UNIVERSITI MALAYSIA PERLIS
International Journal of Nanoelectronics and Materials
IJNeaM
ISSN 1985-5761 | E-ISSN 2232-1535

Effect of laser wavelength on Indium Trioxide (In_2O_3) thin films deposited by pulsed laser deposition method

Sarah M. Taleb [a], Mohammed F. Mohammed [a], and Makram A. Fakhri [b] *

[a]*Al-Nahrain University, College of Engineering, Laser and Optoelectronics Engineering, Iraq*
[b]*Laser and Optoelectronic Department, University of technology-Iraq, Baghdad, Iraq*
* *Corresponding author. E-mail: mokaram_76@yahoo.com & makram.a.fakhri@uotechnology.edu.iq*

Received 22 September 2023, Revised 24 June 2024, Accepted 24 August 2024

ABSTRACT

The pulse laser deposition (PLD) technique was used to prepare and deposit indium trioxide (In_2O_3) as thin films with a nanocrystalline structure on the silicon porous and quartz substrates. The laser wavelength effect on the optical and structure properties of these films was investigated. The PLD technique is accomplished using the following constant parameters: temperature (300°C), frequency (3 Hz), number of pulses (250 pulses), and voltage (900 V), but with three different laser wavelengths: 1064 nm, 532nm, and 355nm. To characterize and analyze these nanostructure thin films Ultra-Violet Visible (UV-vis) and X-ray diffraction (XRD) were used. The UV-vis analysis shows that: the laser wavelength does not have a significant effect on the transmission, absorption, and energy band gap values, it can be seen that when the laser wavelength increases the transmission values of thin films increase. While the values of absorption and energy band gap appear the random behavior with this increase. Also, it can be noted the laser wavelength does not have a significant effect on the refractive index values, since it achieves close values when the three different laser wavelengths were used. The XRD analysis shows that: the structure of In_2O_3 thin film will be purer and more crystalline with increasing laser wavelength because the intensity of phase 2θ at values of 31.8°, 34.06° and 63.48° correspond to (222), (400), and (662) planes increased when the laser wavelength increases.

Keywords: *Indium trioxide (In_2O_3), Laser wavelengths, Pulse laser deposition, Quartz substrates, Silicon porous, Thin films*

1. INTRODUCTION

With the atomic number 49 and the symbol In_2O_3, indium trioxide is a chemical element [1-4]. The softest metal that is not an alkali metal is indium trioxide. It is a silvery-white metal that looks similar to tin [5-9]. The Earth's crust has 0.21 parts per million of this post-transition metal [10-12]. The melting point of indium trioxide is higher than that of sodium and gallium, but lower than that of lithium and tin. In terms of its chemical properties, indium trioxide is primarily halfway between the properties of gallium and thallium [13-18]. Ferdinand Reich and Hieronymous Theodor Richter used spectroscopic techniques to find indium trioxide in 1863 [19-21]. Because it is challenging to create optically excellent samples, the precise optical characteristics of indium trioxide are not yet understood [8, 22-24]. With a few precautions, thin, semi-transparent indium trioxide films can now be created via vacuum deposition [25-27]. Therefore, when the thickness is known, their optical characteristics can be inferred from measurements of both transmittance and reflectance [28-32]. Indium trioxide is used in a wide variety of applications in the electronic industry and engineering [33-46]. Some of these applications include using it in batteries, and transparent thin film infra-red reflectors. It is also commonly doped with tin oxide (SnO_2) to make indium tin oxide (ITO), which is used in transparent thin conductive films, which are used in various types of displays, energy-efficient windows, and photovoltaics. In_2O_3 film is used as a sensing layer in the manufacturing of sensor devices due to its high sensitivity to a variety of gases.

For indium trioxide films, a number of deposition techniques have been investigated, including atomic layer deposition (ALD), pulsed laser deposition (PLD), sol-gel, chemical vapor deposition (CVD), and sputtering [37-42].

Pulsed laser deposition (PLD) is a physical vapor deposition (PVD) technique where a high-power pulsed laser beam is focused inside a vacuum chamber to strike a target of the material that is to be deposited [43-48]. This material is vaporized from the target (in a plasma plume) which deposits it as a thin film on a substrate (such as a silicon wafer facing the target) [49-52]. This process can occur in ultra-high vacuum or in the presence of a background gas, such as oxygen which is commonly used when depositing oxides to fully oxygenate the deposited films [53-57]. The laser material removal method is the primary benefit of PLD. PLD is based on the rapid explosion of the target surface region due to superheating and uses a photon interaction to produce an expelled plume of material from any target [57-61]. The PLD technology has developed as a substitute with the added benefit of maintaining the target phase's stoichiometry. PLD holds great promise for covering implant metals with bioactive glass [62-64].

The motivation behind this research is to show how the laser wavelength affects the optical and structural properties of thin film and achieve thin film with high-quality to use in the different applications.

In this research, a high purity of In_2O_3 nanostructure has been deposited utilizing a straightforward and affordable approach (PLD), and optical with structure analysis has been studied for the fabrication of optoelectronics and sensing devices.

2. EXPERIMENTAL SECTION

On quartz and silicon porous substrates, nanostructured In_2O_3 thin films were prepared and deposited by using a pulsed laser deposition technique (PLD). Before starting the deposition process, the substrates were cleaned, as these cleaning procedures are very important to get rid of impurities and fingerprints on the substrates, which, when present, form an insulating layer between the substrate and the deposited film.

The quartz substrates were cleaned by hand after 10 minutes of immersion in a mixture of soap and water. After that, they are rinsed with water several times before immersing them in ethanol for five minutes. The final step involves drying them with hot air and storing them in a closed container. While, silicon substrates were cleaned by using an ultrasonic device and a mixture between distilled water, alcohol, and hydrofluoric acid.

The most common method for producing porous silicon is to use an electrochemical cell consisting of a solution of hydrofluoric acid and ethanol in different proportions. The proportions in this work are: 12 ml of hydrofluoric acid and 8ml of ethanol, the porous silicon represents a form of silicon, which contains fine and dense pores. Which makes the surface area to volume ratio very high.

The target was made of ultra-pure In_2O_3 from Aldrich Company (United States). The material came in the form of a powder, so it was subjected to 15 tons of pressure to form a disc with a diameter of 2 cm and a height of 1 cm.

After preparing the porous silicon and quartz substrates and the target which is made of pure material (In_2O_3), the thin film deposition process is initiated by PLD as shown in Figure 1. During the deposition process by PLD technique, the following constant parameters were used: temperature 300°C, frequency 3 Hz, number of pulses 250 pulses, voltage 900 V but with three different laser wavelengths: 1064 nm, 532 nm, and 355 nm. All these steps are shown in Figure 2, which represents the flow chart of deposition steps.

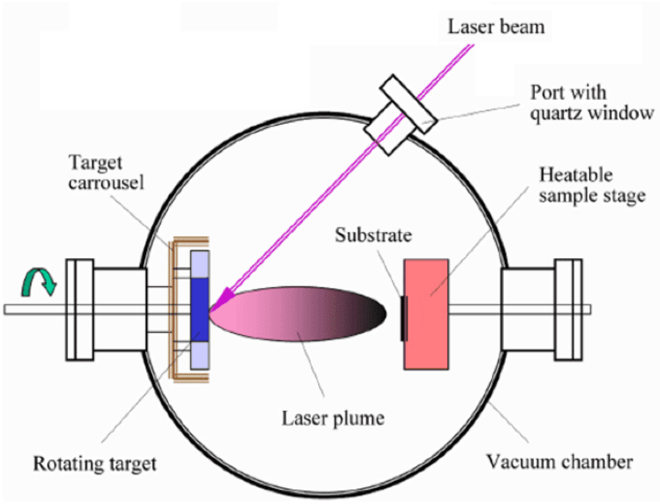

Figure 1. Deposition process by PLD

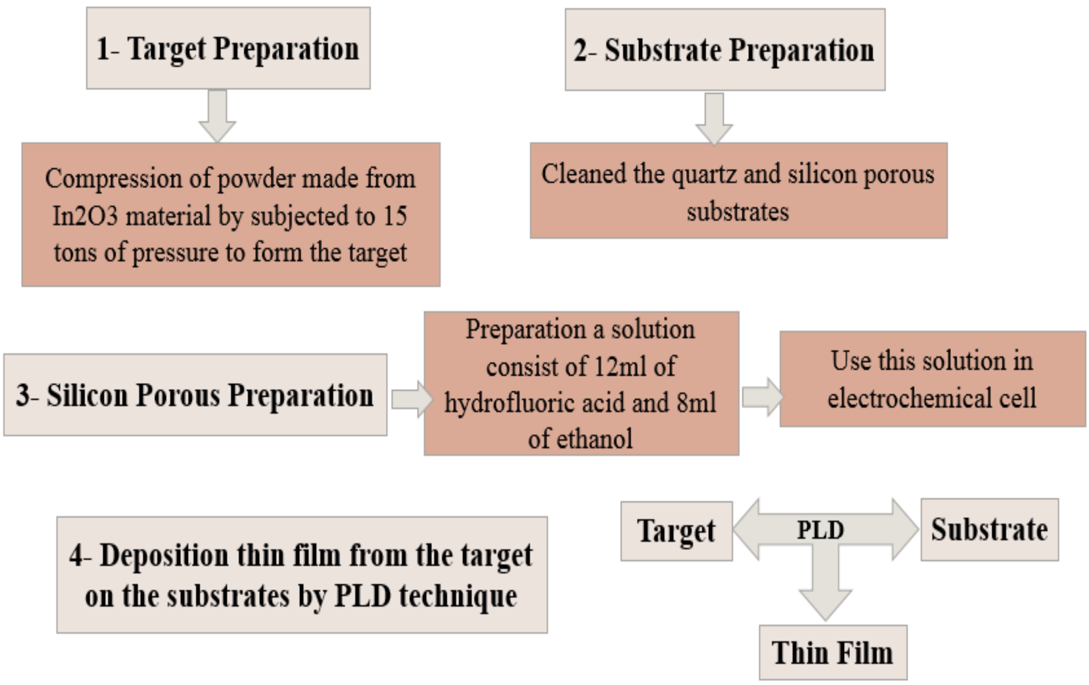

Figure 2. Flow chart of In_2O_3 nanostructure thin film preparation process

To calculate the optical properties of these thin films, including optical transmittance (T), optical absorbance (A), optical band gap (Eg), and refractive index (n) the Ultra-Violet Visible (UV-vis) spectrophotometer (Shimadzu UV-Vis 1800, Japan) in the wavelength range (200-1000 nm) was used [65-69]. Then by the X-ray diffraction (XRD), (X'Pert Pro MRD PW3040 system diffractometer, PANalytical Company, Netherlands) system equipped with Cu-K a-radiation of wavelength k = 0.15418 nm, at 40 kV and 30 mA, we studied the structure properties of these films at same wavelength range.

3. DISCUSSION AND RESULTS

PLD technology was used to deposit In_2O_3 nanostructure thin films on quartz and silicon porous substrates utilizing three different laser wavelengths: 1064 nm, 532 nm, and 355 nm. The UV-Vis spectrophotometer and X-ray Diffraction (XRD) were used to characterize and evaluate the deposited thin films.

From transmission studies in the wavelength range 200–1000 nm, the optical characteristics of In_2O_3 thin films were analyzed [70-72]. Figure 3 shows the optical transmission of In_2O_3 nanostructured thin films. From this figure, we can observe the optical transmission of these films increased with the increased laser wavelength, but this increase is not significant which means the laser wavelength has little effect on the transmission values. Where the values of transmission achieved are (95, 94, and 85) corresponding to laser wavelengths (1064, 532, and 355 nm).

Figure 4 shows the optical absorption (A) of these thin films, the values of absorbance depended on the transmission values mostly, but it can be noted the absorption of these films appears the random behavior with the laser wavelength, where a high value of A appear with the laser wavelength 355 nm which equal to 4.3, while the low value of A appear with the laser wavelength 532 nm which equal to 2.2.

We determined the optical band gap, Eg of these films as a function of photon energy by plotting the curve between $(\alpha h\nu)^2$ and (hv) as shown in Figure 5. The calculated Eg is about (4.6-4.8- 4.2) eV corresponding to laser wavelengths 1064 nm, 532 m, and 355 nm, which means there is also the random behavior of Eg with the laser wavelength.

Additionally, mathematically, the optical reflectance (R) of these nanostructure films can be estimated from the transmission and absorption values in accordance with the formula R+T+A = 1. The refractive index (n) values at the three different laser wavelengths can be calculated as a function of wavelength and from the transmittance values in the range (200-1000 nm). As shown in Figure 6, it can be noted the films deposited achieved convergent values in the range of 2 (2.22, 2.14, and 2.53) corresponding to laser wavelengths 1064 nm, 532 nm, and 355 nm. That means the laser wavelength does not have a high effect on the refractive index value of these films [73-75].

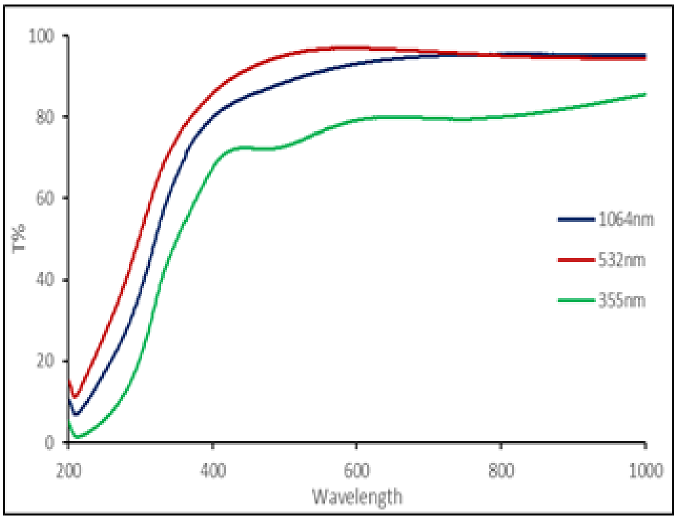

Figure 3. The-optical-transmission-of- In_2O_3 at different-laser wavelength

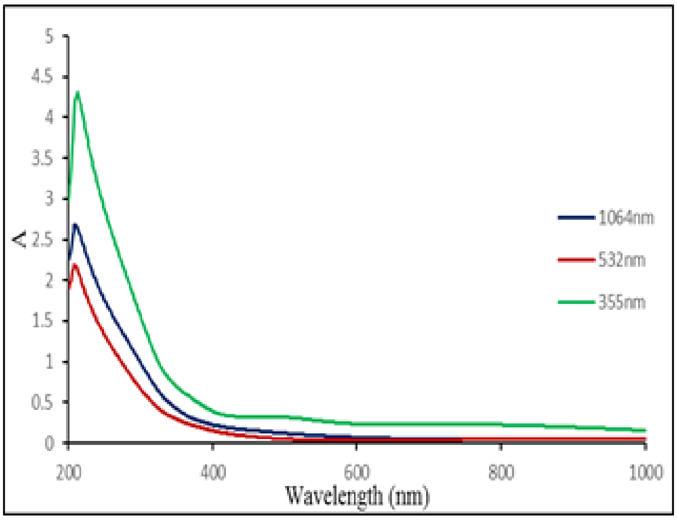

Figure 4. The-optical-absorption-of- In_2O_3 at different-laser wavelength

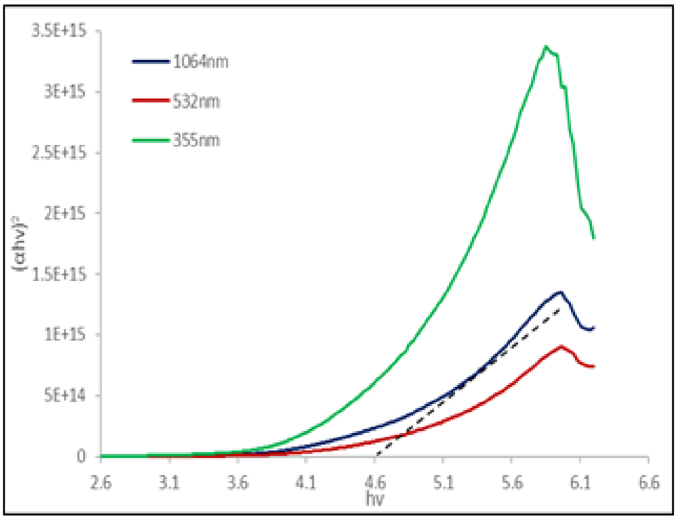

Figure 5. The optical band gap (Eg)-of- In_2O_3 at different-laser wavelength

Through the results of optical analysis, we can conclude that the laser wavelength does not have a strong effect on the transmission values, and it is effect is random on the absorption and energy gap on these films, and it is effect is almost non-existent on the refractive index values.

The effect of laser wavelength on the XRD results of In_2O_3 nanostructured thin films deposited on a silicon porous substrate is shown in Figure 7. It is observed from this data, that the In_2O_3 crystal structure has diffraction peaks at 2θ = 31.8°, 34.06° and 63.48° correspond to (222), (400), and (662) planes, and the intensity of these phases increased when the laser wavelength increased. The intensity of the peak reached a value of 580 at the laser wavelength 1064 nm compared to the intensity of the peak at laser wavelength 532 nm which reached a value of 240, while it reached a value of 180 at the laser wavelength 355 nm at the performed phase has (662) orientation, that means the In_2O_3 structure will be more purity and crystalline when the laser wavelength increasing. Because the crystals will be rearranged and restructured to improve the properties of the structure and to get a high rate of purity of In_2O_3 nanofilm, this leads to an increase in the average particle size that occurs when the wavelength of the laser increases. From these data, we noted that the behavior of nanophotonic at the laser wavelength 1064 nm is much better than its behavior at laser wavelength 532 nm and 355nm, where it found it clearer and more crystallization. Accordingly, the prepared thin film will be better when using laser wavelength 1064 nm.

4. CONCLUSIONS

In_2O_3 nanostructure thin films were prepared and deposited on the quartz and silicon porous substrates by using Q-switched Nd: YAG lasers operating at three different laser wavelengths, including 1064 nm, 532 nm, and 355 nm. From the optical results presented in this paper, we note that: - by changing the laser wavelength, the optical properties of these films did not show a strong change with this effect, and this was confirmed by the values of optical transmission and refractive index, achieved the convergent values at the three different laser wavelengths. While the laser wavelength has a random and incomprehensible effect on the optical absorption and energy gap values. So, we can conclude thought the optical results cannot determine the best laser wavelength for the deposition process. While, from the XRD results, we found the peak at 2θ= 63.48° with (662) orientation has the intensity increasing dramatically with increased the laser wavelength to 1064 nm, which arrived at the value 580. Also, the structure becomes more crystalline and more purity with this increase. That means the improvement in the crystal structure of these films is obtained by increasing the laser wavelength through the deposition process. So, from the XRD results, we can conclude the best laser wavelength for the deposition process of thin film which gives the better results is 1064 nm.

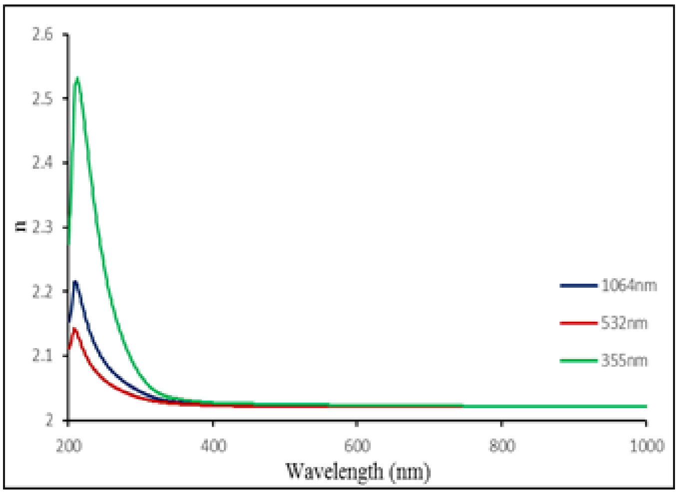

Figure 6. The-refractive-index-of- In_2O_3 at different-laser wavelength

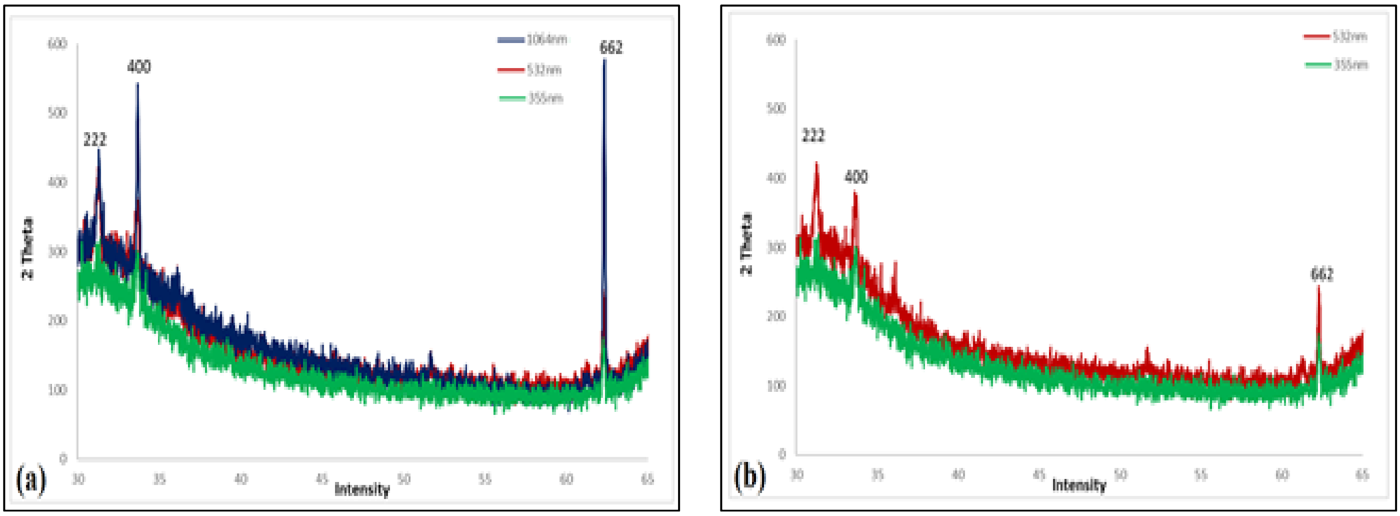

Figure 7. XRD patterns of - In2O3 at:-a) three different-laser wavelengths, b) two different-laser wavelengths

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