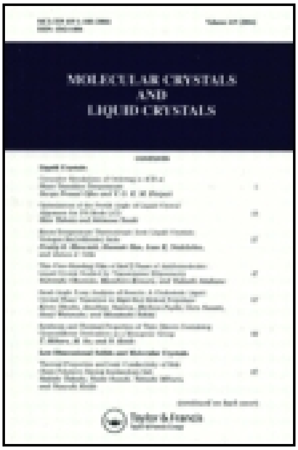

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Electrical and Optical Properties of Multi-Layered $(ITO/Ag/ITO)_n$ Transparent Conductive Films Grown onto PET Substrates

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Transparent conductive films having the sandwich structure of indium tin oxide/silver/indium tin oxide (ITO/Ag/ITO) and indium tin oxide/silver/indium tin oxide/silver/indium tin oxide (ITO/Ag/ITO/Ag/ITO) were grown onto PET substrates by a radio frequency magnetron sputtering method at the room temperature. The electrical and optical properties of the ITO/Ag/ITO films were compared with those of ITO/Ag/ITO/Ag/ITO films. Under optimal deposition conditions, thin films of ITO/Ag/ITO/Ag/ITO with a sheet resistance of 10.8 Ω/γ and an optical transmittance of over 60% in the visible spectrum range and an optical reflectance of under 20% in the visible spectrum range were achieved. This work provides better initial understanding of the relationship between sputter process conditions and their influence on electrical and mechanical performance.

Keywords: Ag; electrical; indium tin oxide; optical; sheet resistance; sputtering

INTRODUCTION

Indium tin oxide (ITO) films have been used widely as transparent electrodes of many devices such as solar cells, touch screen and organic light emitting diodes. The sheet resistance requirements for transparent electrodes in different technical applications of the ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO thin films on PET substrate are listed in Table 1. An overview of the present transparent electrode is given in [1]. The characteristics of the ITO films depend on the

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TABLE 1 Overview of the Sheet Resistance Requirements for Transparent Conductive Films used in Different Technical Applications

Application	Sheet resistance [Ω/γ]
OLED	20
Touch screens	400~700
Solar cells	8~80

preparation conditions and techniques. In particular, the input power during deposition is one of the most important key parameters to prepare highly conductive ITO films; low sheet resistance of 100 Ω/γ is obtained in amorphous ITO films prepared under the condition of the input power of 60 W and below, while such a low sheet resistance seldom achieved under higher input power.

In this paper, we introduce the transparent conductive films having multi-layered structures of ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO prepared by a RF magnetron sputtering method and report that the films with both low-resistivity and high optical transparency in the visible regions can be obtained without heating the substrate in the sputtering deposition process.

EXPERIMENTAL

The ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films with different thickness were deposited on a polyethylene terepthalate (PET) by radio frequency (RF) magnetron sputtering. Multi-layer deposition of ITO and Ag thin films was performed in a RF source that was equipped with two cathodes. The sputtering conditions of the ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO thin films on PET substrate are summarized in Table 2. The flow rates of Ar gas (99.999%) were kept at a

TABLE 2 The Sputtering Conditions for Each Layer

Sputtering parameters	Range (ITO)	Range (Ag)
Applied power [W]	60	40
Base pressure [Torr]	5×10^{-6}	5×10^{-6}
Working pressure [mTorr]	3	3
T-S distance [mm]	70	70
Frequency [Hz]	13.56	13.56
Ar flow rate [SCCM]	20	20
Substrate temp. [γ]	R.T.	R.T.

constant value of 20 sccm controlled by a mass flow controller. Both In_2O_3:SnO_2(90:10 wt%) and Ag targets have a diameter of 3 inch and thickness of 3 mm. The distance between the target and the substrate is about 70 mm. All films were deposited at a working pressure of 3.0 mtorr.

The optical transmittance and reflectance spectra of the films were measured by a UV-Visible spectrophotometer (Shimadzu, UV-1601 PC). The sheet resistance and hall effect of the films were measured using a 4-point probe (Mitsubishi, MCP-T360) and Hall effect system (EGK, HEM-2000).

RESULTS AND DISCUSSION

Table 3 shows the sheet resistance of ITO/Ag/ITO (Series A~I) and ITO/Ag/ITO/Ag/ITO (Series J~L) films deposited by RF magnetron sputtering method. The corresponding sheet resistance values of the ITO/Ag/ITO films prepared with sample series A, B, C, D, E, F, G, H, and I are 720 Ω/γ, 491 Ω/γ, 199 Ω/γ, 605 Ω/γ, 376 Ω/γ, 33 Ω/γ, 14 Ω/γ and 12 Ω/γ, respectively. Meanwhile, the corresponding sheet resistance values of the ITO/Ag/ITO/Ag/ITO films prepared with sample series J, K, and L are 31.6 Ω/γ, 13.5 Ω/γ and 10.8 Ω/γ, respectively. With increasing total deposition thickness from 20.5 nm to 301 nm sheet resistances of the ITO films are decreased. The lowest sheet resistance value of 10.8 Ω/γ is obtained from the sample L with thicker ITO layer.

TABLE 3 Sheet Resistance of the ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO Films

Sample series	Materials	Thickness [nm]	Total thickness [nm]	Sheet resistance [Ω/γ]
A	ITO/Ag/ITO	10/0.5/10	20.5	720
B	ITO/Ag/ITO	20/0.5/20	40.5	491
C	ITO/Ag/ITO	30/0.5/30	60.5	199
D	ITO/Ag/ITO	10/1/10	21	605
E	ITO/Ag/ITO	20/1/20	41	376
F	ITO/Ag/ITO	30/1/30	61	142
G	ITO/Ag/ITO	100/0.5/100	200.5	33
H	ITO/Ag/ITO	125/0.5/125	250.5	14
I	ITO/Ag/ITO	150/0.5/125	300.5	12
J	ITO/Ag/ITO/Ag/ITO	66.7/0.5/66.7/0.5/66.7	201	31.6
K	ITO/Ag/ITO/Ag/ITO	83/0.5/0.5/83	251	13.5
L	ITO/Ag/ITO/Ag/ITO	100/0.5/100/0.5/100	301	10.8

Figure 1 shows the optical transmittance of ITO/Ag/ITO films which have the different deposition thickness with 20.5 nm, 21 nm, 40.5 nm, 41 nm, 60.5 nm, and 61 nm. In visible region from 400 to 700 nm, the average transmittance of ITO thin films without heating is about 59.1~70.5%. The results indicate that the deposition thickness especially the thickness of the silver layer results in a change in the transmittance by 5~10%.

Figure 2 shows the optical reflectance of ITO/Ag/ITO films with different overall thicknesses, indicating that the deposition thickness of the silver layer gives rise to a change in the reflectance by the 2~7%. The transmittance data were used to determine the absorption coefficient of this multi-layered structure. Averaging over multiple reflection effects, the values of transmittance are given by [2,3]

$$T = (1 - R)\exp(-\alpha d) \tag{1}$$

$$\alpha = \left(\frac{1}{d}\right)\ln\left[(1-R)^2/2T + \sqrt{\left(\left(\frac{(1-R)^4}{4T^2}\right) + R^2\right)}\right] \tag{2}$$

where R and α are the reflectance and the optical absorption coefficient, respectively, and d is the total thickness of the sample.

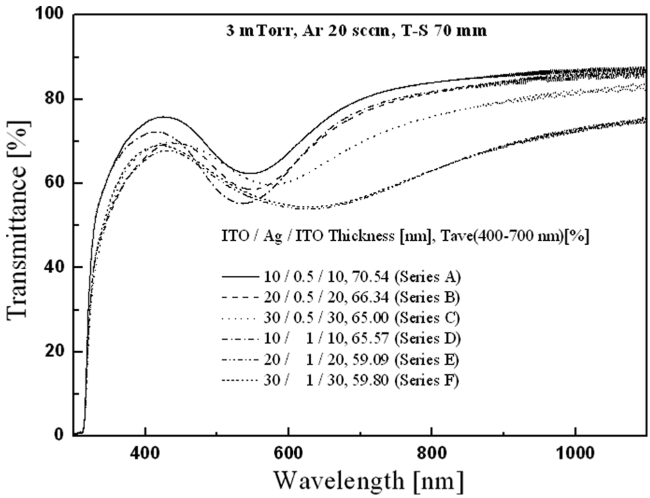


FIGURE 1 Optical transmittance of ITO/Ag/ITO films as a function of the overall thickness.

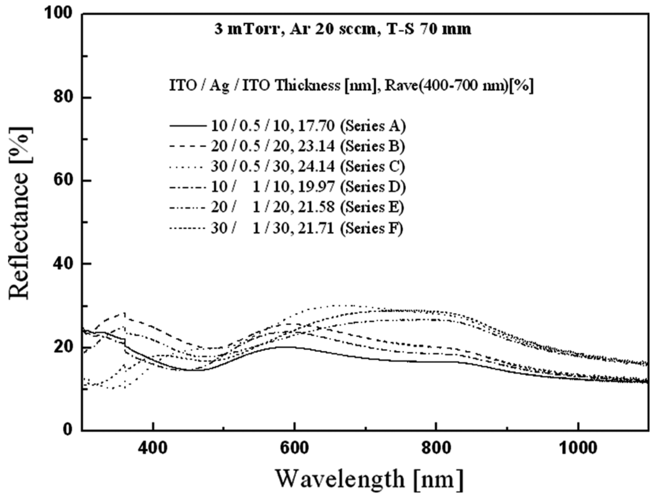


FIGURE 2 Reflectance of ITO/Ag/ITO films versus the overall thickness.

Figure 3 represents the Hall mobility and the carrier concentration of ITO/Ag/ITO films with different overall thickness, showing the inverse relation between them. Higher concentration and mobility of $1.8 \times 10^{20}/cm^3$ and $20.75\,cm^2/Vcm$ were achieved at F sample with the overall thickness of 61 nm.

According to the following relation [4],

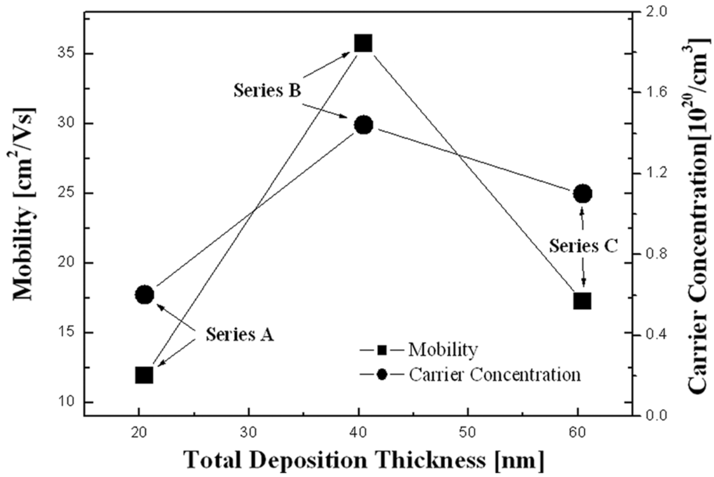
$$\rho = 1/\mathrm{Ne}\,\mu \tag{3}$$

it was suggested that the lower resistivity of ITO/Ag/ITO/Ag/ITO film of series L was due to the higher product of carrier concentration N and mobility μ.

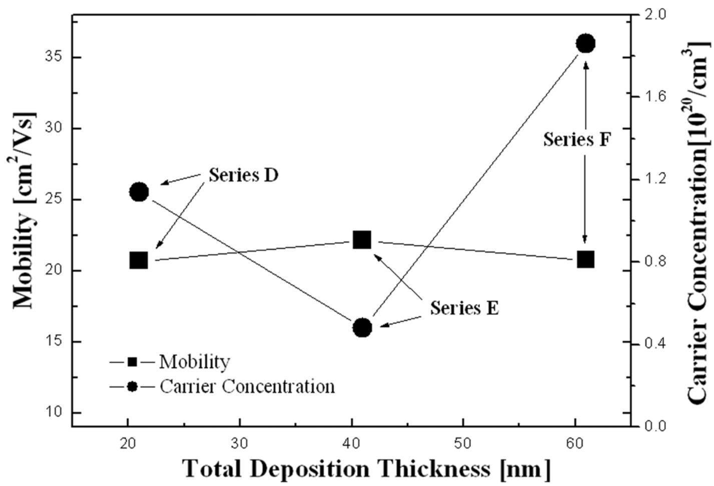
Figure 4 shows the optical transmittance of ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films as a function of the overall thickness. In visible region from 400 to 700 nm, the average transmittance of ITO thin films without heating is about 60~71%. The results indicate that the deposition thickness especially the thickness of the silver layer yields a change in the transmittance by 1~11%.

Figure 5 shows the optical reflectance of ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films with different overall thickness, indicating that the deposition thickness of the silver layer gives rise to a change in the reflectance by the 1~2%.

Figure 6 represents the Hall mobility and the carrier concentration of ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films with different overall



(a)



(b)

FIGURE 3 Hall mobility and carrier concentration of series A, B, C (a) and series D, E, F (b) as a function of the overall thickness.

thickness, showing the inverse relation between them. The mobility and the carrier concentration values of ITO/Ag/ITO/Ag/ITO films of series L on PET substrate are 23 cm^2/Vsec and $3.1 \times 10^{20}/cm^3$, respectively. In general, the sheet resistance of ITO films depends on mobility and carrier concentration, which is mainly determined by oxygen vacancies or concentration of substituted Sn^{4+} on In^{3+} sites [5].

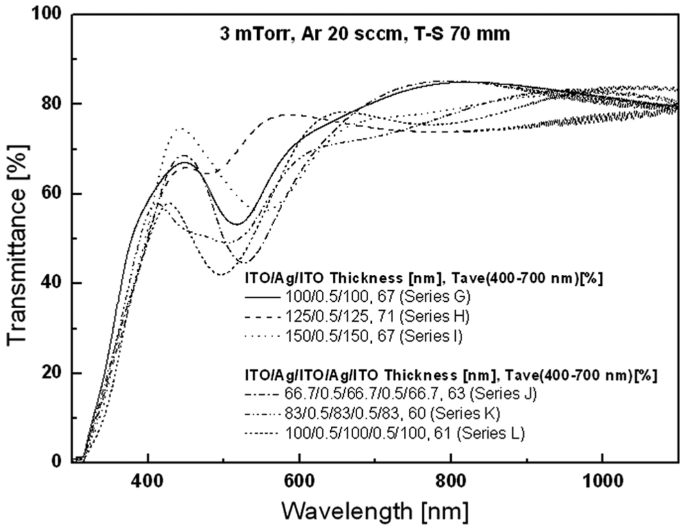


FIGURE 4 Optical transmittance of ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films as a function of the overall thickness.

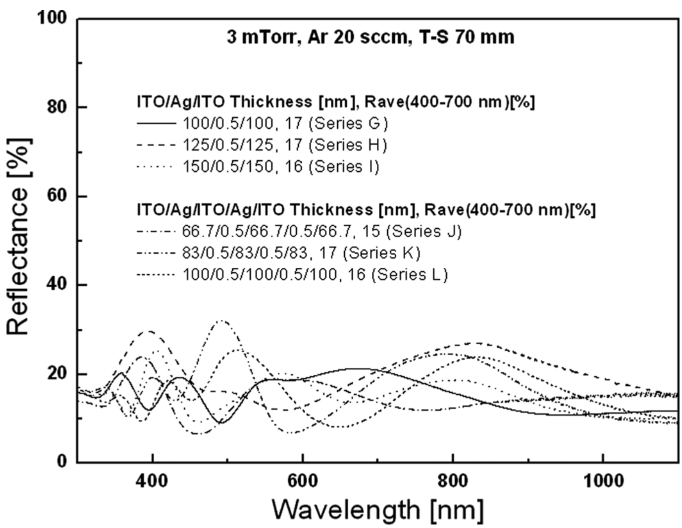
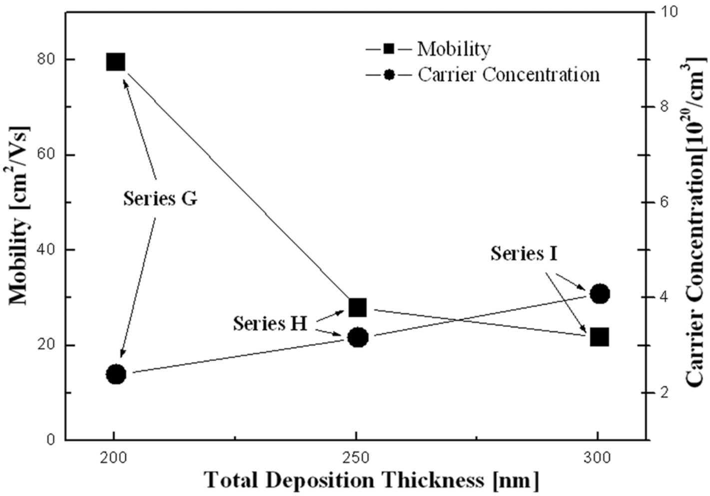
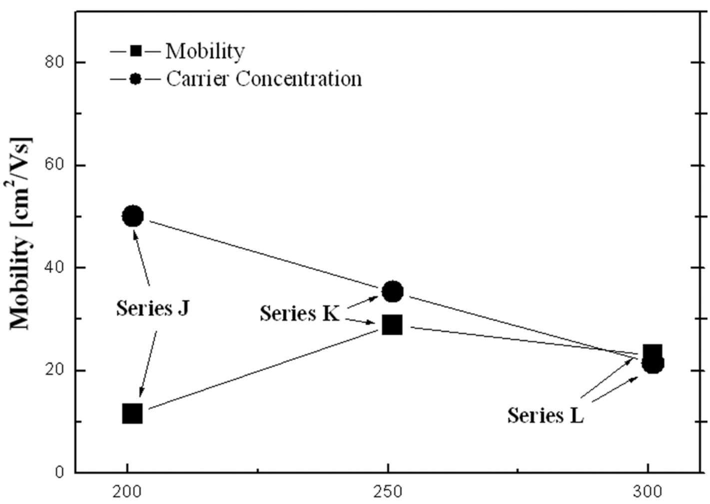


FIGURE 5 Reflectance of ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films versus the overall thickness.



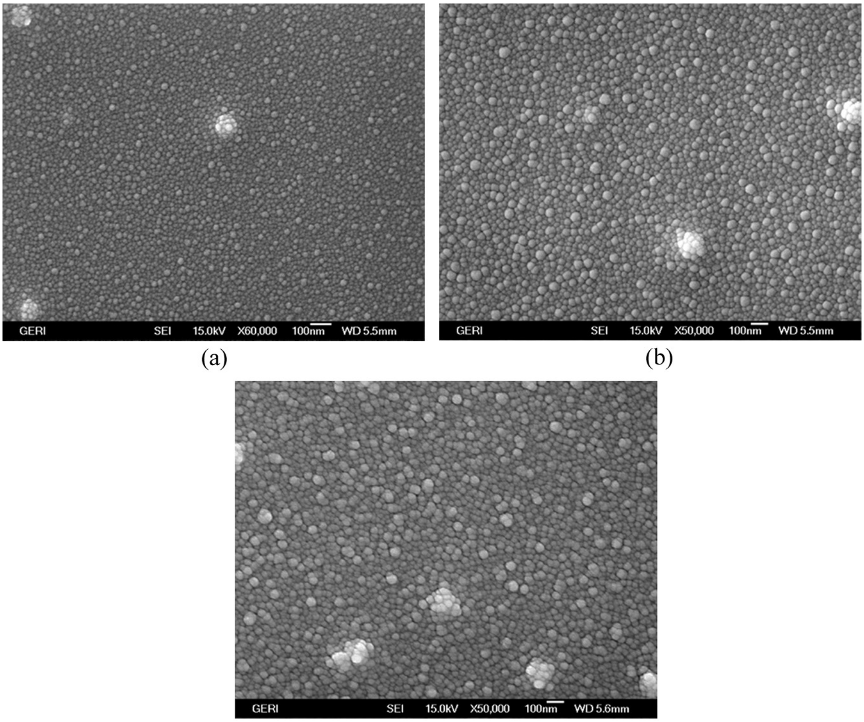
(a)



(b)

FIGURE 6 Hall mobility and carrier concentration of ITO/Ag/ITO (a) and ITO/Ag/ITO/Ag/ITO (b) films as a function of the overall thickness.

Figure 7 shows the SEM surface image of series C, series I and series L films on PET substrate deposited as a function of multi-coating layer by RF magnetron sputtering. Surface of the ITO films deposition at room temperature appear to be almost amorphous. The



(a) (b) (c)

FIGURE 7 SEM surface images of series C(a), series I(b) and series L(c) films.

ITO/Ag/ITO/Ag/ITO films in series L show larger grain sizes than ITO/Ag/ITO film in series C.

CONCLUSIONS

ITO/Ag/ITO films were deposited on the PET substrates by a RF magnetron sputtering using ITO and Ag alloy targets. The ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO films deposited in this method showed high transmittance (60∼71%) and low sheet resistance (10.8∼720 Ω/γ). As deposition thickness is increased, the film electrical resistivity is decreased. Higher concentration and mobility were achieved at a sample without heating the substrate. The resulting ITO/Ag/ITO and ITO/Ag/ITO/Ag/ITO sandwich films had the characteristics of both high transparency and high conductivity in the visible regions.

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