RESEARCH ARTICLE OPEN ACCESS

Investigation of transparent conductive multilayer film with Ag

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ABSTRACT
Recently, transparent conductive oxide films (TCOs) have been used for their application as plastic substrates that can be applied to flexible devices. We focus on transparent conductive films with TCO/metal/TCO multilayer structures. In this study, an AZO/Ag/AZO multilayer film was deposited on a PEN substrate by RF magnetron sputtering (for AZO layer) and electron beam evaporation (for Ag layer). The resistivity of multilayer films decreased considerably compared to single AZO films. The XRD spectra revealed that the resistivity of the multilayer film was barely affected by the crystallinity of AZO. An increase in the carrier concentration caused by Ag affects was also observed. Furthermore, the transmittance of the multilayer film was improved owing to an increase in the oxygen content of the AZO film as well as the uniformity of Ag layer. It was found that the multilayer film of AZO(35 nm)/Ag(13 nm)/AZO(35 nm) exhibited the resistivity of 2.16×10^{-4} Ω cm and an average transmittance of 81%.

Keywords **-** Flexible transparent conductive films, AZO/Ag/AZO multilayer films, Low temperature, RF Magnetron sputtering, Electron beam evaporation

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Date of Submission: 11-12-2017 Date of acceptance: 26-12-2017

I. INTRODUCTION

Transparent conductive oxide films (TCOs) are used in many electronic devices, such as solar cells, flat display, and OLED [1-3]. Recently, TCOs have been reported to be used as plastic substrates that can be applied to flexible devices [4-5]. Among TCOs, Indium tin oxide (ITO) is commonly utilized because of its low resistivity and high transmittance among TCOs [6-7]. However, ITO has a problem of inflexibility. Therefore, various alternative materials for ITO have been studied [8-9]. Among them, ZnO thin films doped with Al or Ga have garnered much attention because ZnO exhibits high mechanical stress and high conductivity when it is deposited at a high temperature (>200 ℃) [10-11]. However, the formation of plastic substrates is usually performed at low temperatures, while substrate distortion could occur during deposition at high temperatures. In this study, we focus on transparent conductive films with AZO/Ag/AZO multilayer structures that can be formed at a lower temperature than that required for ITO formation. Because this multilayer structure film is metallic, high conductivity is achieved when it is formed at a low temperature [12-13]. Moreover, this multilayer film presents more robust mechanical properties than TCO single layers which have been reported previously [14-15]. The effects of the thicknesses of the AZO and Ag layers on optical and electrical properties of the transparent conductive film were also investigated.

II. EXPERIMENTAL

The PEN substrate was cleaned for 20 min using an UV ozone cleaning system (ASM401N, Asumi Giken). AZO layer was deposited by RF magnetron sputtering using an AZO ceramic target (diameter 48 mm, purity 99.99%, Al2O3: 2 wt.%). The deposition conditions of RF magnetron sputtering are shown in Table 1. Ag layer was deposited via electron beam evaporation using Ag grain (purity 99.99%). The deposition conditions of electron beam evaporation are shown in Table 2. The AZO/Ag/AZO film was deposited layer-by-layer without breaking the vacuum. The electrical resistance of the film was measured using a digital multimeter (VOAC7521-H Iwatsu). Optical transmittance of the film in the visible range (380–780 nm) was characterized using a spectrophotometer (U-3900 Hitachi). The orientation and crystallinity of the film were characterized using X-ray diffraction (Ultima-IV Rigaku), while its composition was measured through energy dispersive X-ray spectrometry (Quantax70 BRUKER).

Table. 1 Deposition conditions of RF magnetron sputtering

RF magnetron sputtering	
Sputtering pressure (Pa)	0.4
Substrate temperature (℃)	50
RF power (W)	40
Ar flow rate (sccm)	5
Target to substrate distance (mm)	150
Ultimate vacuum (Pa)	3.0×10^{-5}

Table. 2 Deposition conditions of electron beam evaporation

Electron beam evaporation	
Chamber Pressure (Pa)	3.0×10^{-4}
Substrate temperature (℃)	50
Accelerating voltage (kV)	5.5
Emission current (mA)	50
Evaporation to substrate distance (mm)	440

III. RESULTS & DISCUSSION

3.1. Examination of the thickness of AZO layer

For determining the AZO layer thickness, comparisons were made between single AZO films with different thicknesses and AZO multilayer films with Ag inserted in the middle layer. The thicknesses single AZO layers were 70 and 100 nm. For AZO/Ag/AZO multilayer film, the inserted Ag thickness was 10 nm, while the thicknesses of multilayer films were AZO(50 nm)/Ag(10 nm)/AZO(50 nm) and AZO(35 nm)/Ag(10 nm)/AZO(35 nm). Fig. 1 shows the XRD spectra of the single AZO film and AZO/Ag/AZO multilayer film. The (002) peaks at 34.3° belonging to ZnO were observed in all films. These XRD results indicate that the AZO thin film had a hexagonal structure and were preferentially oriented in a direction perpendicular to the substrate along c-axis. In addition, the (111) peaks at 37.9° belonging to Ag were observed in the AZO with Ag inserted. In comparison with single AZO films, the crystalline and orientation of AZO/Ag/AZO films decreased. This decreased was probably caused because the AZO crystal in multilayer films did not grow continuously because of the inserted Ag.

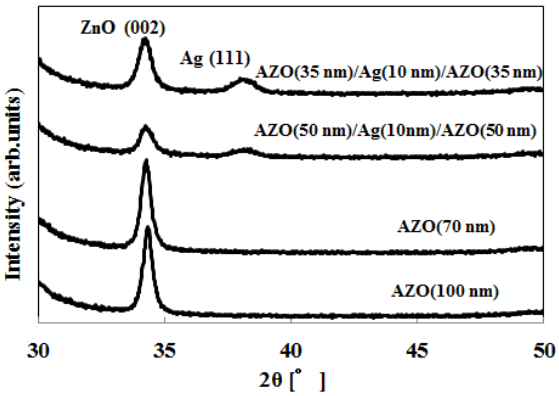

Fig. 1 XRD spectra of single AZO films and AZO/Ag/AZO films with different thicknesses.

Table 3 shows the resistivity, average transmittance, and composition ratio (O/Zn) of single AZO films and AZO/Ag/AZO films, and Fig. 2 shows their transmittance spectra. The transmittance in the visible range decreased with an increase in Ag content. In comparison, the 70-nm thick AZO film contained more oxygen and exhibited higher transmittance than the 100-nm thick AZO film. Previous studies have shown that the transmittance improves as the oxygen content increases since heat is likely to be transferred easily in a thin film, thus increasing the oxygen content [16]. The resistivity of multilayer films decreased considerably regardless of the thickness of AZO; this decrease was possibly due to an increase in carrier concentration [17-18]. Besides, the XRD spectra revealed that the resistivity of the multilayer film was barely affected by the AZO crystallinity and increase in carrier concentration due to Ag affects.

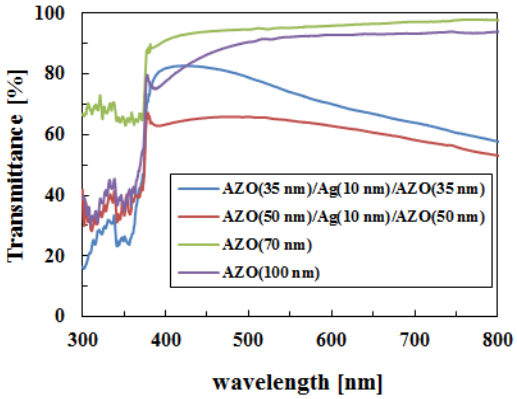

Fig. 2 Transmittance spectra of the single AZO films and AZO/Ag/AZO films.

Table. 3 Resistivity, average transmittance, and composition ratio (O/Zn) of the single AZO films and AZO/Ag/AZO films.

Material	Resistivity (Ω・cm)	Average transmittance (%)	Composition ratio (O/Zn)
AZO (70 nm)	5.64×10^{-3}	95.3	0.762
AZO (100 nm)	2.35×10^{-3}	90.2	0.552
AZO(35 nm)/Ag(10 nm)/AZO(35 nm)	2.22×10^{-4}	71.9	0.716
AZO(50 nm)/Ag(10 nm)/AZO(50 nm)	3.14×10^{-4}	62.0	0.483

3.2. Examination of the thickness of Ag layer

Considering the transmittance and resistivity, the top and bottom AZO layers were maintained constant at 35 nm, and the Ag layer thickness varied between 8 and 15 nm. Fig. 3 shows the XRD spectra of the AZO(35nm)/Ag/AZO(35nm) films as a function of Ag layer thickness. Similarly, the (002) peaks at 34.3° of ZnO and (111) peaks at 37.9° of Ag were observed. The crystallinity and orientation of Ag were improved owing to an increase in the thickness of the Ag layer, while no change was observed in the crystallinity of AZO.

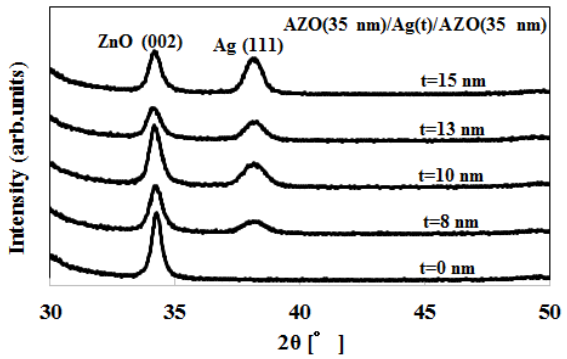

Fig. 3 XRD spectra of the AZO(35nm)/Ag/AZO(35nm) films as a function of Ag layer thickness.

Figs. 4 and 5 show the resistivity and the transmittance spectra of the AZO(35 nm)/Ag/AZO(35 nm) films as a function of Ag layer thickness. The resistivity decreases with an increase in Ag layer thickness because carrier concentration possibly increases with an increase in Ag layer thickness. As shown in Fig. 5, the highest average transmittance of 81.0% is achieved when the Ag thickness is 13 nm. The transmittance was improved when the Ag layer thickness increased from 10 nm to 13 nm. The Ag layer was at an inhomogeneous state at thicknesses of 8 and 10 nm. The separated Ag grains would scatter the incident light; this would decrease the transmittance [19-20]. When the thickness exceeded 13 nm, the Ag layer became uniform and the transmittance was improved. However, a further increase in Ag thickness of up to 15 nm resulted in layer reflection and decreased the transmittance [21].

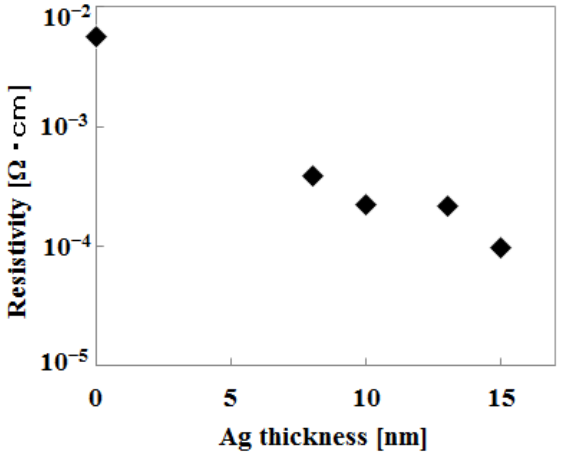

Fig. 4 Resistivity of AZO(35 nm)/Ag/AZO(35 nm) films as a function of Ag layer thickness.

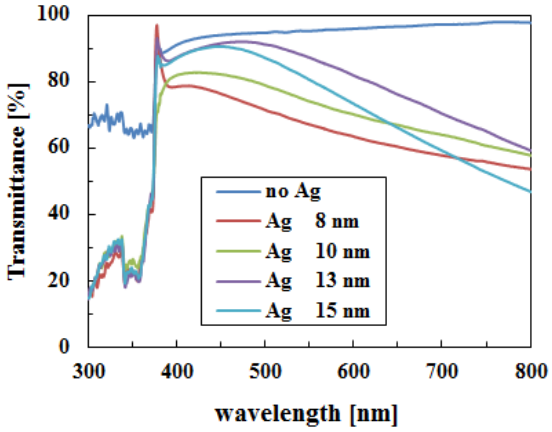

Fig. 5 Transmittance spectra of AZO(35 nm)/Ag/AZO(35 nm) films as a function of Ag layer thickness.

IV. CONCLUSION

The purpose of this study was to develop transparent conductive films with AZO/Ag/AZO structures at lower temperatures than that required for ITO formation. The effect of AZO and Ag thickness on optical and electrical properties of the transparent conductive film was studied. As compared to single AZO, the resistivity decreased when Ag was inserted into the multilayer. The resistivity of the multilayer film is barely affected by the crystallinity of AZO, and the carrier concentration increases owing to Ag effects. In

comparison, AZO with a thickness of 70 nm contains more oxygen and higher transmittance than the AZO with a thickness of 100 nm. When the film thickness of Ag exceeds 13 nm, the Ag layer became uniform and the transmittance was improved. However, when the Ag layer thickness increased up to 15 nm, the transmittance was reduced by Ag layer reflection. The multilayer film of AZO(35 nm)/Ag(13 nm)/AZO(35 nm) exhibited a resistivity of 2.16×10^{-4} Ω・cm and an average of transmittance 81%. Further studies are necessary to improve the transmittance in the near-infrared region.

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Yuichiro Maeda "Investigation of transparent conductive multilayer film with Ag." International Journal of Engineering Research and Applications (IJERA) , vol. 7, no. 12, 2017, pp. 05-09.