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Tae Hoon Kim [a], Bo Hye Choi [a], Jae Soung Park [a], Sang Mun Lee [b], Yun Su Lee [b] & Lee Soon Park [a b c]

[a] Mobile Display Research Center, Kyungpook National University, Daegu, Korea

[b] Advanced Display Manufacturing Research Center, Kyungpook National University, Daegu, Korea

[c] Department of Polymer Science, Kyungpook National University, Daegu, Korea

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Transparent Conductive ITO/Ag/ITO Multilayer Films Prepared by Low Temperature Process and Physical Properties

TAE HOON KIM,[1] BO HYE CHOI,[1]
JAE SOUNG PARK,[1] SANG MUN LEE,[2]
YUN SU LEE,[2] AND LEE SOON PARK[1,2,3]

[1]Mobile Display Research Center, Kyungpook National University, Daegu, Korea
[2]Advanced Display Manufacturing Research Center, Kyungpook National University, Daegu, Korea
[3]Department of Polymer Science, Kyungpook National University, Daegu, Korea

Transparent conductive ITO/Ag/ITO multilayer thin films were successfully deposited on polyethylene terephthalate (PET) substrates by RF sputtering of ITO layer and DC sputtering of Ag layer at room temperature. Electrical and optical properties of ITO/Ag/ITO multilayer films were strongly dependent on the thickness of Ag thin interlayer. The ITO (50 nm)/Ag (17 nm)/ITO (50 nm) multilayer films deposited on PET substrates exhibited lowest sheet resistance ($6.7\,\Omega/\square$) and highest transmittance (83.2% at 550 nm). Especially, the sheet resistance of ITO/Ag/ITO multilayer films was superior to those of the commercial ITO (150 nm) films deposited on rigid glass substrates at high temperature. The ITO/Ag/ITO multilayer films coated PET substrate could be a good candidate for application to a transparent conductive oxide (TCO) electrode material for large size flexible OLED devices.

Keywords Ag thin interlayer; flexible OLED; flexible substrate; indium tin oxide (ITO); transparent conductive oxide (TCO)

Introduction

Transparent conductive oxide (TCO) on polymer substrates plays an important role in flexible electronics especially in flexible displays based on organic light emitting diodes (OLED) [1]. Indium tin oxide (ITO) has been widely used as TCO material due to its excellent electrical and optical properties in the OLED and other flat panel displays [2]. In general, ITO thin film deposited on polymer substrates at room temperature exhibits amorphous state. The electrical and optical properties of

Address correspondence to Lee Soon Park, Department of Polymer Science, Kyungpook National University, Sangyuk-dong Buk-gu, Daegu 702-701, Korea (ROK). Tel.: (+82)53-950-5627; Fax: (+82)53-950-6616; E-mail: lspark@knu.ac.kr

amorphous ITO films are usually inferior to those of crystalline ITO films, which typically are obtained on the rigid substrate through high temperature process (heating of substrate to 250–350°C). Therefore, it is very important to develop a unique technology to deposit ITO thin films on polymer substrates at room temperature (unheated substrate) with a very low sheet resistance ($<20\,\Omega/\square$) and high transmittance ($>85\%$ at 550 nm) for application to high performance of flexible OLED devices [3].

To obtain desired electrical and optical properties of TCO films at low temperature process for flexible OLED applications, many researchers attempted multilayer structure of TCO films with various metal interlayer such as Ag, Pt, Cu, etc. [4–7]. In this study, we adopted the Ag thin layer as interlayer between ITO layers to make high quality TCO films on polyethylene terephthalate (PET) substrate at room temperature by RF sputtering technique for the potential application to the large-sclae flexible OLED. Compared with ITO single layer films, the improvement of electrical and optical properties of ITO/Ag/ITO multilayer films are discussed in relation to the process parameters.

Experimental

ITO/Ag/ITO multilayer films were sequentially deposited on the hard coated PET (thickness $\cong 200\,\mu m$) substrates by in-line RF sputtering (ITO layer) and DC sputtering (Ag layer) at room temperature. Table 1 lists the detailed deposition conditions for ITO/Ag/ITO multilayer films on PET substrate. Both the bottom and top ITO films with thickness 50 nm were deposited utilizing the In_2O_3 ceramic target containing 5 wt% SnO_2. After the sputtering of the bottom ITO layer, Ag thin interlayer was sputtered on the bottom ITO layer by using metallic Ag (99.999% purity). In order to investigate the dependence of Ag thickness on physical properties in ITO/Ag/ITO multilayer structure, the deposition time of Ag interlayer was changed from 12 to 17 sec with one second time interval. Subsequently, the top 50 nm thick ITO layer was deposited on Ag/ITO film under the same deposition conditions to those used for the bottom ITO layer.

The physical properties of ITO/Ag/ITO multilayer films were analyzed layer by layer by using the following equipments. The surface morphology and thickness of films were measured with the scanning electron microscope (SEM, Hitachi S-4200) and 3D optical analyzer (Nano View). The sheet resistance of the films was

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

Deposition parameters	ITO layer	Ag layer
Power (Watt)	RF 1400	DC 240
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	$100 \times 100\,mm^2$	$100 \times 100\,mm^2$

determined by using 4-point probe (Mitsubishi Chem., MCP-T610). The optical transmittance spectra of films were obtained by using UV-VIS spectrophotometer (Konica Minolta CM-3600d).

Results and Discussion

Figure 1 shows the optical transmittance spectra of PET substrate hard coated on both sides and ITO (50 nm)/PET films used in this study. The observed envelope in optical transmittance spectra of hard coated PET substrates was caused by multi reflections between hard coated layer and bare PET film. The total transmittance of ITO (50 nm)/PET films was approximately 80% in the wavelength range of 370–750 nm. We could obtain optically and electrically uniform ITO/PET film by using in-line sputtering system at room temperature under deposition conditions described in Table 1. The average optical transmittance at wavelength of 550 nm and average sheet resistance of ITO (50 nm)/PET film were 81.5% and 419 Ω/□, respectively. When the size of hard coated PET substrate was increased to $200 \times 200\,mm^2$, the optical transmittance and sheet resistance of ITO(50 nm)/PET films showed less than 2% deviations.

Figure 2(a) shows the optical transmittance spectra of Ag/ITO multilayer films as function of Ag layer thickness. The reduction of sheet resistance by increasing the thickness of Ag interlayer should give rise to decrease of the optical transmittance in Ag/ITO films. As the thickness of Ag layer was increased, optical transmittance of Ag/ITO multilayer films decreased rapidly due to the increase of the free carrier concentration (absorption) and metallic behavior of Ag layer [1.8]. In Figure 2(a), the arrow indicates the direction of increasing carrier concentration. Especially, the optical transmittance at longer wavelength was strongly decreased by thicker Ag layer. These critical optical properties seemed to be directly influenced by the carrier concentration assuming mobility is constant. Namely, low sheet resistance and high optical transparency are contrary to each other because photons are strongly absorbed by the high concentration of free charge carriers. Compared with the ITO/PET film, the Ag/ITO multiplayer films exhibited very low sheet resistance

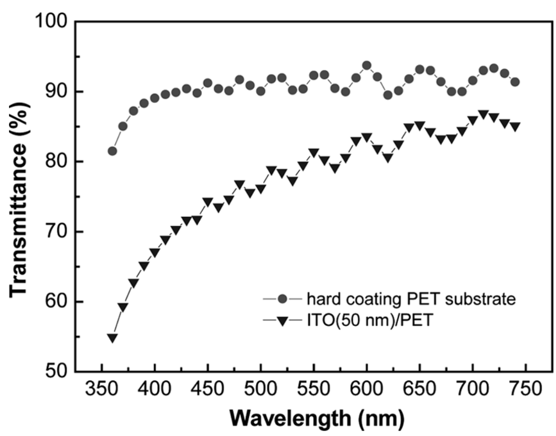


Figure 1. Optical transmittance spectra of PET substrate hard coated on both sides and ITO (50 nm)/PET films.

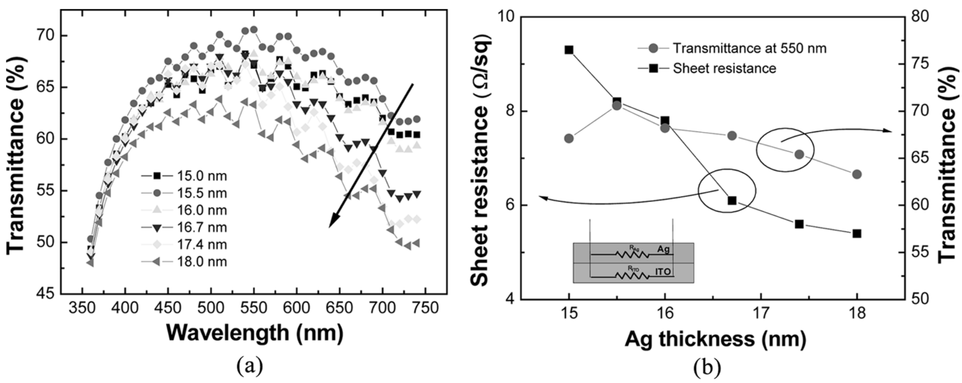


Figure 2. (a) Optical transmittance spectra of Ag/ITO multilayer films as a function of Ag interlayer thickness, (b) Sheet resistance and transmittance of Ag/ITO multilayer films at a wavelength of 550 nm as a function of Ag interlayer thickness. The inset shows the equivalent circuit for Ag/ITO multilayer films.

value ($>10\,\Omega/\square$) as shown in Figure 2(b). When measured repeatedly Ag/ITO multilayer films exhibited the lowest value of 5.4 $\Omega/\square$ at Ag layer thickness of about 18 nm. The reduction of sheet resistance in Ag/ITO multilayer films can be explained by Eq. (1) following the parallel circuit model [9] as shown in inset of Figure 2(b)

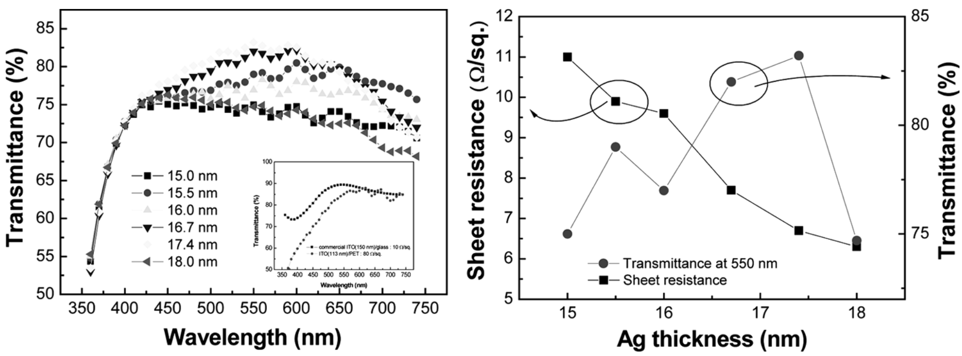
$$1/R_T = 1/R_{ITO} + 1/R_{Ag}. \quad (1)$$

Where R_T is the total sheet resistance of Ag/ITO films, and R_{ITO} and R_{Ag} are the sheet resistance of ITO and Ag layer. Assuming that the sheet resistance of ITO layer is much larger than that of Ag layer, the total resistance of Ag/ITO multilayer films can be expressed by Eq. (2).

$$\begin{aligned} R_T &= R_{ITO}R_{Ag}/(R_{ITO} + R_{Ag}) \\ &\approx R_{Ag}(\text{if},\ R_{ITO} \gg R_{Ag}). \end{aligned} \quad (2)$$

Therefore, Ag thin layer is the dominating conductive layer in the Ag/ITO multilayer films as observed by the experimental results.

Figure 3(a) shows the optical transmittance spectra of ITO/Ag/ITO multilayer films with increasing Ag interlayer thickness. The optical transmittance of ITO/Ag/ITO multilayer films were remarkably increased in the visible region compared to those of Ag/ITO multilayer films. The highest transmittance of 83.2% at a wavelength of 550 nm was observed in ITO/Ag/ITO multilayer films with Ag interlayer thickness of about 17 nm. This transmittance value is similar to that of 113 nm thickness of ITO single layer films fabricated under the same conditions of ITO/Ag/ITO multilayer films as shown in inset of Figure 3(a). However, further increase of Ag interlayer thickness above 17 nm resulted in decrease in the transmittance even though ITO/Ag/ITO multilayer films showed lower sheet resistance as shown in Figure 3(b). From experimental results above, it was considered that the top ITO layer acted as anti-reflection layer in ITO/Ag/ITO multilayer films [6,9]. The analysis of sheet resistance based on the parallel circuit model was also consistent with



(a) (b)

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

experimental results in ITO/Ag/ITO multilayer films. At 17 nm thick Ag layer, the low sheet resistance (6.7 Ω/□) of ITO/Ag/ITO multilayer films (total thickness of films is about 117 nm) was superior to that of the commercial single layer ITO (150 nm) films deposited on glass substrates at high temperature.

To determine the optimum Ag interlayer thickness in the ITO/Ag/ITO multilayer films, we calculated the figure of merit ($\phi = T^{10}/R_s$), as defined by Haacke [10], by using the optical transmittance (T) at 550 nm and sheet resistance (R_s) data as shown in Figure 4. The figure of merit value provides a criterion for the performance of a TCO films. The maximum ϕ value (0.024 Ω^{-1}) of the ITO/Ag/ITO multilayer films was obtained at about 17 nm Ag thickness. This value was similar to that of recently reported the Ag thickness for obtaining the maximum ϕ value in the InZnSnO/Ag/InZnSnO multilayer films [11]. However, the ϕ value decreased rapidly above the optimized Ag interlayer thickness 17 nm due to the decrease of transmittance of ITO/Ag/ITO multilayer films as shown in Figure 4.

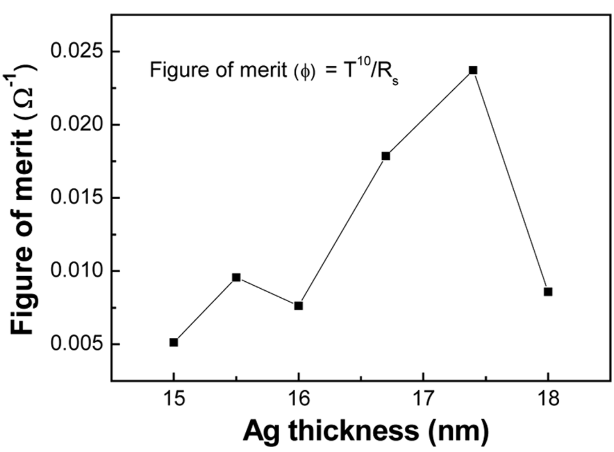


Figure 4. Figure of merit of ITO/Ag/ITO multilayer films as a function of Ag interlayer thickness.

Conclusions

In summary, we successfully fabricated ITO/Ag/ITO multilayer films on PET substrate with good transparency (>80%) and low sheet resistance (>10 Ω/□) on PET substrates at low temperature process by using sputtering technique. The optimized ITO (50 nm)/Ag (17 nm)/ITO (50 nm) multilayer films exhibited both good transmittance (83.2% at 550 nm) and low sheet resistance(6.7 Ω/□). This results maybe attributed to the combined effects of top ITO layer acting as anti-reflection layer and Ag thin interlayer acting as conductive layer in ITO/Ag/ITO multilayer films. The high quality transparent conductive ITO/Ag/ITO multilayer film could be a good candidate for TCO electrode material for large-scale flexible OLED devices.

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References

[1] Crawford, G. P. (2005). *Flexible flat panel displays*, John Wiley & Sons.
[2] Minami, T. (2005). *Semicond. Sci. Technol.*, *20*, S35.
[3] Fahland, M., Vogt, T., Schoenberger, W., & Schiller, N. (2008). *Thin Solid Films*, *516*, 5777.
[4] Sahu, D. R. & Huang, J. L. (2006). *Appl. Surf. Sci.*, 253, 827.
[5] Sahu, D. R. & Huang, J. L. (2006). *Thin Solid Films*, *515*, 876.
[6] Cheng, C. H. & Ting, J. M. (2007). *Thin Solid Films*, *516*, 203.
[7] Park, H. K., Jeong, J. A., Park, Y. S., Na, S. I., Kim, D. Y., & Kim, H. K. (2009). *Electrochem. Solid-State Lett.*, *12*, H309.
[8] Kostlin, H., Jost, R., & Lems, W. (1975). *Phys. Status Solidi(a)*, *29*, 87.
[9] Martin, E. J. J., Yan, M., Lane, M., Ireland, J., Kannewurf, C. R., & Chang, R. P. H. (2004). *Thin Solid Films*, 461, 309.
[10] Haacke, G. (1976). *J. Appl. Phys.* *47*, 4086.
[11] Choi, K. H., Nam, H. J., Jeong, J. A., Cho, S. W., Kim, H. K., Kang, J. W., Kim, D. G., & Cho, W. J. (2008). *Appl. Phys. Lett.*, *92*, 223302.