Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Effects of Temperature on ITO Films Grown by Low Frequency (60 Hz) Magnetron Sputtering

Sang Kooun Jung [a], Sung Ho Lee [b], Yun Su Lee [c], Sang Mun Lee [d], Lee Soon Park [e] & Sang Ho Sohn [a]

[a] Department of Physics, Kyungpook National University, Daegu, Korea

[b] Nano Practical Application Center, Daegu, Korea

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

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

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

Version of record first published: 18 Mar 2009

To cite this article: Sang Kooun Jung, Sung Ho Lee, Yun Su Lee, Sang Mun Lee, Lee Soon Park & Sang Ho Sohn (2009): Effects of Temperature on ITO Films Grown by Low Frequency (60 Hz) Magnetron Sputtering, Molecular Crystals and Liquid Crystals, 499:1, 316/[638]-324/[646]

To link to this article: http://dx.doi.org/10.1080/15421400802619719

Mol. Cryst. Liq. Cryst., Vol. 499, pp. 316/[638]–324/[646], 2009

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400802619719

Effects of Temperature on ITO Films Grown by Low Frequency (60 Hz) Magnetron Sputtering

Sang Kooun Jung[1], Sung Ho Lee[2], Yun Su Lee[3], Sang Mun Lee[4], Lee Soon Park[5], and Sang Ho Sohn[1]
[1]Department of Physics, Kyungpook National University, Daegu, Korea
[2]Nano Practical Application Center, Daegu, Korea
[3]Advanced Display Manufacturing Research Center, Kyungpook National University, Daegu, Korea
[4]Mobile Display Research Center, Kyungpook National University, Daegu, Korea
[5]Department of Polymer Science, Kyungpook National University, Daegu, Korea

In this study, we have tried the growth of indium tin oxide (ITO) thin films by using low frequency (60 Hz) magnetron sputtering method (LFMSM). Characteristics of ITO thin films deposited at different substrate temperatures (T_s) from room temperature (RT) to 300°C were investigated. The optical transmittance of more than 83% in the visible region and a resistivity of $1.7 \times 10^{-4}\,\Omega\cdot cm$ have been obtained in the films prepared at a deposition rate of 100Å/min. The peak to valley roughness ($R_{p\text{-}v}$) value measured by an atomic force microscopy was 4.2–7.8 nm. Structural and electrical characteristics are superior in the films deposited at 250°C. The sheet resistance was 11 Ω/sq for 150-nm-thick films grown at 250°C. LFMSM is a way to obtain ITO films with a smooth surface, high transmittance and high electrical conductivity.

INTRODUCTION

Transparent conducting oxide (TCO) thin films have been widely used as transparent electrodes for various applications, such as organic light emitting diodes (OLEDs), solar cells, touch panels, flat panel display (FPD) and optoelectronics because of its characteristics of

This work was supported by the Mobile Display Research Center (MDRC) Program of the Ministry of Commerce, Industry and Energy of Korea.

Address correspondence to Prof. Sang Ho Sohn, Department of Physics, Kyungpook National University, Sangyuk-dong, Buk-gu, Daegu 702-701, Korea (ROK). E-mail: shsohn@mail.knu.ac.kr

good conductivity and high transparency. Among TCO, ITO is an important material in the fabrication OLED because it combines many technologically interesting properties such as high optical transmittance over the visible wavelength region and good electrical conductivity. Moreover, in OLED where the organic thin film is in direct contact with the ITO anode, the electroluminescent characteristics of OLED is greatly influenced by the surface morphology of the ITO thin film.

Up to date, ITO films have been deposited by a variety of methods such as radio frequency (RF) magnetron sputtering, direct current (DC) magnetron sputtering and ion plating [1]. Recently, studies on the thin film growth by a low frequency (60 Hz) magnetron sputtering method (LFMSM) have been carried [2,3]. In LFMSM, the sputtering process has peculiar properties such as non-continuous discharge, relatively high electron temperature, and small bombarding damage [2,3].

The purpose of this paper is to describe a detailed investigation on the influence of low frequency magnetron sputtering parameter as a function of substrate temperatures and another aim is to deposit the high quality ITO thin films which are used in FPD without any post treatments of films, especially in OLED.

EXPERIMENTAL

ITO films were deposited on glass substrates by a LFMSM. The sputtering conditions of ITO thin films on sodalime glass substrate are summarized in Table 1. The sputtering system was evacuated using a turbo molecular pump. The alloy target was In_2O_3:SnO_2 (90:10 wt%) with a diameter of 3 inch and thickness of 6 mm. The distance between

TABLE 1 The Sputtering Conditions and the Optical Bandgap of ITO Films Deposited at Different Substrate Temperatures

Sputtering parameters	Range
Input voltage [V]	320
Base pressure [Torr]	8×10^{-6}
Working pressure [mTorr]	3.3
T-S distance [mm]	100
Frequency [Hz]	60
Deposition time [min.]	15
Ar flow rate [SCCM]	25
Substrate temp. [°C]	RT/100/150/200/250/300
Film thickness [nm]	≈150
Optical bandgap [eV]	3.78/3.74/3.81/3.81/3.82/3.82

the target and the substrate is about 100 mm. The vacuum chamber was evacuated down to pressure 6×10^{-6} mtorr prior to deposition. The flow rates of Ar gas (99.999%) were kept at a constant value of 25 sccm controlled by a mass flow controller. The discharges were generated at input power of 320 V (60 Hz). The deposition pressure was 3.3 mtorr and the sputtering time was 15 min. ITO thin film deposited at different substrate temperatures of RT, 100°C, 150°C, 200°C, 250°C and 300°C, respectively.

The thickness of the films was determined by anode in OLED. In the present study all the measurements were performed on the films having the thickness of 150 nm [4].

The crystal structure and phase of the ITO films were measured using X-ray diffraction (XRD) with a CuKα source under an applied voltage of 40 kV and a current of 30 mA. The thickness of the films was determined by a α-step (Veeco, Dektak 3). The surface roughness of the films was investigated with atomic force microscope (AFM, Digital Instrument, Nanoscope IIIa). The optical transmittance spectra of the films were recored by a UV-Visible spectrophotometer (Shimadzu, UV-1601PC). The sheet resistance of the films was measured using a 4-point probe (Mitsubishi, MCP-T360). The mobility and carrier density of the ITO thin films were measured using Hall effect system (EGK, HEM-2000).

RESULTS AND DISCUSSION

Structural Properties

The XRD patterns of the ITO films at different substrate temperature are shown in Figure 1. The intensities of the four major peaks, (222), (400), (440), (622) of ITO film, were significantly affected by substrate temperatures. Crystallinity of the films improved after being heated at a substrate temperature of above 100°C and (222) peak becomes higher than (400) peak at a substrate temperature of 300°C. The amorphous films of RT have many defects in structure and nonstoichiometry in composition. Therefore, the crystalline structure becomes more perfect as can been seen from the peak intensity of the heated ITO films.

Figure 2 shows the cross-section analysis of ITO films deposited at different substrate temperatures. The definition of R_{rms} is the root-mean-square value of the surface roughness profile from the centerline, and peak-to-valley roughness ($R_{p\text{-}v} = R_{max}$) is the vertical distance between the highest and lowest points. As shown in Figure 2, the ITO films have relatively lower roughness ($R_{p\text{-}v}$) at a temperature of about 250°C even if they were suffered from any post treatment such as the

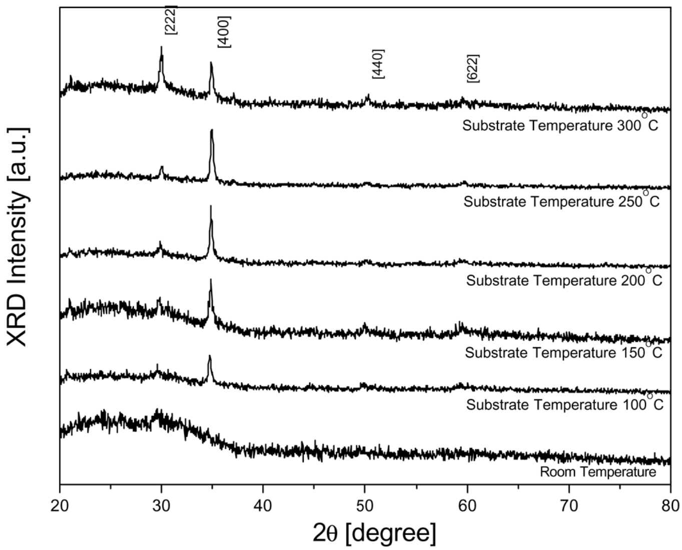

FIGURE 1 XRD pattern of ITO films deposited at different substrate temperatures.

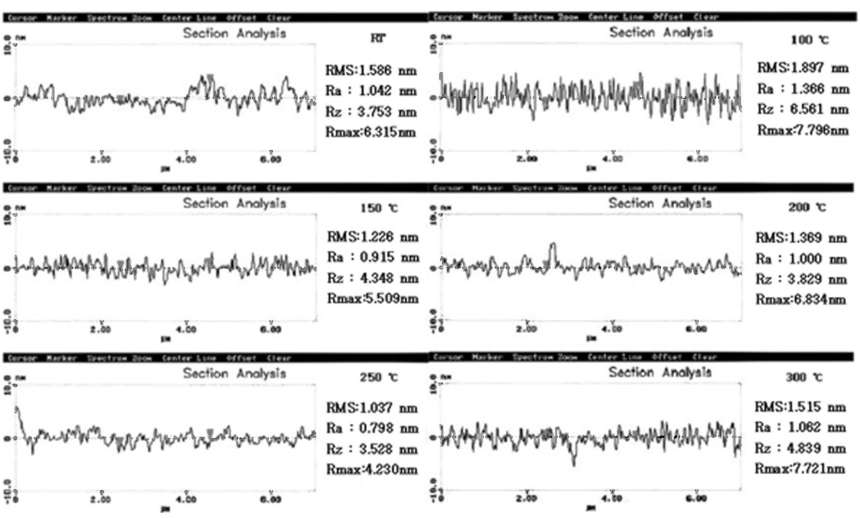

FIGURE 2 Roughness of ITO films deposited at different substrate temperatures.

ion showering. The corresponding $R_{p\text{-}v}$ values of the ITO films prepared at substrate temperatures 100°C, 150°C, 200°C, and 250°C are 7.796 nm, 5.509 nm, 6.834 nm, and 4.230 nm, respectively. With increasing substrate temperatures from 100°C to 250°C, $R_{p\text{-}v}$ values of the ITO films are decreased.

The effect of substrate temperatures on film morphology could be used to produce smooth ITO films with optoelectrical properties suitable for various applications. The surface morphology of ITO films grown at above temperature of 300°C is not uniform. The surface morphology of non-heated films is much rougher than that of films heated at a substrate temperature of 250°C.

Electrical Properties

Figure 3 shows the grain size of ITO films deposited at different substrate temperatures. As seen in Figure 4, The ITO films grown at a substrate temperature of 250°C showed the lowest sheet resistance, compared with films grown at all other substrate temperatures. The lowest sheet resistance value is 11 Ω/sq.

The crystallite size D of the scattering material is determined from the half-width w of the peaks by means of the Scherrer formula [5],

$$D = \frac{K\lambda}{w' \cos(\theta)}$$

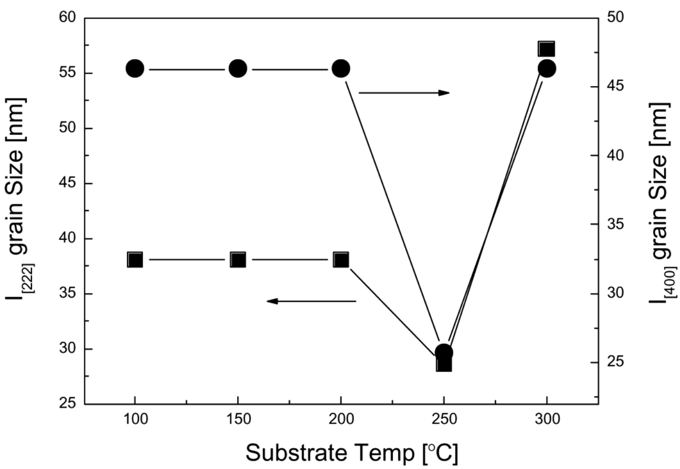

FIGURE 3 $I_{[222]}$ and $I_{[400]}$ grain size of ITO films deposited at different substrate temperatures.

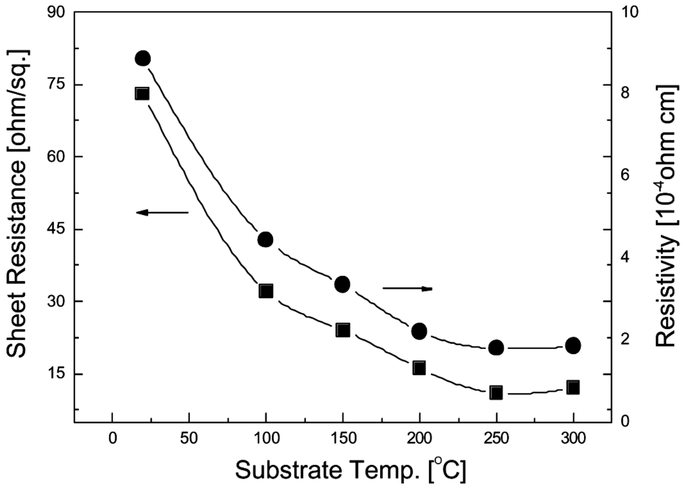

FIGURE 4 Sheet resistance and resistivity of ITO films deposited at different substrate temperatures.

$$w' = \sqrt{w^2 - \delta^2}$$

with δ being the resolution of the spectrometer, here 0.01°K is a shape factor and assumes a value of 0.9 for spherical particles. As revealed in Figure 3, for all orientations, D decreases with increasing substrate temperatures.

The resistivity and sheet resistance of the ITO films were found to decrease with increase of substrate temperatures in Figure 4. The low resistivity value of ITO thin films is due to a high carrier density because the Fermi level is located above the conduction level. It is well known that the resistivity is related with the carrier density and its mobility [6]. In ITO thin films, the main source of the charge carrier is both tin dopants and ionized oxygen vacancies. Several authors have proposed that the main conduction mechanism of ITO thin films deposited at low substrate temperature is due to the creation of oxygen vacancies, which donate two electrons for each vacancy [7].

Figure 5 shows the resistivity, Hall mobility, and carrier density of ITO films deposited at different substrate temperatures. With increasing substrate temperatures, the carrier density increased rapidly at a substrate temperature of 200°C and reached $9.3 \times 10^{20}/\text{cm}^3$ at 250°C. The resistivity showed the lowest value of $1.7 \times 10^{-4}\,\Omega\,\text{cm}$ at a substrate temperature of 250°C. Above 250°C, the mobility and the carrier density decreased according to the substrate temperatures. When the

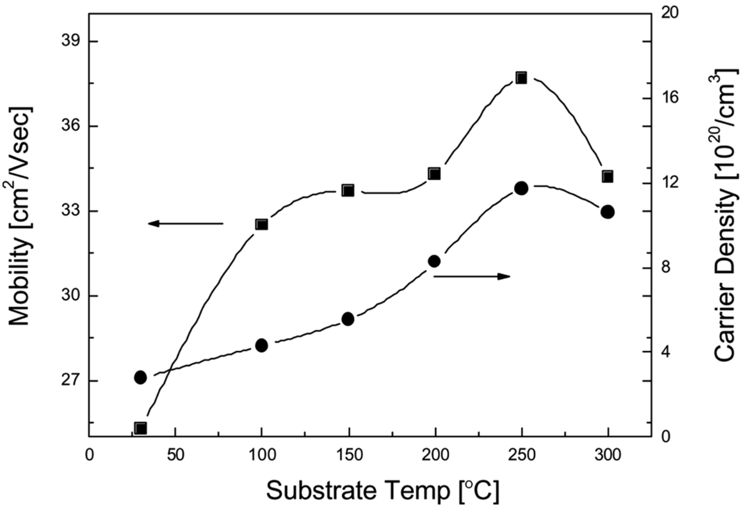

FIGURE 5 Hall mobility and carrier density of ITO films deposited at difference substrate temperatures.

substrate temperatures are low, both the mobility and carrier density are reduced.

Optical Properties

Figure 6 shows the optical transmittance of ITO films deposited at different substrate temperatures of RT, 100°C, 150°C, 200°C, 250°C and 300°C. In the visible region from 400 to 700 nm, the average transmittance of as-depo ITO thin films is about 80%, and increases up to over 85% after heating at 200°C. The results indicate that the substrate temperature gives an increase of the transmittance by 1 ~ 5%. Combined with the effect of heating on crystallization, we can see that the optical transmittance of the crystalline film is much higher than the optical transmittance of amorphous films as shown in Figure 6. The optical bandgap (E_g) of ITO thin film can be deduced from this graph. E_g can be calcultated using Cody' relation[6],

$$\alpha^2 = (h\nu - E_g)$$

where α is an optical absorption coefficient and $h\nu$ is a photon energy. Absorption coefficients of the films in different wavelengths have been calculated from transmission and reflection data. Extrapolation of the straight regions of the plots to $\alpha = 0$ gives E_g. E_g according to different

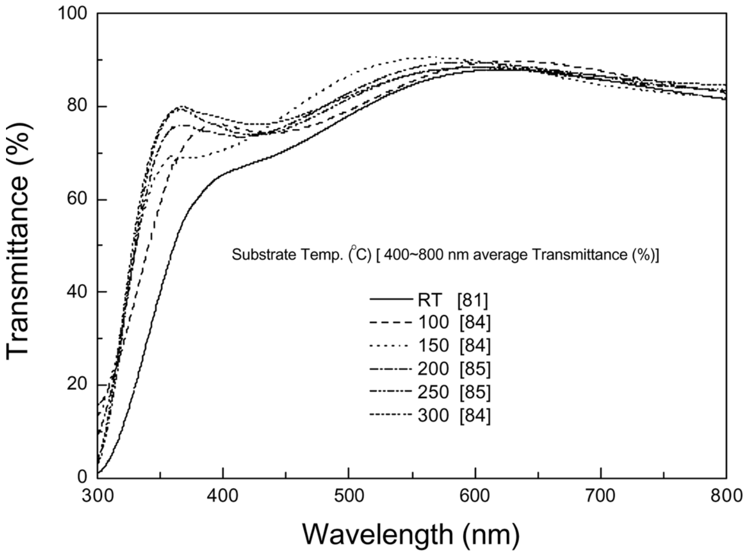

FIGURE 6 Transmittance of ITO films deposited at difference substrate temperatures.

substrate temperatures is listed on last row in Table 1. It was observed that the direct band gap of the ITO thin films increased from 3.78 to 3.82 eV with increasing deposition substrate temperatures from RT to 300°C [8]. This increase in the band gap is due to an increase in the carrier density of the films. This shift of the band gap with an change in the carrier density can be explained by the Burstein-Moss (B-M) shift [9].

CONCLUSIONS

Characteristics of ITO films deposited by a LFMSM at different substrate temperatures from room temperature to 300°C were investigated. The optical, electrical and structural properties of the films grown at the substrate temperature of 250°C are much better, being compared when they prepared at other temperatures and it is confirmed that this LFMSM is a way to obtain ITO films with the smooth surface morphology ($R_{p\text{-}v}$:4.23 nm), high transmittance (>85%) and low sheet resistance (≈11 Ω/sq). The experimental results reveals that transparent conducting ITO thin films can be prepared by LFMSM and they can be used for display applications without any post surface treatment.

REFERENCES

[1] Sun, X. W., Wang, L. D., & Kwok, H. S. (2000). *Thin Solid Films*, *360*, 75.
[2] Lee, S. H. & Park, D. K. (2004). *J. Korean Phys. Soc.*, *44*, 950.
[3] Kim, H. T. & Park, D. K. (2003). *J. Korean Phys. Soc.*, *42*, S916.
[4] Park, N. G., Kwak, M. Y., Kim, B. O., & Kwon O. K. Kwon, (2002). *Jpn. J. Appl. Phys.*, *41*, 1523.
[5] Mergel, M., Schenkel, M., Ghebre, M., & Sulkowski, M. (2001). *Thin Solid Films*, *392*, 91.
[6] Zhang, K., Zhu, F., Huan, C. H. A., & Wee, A. T. S. (2000). *Thin Solid Films*, *376*, 255.
[7] Adurodijia, F., Izumi, H., & Ishihara, T. (2000). *J. Appl. Phys.*, *86*, 814.
[8] Kim, H., Horwitz, J. S., Pique, A., Gilmore, C. M., & Chrisey, D. B. (1999). *Appl. Phys.*, *A 69*, S447.
[9] Kim, H. & Gilmore, C. M. (1999). *J. Appl. Phys.*, *86*, 6451.