

# Improving the structural, optical and electrical properties of ITO Nano-layered thin films by gas flow Argon

Mehdi Q.Z<sup>1</sup>, Gurumurthy Hegde<sup>1a</sup>, Mohamad Ashry Bin Juusoh<sup>1</sup>, Jinan B. Al-Dabbagh<sup>1</sup>, Naser Mahmoud Ahmed<sup>2</sup>

<sup>1</sup> Faculty of Industrial Sciences and Technology, University Malaysia Pahang, Kuantan, 26300, Malaysia

<sup>2</sup> School of Physics / University Sains Malaysia/ Penang/11800 / Malaysia

<sup>1a</sup>Author for Correspondence: [murthyhegde@gmail.com](mailto:murthyhegde@gmail.com)

**Keywords:** ITO Argon gas flow, transparent films, resistance.

**Abstract.** Indium tin oxide (ITO) thin films of 150 nm thickness were deposited on quartz glass substrates by RF sputtering technique, followed by thermal annealing treatment. In this technique, the samples have been annealed at different temperature, 300°C, 400°C, 500°C respectively in Argon gas flow. Structural and surface morphological properties were analyzed by X-ray diffraction (XRD) and Atomic Force Microscopy (AFM) after annealing. The XRD showed a polycrystalline structure of ITO film with maximum peak intensity at  $2\theta = 30.54$ , <222> orientation without any change in the cubic structure. Continuous and homogeneous films obtained by the AFM after annealing treatment. The visible spectrum from the spectrophotometer showed high transparency between 81% and 95% in the. Increasing the annealing temperature yields evenly distributed pyramidal peaks shaped particles with low roughness. Resistance of ITO thin film was significantly improved from 8.75 k $\Omega$  to 1.96 k $\Omega$  after 10 minute from 300°C to 500°C annealing temperatures respectively under Argon gas flow. ITO films physical properties would be well improved by this method which is highly suitable for cost effective photonic devices.

## Introduction

The ITO Thin films are crystalline and also nanostructure materials on the surface of quartz glass substrate enhanced by a physical method, which play important role in virtually all electronic and optical devices. Having long been used as anti-reflection coatings on window glass, video screens camera lenses and other optical devices [1-5], ITO films already acquired major electronic market shares. These films are less than 150 nm thickness made from conducting transparent material having refractive indices less than those of the reported substrates [6-8]. Although recently applied techniques with the ITO films are having great applications in solar energy conversion, semi-transparent films in schottky barrier solar cells combinations, thin films in photo-thermal devices, which generate low or high grade heat, while thin semiconductor films on ether glass or material substrate are promising low cost solar cells but they are too much expensive due to their fabrication process. Application of thin films deposition for optical, electronic and optoelectronic device has become an industrial technique in most advanced countries with high technological and sophisticated facilities [9-18]. The structural, morphological, optical, and electrical properties of the ITO films have been analyzed as a function of the annealing time during thermal argon gas flow [19-22]. In this present work, investigation of the influence of annealing temperatures on the characteristics of the ITO thin films while treating with Argon to develop better cost effective thin films.

## Experimental

### Thin Film Preparation

The ITO target (99.9% purity) a commercially available hot-pressed pallet composed of 90 wt. % In<sub>2</sub>O<sub>3</sub> with 10 wt. % SnO<sub>2</sub>. The ITO films with 150 nm thickness were deposited at 200°C of

temperature at basic vacuum pressure of about  $3 \times 10^{-5}$  Torr when Argon gas plasma was introduced, pressure increases to  $6 \times 10^{-3}$  Torr.

The ITO thin film on the quartz glass substrate was inserted in to the heat tube furnace which was subjected to argon gas flow, later treated at deferent temperatures of about 300°C, 400°C and 500°C respectively. The thermal annealing treatment was carried out for each sample in the argon gas flow for about 10 minutes annealing time.

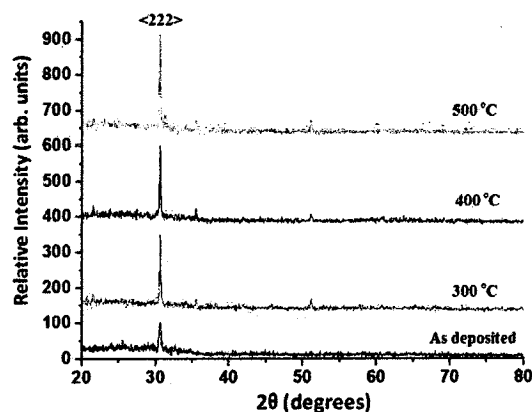
### Thin film measurement

Different time scales were determined using XRD technique with a Rigaku Mintron X-ray diffraction at room temperature, with monochromatic Cu K $\alpha$  ( $\lambda = 1.540562 \text{ \AA}$ ) in the scan range  $2\theta$  between 15° to 90°. Prior to the AFM measurements, the films were ultrasonically washed in methanol and dry with nitrogen. Their phase composition was annealed at different temperatures. After thermal annealing process, each sample was taken out for Atomic force microscopy (AFM) measurements to study the surface morphology of the films. Surface morphology of the AFM films were measured using Dimension Edge, Bruker instrument in tapping mode and nano-drive dimension-edge-tapping image-processing software were used to measure the surface roughness of the film. Optical transmittance measurements were measured in the wavelength range of 200–1100 nm using an OCEAN OPTICS, HR 2000+ miniaturized spectrophotometer. Electrical properties of the films were measured using DIGITAL MULTIMETER (VICTOR VC97).

## Result

### Structural properties

The XRD diffraction was carried out to evaluate the crystal structure of ITO films deposited on the quartz glass substrate under various conditions and can be seen in Fig. 1. The representative XRD diffraction measurement profiles of ITO thin films annealed at temperature 300°C, 400°C, 500°C with as deposited sample. It shows the measurement of the samples after treatment by argon gas flow, and fixed time is 10 minutes. The film has one strong  $\langle 222 \rangle$  crystal orientation where the diffraction intensity increasing with changes in the annealing temperature. The oxidation of the growing film obtained from RF sputtering technique affects the modification of the preferred



**Fig. 1, XRD diffraction by used thermal annealing technique with argon gas flow.**

orientation growth from  $\langle 222 \rangle$ . A higher temperature annealing is known to provide a greater driving force of the nucleation and growth of the ITO films. However, it is not found on the coupling agent distinctive peak coupling agents because the low concentration cannot be tested. Furthermore, it did not change the crystal structures of the ITO film by coupling agent compared with previous studies. The film XRD chart that shows peak at value  $2\theta = 30.54$ ,  $\langle 222 \rangle$  the characteristic peak of cubic  $\text{In}_2\text{O}_3$  structure. These XRD results indicate that crystalline ITO thin films can be obtained at high  $\langle 222 \rangle$  peak by using thermal annealing with argon gas flow treatment.

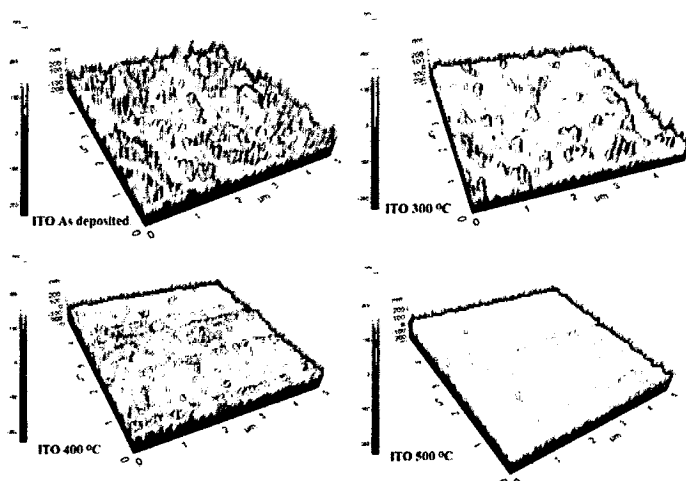
### Morphology properties

Among the important benefits that can be obtained from atomic force microscopy (AFM) were used to investigate the nanostructure with the roughness degree for the films [23]. Both the X-ray and AFM after the annealing temperature of the samples, we found that both were improved as compared with as-deposited coating films.

**Table 1, Shows measurements for the as-deposited and annealing temperature 300°C, 400°C, 500°C by used argon gas flow technique.**

| Annealing Temperatures | $R_{rms}$<br>nm | $R_a$<br>nm | $R_{max}$<br>nm | R<br>k $\Omega$ | T%     |
|------------------------|-----------------|-------------|-----------------|-----------------|--------|
| As deposited           | 1.96            | 1.74        | 22.45           | 8.75            | 82.231 |
| 300 °C                 | 1.21            | 1.01        | 14.34           | 4.6             | 86.505 |
| 400 °C                 | 0.73            | 0.85        | 12.94           | 2.79            | 91.825 |
| 500 °C                 | 0.57            | 0.67        | 7.16            | 1.96            | 94.484 |

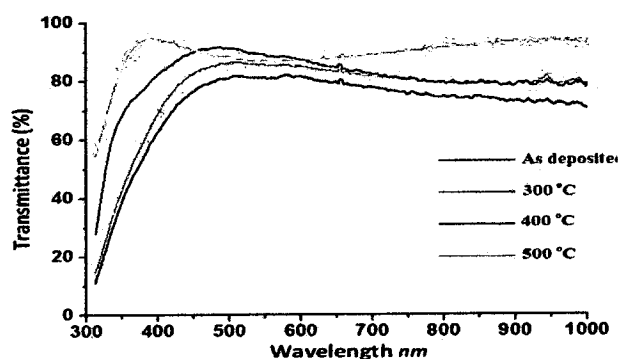
Charge carriers traveling on the rough surface may impede the path along the greater portion of their free medium, and thus leads to a decrease in mobility. The surface morphological of the ITO films those were treated with the annealing times were observed as a function of annealing temperature by using AFM (Fig.2). Grain size improvement is very important for the layered prepared by using argon gas flow, which shown the smallest grains as comparable with as-deposited sample. With an increase in the annealing temperature, the surface morphologies of the films seem to change from a rough surface to smoothness pattern. Such smoothness morphologies of the ITO films were observed up to the root mean square  $R_{rms} = 0.57$  nm at temperature treatment of 500°C, also a surface roughness decreasing was observed from the table 1. The annealing treatment using argon gas flow, seems the  $R_{rms}$  surface roughness found to be 1.21, 0.73, 0.57 nm at 300°C, 400°C, 500°C annealing temperature, respectively in Fig. 2 and table 1. However, the particle size for surface morphology of the ITO also is of great significance in bounding the hall mobility.



**Fig. 2, Shows AFM images samples for the as-deposited and annealing temperature 300°C, 400°C, 500°C by used argon gas flow technique.**

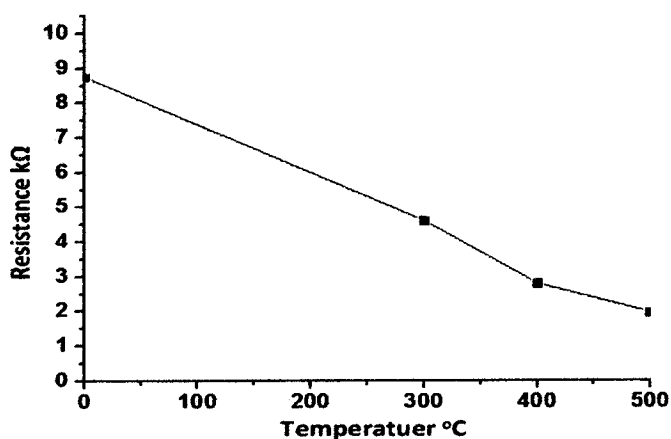
### Optical and electrical properties

The optical properties of the Nano-layered ITO thin films at thickness 150 nm were produced on quartz glass substrates. The transmittance spectra using UV, VIS and NIR spectral ranges are given in Fig.3. In argon gas flow and thermal treatment, the film showed high transparent properties in the visible and near infrared range because of the long spectrophotometer wavelength. The increasing values of annealing temperature cause a low electrical resistance with high transmittance as the show in table 1.



**Fig.3, the relationship between Transmittance and wavelength for the ITO films in Argon gas flow technique.**

The as-deposited sample was amorphous ITO film has a dark brownish form, with optical transmission in the visible range being about 82.231 %. The resistance R value for as-deposited ITO film without annealing treatment are 8.75 K  $\Omega$ , but after raising the annealing temperature causing for a decrease in the resistance as show in Fig.4 and table 1. However, the observed variation in resistance with annealing temperature is related to a change in transmission in the visible range. It is observed that at 500°C annealing temperature with argon gas flow, increase in high transmittance and resistance were obtained say, 94 % and 1.96 K $\Omega$ , respectively as shown in fig.4 and table 1.



**Fig. 4, the Sheet resistance for the ITO thin film by used Argon flow treatment.**

The effect of argon gas flow during the annealing treatment on the optical and electrical properties of the ITO thin films is explained in the table 1 at the different annealed temperature. However, the transparent of ITO film on quartz glass which used is wider than in argon gas flow. These changes

are under investigation in the sense that the high transmittance should decrease the resistance. This study is having great potential to use for many photonic devices.

### Summary

Indium tin oxide (ITO) thin films of 150 nm thickness were deposited on quartz glass substrates by RF sputtering technique, followed by thermal annealing treatment using different annealing temperature were studied using an Argon gas flow. The XRD showed a polycrystalline structure of ITO film with maximum peak intensity at  $2\theta = 30.54^\circ$ ,  $\langle 222 \rangle$  orientation without any change in the cubic structure. Increasing the annealing temperature yields evenly distributed pyramidal peaks shaped particles with low roughness. Resistance of ITO thin film was significantly improved from 8.75 k $\Omega$  to 1.96 k $\Omega$  after 10 minute from 300°C to 500°C annealing temperatures respectively under Argon gas flow. Obtained thin films were highly useful in electronic industries.

### References

- [1] Gurusurthy Hegde, Yuvaraj A R, Gan Siew Mei , Siti Aisyah Mustapha, Mashitah M Yusoff, V.G.Chigrinov, Proc. of SPIE Vol. 9005 (2014) 90050B-90050B-6.
- [2] RA Alla, G Hegde, L Komitov, Applied Physics Letters 102 (23) (2013) 233505; M Qasim, JB Al-Dabbagh, AN Abdalla, MM Yusoff, G Hegde, Nano Hybrids 4 (2013) 21-31.
- [3] O Elamain, G Hegde, L Komitov, Applied Physics Letters Vol 103 (2) (2013) 023301.
- [4] Ramesh M. Krishna, Materials Letters 66 (2012) 233–235.
- [5] S. Worasukkhunga, Procedia Engineering 00 (2012) 780 – 786.
- [6] Sung Mook Chung, Ceramics International 38S (2012) S617–S621.
- [7] A. Solieman, M.A. Aegerter, Thin Solid Films 502 (2006) 205–211.
- [8] L.R. Cruz, C. Legnani, I.G. Matoso, C.L. Ferreria, H.R. Moutinho, Materials Research Bulletin 39 (2004) 993–1003.
- [9] H. Han, J.W. Mayer, T.L. Alford, Journal of Applied Physics 100 (2006) 083715,1–083715,6.
- [10] T. Minami, Thin Solid Films 516 (2008) 5822–5828.
- [11] D.-S. Liu, C.-S. Sheu, C.-T. Lee, C.-H. Lin, Thin Solid Films 516 (2008) 3196–3203.
- [12] C.-H. Yang, S.-C. Lee, T.-C. Lin, W.-Y. Zhuang, Materials Science Engineering B 134 (2006) 68–75.
- [13] Y.M. Kang, S.H. Kwon, J.H. Choi, Y.J. Cho, P.K. Song, Thin Solid Films 518 (2010) 3081–3084.
- [14] M.M. El-Nahass, Materials Science and Engineering B 177 (2012) 145– 150.
- [15] A. Solieman, Materials Chemistry and Physics 134 (2012) 127– 132.
- [16] B. Leif Hanson, Radiation Measurements 46 (2011) 1595e1597.
- [17] C.J. Lee, Thin Solid Films 522 (2012) 330–335.
- [18] C. Guillén, Materials Chemistry and Physics 112 (2008) 641–644.
- [19] Chang S. Moon, Jeon G. Han, Thin Solid Films 516 (2008) 6560–6564.
- [20] Y. Masuda, N. Saito, R. Hoffmann, M.R. De Guire, K. Koumoto, Sci. Technol. Adv. Mater. 4 (2003) 461.
- [21] E. Williams, K. Haavisto, J. Li, G. Jabbour, Adv. Mater. 19 (2007) 197.
- [22] A.R. Duggal, J.J. Shiang, C.M. Heller, D.F. Foust, Appl. Phys. Lett. 80 (2002) 3470.
- [23] Y.-H. Tak, K.-B. Kim, H.-G. Park, K.-H. Lee, J.-R. Lee, Thin Solid Films 411 (2002) 12.