# Effect of Sputtering Condition on Electrical and Optical Properties of Indium-Tin Oxide Thin Films

Nguyen Thi Thu Hien<sup>1,2</sup>, Tran Quoc Hoan<sup>1,2</sup>, Dang Viet Anh Dung<sup>1</sup>, Tran Duc Huy<sup>1</sup>, Pham Anh Tuan<sup>1,2</sup>, Nguyen Huu Dung<sup>1</sup>, Vu Ngoc Phan<sup>1</sup>, Nguyen Duy Cuong<sup>1\*</sup>

<sup>1</sup>Hanoi University of Science and Technology – No. 1, Dai Co Viet Str., Hai Ba Trung, Ha Noi, Viet Nam <sup>2</sup>Electric Power University - No.235, Hoang Quoc Viet, Hanoi.

Received: June 12, 2018; Accepted: June 24, 2019

## Abstract

Indium-tin oxide (ITO) thin films were deposited by radio frequency (rf)-magnetron sputtering at different substrate temperatures and oxygen concentrations. Oxygen concentration affects significantly on the electrical and optical properties of ITO films. The best sample was observed at 1% oxygen with the sheet resistance of 227  $\Omega/\Box$ , resistivity of 56x10<sup>-4</sup>  $\Omega$ cm, and transmittance (at 550 nm) of 73%. The substrate temperature affects strongly on the surface morphology and electrical properties of ITO films. The size of ITO nanocrystalites increased with the increasing substrate temperature, indicating an improvement of the crystallinity. The sheet resistance and resistivity of ITO films are decreased with raising the substrate temperature, and are around 17  $\Omega/\Box$  and  $4x10^{-4}$   $\Omega$ cm at 400 °C, respectively. The higher substrate temperature shows better optical property.

Keywords: ITO thin films, substrate temperature, oxygen concentration, sheet resistance, transmittance.

#### 1. Introduction

Recently, transparent conductive oxide (TCO) has received the consideration around the world because of their attractive properties such as low resistivity and high transmittance in the range of visible light range [1-4]. Because of this advantage, TCO materials are widely applied in the fields such as liquid display, light emitting diodes and photovoltaic [5-7]. In addition, TCO is also used in other fields such as gas sensors, catalytic and electronic etc [8-10].

Excepting the applications as mentioned above, some TCOs also reflect the light in the infrared range. With high infrared reflective properties and high transmittance in the visible area, TCO materials are applied to low emission glass in buildings or cars [11]. One of the most used materials is the Indium-Tin Oxide (ITO) because it is quite durable with the environment, high transmittance and low resistivity.

In this study we fabricate ITO films by sputtering and analyze to improve the optical and electrical properties of the ITO films by controlling the oxygen concentration during sputtering and the substrate temperature.

#### 2. Experimental

ITO films were deposited on slide glass substrates with a power of 80 W at working pressure of 5 mtorr by rf-magnetron sputtering. Base pressure is 4 x  $10^{-7}$  torr. An ITO target (weight ratio of In<sub>2</sub>O<sub>3</sub>:SnO<sub>2</sub>= 90:10; purity of 99.99%) with diameter of 2 inch was used as source material for sputtering ITO films. The substrate was heated at different temperatures of room, 100, 200, 300, and 400 °C. The oxygen concentrations (O<sub>2</sub>/(O<sub>2</sub>+Ar) x 100%) during sputtering are 0, 1, 1.5, 2, 2.5, 3, 4, and 5%. The thickness of all ITO films in this paper is ~ 260 nm.

The surface morphology and thickness of ITO films were measured by field emission scanning electron microscopy (FESEM) (JSM-7600F, Jeol) at Laboratory of Electron Microscopy and Microanalysis (BKEMMA). The optical properties were measured by UV-Vis-NIR spectrophotometer (Cary 5000). The sheet resistance of ITO films was measured by four-probe equipment.

#### 3. Results and discussion

In order to investigate the effect of oxgen concentration on the surface morphology of ITO films we measured FESEM, and the results are shown in Fig.1. Based on the surface SEM images, the size of the nanoparticles on the surface of the ITO films has varied with the change of oxygen concentration from 0 to 5%. ITO films deposited with 0% oxygen showed that the nanoparticles on the surface were

<sup>\*</sup> Coresponding author: Tel.: (+84) 164.980.5375 Email: cuong.nguyenduy@hust.edu.vn

quite large. However, if we view closely, the nanoparticles on the surface are formed by smaller particles, not single crystallites. When increasing the oxygen concentration by 1-5%, the size of the nanoparticles (formed by smaller particles) seems to decrease. The size of small nanoparticles is below 10 nm.



**Fig. 1.** Surface FESEM image of ITO films deposited at room temperature with various oxygen concentrations

To analyze the change of the electrical properties of ITO films at different oxygen concentrations, we measured sheet resistance using the four-probe method. Fig.2 is the variation of sheet resistance and resistivity via the oxygen concentrations. In the range of 0-1% oxygen, the sheet resistance and resistivity values decrease with the increase in oxygen concentration. However, when the oxygen concentration is higher than 1%, the sheet resistance and resistivity become greater at higher oxygen concentrations. Namely, the sheet resistance and resistivity of ITO films with oxygen concentration of 0% are 250.4  $\Omega/\Box$  and 62.6 x  $10^{-4} \Omega$ .cm, respectively. When 1% oxygen was added to sputtering gas, ITO films shows a sheet resistance of 226.8  $\Omega/\Box$  and resistivity of 56.7 x 10<sup>-4</sup>  $\Omega$ .cm. With the oxygen concentration of 2.5, 3, 4, and 5% the sheet resistance and resistivity increased drastically; these values of 5%O2 ITO films are 630.1  $\Omega/\Box$  and 157.5 x 10<sup>-4</sup>  $\Omega$ .cm. The increase of sheet resistance at and resistivity with oxygen concentration may be due to the decrease in

crystallite size as shown in Fig.1. In general, the sheet resistance and resistivity values of all ITO films deposited at different oxygen concentrations and room temperature are still rather high. The reason can be attributed to low crystallinity with small crystallites.



**Fig. 2.** Variation of sheet resistance and resistivity of ITO films via oxygen concentrations.



**Fig. 3.** Transmittance of ITO films deposited by rfmagnetron sputtering at room temperature with different concentrations of oxygen.

Fig.3 shows the transmittance spectra of ITO films deposited with various oxygen concetrations. The ITO film with 0% oxygen depicts low transmittance; the color of this ITO films is dark orange. As the oxygen concentration increased, the ITO transmittance of the films increased dramatically. Specifically, at 550 nm, the transmittance of the ITO film with 0% oxygen was  $\sim$  58%, while the films with an oxygen concentration of 1-5% showed a transmittance of  $\sim$  73%. Low transmittance of 0% oxygen film may be due to the lack of oxygen during sputtering to form the ITO

compound. Generally, the transmittance of ITO deposited at room temperature is still low; it should be more improved.

For application as transparent conductive electrodes in display and photovoltaic devices, the sheet resistance and transmittance should be  $\sim 10 \ \Omega/\Box$  and 80%, respectively. To compare with this requirement, the sheet resistance of room temperature-deposited ITO films is so high.

As mentioned above, the high sheet resistance of the ITO films is due to the size of the ITO nanoparticles too small, resulting in many boundaries formed in the films, hindering the transfer of free electrons. This is the main reason causing high sheet resistance of room temperature-deposited ITO films. To improve the sheet resistance, the size of the ITO crystallites needs to be increased. Therefore, we have investigated ITO films at different substrate temperatures.

Fig.4 is the surface SEM image of ITO films deposited with the oxygen concentration of 1% at room temperature, 100, 200 300 and 400 °C. Here we choose 1% oxygen because the ITO film has the lowest sheet resistance as shown in Fig.2. The size of the ITO crystallites has increased steadily with the increase of the substrate temperature. ITO nanoparticles are deposited at room temperature for only a few nanometers. However, the size of ITO nanoparticles was in the range of 20-100 nm at 400 °C.



**Fig. 4.** Surface SEM image of ITO films deposited at different substrate temperatures with oxygen concentration of 1% under working pressure of 5 mtorr.

Fig.5 shows the variation of sheet resistance and resistivity via the substrate temperature. Both sheet resistance and resistivity have decreased sharply with increasing temperature. Sheet resistance and resistivity decreased from 250.4  $\Omega/\Box$  and 62.6 x 10<sup>-4</sup>  $\Omega$ .cm (room temperature) to 17.6  $\Omega/\Box$  and 4.4 x 10<sup>-4</sup>  $\Omega$ .cm (400 °C).



**Fig. 5.** Variation of sheet resistance and resistivity with substrate temperatures.



**Fig. 6.** Transmittance of ITO films deposited by rfmagnetron sputtering with oxygen concentration of 2% at different substrate temperatures.

The sharp decrease in sheet resistance as well as the resistivity is explained by the increase in crystallite size as seen in Fig.4. The sheet resistance is quite close to the requirement for transparent conductive electrodes.

To analyze the substrate temperature on the optical properties of ITO films, the transmittance

spectra were recorded and the results are shown in Figure 6. With the increase of substrate temperature, transmittance of ITO films in the range of 520 - 950 nm tends to increase and the transmittance edge shifts to shorter wavelength. At the temperatures of 100-300 °C, the transmittance changes insignificantly; however when the substrate temperature increases to 400 °C, the transmittance is quite high. The transmittance of 400 °C-ITO films is 84.3% at the wavelength of 550 nm. The increase of transmittance and the shift of transmittance edge towards to shorter wavelength are due to the improvement of the crystallinity of ITO films.

### 4. Conclusions

ITO films were deposited with the power of 80 W and different oxygen concentrations at temperatures by rf-magnetron sputtering. The oxygen concentration strongly affects on the sheet resistance and resistivity. Sheet resistance of ITO films deposited at room temperature is rather high. The electrical and optical properties were improved significantly by increasing substrate temperature. ITO film fabricated with oxygen concentration of 1% at 400 °C under pressure of 5 mtorr shows a sheet resistance of 17.6  $\Omega/\Box$ , resistivity of 4.4 x 10<sup>-4</sup>  $\Omega$ .cm, and transmittance of 84.3% at the wavelength of 550 nm. The electrical and optical properties of 400 °C-deposited ITO films are appropriate for application as transparent conductive electrodes. The optical and electrical properties of ITO films may be further improved at the deposition temperatures of higher 400 °C, however, the substrate temperature of our sputter equipment is limited  $\leq 400$  °C.

### Acknowledgement

This research is funded by Vietnam National Foundation for Science and Technology Development (NAFOSTED) under grant number 103.02-2017.45.

#### References

- F. Niino, H. Hirasawa, and K. Kondo; Deposition of low-resistivity ITO on plastic substrates by DC arcdischarge ion plating; Thin Solid Films 411 (2002) 28-31.
- [2] J. Lee, S. Lee, G. Li, M. A. Petruska, D. C. Paine, and

S. Sun; A Facile Solution-Phase Approach to Transparent and Conducting ITO Nanocrystal Assemblies; J. Am. Chem. Soc., 134 (2012) 13410–13414.

- [3] C.G.Granqvist and A.Hultaker; Transparent and conducting ITO films: new developments and applications; Thin Solid Films 411 (2002) 1-5.
- [4] T. Minami; Present status of transparent conducting oxide thin-film development for Indium-Tin-Oxide (ITO) substitutes; Thin Solid Films 516 (2008) 5822-5828.
- [5] U.Betz, M.K. Olsson, J. Marthy, M.F.Escola, F.Atamny; Thin films engineering of indium tin oxide: Large area flat panel displays application; Surface and Coatings Technology 200 (2006) 5751-5759.
- [6] D. Aaron, R. Barkhouse, O. Gunawan, T. Gokmen, T. K. Todorov, D. B. Mitzi; Device characteristics of a 10.1% hydrazine processed Cu<sub>2</sub>ZnSn(Se,S)<sub>4</sub> solar cell; Prog. Photovolt: Res. Appl. 20 (2012) 6–11.
- [7] Z. K. Tan, R. S. Moghaddam, M. L. Lai, P. Docampo, R. Higler, F. Deschler, M. Price, A. Sadhanala, L. M. Pazos, D.Credgington, F. Hanusch, T. Bein, H. J. Snaith, and R. H. Friend; Bright light-emitting diodes based on organometal halide perovskite; Nature Nanotechnology 9 (2014) 687–692.
- [8] S. K.Mishra, S. Rani, and B. D.Gupta; Surface plasmon resonance based fiber optic hydrogen sulphide gas sensor utilizing nickel oxide doped ITO thin film; Sensors and Actuators B: Chemical 195(2014) 215-222.
- [9] M. Kato, T. Cardona, A. W. Rutherford, and E. Reisner; Photoelectrochemical Water Oxidation with Photosystem II Integrated in a Mesoporous Indium– Tin Oxide Electrode; J. Am. Chem. Soc. 134 (201) 8332-8335.
- [10] M. G. Helander, Z. B. Wang, J. Qiu, M. T. Greiner, D. P. Puzzo, Z. W. Liu, Z. H. Lu; Chlorinated Indium Tin Oxide Electrodes with High Work Function for Organic Device Compatibility; Science, 332 (2011) 944-947.
- [11] Brian G. Lewis and David C. Paine; Applications and Processing of Transparent Conducting Oxides; MRS Bulletin 25 (2000) 22-27.