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Influence of Electron Beam Irradiation on the Structural, Optical, and Electrical Properties of ZTO/Ag/ZTO Trilayer Films

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We deposited transparent conductive ZTO/Ag/ZTO trilayer thin films on glass substrates through magnetron sputtering, and then conducted intense electron beam irradiation on their surfaces to investigate the effects of electron irradiation on the structural, optical, and electrical properties of these films. After deposition, we electron irradiated the ZTO/Ag/ZTO films for 10 min at electron energies of 300, 500, and 700 eV. The films that were electron irradiated at 700 eV showed a higher optical transmittance (84.2%) in the visible wavelength region and a lower resistivity (7.2×10^{-5} Ωcm) compared with the other films. The figure of merit revealed that the ZTO/Ag/ZTO films that were electron irradiated at 700 eV had a higher optical and electrical performance than the other films prepared in this study.

Keywords : ZTO, Ag, Thin film, Magnetron sputtering, Figure of merit

1. INTRODUCTION

The rapidly increasing use of transparent conductive oxide (TCO) films for large displays and solar cells has prompted the research and development of inexpensive TCO materials that possess appropriate optical and electrical properties [1]. Recently, Ga-doped ZnO (GZO) films have attracted attention, due to their high optical transmittance in the visible wavelength range, low electrical resistivity, and relatively low cost compared to conventional Sn-doped In2O3 (ITO) films [2]. In addition, researchers know well that most metal-doped ZnO films require relatively high substrate temperatures (≥ 400℃) to simultaneously increase electrical conductivity and optical transmittance in the visible wavelength range [3]. However, for flexible display applications, high substrate temperatures are undesirable, owing to the low thermal resistivity of polymer substrates. Thus, many researches [4–6] have focused on nonthermal annealing technologies that ensure the high optoelectrical performance of TCO films without causing thermal damage to the polymer substrate.

Thus, many researches have focused on TCO/Ag/TCO trilayer films [4] to enhance electrical conductivity without substrate heating and also on nonthermal annealing technologies that ensure high optoelectrical performance of TCO films without causing thermal damage to the polymer substrate [5,6].

In a previous study, Y. Kim et al. reported on the effect of electron irradiation on the properties of GZO films deposited on PC substrates, with the optoelectrical properties of the films being effectively enhanced by intense electron irradiation without causing thermal damage [7].

In this study, we deposited Sn-doped ZnO (ZTO) films with a 15-nm-thick Ag interlayer through RF and DC magnetron sputtering. After post-deposition, we irradiated the films with an electron beam to investigate the effects of electron irradiation on the structural, electrical, and optical properties of the films, using an X-ray diffractometer (XRD), an atomic force microscope (AFM), and an UV-visible (UV-vis) spectrophotometer, respectively.

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2. EXPERIMENTS

Figure 1 shows the DC and RF magnetron sputtering system with an electron beam source (INFO-RFE-60G, Infovion) used to deposit the ZTO/Ag/ZTO trilayer thin films.

We used transparent glass (40 × 40 mm^2, Corning 1797) as substrate for all depositions. We prepared the ZTO films (50 nm thick) by RF magnetron sputtering of ZTO targets (Zn:SnO_2 =50:50 at%; purity 99.99%, 3-inch diameter, and we prepared the Ag interlayer by DC magnetron sputtering of pure Ag targets (purity 99.95%, 3-inch diameter), respectively. Prior to deposition, we evacuated the chamber to 6.0×10^{-7} Torr, and then injected inert argon (Ar) to obtain a deposition pressure of 1.0×10^{-3} Torr. After deposition, we applied an electrical power of 200 W on an RF (13.56 MHz) coil antenna in the electron beam source.

Table 1 summarizes the experimental conditions used in this study. We measured the thickness of the ZTO and Ag films with a surface profilometer (Dektak 150, Veeco) and an AFM (XE-100, Park Systems), respectively. Crystallinity was measured by using an XRD (X'pert PRO MRD, Philips) with Cu-Kα (0.154 nm) radiation at the Korea Basic Science Institute (KBSI), Daegu Center. We measured optical transmittance and electrical resistivity with a UV-visible spectrophotometer (Carry 100 Cone, Varian) and a Hall Effect measurement system (HMS-3000, Ecopia) in the Van der Pauw configuration, respectively. The glass substrates had a visible transmittance of 90%. The surface roughness was analyzed with an AFM (XE-100, Park Systems) and the optoelectrical performance of the films as transparent conducting electrodes was weighed using a figure of merit (FOM) [8].

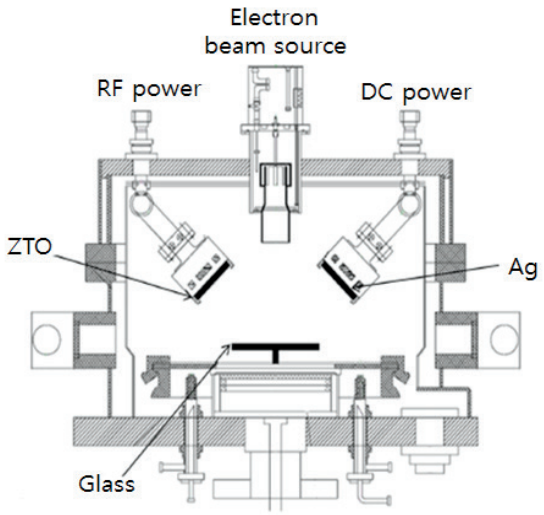

Fig. 1. Experimental RF and DC magnetron sputterer with an electron beam source.

Table 1. Deposition and irradiation conditions of ZTO/Ag/ZTO trilayer films.

Parameter	Conditions
Base pressure (Torr)	6.0×10^{-7}
Deposition pressure (Torr)	1.0×10^{-3}
Target power (W/cm^2)	ZTO; RF 2.5, Ag; DC 2.0
Ar gas flow rate (sccm)	10
Film thickness (nm)	ZTO; 50, Ag; 15
Deposition rate (nm/min)	ZTO; 10, Ag; 40
Electron beam energy (eV)	300, 500, 700

3. RESULTS AND DISCUSSION

Figure 2 shows XRD plots obtained from the ZTO/Ag/ZTO trilayer films for different electron irradiation energies. All XRD plots, regardless of the electron irradiation energy applied, exhibited a weak diffraction peak corresponding to the intermediate Ag (222) thin film, while diffraction peaks for ZnO or SnO_2 crystalline did not appear in the XRD plots.

Recently, Hayashi et al. [9] reported on the growth mode of ZTO thin films, suggesting that ZTO films keep a stable amorphous structure despite being heated to 600℃. Based on the aforementioned report [9] and the XRD patterns shown in Fig. 1, we suppose that crystal growth did not occur for the ZTO films created by RF magnetron sputtering at room temperature conditions.

Figure 3 shows a cross-sectional SEM image of a ZTO/Ag/ZTO trilayer film and Fig. 4 shows the optical transmittance of ZTO/Ag/ZTO films in the visible wavelength region. While the as-deposited films and the films that were electron irradiated at 300 and 500 eV did not differ in optical transmittance, the films that were electron irradiated at 700 eV showed a higher optical transmittance of 84.2%.

Table 2 shows the electrical properties of the electron-irradiated ZTO/Ag/ZTO films. We attribute the increased carrier mobility caused by higher electron irradiation energy to the flat surface morphology observed by AFM analysis. While the as-deposited films had a resistivity of 8.5×10^{-5} Ωcm, the films that were electron irradiated

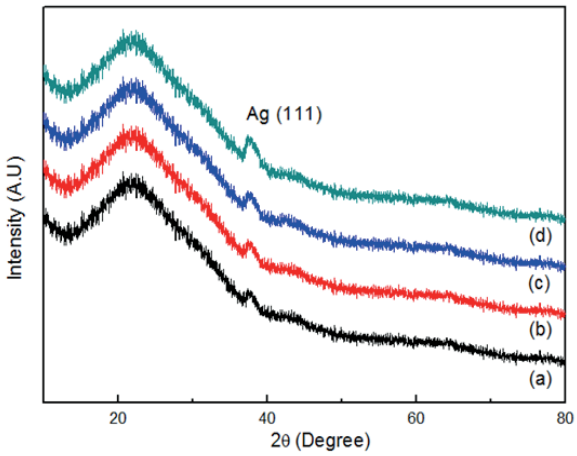

Fig. 2. XRD pattern of the ZTO/Ag/ZTO trilayer films for electron beams irradiated at different energies. (a) As deposited, (b) 300 eV, (c) 500 eV, and (d) 700 eV.

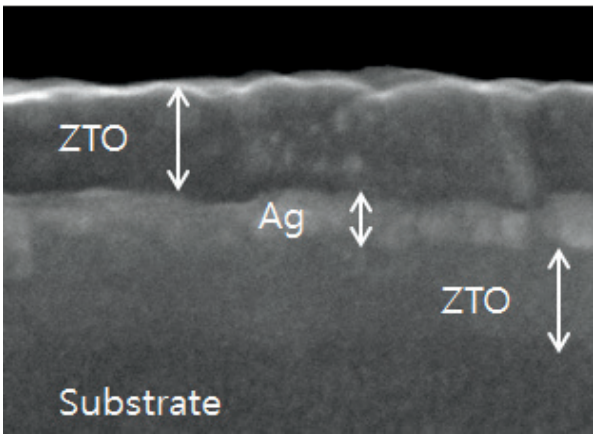

Fig. 3. Cross-sectional SEM image of ZTO/Ag/ZTO trilayer film.

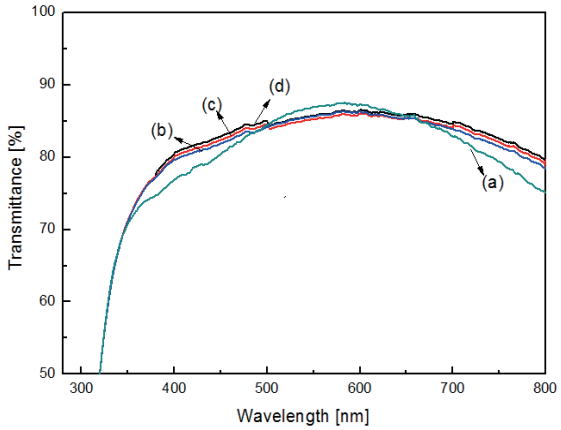

Fig. 4. Optical transmittance spectra in the visible wavelength region for the as-deposited and the electron-irradiated ZTO/Ag/ZTO films. (a) As deposited, (b) 300 eV, (c) 500 eV, and (d) 700 eV.

Table 2. Electrical properties of ZTO/Ag/ZTO films that were electron irradiated at different energies.

Irradiation energy (eV)	Carrier density ($\times 10^{21}$ cm^{-3})	Carrier mobility (cm^2/Vs)	Electrical resistivity ($\times 10^{-5}$ Ω cm)
As deposition	2.87	25.0	8.5
300	2.85	26.9	8.1
500	2.81	28.8	7.7
700	2.73	31.5	7.2

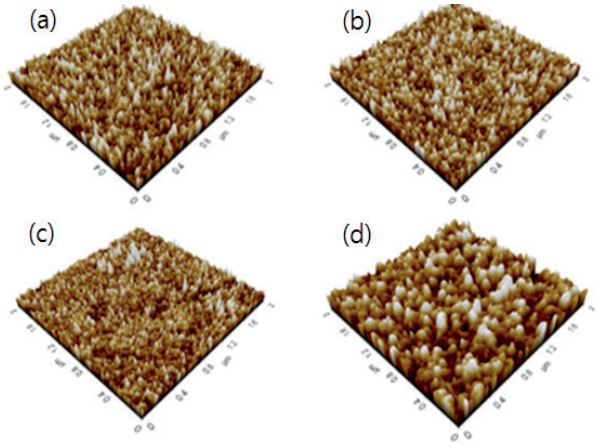

Fig. 5. Surface morphology and RMS roughness of the ZTO/Ag/ZTO trilayer films electron beam irradiated at different energy. (a) As deposited, RMS roughness: 0.75 nm, (b) 300 eV, RMS roughness: 0.73 nm, (c) 500 eV, RMS roughness: 0.62 nm, and (d) 700 eV, RMS roughness: 0.51 nm.

at 700 eV had the lowest resistivity of 7.2×10^{-5} Ωcm.

Figure 5 shows AFM images of the as-deposited and the electron beam-irradiated ZTO/Ag/ZTO films. We found that surface roughness decreased as electron irradiation energy increased. While the as-deposited films showed an RMS roughness of 0.75 nm, the films that were electron irradiated at 700 eV showed the lowest RMS roughness, measured at 0.51 nm.

In a previous study, V. Teixeira reported that two factors, namely absorption and scattering, can cause a reduction of visible light passing through optical films, with light scattering caused by either surface micro-roughness or by scattering at the film grain boundaries

Table 3. Figure of merit of ZTO/Ag/ZTO films that were electron irradiated at different energies.

Irradiation energy (eV)	Sheet resistance (Ω/□)	Optical transmittance (%)	Figure of merit (10^{-2} Ω^{-1})
As deposition	7.4	83.2	2.1
300	7.0	83.5	2.3
500	6.7	83.7	2.5
700	6.3	84.2	2.8

[10]. This report may mean that we could attribute the enhanced optical transmittance of the annealed ZTO/Ag/ZTO films to a flatter surface caused by the post-deposition electron irradiation.

Table 3 shows the mean optical transmittances in the visible wavelength region, sheet resistances (Rs), and figures of merit (FOM) as a function of electron irradiation energy. The FOM is defined as FOM = T^{10} / R_{sh}, where T is the optical transmittance in the visible wavelength region, and R_{sh} is sheet resistance [11,12]. The FOM reached a maximum value of 2.8×10^{-2} Ω^{-1} for the electron-irradiated films at 700 eV, higher than that of the as-deposited films, which was measured at 2.1×10^{-2} Ω^{-1}.

4. CONCLUSIONS

We deposited ZTO/Ag/ZTO films onto glass substrates by RF and DC magnetron sputtering and then superficially irradiated the films with an electron beam to investigate the effects of electron irradiation on the structural, optical, and electrical properties of these films.

Surface roughness decreased with electron irradiation, and we attributed the fairly high carrier mobility of the films to their flat surface morphology caused by intense electron irradiation. The FOM reached a maximum of 2.8×10^{-2} Ω^{-1} for electron-irradiated films at 700 eV, which was higher than the FOM of the as-deposited films, which was 2.1×10^{-2} Ω^{-1}.

Thus, we suppose that electron irradiation is effective for improving the optical and electrical performance of ZTO/Ag/ZTO films.

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