Duaa A. Mohammed
Applied Science Department,
University of Technology,
Baghdad, Iraq.
duaa.adeeb93@gmail.com

Muslim F. Jawad
Applied Science Department,
University of Technology,
Baghdad, Iraq
drmuslim11@yahoo.com

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Studying the Effect of Annealing Temperature on some Physical Properties of In_2O_3 Thin Films

Abstract-*In this study, In_2O_3 thin films were deposited on quartz substrates by pulsed laser deposition technique at room temperature and followed by thermally annealing at 300 ℃, 400 ℃ and 500 ℃ for 1 hour. The optical band gap was found to increase with the annealing temperature from 3.5 to 3.85 eV and the transmittance was observed above 90%. XRD results show that the films are polycrystalline in nature and crystallizes with preferred orientation (222). SEM images show that the films are high homogenous and they contained uniformly distributed small grains.*

Keywords- *PLD, In_2O_3 thin films, SEM, morphological properties.*

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1. Introduction

Conducting, transparent, electrically electrodes are one of the most significant optoelectronics materials .These optoelectronics materials are in prospect to show high transmittance overcoming 90% in the visible region. Various studies have been accomplished over many years to efficiently and economically present indium oxide based thin films due to their highly conductive, transparent nature [1]. Lowest resistivity was observed in indium oxide based transparent conducting oxides of many diverse metal oxides [2]. Unique shape and size dependent properties were noticed in degenerate indium oxide thin films and have been formed by number of different deposition techniques, which include PLD [3], electron beam evaporation [4], spray pyrolysis [5], thermal evaporation [6] and sol-gel [7]. Usually, In_2O_3 crystallizes into a cubic bixtype structure and has a melting point temperature of 1910°C. It displays a direct band gap between (3.55 and 3.75) eV and that is highly conductive, which is an uncommon property for wide band gap material [8], in its stoichiometric state, it is an insulator but it becomes more conductive when its oxygen deficient is increasing [9]. Simple and inexpensive vacuum evaporation techniques were employed to deposit metal indium thin films, performing this technique by laser radiation heating local oxidization will be permitted of indium thin films, enabling the conductivity and transparency of thin films to be controlled [10].

The ultimate goal of our study is to acquire a homogeneous deposited In_2O_3 thin film on flat optical quartz substrate with aid of pulsed laser deposition technique. In this research, some physical characteristics of these films are characterized by XRD, SEM and UV-visible measurements.

2. Experimental Work

Fabrication of In_2O_3 thin films by pulsed laser deposition was achieved by using a vacuum chamber that was evacuated down to a pressure of 4×10^{-3} mbar. Indium oxide targets of 1 cm diameter were prepared by cold pressing the In_2O_3 high purity (99.999%) powder using a hydraulic press of 5 tons. Thin films were deposited on optically flat quartz substrate that is well cleaned by soaking for 1 hour in ethanol and acetone then dried by hot air. Q-switched Nd:YAG laser with 7 ns pulse duration and wavelength λ=1.064 nm at 10Hz pulse rate was focused through 10 cm focal length lens to irradiate the In_2O_3 target. Target is put at 45°angle of incidence delivering (600,700 mJ/pulse) laser pulse energy at the surface using 100 pulse while the substrate was placed at a distance of 3cm from the target at room temperature (27°C). Thickness of the films was measured by optical interferometer method and found to be (158,244) ±1.5 nm, changing with the pulse energy. Films were post- deposition annealed at (300, 400 and 500) ℃ for 1 hour by nabertherm furnace.

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2412-0758/University of Technology-Iraq, Baghdad, Iraq

The microstructure of the In_2O_3 films was examined by X-ray diffraction (XRD) employing SHIMADZU-6000 X-ray diffractometer equipped with CuK_α radiation (λ=1.54506 A°) with scanning speed of 5 deg/min in 2Ɵ mode. Optical transmittance spectra are recorded by UV-visible spectrophotometer (Shamadzu type) in the range of (300-1100) nm.

Morphological studies and particle size of In_2O_3 films are carried out using a Scanning Electron Microscope (SEM) tescan VEGA3 (AA-3000 model, Angstrom Advanced Inc, USA).

By using tauc relationship [15] the optical band gap is evaluated:

$$(\propto \mathrm{hv})^2 = \mathrm{A}\left(\mathrm{h\,v} - \mathrm{E_g}\right) \quad (1)$$

Where A is the edge width parameter (constant), E_g band gap and α optical absorption coefficient.

3. Results and Discussion

I. structural studies

All films prepared by PLD on the quartz substrates are dense and homogeneous. One of the substantial parameter that influence the films' physical properties is film thickness as its increases. The grain size increasing and the crystallinity is improving [9]. As shown in XRD spectra patterns of the films tell about their crystallinity where it can be concluded that the films showed polycrystalline nature. Strong and sharp peaks correspond to the body centered cubic (bcc) phase are present at 2Ɵ=30.6°, 2Ɵ=35.6° and 2Ɵ=51.2° corresponding to (222), (400) and (440) planes, respectively. The prominent peaks are in good agreement with JCPDS card (06-0416) for the 2Ɵ scans between 10° and 60°. Results of XRD analysis as shown in Figure 1. It was revealed that the In_2O_3 thin film with 158 ± 1.5 nm thickness and annealed at 300 ℃ and 400 ℃ gave no spotted diffraction peaks which mean it's in the amorphous state and annealing at 500 ℃ crystallize the films to cubic In_2O_3. Furthermore, film's oxygen deficiency increases when films are annealed [9]. Scherrer equation was used to estimate the crystallite size 125.8 nm, 94.4 nm and 16 nm for the films annealed at 500 ℃, 400 ℃ and 300 ℃, respectively as it increases with increasing annealing temperature as reported in [11] as a result of the improvement and enhancement in the films' crystallinity.

The locations of peak coincide to an average lattice parameter of 10.081 Å ± 0.005 Å that is slightly smaller than (10.118 Å) of the bulk material [12].

II. Optical properties

Optical transmission spectra from 300-1100 for the films deposited on quartz substrates and air annealed at different temperatures are given in Figure 2. It is observed that the thickness and annealing temperature strongly effect the transmittance of the In_2O_3 films as well as the nature of the surface and its crystal structure as in agreement with [13]. The film deposited at room temperature (27C°) has low optical transparency. Improvement in the optical transparency from 70% to 95% in the visible and near infrared regions is observed when the annealing temperature increasing as shown in Figure 2. Due to its high absorbing properties at short transmittance values that related to temperature it was noticed that optical transmission of the films are higher at longer wavelengths that's mean the film behave as opaque material [14]. $(\propto h\nu)^2$ relation of dependence with the photon energy $(h\nu)$ is exhibited in Fig.3. The electronic transitions was found to be direct type, the values of the band gap energy are specified by extrapolation of the linear part of the curves into the $(h\nu)$ axis and found to be varies in range (3.8-3.86) eV for annealed films and (3.45-3.68) eV for the films before any post-annealing at different thickness which is in good agreement with reported results in [9, 16, 17]. Conformity of the surface smoothness and it's uniform morphology for the films, are indicated in the high values of the band gap. While film thickness decreases as shown in Figure 3 (b), the band gap energy decreases while the transmittance increasing, that's related to the decreasing in absorption and Bragg reflection engendered at different combinations of thickness and wavelength as in references [4, 2]. From transmittance (T) measurement, the absorption coefficient (α) was found to be in order of 10^4 cm^{-1} and it dependence with the photon energy $(h\nu)$ obeys the tauc relation as shown in ref. [15].

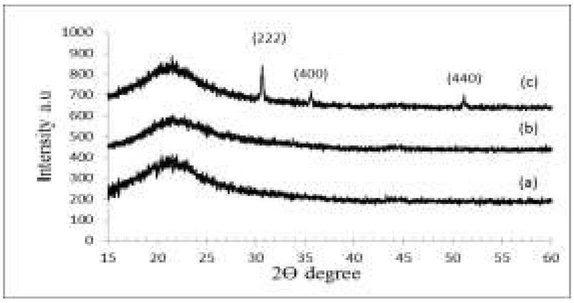

Figure 1: XRD patterns of In_2O_3 deposited thin films with 158 nm thickness and annealed at: (a) 300℃ (b) 400℃ (c) 500℃.

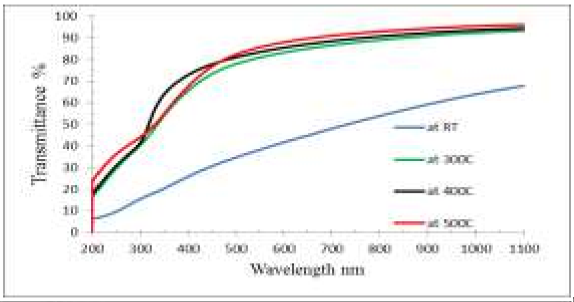

Figure 2: Optical transmission spectra as a function of wavelength for the In_2O_3 films annealed at different temperatures.

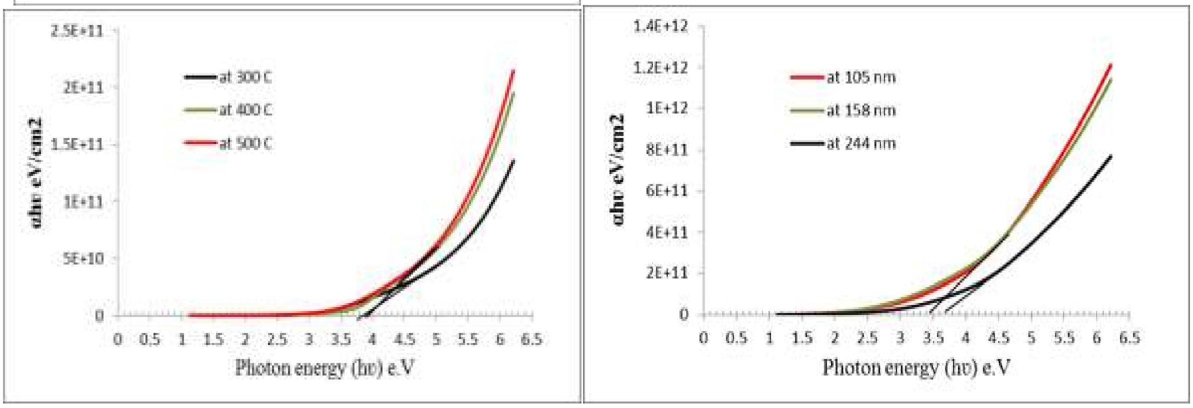

Figure 3: $(\alpha h\upsilon)^2$ versus $(h\upsilon)$ of In_2O_3 thin films at:
(a) Fixed thickness (158 nm) and different annealing temperatures.
(b) Different thickness before any post-annealing.

III. Surface morphological studies

The morphology of the annealed films of was researched by SEM. Generally, air annealing enhances both coalescence and atomic rearrangement processes that is why bigger sizes particles consider predictable as annealing temperature increases. Typical SEM images of annealed In_2O_3 thin films are shown in Figure 4. It shows uniform surface morphology with dense homogeneous grains distribution. Particulates of submicron sizes and droplets have been spotted over the film surface, due to combination and growth of the small grains, films annealed at higher temperatures will be composed of more number of particulates with bigger sizes and shows improvement in structure and uniformity enhanced. With the increase in the annealing temperature, an increase in grain size was observed as a result of the reduction in grain boundary density. The latter results in the grains agglomeration and hence leading to the scattering of grain boundary of the carriers to decrease as in [18].

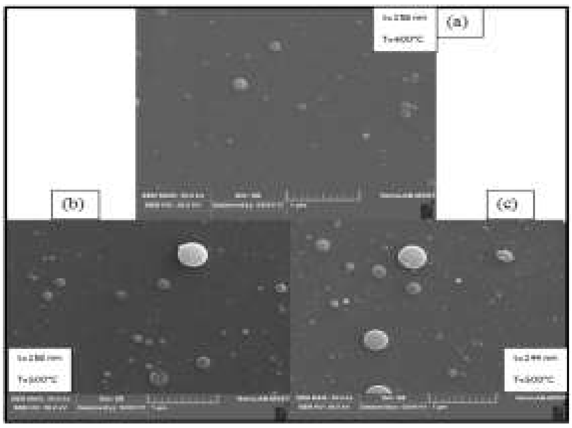

Figure 4: SEM images of In_2O_3 thin films prepared by PLD

From Figure 4(b)(c) the two images of the In_2O_3 thin films at different thickness both annealed at 500 ℃, both images shows the textures consisting of both small and large grains for film with more thickness, the grains are larger in sizes and in numbers. Crystallite size as obtained from the XRD results, for the In_2O_3 film with 244 nm thickness exceeded the 158 nm film, due to using higher laser energy at the preparation.

4. Conclusion

High-quality transparent homogenous In_2O_3 thin films were successfully deposited by PLD. The research of annealing process reveled the deposited thin film crystallizes to (bcc) structure and preferred oriented along (222) plane after air annealing at higher temperatures above 400℃. From SEM images, films with 158 nm thickness annealed at 500℃ exhibited good crystallographic structure with the presence of fewer submicron particles. Post-deposition annealing temperatures enhanced the physical and morphological properties of the prepared films to be used in optoelectronics applications like transparent electrodes.

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