doped ZnO films are dependent on the film thickness. The crystal structure of the Ga-Al doped ZnO films was hexagonal wurtzite and films were highly oriented along the c-axis perpendicular to the substrates. As thickness of film increased, the crystallinity improved and crystal sizes became larger. When the thickness of film increased from 100 nm to 500 nm, the resistivity of films decreased from $1.29\times10^{-3}\Omega$.cm to 4.17×10^{-4} Ω.cm. The average optical transmittance of all deposited films was above 85% in visible range.

Acknowledgment

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