Effect of thickness on the electrical and optical properties of ITO films deposited on plastic substrates

Weifeng Zheng, Limei Lin, Peiwei Lv, Rongquan Gai, Fachun Lai
School of Physics and OptoElectronics Technology
Fujian Normal University
Fuzhou 350007, PR China
zhengwf3@fjnu.edu.cn

Abstract—**Indium tin oxide (ITO) thin films with different thicknesses were prepared on polyethylene terephthalate (PET) substrates by RF magnetron sputtering at room temperature. The structure, morphology, electrical, and optical properties of the films were investigated by X-ray diffraction, atomic force microscopy, spectrophotometer, and van der pauw method, respectively. The experimental results show that the surface grain size, roughness, and carrier concentration of the films increase with the increase of thickness. The sheet resistance, Hall mobility, and transmittance decrease as the film thickness increases. When the thickness is 148 nm, the sheet resistance is 26.5 $\Omega\square^{-1}$, mobility is 19.1 $cm^2V^{-1}s^{-1}$, and carrier concentration is 8.43×10^{20} cm^{-3}.**

Keywords-Indium tin oxide (ITO); PET substrates; Magnetron sputtering; Optical properties; Electrical properties

I. INTRODUCTION

Indium tin oxide (ITO) film has been widely used in solar sells, flat panel displays, sensors, and organic light emitting diodes, because it has a highly optical transmittance in the visible optical region, a high reflectance in infrared spectra, and a low resistance [1]. The growth of ITO films on flexible polymer substrates is more and more important as the displayer will be flexible and thinner for the user. The investigation of the growth of ITO films, therefore, changes from on glass substrates to on polymer substrates. ITO films on flexible substrates have many advantages such as can be bent, light weight, not easy broken, large scale production, and good for transportation [2].

Previously, a lot of techniques have been applied to prepare ITO films, such as sputtering, thermal evaporation, chemical vapor deposition, pulsed laser deposition, and sol-gel process [3-8]. However, the most difficulty for the deposition ITO film on polymer substrate is the substrate distortion and low adhesion of film on substrate, because the polymer can not stand the high temperature [5,9]. Therefore, there are many investigations about the deposition ITO films on polymer substrates had been reported [10-13]. The effect of deposition condition, such as sputtering pressure, oxygen partial pressure, deposition rate, and substrate temperature, on the structure, optical, and electrical properties of ITO films on flexible substrates has been studied. For example, Kim et al [14] had prepared ITO films on PET substrates by direct current magnetron sputtering, it was found that when the sputtering pressure was 3 mτ and sputtering power was 30 W, the sheet resistance was 22 $\Omega\square^{-1}$ and transmittance is higher than 80%. ITO films on PES substrates had been studied by Park et al [1] by RF magnetron sputtering and sheet resistance was about 20–25 $\Omega\square^{-1}$.

In this paper, ITO films with different thicknesses have been deposited on commercial PET substrates by RF magnetron sputtering at room temperature. The structure, surface morphology, electrical, and optical properties of the films were investigated.

II. EXPERIMENTAL DETAILS

ITO films were deposited on commercial PET substrates at room temperature by RF magnetron sputtering in a commercial sputtering system (JGP560BⅡ). The sputter target was an oxide ceramic (In_2O_3:SnO_2=90:10 at wt.%). The vacuum chamber was initially evacuated to a base pressure of 4.0×10^{-4} Pa and the total pressure during the sputter process was kept at 0.5 Pa. The Ar flow rate was controlled by a mass-flow controlled regulator. The sputtering power was 60 W. Before each deposition, the target was pre-sputtered for 5 min to clean the target. In order to vary the thickness of the ITO films, the deposition time was varied between 400 s and 1000 s in steps of 200 s. All the sputtering conditions were nearly identical expect the sputtering time. The samples were labeled as A, B, C, and D, for the deposition time of 400 s, 600 s, 800 s, and 1000 s.

The crystal structure of film was examined by X-ray diffraction (XRD). XRD study was carried out on an X-ray diffractometer (Y-2000) with high-intensity Cu Kα radiation (λ=1.5419 Å). The morphology of the films was observed by an atomic force microscopy (AFM) (CSPM-4000) under ambient conditions. The sheet resistance was measured by using four-point probe. The carrier mobility and concentration were characterized by Hall effect measurements using the van der Pauw technique. The normal incidence transmittances of samples were recorded by a double-beam spectrophotometer (UV-2450) in the wavelength range 200-900 nm. The refractive indices, extinction coefficients and thicknesses (*d*) of the films were retrieved by optimizing all of transmittance data [15], and the thicknesses of the films were showed in Table I.

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III. RESULTS AND DISCUSSION

A. *Crystal structure*

Fig. 1 shows the XRD pattern of the sample C. The diffraction peaks at 30.2°, 35.1°, 45.3°, 50.8°, and 60.3° correspond to the (2 2 2), (4 0 0), (5 1 1), (4 4 0), and (6 2 2) plans respectively of In_2O_3 cubic crystalline structure. None of the spectra indicated any characteristic peaks of Sn, SnO, or SnO_2, which means that the tin atoms were doped substitutionally in In_2O_3. ITO film showed noncrystalline structure at initial phase when the thickness was thin. Kumar et al [3] considered that no characteristic diffraction peak of ITO was identified and only amorphous nature of polymer substrate was observed as the thickness of films were lower than 165 nm. However, the study in reference [5] indicated that In_2O_3 cubic crystalline structure was appeared when the thickness of films were more than 80 nm.

Figs. 2 (a)–(d) give AFM images of ITO films with d = 118 nm, 148 nm, 235 nm and 300 nm, respectively. The surface root mean square roughness of sample A, B, C, and D are 2.19 nm, 2.57 nm, 3.36 nm, and 3.04 nm, respectively. Correspondingly, the average surface grain sizes are 66.1 nm, 69.0 nm, 77.12 nm, and 90.7 nm and surface grain can be seen obviously. The increase of surface roughness with increasing thickness is the result of the aggregation of grains or the increase of crystallinity.

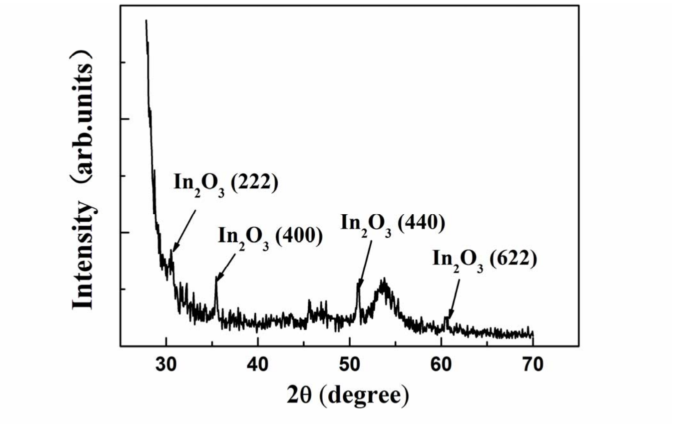

Figure 1. X-ray diffraction pattern of sample C.

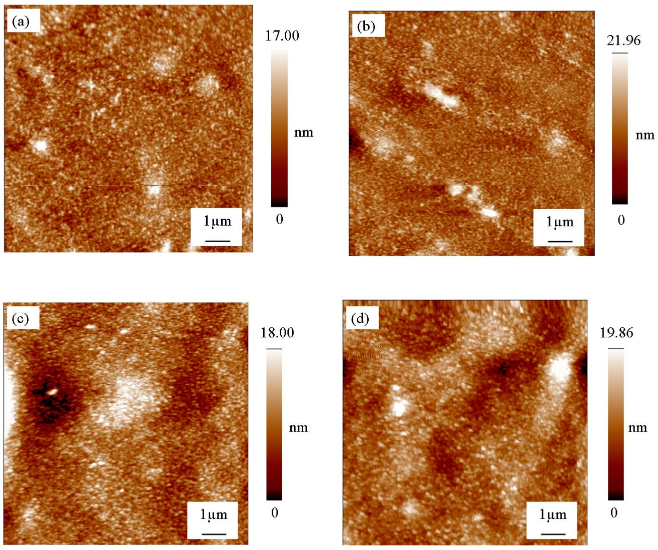

Figure 2. AFM-images (10 μm×10 μm) of ITO thin films with different thicknesses.

B. *Optical properties*

The transmittance spectra of PET substrate and ITO films with different thicknesses are shown in Fig. 3. The maximum transmittance and the average transmittance at the wavelength between 400 nm and 800 nm are 88.7% and 84.6% for the bare PET substrate. Comparing with the bare substrate, ITO film on PET substrate has a lower transmittance. Moreover, the transmittance of ITO films decreases and short wavelength threshold shifts to longer wavelength (red shift) gradually when ITO film thickness increases. It indicates that the optical band gap of sample is smaller as the film thickness increases.

Fig. 4 presents the average transmittance at wavelength between 400 nm and 800 nm and the maximum transmittance as a function of ITO film thickness. The average transmittance decreases from 84.5% to 77.9%, and the maximum transmittance decreases from 83.8% to 79.6% when thickness increases from 118 nm to 300 nm. The surface roughness increases with increasing thickness as discussed in AFM results and the change of film structure, which result in the increase of optical surface scattering and volume scattering [16]. Therefore, the optical losses and optical absorption increase, and transmittances decrease.

Refractive indices (n) and extinction coefficients (k) of ITO films with different thickness were calculated by simulating all the transmittance data of the films. An optical model which combines the modified Drude model and Forouhi-Bloomer model had been used in the simulation process [15]. Fig. 5 shows refractive indices as a function of wavelength for the films with different thickness. The obtained refractive indices are in agreement with our previous research report [15] and decrease with the increase of film thickness. The extinction coefficients as a function of wavelength are shown in Fig. 6. The extinction coefficients are lower than 10^{-1} in the visible optical region and their change is small when the thickness changes.

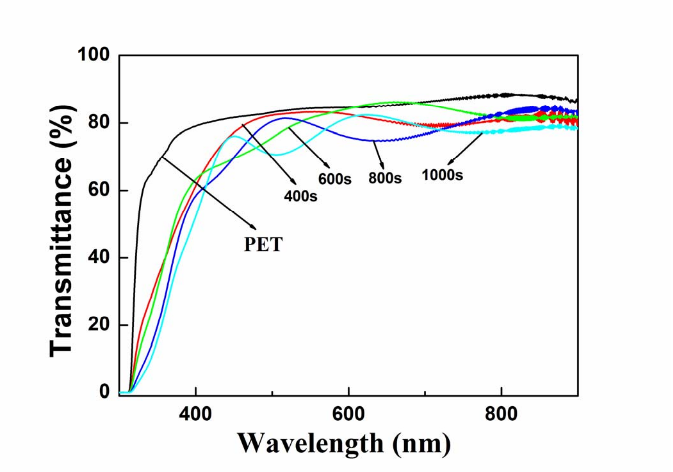

Figure 3. Transmittance spectra of ITO films with different thicknesses.

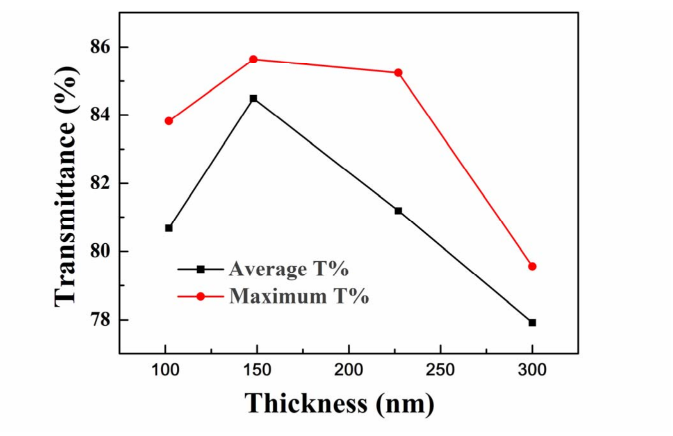

Figure 4. Average transmittance (400–800 nm) and maximum transmittance of ITO films on PET as a function of thickness.

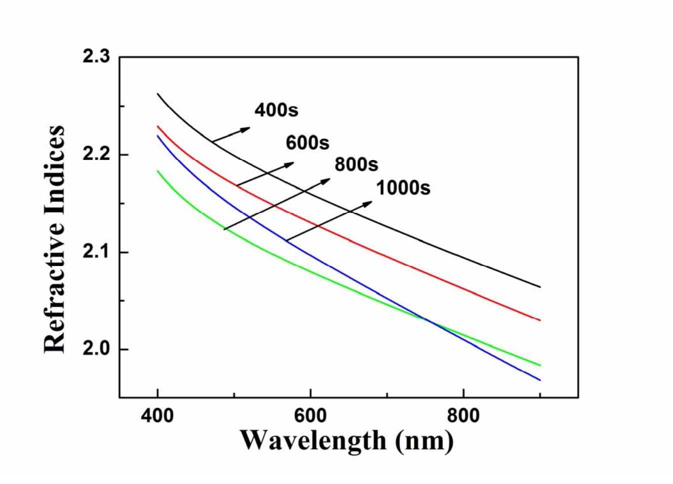

Figure 5. Variation of refractive index as a function of wavelength for different thicknesses of ITO films.

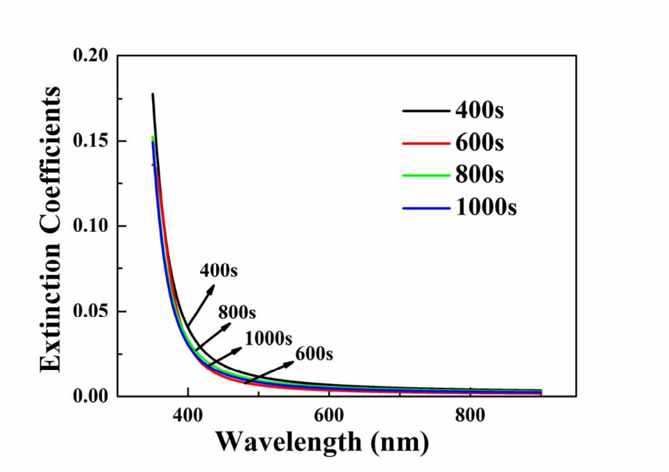

Figure 6. Variation of extinction coefficients as a function of wavelength for different thicknesses.

C. Electrical properties

The sheet resistance, resistivity, carrier concentration, and mobility of samples are listed in Table I. The sheet resistance and resistivity of the films decrease as the thickness increases from 118 nm to 235 nm. The minimum sheet resistance is about 17.01 $\Omega\square^{-1}$ for the film with 235 nm thickness. The decrease of resistivity with the increase of film thickness could be attributed to the improvement of crystalline quality of ITO film. These results indicate that in order to get high-quality ITO films with low resistivity, one need to enhance the crystalline quality of the films from the initial stage of deposition. But for the sample D which deposition time is about 1000 s, the middle of film extrude because the substrate temperature is too high during sputtering and PET substrate distorts. Accordingly, the electrical and optical properties of sample D are worse than those of the other samples. In order to improve the crystalline performance of ITO film, the treatment of PET substrate before deposition has been studied by some investigators. Kim et al [14] applied ion to radiate PMMA substrate before deposition and decreased the resistivity. Park et al [1] heat PET substrate when they prepared ITO film by RF magnetron sputtering.

TABLE I. SHEET RESISTANCE, RESISTIVITY, CARRIER CONCENTRATION AND MOBILITY OF ITO/PET FILMS

Sputtering time	**d (nm)**	**Rs ($\Omega\square^{-1}$)**	**μ ($cm^2V^{-1}s^{-1}$)**	**Ne ($10^{20}cm^{-3}$)**	**ρ ($10^{-4}\Omega$ cm)**
400 s	118	32.26	27.70	5.567	3.81
600 s	148	26.50	19.13	8.434	3.92
800 s	235	17.07	17.60	8.704	4.08
1000 s	300	25.29	12.46	6.604	7.58

As shown in Table I, the Hall mobility decreases, and the carrier concentration increases when the film thickness increases. These results can be explained below. Each doubly charged oxygen vacancy contributes two free electrons [17], so the free carriers in ITO films mainly originate from the formation of oxygen vacancies. Therefore, it is believed that the oxygen vacancies increase as the film thickness increases. The mobility of ITO films is close related to microstructure features and impurities [17]. As film thickness increases, the grain scattering decreases because the surface grain size increases, but the ionized-impurity scattering increases as the increase of oxygen vacancy, which results in the decrease of mobility.

IV. CONCLUSION

Without intentionally heating the substrates, ITO films with various thicknesses have been prepared on PET substrates by RF magnetron sputtering. It is found that the structural, electrical, and optical properties clearly depend on the film thickness. The average surface grain size increases, but the transmittance, the resistivity, and sheet resistance decrease when the film thickness increases from 118 nm to 235 nm. When the thickness is 148 nm, the sheet resistance is $26.5\ \Omega\square^{-1}$, mobility is $19.1\ \mathrm{cm^2V^{-1}s^{-1}}$, and carrier concentration is $8.43\times10^{20}\ \mathrm{cm^{-3}}$. All samples are good transparent in visible optical region. The average transmittance at wavelength between 400 nm and 800 nm is about 80%, and the maximum transmittance is higher than 85%.

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REFERENCES

[1] S. K. Park, J. I. Han, W. K. Kim, and M. G. Kwak, "Deposition of indium-tin-oxide films on polymer substrates for application in plastic-based flat panel displays, " Thin Solid Films, vol. 397, pp. 49-55, October 2001.

[2] S. Y. Tsai, Y. M. Lu , J. J. Lu , and M. H. Hon, "Comparison with electrical and optical properties of zinc oxide films deposited on the glass and PET substrates," Surf. Coat. Technol., vol. 200, pp. 3241-3244, February 2006.

[3] K. J. Kumar, N. R. C. Raju, and A. Subrahmanyam, "Thickness dependent physical and photocatalytic properties of ITO thin films prepared by reactive DC magnetron sputtering," Appl. Surf. Sci., vol. 257, pp. 3075-3080, January 2011.

[4] D. Raoufi, A. Kiasatpour, H. R. Fallah, and A. S. H. Rozatian, "Surface characterization and microstructure of ITO thin films at different annealing temperatures," Appl. Surf. Sci., vol. 253, pp. 9085-9090, September 2007.

[5] D. H. Kim, M. R. Park, H. J. Lee, and G. H. Lee, "Thickness dependence of electrical properties of ITO film deposited on a plastic substrate by RF magnetron sputtering," Appl. Surf. Sci., vol. 253, pp. 409-411, November 2006.

[6] M. Nisha, and M. K. Jayaraj, "Influence of RF power and fluorine doping on the properties of sputtered ITO thin films," Appl. Surf. Sci., vol. 255, pp. 1790-1795, December 2008.

[7] J. H. Kim, K. A. Jeon, G. H. Kim, and S. Y. Lee, "Electrical, structural, and optical properties of ITO thin films prepared at room temperature by pulsed laser deposition," Appl. Surf. Sci.,vol. 252, pp. 4834-4837, April 2006.

[8] L. Hao, X. G. Diao, H. Z. Xu, B. X. Gu, and T. M. Wang, "Thickness dependence of structural electrical and optical properties of indium tin oxide (ITO) films deposited on PET substrates," Appl. Surf. Sci., vol. 254, pp. 3504-3508, March 2008.

[9] M. Boehmea, and C. Charton, "Properties of ITO on PET film in dependence on the coating conditions and thermal processing," Surf. Coat. Technol., vol. 200, pp. 932-935, October 2005.

[10] D. H. Lee, S. H. Shim, J. S. Choi, and K. B Yoon, "The effect of electro-annealing on the electrical properties of ITO film on colorless polyimide substrate," Appl. Surf. Sci., vol. 254, pp. 4650-4654, May 2008.

[11] C. H. Yang , S. C. Lee , T. C. Lin , and S. C. Chen, "Electrical and optical properties of indium tin oxide films prepared on plastic substrates by radio frequency magnetron sputtering," Thin Solid Films, vol. 516, pp. 1984-1991, February 2008.

[12] Y. Q. Li, Z. N. Yu, J. Leng, W. Xue, X. Fan, and Z. Ding, "The effects of inorganic buffer layers on the flexible indium-tin-oxide film's photoelectric properties and bending resistance performance," Acta Optica Sinica, vol. 30, pp. 1205-1210, April 2010.(In Chinese)

[13] E. Fortunato, P. Nunes, A. Marques, D. Costa, H. Águas, and I. Ferreira, et al., "Transparent conductive ZnO:Al thin filmdeposited on polymer substrates by RF magnetron sputtering," Surf. Coat. Technol., vol. 151 – 152, pp. 247–251, March 2002.

[14] H. H. Kim, M. Y. Lee, K. T. Kim, and S. H. Yoon, "Transparent conducting ITO films reactively sputtered on polyethylene terephtalate substrates without heat treatment," Int. J. Mod. Phys. B, vol. 17, pp. 1242-1247, April 2003.

[15] F. C. Lai, L. M. Lin, R. Q. Gai, Y. Z. Lin, and Z. G. Huang, "Determination of optical constants and thicknesses of In2O3:Sn films from transmittance data," Thin Solid Films, vol. 515, pp. 7387-7392, June 2007.

[16] F. C. Lai, M. Li, H. Q. Wang, H. L. Hu, X. P. Wang, and J. G. Hou, et al., "Optical scattering characteristic of annealed niobium oxide films, " Thin Solid Films, vol. 488, pp. 314-320, September 2005.

[17] L. M. Lin, F. C. Lai , Y. Qu, R. Q. Gai, and Z. G. Huang, "Influence of annealing in N_2 on the properties of In_2O_3:Sn thin films prepared by direct current magnetron sputtering," Mater. Sci. Eng., B, vol. 138, pp. 166–171, March 2007.