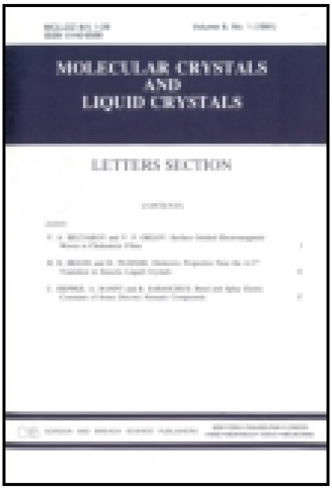

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Characteristics of Ga-Al Doped Zinc Oxide Thin Films Deposited by Facing Targets Sputtering

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Characteristics of Ga-Al Doped Zinc Oxide Thin Films Deposited by Facing Targets Sputtering

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Gallium-aluminum doped zinc oxide (GAZO) thin film is evaluated as an alternative to ITO for use as transparent electrodes for display devices. GAZO transparent conducting thin films were deposited using a facing targets sputtering (FTS) system with a hetero-targets. The FTS system can generate high plasma density, suppress the bombardment of high-energy particles to the substrate, and reduce the working pressure and substrate temperature. The average transmittance achieved with these films was as high as 80% in the visible range. Using a working pressure of 0.4 Pa, the lowest resistivity achieved with the deposited film was 5.342×10^{-4} *Ω·cm.*

Keywords GAZO; thin film; facing target sputtering; transparent electrode; working pressure

Introduction

Transparent conductive oxides (TCOs) have been vital in the development of cutting edge technology such as photovoltaic devices, display devices, and touch panels. Metal oxide thin films possess high optical transparency and electrical conductivity resulting from oxygen vacancies or impurity dopants [1, 2]. Consequently, these materials are potentially applicable as transparent electrodes for various devices. ITO is the metal oxide most widely utilized for transparent electrodes owing to its superior optical and electrical properties. However, the use of ITO suffers from certain limitations, including the increasing demand for ITO due to the expansion of markets for display and solar cells. Stable supplies of indium are restricted by the limited reserves, and the cost of indium also limits it use [3]. Consequently, the development of alternatives to ITO transparent electrodes is required. Numerous studies have focused on utilizing ZnO in optoelectronic devices as a replacement for the ITO electrode. Based on the ubiquitous supply of ZnO, the drawbacks of ITO associated with cost or limited resources could be negated [4]. Furthermore, ZnO can be deposited at low temperature [5] and maintains its stability in hydrogen plasma [6, 7]. However, the conductivity and transmittance of ZnO are lower than those of commercial ITO, making application of ZnO as an electrode for optical devices difficult. Consequently,

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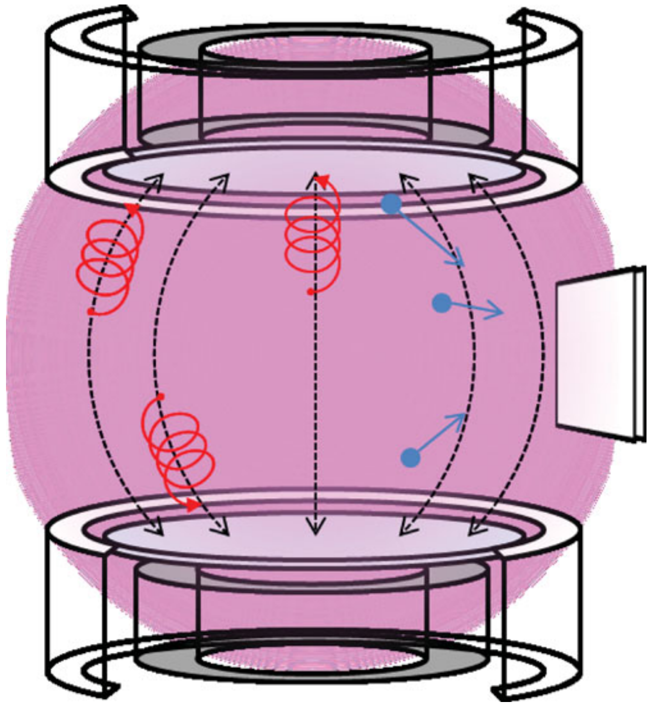

Figure 1. A schematic diagram of the facing targets sputtering system.

the properties of ZnO must be augmented for this purpose. Doping with group III elements such as B [8], Al [9], Ga [10], and In [11] is a prospective means of improving the conductivity of ZnO. As a dopant for ZnO, group III elements possess one additional valence electron. Ga and Al have been found to be the most effective for improving the electrical properties of ZnO, and co-doping with these dopants may enhance the ZnO properties [12]. Herein, Ga-Al co-doped ZnO thin films are deposited via the facing target sputtering (FTS) method using various pressures for the purpose of investigating the influence of pressure during deposition on the properties of the doped ZnO thin films.

Experimental

Gallium-aluminum doped zinc oxide (GAZO) thin films were deposited on a glass substrate via the FTS system using GZO (3 wt.%) and AZO (2 wt.%) targets. Figure 1 shows a schematic diagram of the FTS system, where the two targets face each other. This unique cathode arrangement gives rise to many advantages during deposition. Dispersion of the charged energetic particles is prevented by the magnetic field between each cathode and a high-density, spiral-shaped plasma is readily generated, allowing for an improved deposition rate and uniformity of the thin film under low temperature and pressure conditions. Furthermore, the substrates are placed out of the plasma region. Consequently, bombardment of the substrate by energetic particles is suppressed and the quality of the films is thus improved [13]. Before deposition, the substrates were ultrasonically cleaned in acetone and ethyl alcohol and rinsed in deionized water. The cleaned wet substrates were dried under a jet of nitrogen gas. The chamber was evacuated to a base pressure of 5.33×10^{-4} Pa, and

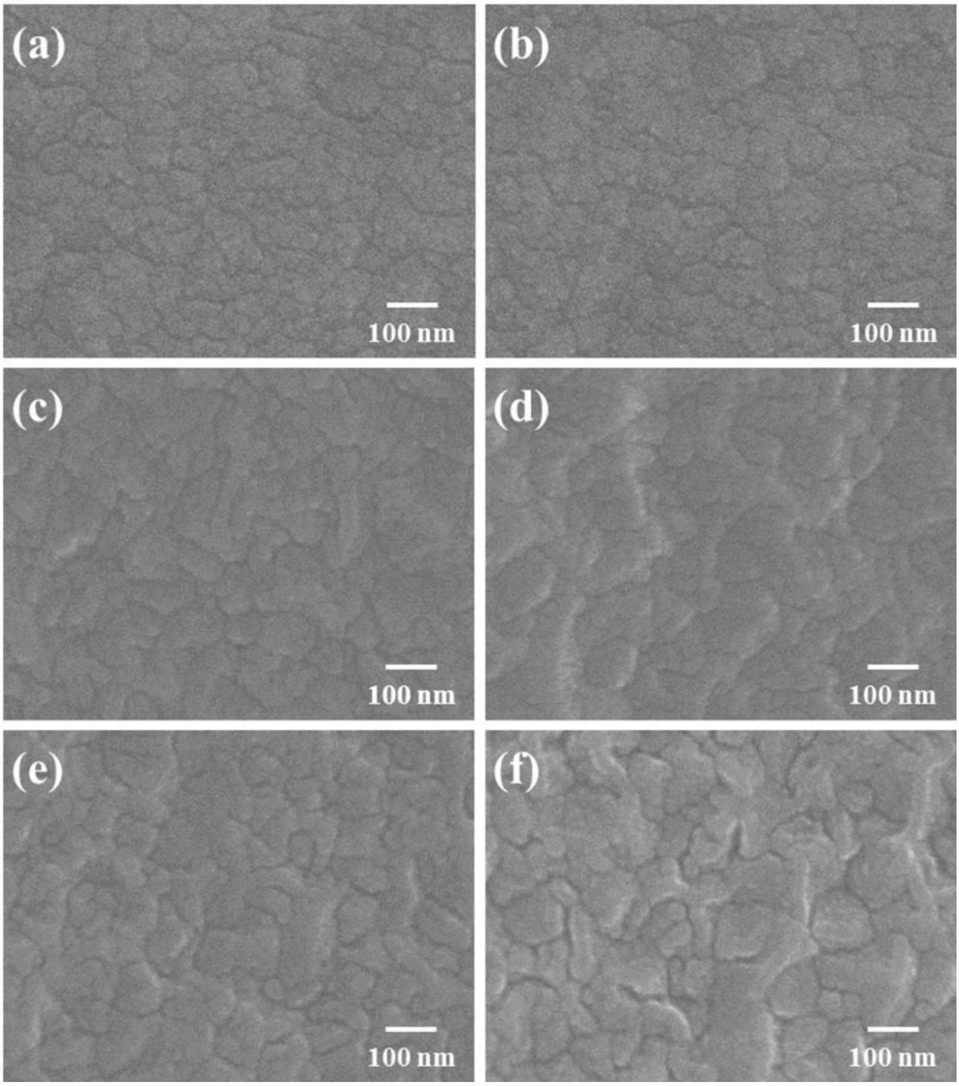

Figure 2. SEM images for GAZO films deposited on a glass substrate at different working pressures: (a) 0.013 Pa, (b) 0.066 Pa, (c) 0.13 Pa, (d) 0.4 Pa, (e) 0.66 Pa, (f) 0.93 Pa.

deposition processes were performed within the pressure range of 0. 013–0.93 Pa under argon. All other sputtering parameters were kept constant. Based on previous research, the thickness of the films was fixed at 500 nm, which gives rise to better electrical properties [14]. To improve the crystallinity and adhesion of the films, the substrates were heated to 250° during the deposition process. The electrical properties of the samples were evaluated using Hall effect measurements (Ecopia, HMS-3000) and calculated using van der Pauw geometry. The structural characteristics were analyzed via X-ray diffraction (Rigaku, D.MAX 2200) within the range of 20° to 80°. The optical properties were evaluated via UV-Vis spectrometry (Agilent, 8453), and the surface morphology of the samples was evaluated using scanning electron microscopy (Hitachi, S-4700).

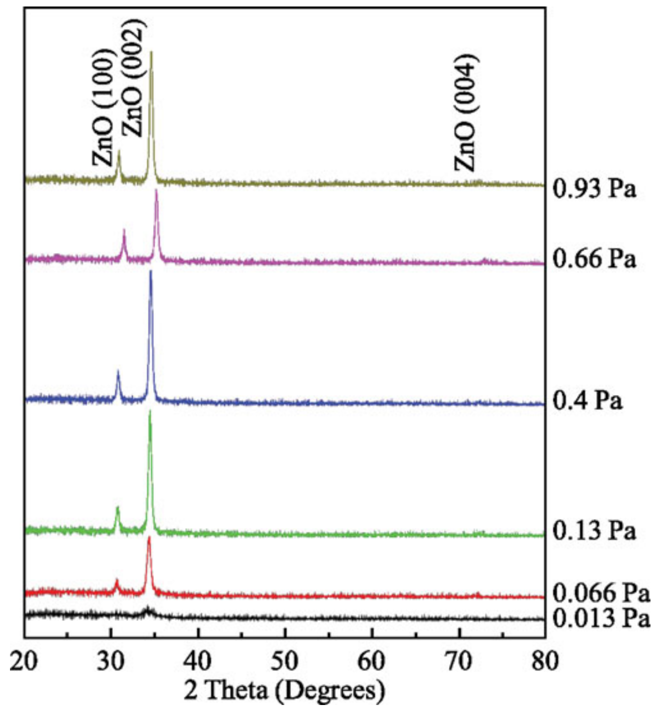

Figure 3. XRD patterns of GAZO films with various working pressure.

Results and Discussion

Figure 2 shows the scanning electron microscopy (SEM) images of the surface of the GAZO films deposited on a glass substrate at different working pressures. The GAZO thin film that was deposited at low pressure was characterized by a dense and continuous surface. In contrast, grains on surface that deposited at high pressure have been separated by voids. These observations demonstrate that the working pressure has a significant impact on the quality of the thin film. At low working pressure, the molecules may travel through a lengthy mean free path and could also reach the surface with higher energy owing to fewer collisions with residual gas particles [15]. The structural properties of the GAZO films are shown in Figure 3. The XRD patterns of all films are characterized by a ZnO (002) peak near 34° and a ZnO (004) peak near 70°. The preferred orientation of thin films is affected by the surface free energy. ZnO may form a hexagonal wurtzite phase; the (001) plane has the lowest surface free energy in this structure [16]. Therefore, during film deposition, the crystals grow preferentially with the (002) orientation. The XRD patterns also show a peak near 31° that was not observed in the patterns of the films deposited at room temperature. This peak is assigned as a (100) oriented peak. During film deposition, particles on the substrate may receive enough energy from the heated substrate, so deposited particles could easily migrate along the surface of the substrate [17]. Consequently, lateral growth of the ZnO crystal may be facilitated. Neither metallic peaks nor other oxide peaks were detected. The absence of these peaks is indicative of the substitution of zinc by group III dopants in the ZnO lattice or segregation of metallic atoms into the noncrystalline region in the grain boundary [10], [18]. A positive shift of the (002) peaks was also observed. This minor shift was attributed to lattice stress. The atomic radii of aluminum and gallium are slightly smaller than that

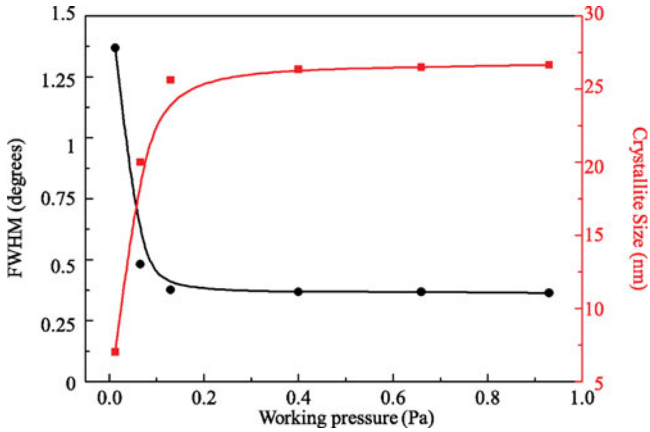

Figure 4. FWHM values and grain size of (002) peaks of GAZO films.

of zinc, and this mismatch of the atomic radii may be the source of lattice stress [19, 20]. Figure 4 shows the full width at half maximum (FWHM) of the (002) peak and the grain size of the GAZO films. The grain size of the films was estimated using Scherrer's equation. The film deposited under a working pressure of 0.4 Pa had the highest crystallinity based on the FWHM or the grain size computed from this data. At low working pressure, bombardment of the sample by energetic particles was increased by fewer collisions owing to the low density of gas molecules in the sputtering chamber. Thus, growth of the crystal could be hampered by this damage. Consequently, increasing the pressure could enhance the crystallinity of the sample because of less damage. However, the frequency of collisions could increase under excessively high pressure leading to deterioration of the crystallinity under the more severe pressure conditions [21]. The electrical properties of the GAZO thin films were evaluated as shown in Figure 5. The lowest resistivity was achieved at a working pressure of 0.4 Pa. Within the range of working pressures utilized, the mobility varies to a greater extent than the carrier concentration. The resistivity is mainly affected by the mobility. Enhanced crystallinity of the thin films may diminish the scattering in the grain

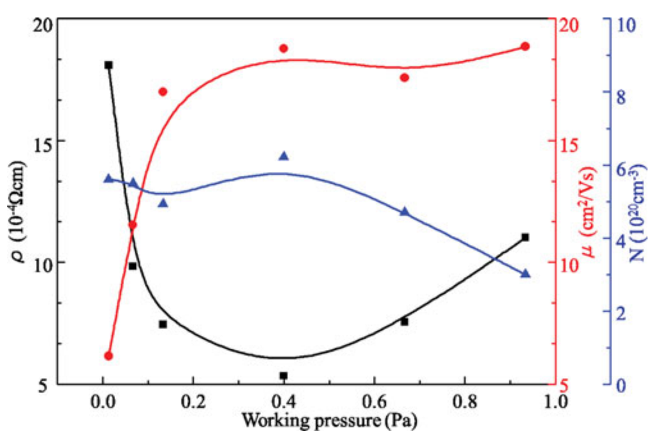

Figure 5. Electrical properties of GAZO films with various working pressure.

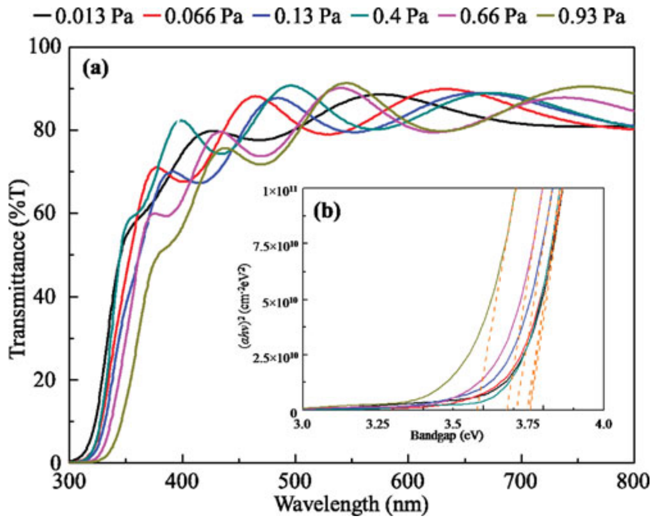

Figure 6. Optical properties of GAZO thin films: (a) Optical transmittance, (b) optical band gap.

boundary and increase the mobility. The lifetime of the carrier may consequently be prolonged with abatement of scattering. Hence, the carrier concentration would concomitantly increase [22]. This discussion is consistent with the observed highest crystallinity being obtained at 0.4 Pa. The optical properties of GAZO films were investigated via UV-Vis spectroscopy. The wide bandgap of the films facilitated transmission of visible light. All of the deposited films exhibited over 80% transmittance in the visible wavelength range. Increasing the working pressure during film deposition narrowed the optical band gap of the films and decreased the transmittance. This may be due to increased optical scattering or formation of donor levels in the band structure by the grain boundary.

Conclusion

GAZO thin films were deposited from hetero-targets by variation of the working pressures in the FTS system. The GAZO thin films had a preferred orientation in the (002) direction. In addition, a (001) oriented peak was detected; these peaks are associated with the temperature effect of lateral growth. However, no peaks corresponding to Ga or Al were observed. The films exhibited an average transmittance of over 80% in the visible region. The highest mobility of 18.78 cm^2/Vs and the lowest resistivity of 5.342×10^{-4} Ω·cm were achieved by deposition of the thin film at 0.4 Pa. In summary, GAZO thin films deposited via the FTS system exhibit adequate properties for application as transparent electrodes in optoelectric devices, and offer the advantages of lowered production cost and a simple generation process.

Funding

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