Indium Tin Oxide (ITO) films on flexible substrates for organic light emitting diodes

Vandana Singh, C.K. Suman and Satyendra Kumar

Department of Physics and Samtel Center for Display Technologies, Indian Institute of Technology Kanpur, India vandanas@iitk.ac.in

Abstract

Flexible electronics opens up new possibilities in applications such as flat panel displays, solar cells, RFID tags, low cost sensors and other disposable electronics devices. For flexible polymer light emitting diodes (PLEDs), transparent and conducting electrodes are required on flexible plastic substrates. In this work we report the results on the growth of smooth ITO films using RF magnetron sputtering. To avoid any thermal distortion, ITO films are deposited at low temperature on various plastic substrates (Mylar and Transphan). ITO films with room temperature conductivity of the order of $10^{-4} \Omega$ -cm having a work function of 4.6 eV were obtained. Atomic force microscopy measurements on these films show a low average roughness of ~1.5 nm (over a scan area of 1500 nm x 1500 nm). While the films deposited at low substrate temperature were amorphous in nature, some crystallinity was observed for those deposited at higher temperatures. These ITO films could be comfortably bent to an angle of 30° and did not reveal any degradation in properties with time. PLEDs were successfully fabricated on these ITO coated plastics with low turn-on voltage.

Keywords: ITO; OLED; flexible; AFM; low temperature processing; RF sputtering.

1. Introduction

Various kinds of transparent conducting oxide (TCO) thin films such as zinc oxide, impurity-doped indium oxides and tin oxides have been widely used as transparent conductors for many opto-electronic applications [1]. Indium tin oxide (ITO) provides good electrical conductivity and high transparency in the visible region hence it is the most commonly used electrode material in flat panel displays. Moreover for organic electronics such as organic light-emitting diodes (OLEDs), organic solar cells and organic thin film transistors, ITO is the best suited anode material as ITO provides good energy-level matching for the efficient injection of holes into the organic layers [2–4]. These OLEDs have more brightness, high efficiency, a wide viewing angle, and quick response time. Currently, organic devices are fabricated on rigid substrates and then encapsulated with glass lid. fabricating the devices on plastic Alternatively, substrates increases the number of OLED applications. Transparent conducting indium tin oxide (ITO) thin films on flexible substrates have many applications. They can be used in plastic liquid crystal display devices, transparent electron-magnetic shielding

materials, flexible electro-optical devices, heat reacting mirrors, etc. These flexible substrates may be paper, thin aluminum foil or polymer (PET, PEN, MYLAR, Transphan and polycarbonate).

Due to the poor thermal endurance of plastic substrates. ITO films are deposited at low temperature. Also, low temperature deposition improves the electrical as well as optical properties of ITO, increases the efficiency of devices. A few researchers have reported ITO film deposited at low substrate temperature by different techniques, especially by sputtering [5-8]. Fan reported a resistivity as low as $5.5 \times 10^{-4} \Omega$ - cm and the transmittance over 80% for ITO films deposited at substrate temperature below 100°C by ion-beam sputtering [5]. For the first time in 1988, Mansingh and Vasant Kumar reported a resistivity of $1 \times 10^{-3} \Omega$ - cm for the ITO films deposited on water-cooled Mylar by RF sputtering [9]. We have also report the conductivity of the order of $10^{-4} \Omega$ - cm and transmission upto 80% of the flexible ITO substrate [10].

In this paper we report the low temperature deposition of ITO on flexible substrate (Mylar and Transphan). This deposition is done by RF magnetron sputtering technique at low RF power. ITO thin films are characterized by AFM, XRD, Kelvin probe, and four point probe and transmission spectra.

2. Experimental Details

ITO thin films were deposited on Mylar and Transphan plastic substrates by RF magnetron sputtering. Transphan sheets are provided by LOFO High Tech Film Germany. [11] The ITO target used was In₂O₃ having 10% SnO₂ and procured from Kurt J. Leskar. The distance between target and substrate was approximately 4 cm. All the substrates were ultrasonically cleaned in iso-propyle alcohol (propan-2ol) and dried. Before deposition, all the samples were annealed in microwave oven at 80°C for 30 min. The deposition process was carried out at 15 sccm of Argon gas, which was controlled through mass flow-meter. The base pressure of sputtering system was $3x10^{-6}$ mbar and process pressure was 8.2×10^{-4} mbar. The applied RF power was 60 W and constant during all the processes. The depositions were done at different temperatures for 20 minutes. As deposition was to be done at low temperature, the highest deposition temperature was 80°C to avoid thermal distortion of the substrate. An increase of 5-8 °C was observed due the plasma.

V. Singh

Finally, optical, electrical and structural properties of the ITO films were studied. Transmission was measured with Perkin-elmer-UV/VIS Lambda 40 spectrometer. Work function measurement was done with Kelvin probe; sheet resistance was measured with 4 point probe; surface profilometer was used for thickness measurement; AFM and XRD were used for nano structural analysis of ITO thin film.

3. Result and Discussion

For any organic device, electrodes are chosen to facilitate charge injection; ITO has a relatively high work function 4.6 eV and is therefore suitable for use as a hole-injecting electrode, and low work-function metals such as Al, Mg, or Ca are suitable for injection of electrons. This work function is well matched with various organic materials (conjugated polymers and small molecules). Fig. 1 shows the energy levels and thicknesses of various layer for a polymer LED formed with a layer of the conducting polymer system PEDOT-PSS, which acts as hole-injecting electrode, formed on indium-tin oxide (ITO) coated substrate. From table 1, it is clear that the work function of deposited ITO is ~ 4.54 eV which is comparable to the commercially available ITO plastic sheet. Results indicate that the work function of deposited ITO is matched properly with energy band gap for PEDOT: PSS for OLED fabrication.

Deposition rate is defined as the film thickness divided by disposition time, and it is important in film thickness control, especially for precise multi-layer coating. Fig.2 shows the deposition rate of the ITO films as a function of temperature at an RF power of 60 W. In RF sputtering, the deposition rate of the films strongly depends on the energy of the sputtered particles arriving at the substrate. The ejected target atoms or molecules undergo collisions with the ambient gas atoms and other sputtered atoms and lose a part of their energy during their transit to the substrate. Deposition rate decreases with temperature.

Sheet resistance and Resistivity of deposited ITO films are shown in Fig.3 and Fig.4 respectively. At room temperature (25° C) we get thick film. Due to high roughness the value of sheet resistance is higher. As the deposition temperature increases sheet resistance of the film increases. Average resistivity is of the order of 8.23 $\times 10^{-4}$ Ω -cm. Yang et al. [12] reported the resistivity of the order of 10^{-4} Ω -cm for the flexible substrate.

Structural characterization of the films has been done by XRD. Fig. 4 shows the diffraction patterns of the films deposited at different temperature. Films deposited without substrate heating were amorphous and the crystallinity improved with the substrate temperature. An XRD spectrum of plastic has a (111) peak at 29.90° angle. ITO on plastic substrates shows a prominent peak at around 29.90° angle. The

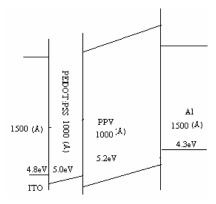


Figure1. Schematic diagram of the energy levels for a polymer LED formed with a layer of the conducting polymer system PEDOT-PSS, which acts as hole-injecting electrode, formed on glass coated with indium-tin oxide (ITO).

Table1. Work function and thickness of ITO on Mylar and plastic substrate at different deposition temperature. Work function of commercial ITO is also given for the comparison

			Work function (eV)	
	Temp.(⁰ C)	Thickness(Å)	Mylar	Transphan
1.	80	4016	4.55	4.56
2.	70	4185	4.56	4.56
3.	60	4216	4.52	4.54
4.	RT(25)	4828	4.55	4.53
5.	Commercial		4.45	

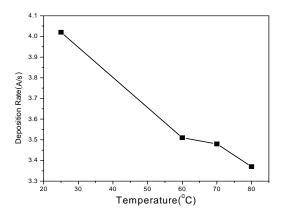


Figure 2. Deposition rate of the ITO films as a function of temperature.

diffraction patterns show the presence of (400) phase in the films. As the temperature was increased the sharpness and intensity of (400) diffraction peak also increased which indicates a preferred orientation along (100) direction. It is clear from the literature also that (100) as preferred orientation is observed when the films are deposited at high temperatures [13].

V. Singh

Surface morphology of the ITO films is studied by AFM. Fig. 6 and Fig. 7 are the typical AFM

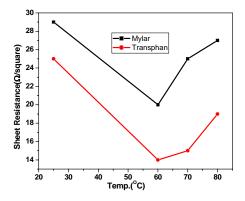


Figure 3. Sheet resistance of ITO on Mylar and Transphan substrate at different Temperature.

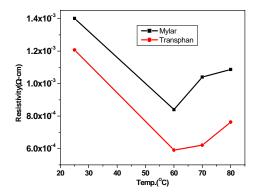


Figure 4. Resistivity of ITO on Mylar and Transphan substrate at different Temperature.

images of ITO films prepared with argon at 80° C. Average roughness of ITO film found was 1.32 nm (1.5 x1.5 µm range) for Mylar and 1.88 nm (1000x 1000 nm range) for Transphan substrates respectively. Transmittance found was 75.0% at 80° C from ITO films on plastic in the visible region [14].

4. Conclusion

An ITO anode with a smooth surface can minimize electrical shorts in the thin functional organic layers in OLED that are very often in the range of 100–200 nm. Our results demonstrate that the process developed in this work may provide a practical approach to fabricate ITO film with a smoother morphology and lower sheet resistance on plastic films at a low processing temperature and RF power. As we reported ITO deposition on two flexible substrates, similar results obtain in both the cases verifies the consistency of the deposition process. Smooth ITO films with a resistivity of $8.23 \times 10^{-4} \Omega$ -cm and work function of 4.54 eV were obtained. These films are highly oriented in (100) direction. At RT we get high deposition rate but sheet resistance is very high. It indicates that for good quality

ITO film some heating is required. We have earlier use similarly fabricated ITO

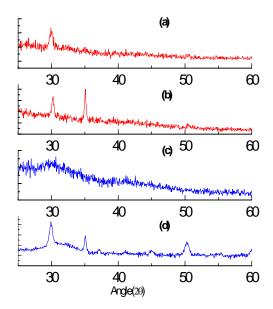


Figure 5. XRD of ITO at different temperatures.(a) Mylar substrate at RT (b) At 80°C (c) Transphan substrate at RT (d) At 80°C

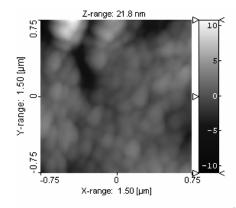


Figure 6. AFM image of ITO on Mylar at 80°C

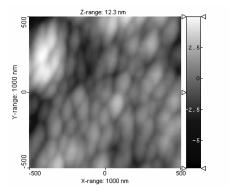


Figure 7. AFM image of ITO on Transphan at 80°C

V. Singh

films as the anode in OLEDs and studied the device performance [15]. The EL performance of these OLEDs was identical with the devices made with commercial ITO-coated rigid glass substrates. We expect the similar results with OLED prepared with films made and reported in this work.

5. Acknowledgements

Authors acknowledge the support from Department of Science and Technology, India and Samtel center for display technologies, IIT Kanpur India for conducting the reported work. Author also acknowledge to the company providing plastic substrate.

References

- K. L. Chopra, S. Major and D. K. Pandya, "Transparent conductors—A status review," *Thin Solid Films*, Vol. 102, pp 1-46, 1983
- F. Nuesch, E. W. Forsythe, Q. T. Le, Y. Gao, and L. J. Rothberg, "Importance of indium tin oxide surface acido basicity for charge injection into organic materials based light emitting diodes", J. Appl. Phys., vol. 87, pp 7973-7980, 2000.
- 3. Q. T. Le, F. Nuesch, L. J. Rothberg, E. W. Forsythe, and Y. Gao, "Photoemission study of the interface between phenyl diamine and treated indium–tin–oxide" *Appl. Phys. Lett.*, vol. 75, pp 1357-1359, 1999.
- M. G. Mason, L. S. Hung, C. W. Tang, S. T. Lee, K. W. Wong, and M. Wang, "Characterization of treated indium-tin-oxide surfaces used in electroluminescent devices," *J. Appl. Phys.*, vol. 861,688-1692, 1999.
- 5. J.C.C. Fan, "Preparation of Sn doped In₂O₃ (ITO) films at low deposition temperatures by ion beam sputtering," *Appl. Phys. Lett.*, vol .34, pp 515-517, 1979.
- W.F. Wu, B.S. Chiou, "Properties of radiofrequency magnetron sputtered ITO films without in-situ substrate heating and post-deposition annealing," Thin Solid Films, vol. 247,pp 201-207, 1994.

- L Kerkache, A Layadi, E Dogheche and D R'emiens, J. Phys. D: Appl. Phys., vol.39, pp 184-189,2006.
- 8. Y. Hoshi, R. Ohki, "Low energy rf sputtering system for the deposition of ITO thin films," *Electrochimica Acta*, vol. 44, pp 3927-3932, 1999.
- A. Mansingh and C. V. R. Vasant Kumar, "R.f.sputtered indium tin oxide films on water-cooled substrates" *Thin Solid Films*, vol.167, pp Lll-L13, 1988.
- 10. V. Singh, B. Saswat and S. Kumar, "Low temperature deposition of Indium tin oxide (ITO) films on plastic substrates," *Material Res. Soc. Symp. Proc.*, vol. 869, pp D2.9.1-D2.9.6, 2005.
- 11. http://www.lofo.com
- Z. W. Yang, S.H. Hana, T.L. Yanga, Lina Yea, D.H. Zhangb, H.L. Maa, C.F. Cheng, Bias voltage dependence of properties for depositing transparent conducting ITO films on flexible substrates, *Thin Solid Films*, vol.366, pp 4-7, 2000.
- 13. E. Shanthi, A. Bannerjee, V. Dutta and K.L. Chopra, "Annealing characteristics of tin oxide films prepared by spray pyrolysis" *Thin Solid Films*, vol. 71,pp 237-244, 1980.
- 14. V. Singh, Arpit and S. Kumar, "Structural and electrical properties of Indium Tin Oxide (ITO) film on flexible and glass substrates for the fabrication of organic light emitting devices" Proc. Of *Thirteenth International Workshop on The Physics of Semiconductor Devices*, pp 1535-1538, 2005
- 15. V. Singh, S. Bharat and S. Kumar, "Fabrication of Organic Light Emitting Displays on Transparent conducting Oxide film deposited at low temperature on plastic substrates," Communicated to *Vacuum*.