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Enhanced Optical and Electrical Properties of TiO_2 Buffered IGZO/TiO_2 Bi-Layered Films

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Abstract: In and Ga doped ZnO (IGZO, 100-nm thick) thin films were deposited by radio frequency magnetron sputtering without intentional substrate heating on a bare glass substrate and a TiO_2-deposited glass substrate to determine the effect of the thickness of a thin TiO_2 buffer layer on the structural, optical, and electrical properties of the films. The thicknesses of the TiO_2 buffer layers were 5, 10 and 15 nm, respectively. As-deposited IGZO films with a 10 nm-thick TiO_2 buffer layer had an average optical transmittance of 85.0% with lower resistivity (1.83×10^{-2} Ω cm) than that of IGZO single layer films. The figure of merit (FOM) reached a maximum of 1.44×10^{-4} Ω^{-1} for IGZO/10 nm-thick TiO_2 bi-layered films, which is higher than the FOM of 6.85×10^{-5} Ω^{-1} for IGZO single layer films. Because a higher FOM value indicates better quality transparent conducting oxide (TCO) films, the IGZO/10 nm-thick TiO_2 bi-layered films are likely to perform better in TCO applications than IGZO single layer films.

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Keywords: thin films, sputtering, optical properties, electrical properties, AFM.

1. INTRODUCTION

There is considerable interest in using metal-doped ZnO films such as AZO [1], GZO [2], and IGZO [3] films as transparent conducting oxide (TCO) layers for transparent electrodes because of their high optical transmittance in the visible wavelength range and greater chemical stability compared to conventional Sn-doped In_2O_3 (ITO) films.

Among these films, IGZO films have been used as an active layer in transparent thin film transistors (TTFTs) because the films can be deposited onto large area substrates as a uniform amorphous phase while retaining high carrier mobility [4]. However, it is has also been reported that a relatively high substrate temperature or post-deposition annealing is required to obtain IGZO films with simultaneous optimal transparency and low resistivity [5].

One way to improve the optical and electrical properties of IGZO films without substrate heating is to use IGZO/metal/IGZO (OMO) structures [6], which have lower resistivity than IGZO single layer films. Another strategy that has been used to improve the electrical properties of IGZO films has been to insert an oxide buffer layer under the IGZO film [3].

In this study, IGZO thin films were deposited by radio frequency (RF) magnetron sputtering on glass substrate with and without a TiO_2 buffer layer, and then the effects of the TiO_2 buffer layer thickness on the optical and electrical properties of the IGZO films were investigated using X-ray diffraction (XRD), atomic force microscopy (AFM), Hall effect measurements, UV-visible spectrophotometry, and UV photoelectron spectroscopy (UPS).

2. EXPERIMENTAL PROCEDURES

IGZO single layer and IGZO/TiO_2 bi-layered films were deposited by RF magnetron sputtering on glass substrate (Corning 1797, 2×2 cm^2) without intentional substrate heating. Both IGZO (In_2O_3 : Ga_2O_3 : ZnO = 1:1:1 at%) and TiO_2 targets had 3-inch diameters. Glass substrates were degreased in a dilute detergent solution, rinsed in de-ionized water, and blown dry in nitrogen gas before they were set on the substrate holder in the chamber. The distance between the target and substrate was kept at 60 mm. Prior to deposition, the sputter chamber was evacuated to 6.0×10^{-7} Torr and then 10 sccm Argon (Ar) gas was injected to achieve an optimized working pressure of 1.0×10^{-3} Torr. Table 1 shows the experimental conditions for IGZO film and TiO_2 buffer layer

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Table 1. The experimental conditions for IGZO film and TiO_2 buffer layer deposition.

Parameters	Condition
Base pressure (Torr)	6.0×10^{-7}
Deposition pressure (Torr)	1.0×10^{-3}
IGZO target RF Power (W/cm^2)	2.5
TiO_2 target RF Power (W/cm^2)	2.5
Ar gas flow rate (sccm)	10
Film thickness (nm)	IGZO: 100 TiO_2: 5, 10, 15
Deposition rate (nm/min)	IGZO: 3.4, TiO_2: 1.1

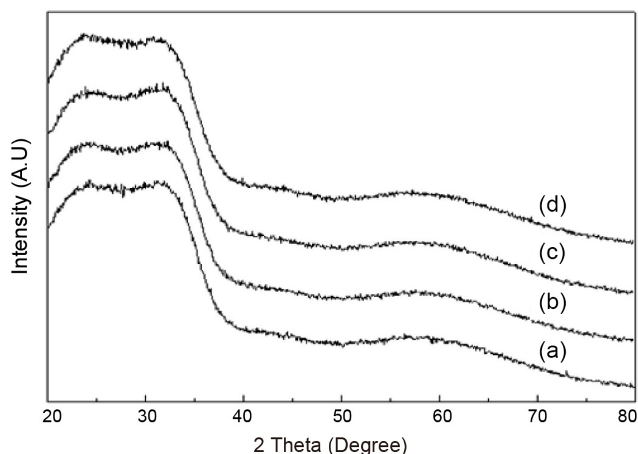

Fig. 1. The XRD patterns of the IGZO single layer and IGZO/TiO_2 bi-layered films: (a) 100 nm-thick IGZO films, (b) IGZO/5 nm-thick TiO_2 films, (c) IGZO 10 nm-thick TiO_2 films, and (d) IGZO/15 nm-thick TiO_2 films.

deposition. The deposition rates of the IGZO and TiO_2 films were evaluated with a surface profiler (Dektak-150, Varian) and the crystallinity of the films was investigated by XRD (X'pert PRO MRD, Phillips) analysis at the Korea Basic Science Institute (KBSI, Daegu Center). Optical transmittance in the visible wavelength region (400-700 nm) was determined using a UV-Vis. spectrophotometer (Cary 100 Cone, Varian). Variations in surface morphology (2×2 μm^2 scanning area) and root mean square (RMS) roughness of the films were characterized by AFM [7] with the microscope operated in contact mode (XE-100, Park Systems). Electrical properties of the films were investigated by Hall effect measurements employing the Van der Pauw technique (HMS-3000, Ecopia). The opto-electrical performances of IGZO and IGZO/TiO_2 films as TCO electrodes were compared using the figure of merit (FOM). In addition, the influence of the TiO_2 buffer layer on the work function of IGZO films was evaluated by UPS analysis.

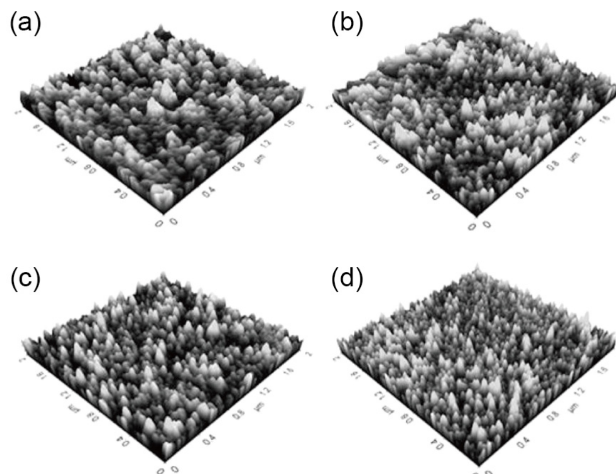

Fig. 2. Surface morphology and RMS roughness of the IGZO single layer and IGZO/TiO_2 bi-layer films: (a) 100 nm-thick IGZO films, (b) IGZO/5 nm-thick TiO_2 films, (c) IGZO/10 nm-thick TiO_2 films, and (d) IGZO/15 nm-thick TiO_2 films.

3. RESULTS AND DISCUSSION

Figure 1 shows the XRD patterns of the IGZO single layer and IGZO/TiO_2 bi-layered films. The observed broad peak in the range of 20° to 40° is an amorphous halo-peak contributed by the IGZO film [3]. This means that IGZO films with the composition of In:Ga:Zn =1:1:1 at% have an amorphous crystal structure.

Kim et al. reported that when the annealing temperature was below 600 ℃, no crystalline peaks were present in the XRD pattern of IGZO films [8]. Based on these previous reports and the XRD patterns shown in Fig. 1, it appears that crystal growth did not occur for the IGZO/TiO_2 films deposited at room temperature.

Surface morphologies and RMS roughness of the IGZO single layer and IGZO/TiO_2 films with different TiO_2 buffer layer thicknesses are shown in Fig. 2. Surface roughness of the TCO films is an important determinant of their optical and electrical properties. The RMS roughness of the IGZO single layer film (1.5 nm) was higher than that of the IGZO/TiO_2 films. The presence of a TiO_2 buffer layer appeared to

Table 2. The electrical properties of the IGZO single layer and $IGZO/TiO_2$ bi-layered films.

Films	Carrier density ($\times 10^{19}$ cm^{-3})	Mobility (cm^2 $V^{-1}S^{-1}$)	Resistivity ($\times 10^{-2}$ Ω cm)
IGZO 100 nm	4.98	4.11	3.05
$IGZO/TiO_2$ 5 nm	5.35	4.53	2.58
$IGZO/TiO_2$ 10 nm	6.14	5.54	1.83
$IGZO/TiO_2$ 15 nm	2.24	8.77	3.18

Table 3. The comparison of optical and electrical properties of IGZO single layer and $IGZO/TiO_2$ bi-layered films.

Films	Sheet resistance [Ω/□]	Transmittance [%]	Figure of merit [$\times 10^{-5}$ Ω^{-1}]
IGZO 100 nm	3050	85.5	6.85
$IGZO/TiO_2$ 5 nm	2457	86.9	9.99
$IGZO/TiO_2$ 10 nm	1664	86.6	14.4
$IGZO/TiO_2$ 15 nm	2765	85.8	7.82

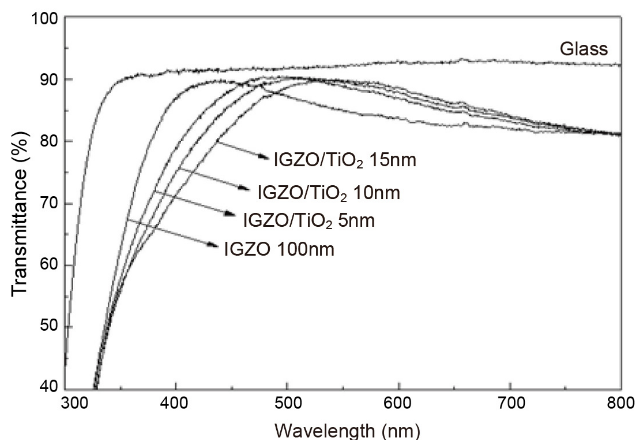

Fig. 3. The optical transmittance for IGZO single layer and $IGZO/TiO_2$ bi-layered films.

enhance the flatness of the $IGZO/TiO_2$ films. This indicates that the TiO_2 buffer layer grew preferably in sunken and defective regions of the substrate during deposition.

Thus, the $IGZO/TiO_2$ films had a flatter surface than the IGZO single layer films. The IGZO films with a 15 nm-thick TiO_2 buffer layer had a lower RMS roughness value (0.8 nm) than the other films prepared in this study. In a previous study, we reported similar results; a TiO_2 buffer layer resulted in the decreased RMS roughness of the upper GZO film in the context of GZO/TiO_2 bi-layered films [9].

Table 2 shows the influence of the TiO_2 buffer layer on the electrical properties of the films. IGZO/10 nm-thick TiO_2 films had a lower resistivity (1.83×10^{-2} Ω cm) than IGZO single layer films due to increases in both carrier concentration and mobility in the former. The highest mobility of 8.77 cm^2 V^{-1} s^{-1} was observed for IGZO/15 nm-thick TiO_2 film; we attributed this to the flat surface morphology of the films, as shown in Fig. 2.

Figure 3 shows the optical transmittance of the IGZO and $IGZO/TiO_2$ films. The average transmittance in the visible range was about 85.5% for the IGZO film, while the transmittance of IGZO/10 nm-thick TiO_2 film was about 86.6%.

Optical and electrical properties of the films are compared in Table 3. The figure of merit (FOM) is an important index for evaluating the performance of transparent conducting oxide (TCO) films [10]. FOM is defined as

$$FOM = T^{10} / R_s$$

where T is the optical transmittance and R_s is the sheet resistance. The FOM reached a maximum of 1.44×10^{-4} Ω^{-1} for IGZO films with a 10 nm-thick TiO_2 buffer layer, which is higher than the FOM of 6.85×10^{-5} Ω^{-1} for IGZO single layer films. Because a higher FOM value indicates a better quality TCO film, the IGZO film with a 10 nm-thick TiO_2 buffer layer will likely perform better in TCO applications than IGZO single layer films.

The high work function of TCO films, which is close to the value of the highest occupied molecular orbital (HOMO) of the organic layer, allows hole injection from TCO to the organic layer of an organic light-emitting diode (OLED), which results in a decrease in the turn-on voltage of the OLED [9]. However, the work function of conventional ITO films is lower than the HOMO of the organic layer of OLEDs. Thus, several techniques have been developed to increase the work function of ITO [11].

Figure 4 shows the kinetic energy cut-off spectra obtained for the IGZO/10 nm-thick TiO_2 film. This allowed determination of the work function values directly from the spectra by fitting straight lines into their kinetic energy cut-offs and determining the intersections with the baselines of the

Table 4. Comparison of work function of the ITO/Ni/ITO trilayer, IGZO single layer and $IGZO/TiO_2$ bi-layered films.

Films	Work function (eV)
ITO/Ni/ITO	4.51 [11]
IGZO	4.10 [This study]
$IGZO/TiO_2$ 10 nm	4.61 [This study]

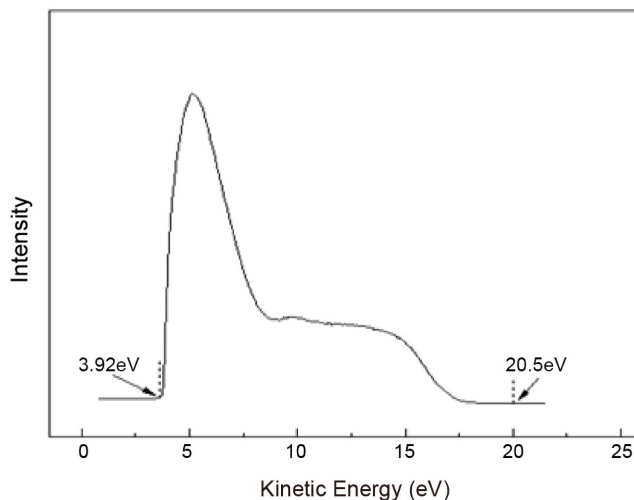

Fig. 4. The kinetic energy cut-off spectra obtained from the IGZO/10 nm-thick TiO_2 bi-layered films.

spectra. The work functions of ITO/Ni/ITO trilayer [11], IGZO single layer, and $IGZO/TiO_2$ bi-layered films are compared in Table 4. The IGZO/10 nm-thick TiO_2 films had a higher work function (4.61 eV) than the other films. Thus, the addition of a TiO_2 buffer layer is a useful strategy to increase the work function of IGZO films.

4. CONCLUSIONS

Both IGZO single layer and $IGZO/TiO_2$ bi-layered films were deposited by RF magnetron sputtering on glass substrates without intentional substrate heating.

From AFM observations, it was apparent that the TiO_2 buffer layer enhanced the flatness of the $IGZO/TiO_2$ films, and the figure of merit of the $IGZO/TiO_2$ films reached a maximum value of $1.18\times10^{-4}\ \Omega^{-1}$, which was higher than that of the IGZO films. In addition, IGZO/10 nm-thick TiO_2 films had a higher work function than IGZO single layer films.

The observed results indicated that the structural, optical, and electrical properties of the IGZO films were dependent on the TiO_2 buffer layer. The 10 nm-thick TiO_2 buffer layer in $IGZO/TiO_2$ films yielded films with better opto-electrical performance than IGZO single layer films.

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