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High Quality Transparent Conductive ITO/Ag/ITO Multilayer Films Deposited on Glass Substrate at Room Temperature

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In this study, we report the electrical and optical properties of ITO/Ag/ITO multilayer films deposited on glass substrate by in-line RF sputtering of ITO and DC sputtering of Ag at room temperature. The physical properties of the ITO/Ag/ITO multilayer films changed significantly with the thickness of Ag interlayer. The ITO (43 nm)/Ag (16.1 nm)/ITO (43 nm) multilayer films exhibited both good transmittance (79.4% at 550 nm) and low sheet resistance (8.9 Ω/sq.) due to the combined effects of top ITO layer acting as anti-reflection layer and Ag thin interlayer acting as conductive layer. Furthermore, the maximum figure of merit value of the ITO/Ag/ITO multilayer films was obtained at a Ag thickness of 16.1 nm.

Keywords Ag thin interlayer; indium tin oxide (ITO); multilayer; transparent conductive oxide (TCO)

Introduction

Transparent conductive oxides (TCO), such as indium tin oxide (ITO) have attracted much interest in various optoelectronic devices including solar cells, liquid crystal displays, and organic light emitting diodes (OLED) due to their low sheet resistance and high optical transparency in visible region [1–3]. In general, the sputtering and post annealing as high as 250–350°C is required to obtain high quality ITO electrodes with a low sheet resistance ($<10\,\Omega$/sq.) and a high optical transmittance ($>80\%$) for various display applications [4,5]. However, ITO films deposited on the flexible substrate at room temperature show higher sheet resistance than that of

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ITO electrode fabricated on glass substrate. It is usual that the low temperature processed ITO films have at least one order of magnitude higher electrical characteristics compared to those of the high temperature processed ITO electrodes. This reason limits the use of ITO films in optoelectronic applications. A possible method to overcome this problem is to employ very a thin metal interlayer as conducting layer between ITO layers. Several papers have been reported on the ITO/metal/ITO type multilayer films with sheet resistance lower than the single layer ITO glass in which the metal layer included Ag, Au and Cu [6–9].

In this study, we report the electrical and optical properties of ITO/Ag/ITO multilayer films deposited on glass substrate by in-line RF sputtering of ITO and DC sputtering of Ag at room temperature, and discuss the effect of Ag interlayer on the electrical and optical properties of ITO/Ag/ITO multilayer films.

Experimental

ITO/Ag/ITO multilayer films were sequentially deposited on glass substrate (thickness = 0.7 mm) by in-line RF sputtering of ITO and DC sputtering of Ag at room temperature. The sputtering was conducted at room temperature, but when the deposition process was finished, substrate temperature was increased up to about 60°C. The detailed deposition conditions for ITO/Ag/ITO multilayer films on glass substrate are summarized in Table 1. During the sputtering at room temperature, argon gas flow was finely regulated by a mass flow controller while the desired chamber pressure was maintained by auto pressure controller. First bottom ITO layer was deposited up to 43 nm utilizing the In_2O_3 ceramic target containing 5 wt% SnO_2. Then Ag thin interlayer was sputtered on the top of ITO layer by using metallic Ag (99.999% purity) target. In order to investigate the effect of Ag layer thickness on physical properties in ITO/Ag/ITO multilayer film, the deposition time of Ag interlayer was changed from 10 to 16 sec with time interval of 2 seconds. The thickness of the resulting Ag interlayer was found to be from 13.6 to 17.4 nm. Subsequently, the top ITO layer was deposited on Ag/ITO layer to 43 nm thickness under the same deposition conditions as the bottom ITO layer.

The physical properties of ITO/Ag/ITO multilayer films on glass substrate were analyzed as a function Ag thickness by using the following equipments. The surface morphology and thickness of films were measured by 3D optical analyzer (Nano View). The sheet resistance of the films was obtained by using 4-point probe

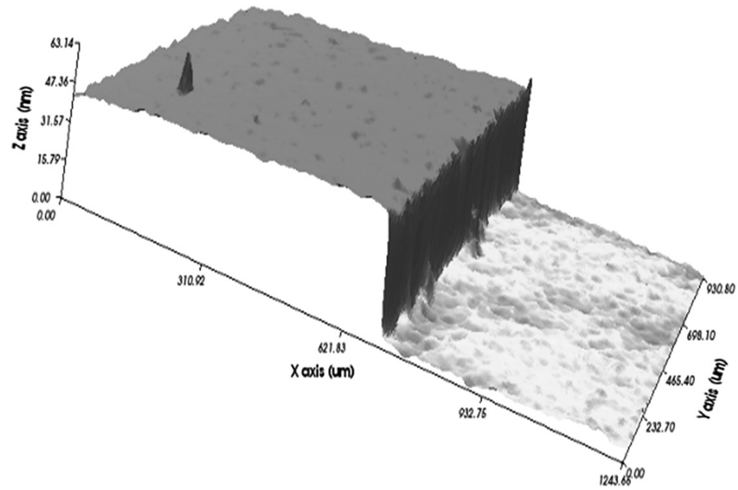
Table 1. Deposition conditions for ITO/Ag/ITO multilayer films on glass substrates at room temperature

Deposition parameters	ITO layer	Ag layer
Power (Watt)	RF 1400	DC 220
Sputtering gas	Ar	Ar
Ar flow rate (sccm)	30	20
Base pressure (Torr)	8×10^{-6}	6×10^{-6}
Working pressure (mTorr)	5	3
Target size	93 W × 430 L × 6 t	4″, 5 t
Substrate size	$100 \times 100\ mm^2$	$100 \times 100\ mm^2$

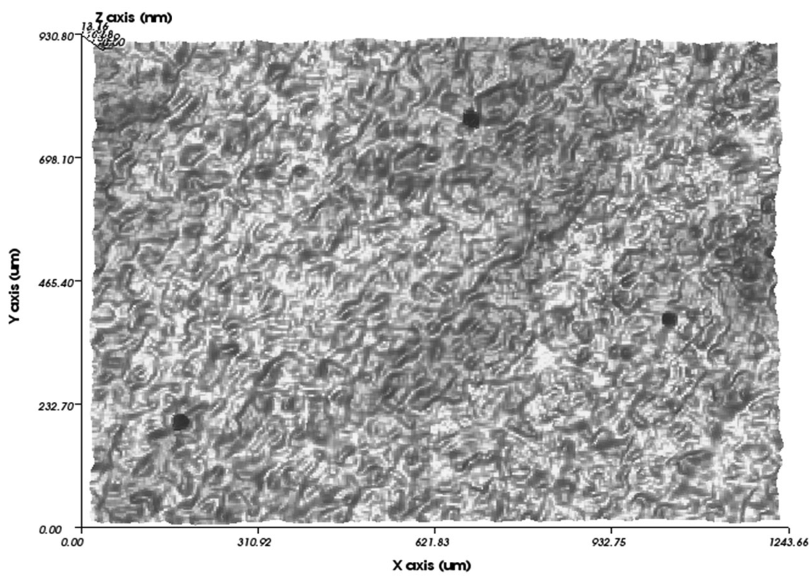
(Mitsubishi Chem., MCP-T610). The optical transmittance spectra of films were obtained with a UV-VIS spectrophotometer (Konica Minolta CM-3600d).

Results and Discussion

Figure 1 shows the thickness and surface roughness of ITO thin films deposited on glass substrate by RF sputter at room temperature. The thickness and surface roughness of ITO films were found to be 43 nm and 1.12 nm, respectively. Figure 2



(a)



(b)

Figure 1. Thickness (a) and surface roughness (b) of ITO thin films deposited on glass substrate at room temperature measured by 3D optical analyzer.

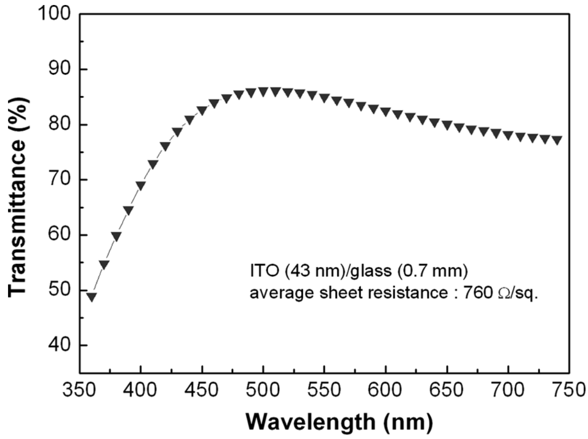


Figure 2. Optical transmittance spectra of ITO thin film deposited on glass substrate at room temperature.

shows the optical transmittance spectra of ITO (43 nm) thin film deposited on glass substrate at room temperature. For the optically and electrically uniform ITO thin films, the optimum conditions are summarized in Table 1. The average optical transmittance at wavelength of 550 nm and the average sheet resistance (R_s) of ITO films were 85% and 760 Ω/sq., respectively. When the size of glass substrate was increased to $200 \times 200\,mm^2$, the average optical transmittance and sheet resistance of ITO films showed less than 3% deviations. The resistivity (ρ) of ITO film was calculated by Eq. (1)

$$\rho = R_s d = (ne\mu)^{-1} \tag{1}$$

where d is the thickness of ITO film, n is the carrier concentration, e is the elementary charge, and μ is the mobility [10]. From the measurement of sheet resistance, the resistivity of ITO films was determined to be $3.3 \times 10^{-3}\ \Omega \cdot cm$ by using Eq. (1).

Figure 3(a) shows the optical transmittance spectra of Ag/ITO multilayer films as a function of Ag layer thickness. The transmittance of Ag/ITO thin films on glass substrate decreased significantly with the increase of the thickness of Ag interlayer, when compared to the single ITO layer due to the metallic characteristics of the Ag interlayer. The sharp drop of transmittance at long wavelength region with the increasing thickness of Ag interlayer resulted in the narrowing of transmittance window as shown in Figure 3(a). This could be attributed to the shift of resonance plasma frequency (ω_p) of the free carrier to the shorter wavelength, because the resonance plasma frequency is proportional to the carrier concentration [5, 11, 12]. Figure 3(b) shows the total sheet resistance (R_T) and transmittance of Ag/ITO multilayer film at 550 nm as a function of Ag interlayer thickness. The total sheet resistance of Ag/ITO films exhibited the lowest sheet resistance value of 3.8 Ω/sq. corresponding to the resistivity of $2.3 \times 10^{-5}\ \Omega \cdot cm$ at the Ag layer thickness of 17.4 nm. The decrease of the total sheet resistance in Ag/ITO multilayer films could be explained by the parallel circuit model proposed by Martin et al. [13]. The total sheet resistance of Ag/ITO multilayer was found to be controlled by the

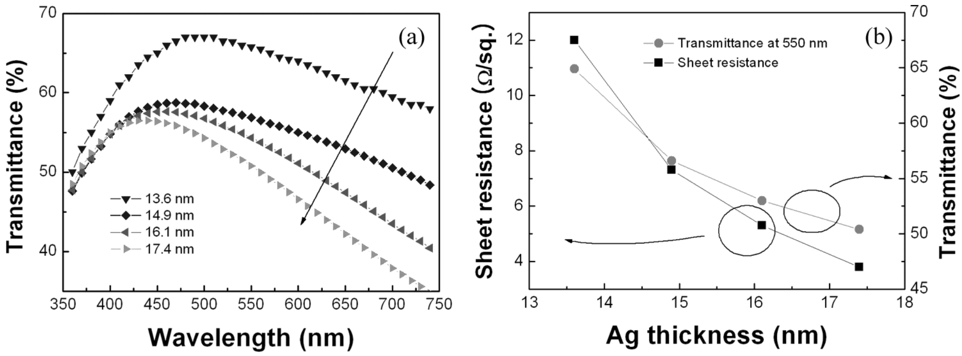


Figure 3. (a) Optical transmittance spectra of Ag/ITO multilayer films as a function of Ag interlayer thickness, (b) total sheet resistance and transmittance of Ag/ITO multilayer films at 550 nm as a function of Ag interlayer thickness.

Ag interlayer. Namely, the total sheet resistance could be expressed by Eq. (2) as the thickness of the Ag layer was increased.

$$\begin{aligned} R_T &= R_{ITO}R_{Ag}/(R_{ITO} + R_{Ag}) \\ &\approx R_{Ag}(\text{if}, R >> R_{Ag}) \end{aligned} \tag{2}$$

The optical transmittance of the Ag/ITO thin film at 550 nm decreased sharply, which the total sheet resistance of the Ag/ITO film was improved with the increasing thickness of Ag layer as shown in Figure3(b).

Figure 4(a) shows the optical transmittance spectra of ITO/Ag/ITO multilayer films as a function of Ag interlayer thickness. The optical transmittance of ITO/Ag/ITO multilayer film was improved remarkably in the visible region compared to those of Ag/ITO multilayer films. The highest transmittance of 79.4% at 550 nm was observed in ITO/Ag/ITO multilayer film with Ag interlayer thickness of 16.1 nm. However, further increase of Ag interlayer above 16.1 nm resulted in decrease of transmittance even though ITO/Ag/ITO multilayer films showed lower sheet resistance as shown in Fig. 4(b). From these experimental results, it was

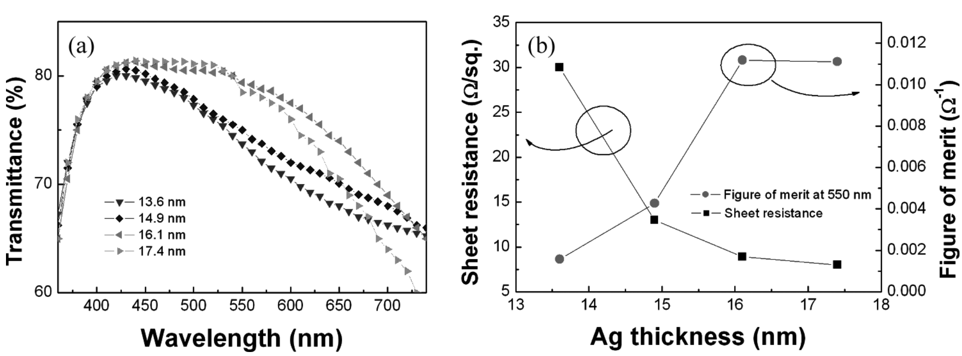


Figure 4. (a) Optical transmittance spectra of ITO/Ag/ITO multilayer films as a function of Ag interlayer thickness (b) total sheet resistance and figure of merit of ITO/Ag/ITO multilayer films as a function of Ag interlayer thickness.

considered that the top ITO layer acted as anti-reflection layer in the ITO/Ag/ITO multilayer structure [3,13,14]. The total sheet resistance of ITO/Ag/ITO multilayer films increased slightly compared to that of Ag/ITO layer due to the contribution of high sheet resistance of ITO top layer. The analysis of total sheet resistance based on the parallel circuit model was also consistent with experimental results in the ITO/Ag/ITO multilayer films. The total sheet resistance (8.0 Ω/sq.) of ITO/Ag/ITO multilayer films (with 17.4 nm Ag layer and total thickness of about 113 nm) was superior to that of the commercial single layer ITO (150 nm, 10 Ω/sq.) film deposited on glass substrates under high temperature condition. At a 17.4 nm Ag thickness, the resitivity of the ITO/Ag/ITO multilayer films was estimated to be $9.0 \times 10^{-5}\ \Omega \cdot cm$. To determine the optimum Ag interlayer thickness in the ITO/Ag/ITO multilayer films, we used the figure of merit ($\phi = T^{10}/R_s$), as defined by Haacke [15]. The maximum ϕ value (0.011 Ω^{-1}) was obtained at a 16.1 nm Ag thickness in the ITO/Ag/ITO multilayer films. The optimized Ag thickness to obtain the maximum ϕ value was similar to that of the previously reported result in the InZnSnO/Ag/InZnSnO multilayer films [2]. It was also noted that the figure of merit of ITO/Ag/ITO multilayer films decreased rapidly above the Ag interlayer thickness of 18 nm due to the metallic characteristics of the Ag interlayer.

Conclusions

In summary, we could fabricate transparent conductive ITO/Ag/ITO multilayer films on glass substrate with good transparency ($\geq$80%) and low sheet resistance ($>$10 Ω/sq.) by using sputtering technique without an additional heat treatment after the deposition. The ITO (43 nm)/Ag (16.1 nm)/ITO (43 nm) multilayer films exhibited both good transmittance (79.4% at 550 nm) and low sheet resistance (8.9 Ω/sq.) due to the combined effects of top ITO layer acting as anti-reflection layer and Ag thin interlayer acting as conductive layer. This indicates that the ITO/Ag/ITO multilayer films are promising electrode scheme for transparent conducting electrodes in OLED applications as well as others optoelectronic devices such as touch panel, solar cell, and LED.

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