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Effect of Substrate Temperature on Optical and Structural Properties of Indium Oxide Thin Films Prepared by Reactive PLD Method

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ABSTRACT

In the present Work, effect of substrate temperatures on the optical and morphological properties of In_2O_3 trioxide thin film has been carried out using Reactive Pulsed Laser as a Deposition technique (RPLD). 1.064µm, 7 nsec Q-switch Nd-YAG laser with 400 mJ/cm^2 laser energy's has been used to ablated pure Indium target and deposited on glass substrates . The resulted films show High transparency reached to about (85) % which found to decrease sharply with the substrate temperatures. The estimated optical band gap found to be about 3.6eV at optimum substrate temperatures (70 C^o). The FTIR results insure the formation ofIn-O vibrational bond with different vibrational intensity depending on substrate temperatures.

Keyword: PLD, In_2O_3 Surface morphology, optical properties, FTIR.

تأثير درجة حرارة القاعدة على الخصائص البصرية والتركيبية لاغشية اوكسيد الانديوم الرقيقة المحضرة بواسطة الترسيب بالليزر النبضي

الخلاصة

تم في هذا البحث دراسة تاثير درجة حرارة القاعده على الخصائص البصريه والمورفولوجيه لسطح الاغشية الرقيقة المحضرة بطريقة الترسيب بالليزر النبضي . تم استخدام ليزر نديميوم ياك النبضي يعمل بضابط عامل نوعيه بطول موجي 1.06 مايكرومتر و امد نبضة 7 نانوثانيه وبطاقة 400 ملي جول/سم^2 لغرض اجراء عملية الاستئصال لسطح معدن الندميوم عالي النقاوه ولغرض الترسيب على سطح القواعد الزجاجيه. اظهرت نتائج الخصائص البصريه النفاذية العالية للاغشية المحضرة والتي تصل ال 85% والتي سرعان ما تهبط بشدة مع نقصان درجة حرارة القاعدة . القيمة المحسوبه لفجوة الطاقة وجدت لتكون حوالي 3.6الكترون فولت عند الدرجة الحرارية التي اختيرت لتكون المثلى (70C^o). نتائج النفاذية للمنطقة تحت الحمراء FTIR اكدت وجود الاصرة نوع الاهتزازي بين مادتي الانديوم – الاوكسجين وبشدات اهتزاز مختلفه بالاعتماد على درجة الحرارة.

INTRODUCTION

Indium oxide (In_2O_3) is an important n-type semiconductor with its wide direct band-gaps ranged between 3.55-3.75 eV, it's also found to has an indirect band gap of about 2.6 eV [1-3] . It considered as an important transparent conducting oxide (TCO) material [4-7]. It has interesting properties such as high transparency to visible light, high electrical conductance, and strong interaction between certain gas sensor molecules and its surfaces [8-10]. These properties make In_2O_3 an interesting material for a variety of applications, including solar cells, panel displays, organic light emitting diodes, photo catalysts, architectural glasses, field emission [1- 4] . Studying the optical properties of these thin films is in particular interest because of their use in optical devices and oxide films have been used for microelectronics applications and surface coating of optical and electrical materials [10-13] Moreover, In_2O_3 is an important material for semiconductor gas sensors ,. The technological importance of In_2O_3 thin films is reflected in the fundamental study of their physical properties[9,11,13]. The search for new material to satisfy the practical demand and the use of electronic and optical devices has stimulated considerable interest in the growth and understanding of optical properties of Indium Oxide (In_2O_3) thin films[10-12]. In_2O_3 have been synthesized by several techniques including sol–gel technique ,pulse laser deposition , thermal decomposition , thermal hydrolysis , microemulsion , spray pyrolysis , mechanical chemical processing ,and others [11-13] In this paper we report on the growth of indium oxide thin films on glass substrates by ablation of a pure indium target in an oxygen reactive atmosphere. This fabrication technique allows control of the structural characteristics of the films by changing the substrate temperatures [15].

Experimental

Indium Oxide (In_2O_3) thin films were prepared using pulsed laser deposition technique on glass substrates. Fig. 1 shows the schematic diagram of PLD system used in this study[15-19]. Q-switched Nd:YAG laser with 7 ns, λ= 1.064 nm and 400 mJ/cm^2 laser energy density was employed. Converging lens with 12 cm focal length was used to focusing the laser beam on the rotating target. High purity indium target (99.999%) provided from Fluka com. was fixed at 45^o angle of incidence. The target rotated with frequency of 50 Hz. The pulse energy density of laser at the target surface was maintained within the range 400 mJ/cm^2. All films were produced using 100 laser shots and deposited at glass substrate with different substrate temperature between 40 - $70C^o$, in background oxygen pressure 100 mbar . The film thickness was measured by a stylus profile meter and it found to be about 48 nm. The transmittance of the films was investigated in spectral range (200–800) nm using UV-VIS Shimatzu double -beam spectrophotometer(double-monochromator optical system). The crystal structure of the grown films was analyzed with FTIR system (Philips) using CuKa radiation. The morphology of the films was studied using optical microscope. the Atomic Force Microscope of this films was studied using Shimatzu AAXOO Scanning Probe Microscope.

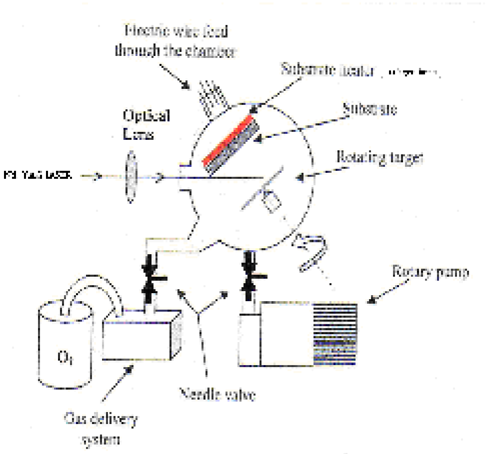


Figure (1) a-schematic diagram of the PLD system used

Results and discussion

The effect of substrate temperature on the optical transmission spectra of In_2O_3 structured thin films with constant number of pulses and optimum laser power at 400 mJ /cm^2 could be shown in Figure (2) , it has been found that the transmission increase with increase the substrate temperatures due to decrees the deposition efficiency and then the thickness of In_2O_3 films.

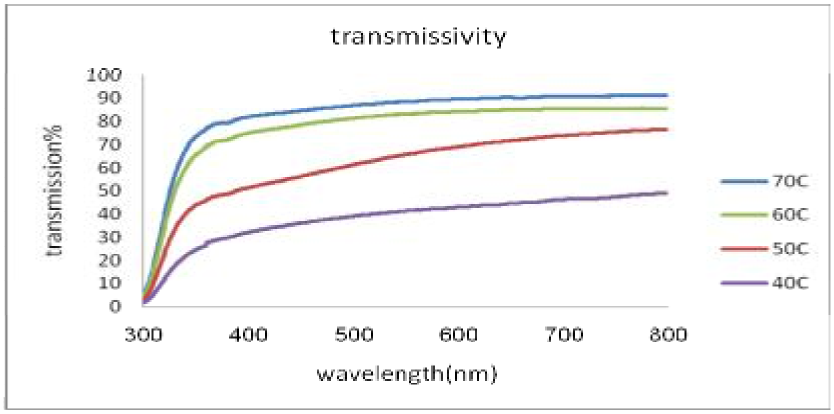
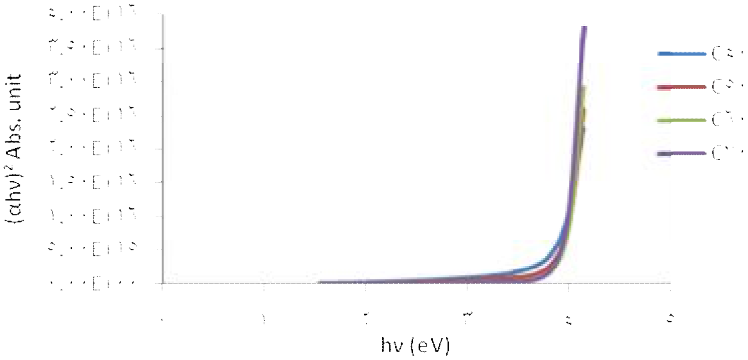


Figure (2):Optical Transmission as a function of wavelength for In_2O_3 at deferent heat of substrate

These peaks have as lightly red shift with the increase of substrate temperature (increase in particle size) . In fact, increasing the heating means delivering more energy that implies ablation large amount of material, because of the plasma plume becomes more intense and the In_2O_3 particles cloud becomes smoothly . Most likely, this means that big particles will be present due to longer growth time and to the high probability of deposited particles muster . In other words, atoms and nanoscale particles deposited under heating radiation tend to muster during and after the laser pulse . This reality leads to generate of larger particles that becomes more distinguished when the density of the In_2O_3 particles increases further with increasing the heating .

The band gap energy Eg is found by plot $(\alpha h\nu)^2$vs hν as shown in Figure (3)[15-19]. The calculated band gap energies are (3.65, 3.75, 3.8, 3.82) corresponding to the (40,50,60,and 70 C° substrate temperature) are higher than the bulk In_2O_3, Indium oxide (In_2O_3) direct band-gaps ranged between 3.55-3.75 eV, and indirect band gap of about 2.6 eV [9,14] because of reduction in particle sizes reason due to the quantum confinement effect and increase of surface/volume ratio. As a result surface atom has lower coordination number and atomic interaction which increases the highest valance band energy and decreases the lowest unoccupied conduction band energy ,This leads to increase in band gap energies[18].



Figure(3) (α hυ)2 (eV/ cm)2 vs. hυ(ev) of In_2O_3 NPs at different laser power intensity

Figure (4) shows IR spectra of the samples from The band at 1050 cm^{-1} is attributed to the absorption of C-O vibration, while the absorptions around 500 cm^{-1} are due to the In-O vibrations . As an exception, a weak absorption at 1568 cm^{-1} appeared on the IR spectrum of the nanocrystals from toluene, which should be attributed to the C=O vibrations from the acetylacetone species .Figure (4-a) give the FTIR result for film prepared at Substrate temperature of (60C°), we can recognize the absorption peak at (500 ,1550) cm^{-1} which related to the In metal atoms, beside (632.6,860.7,1615.3,172697) cm^{-1} absorption spectra which related to the formation of In_2O_3 molecule. An increasing the formation ability of the In_2O_3 molecule could be recognize obviously by decreasing of substrate temperature to(50)C° Figure (4-b) where (632.6,860.7,1615.3,1640,172697,1740)cm^{-1} absorption peak could be found that belong to the for formation of the In_2O_3 molecule beside that ,the presences of the (987.49,918.05,817.7) cm^{-1} peak which related to In atom that still unoxide sized.

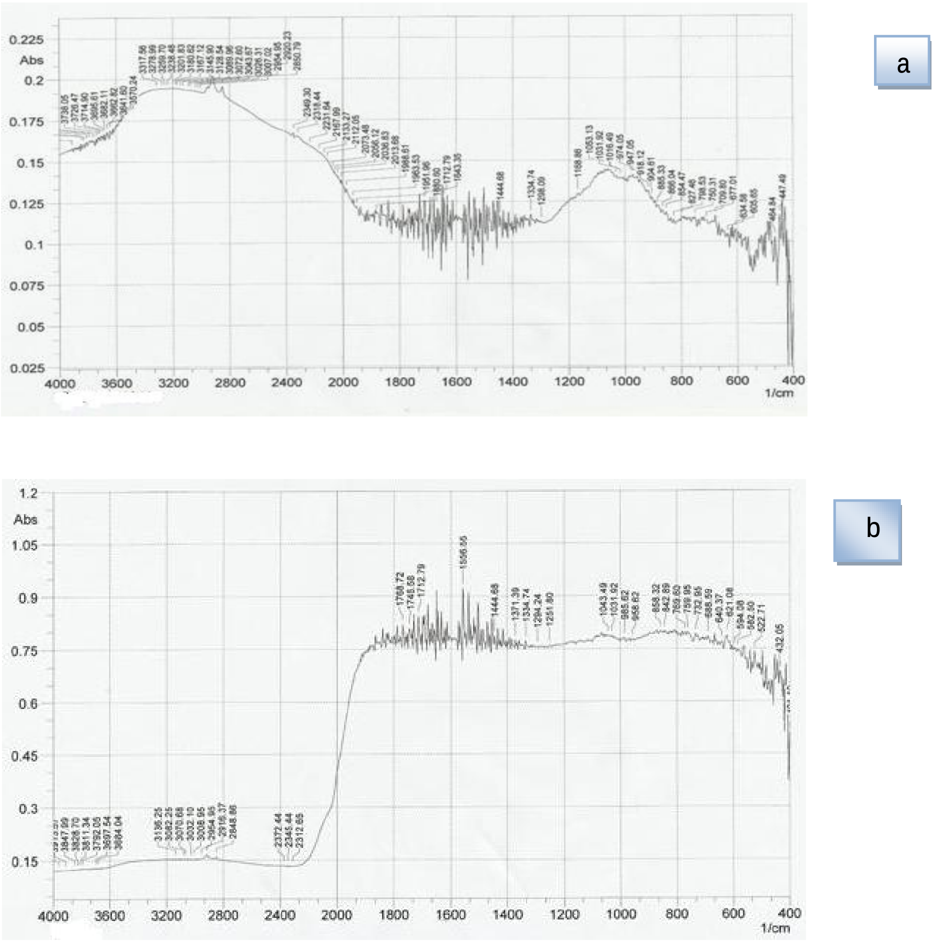


Figure (4) FTIR spectrum of In_2O_3 thin film deposited at a- 60C^o,b-50C^o

The morphologies and grain sizes of the In_2O_3 thin films were examined by tapping-mode atomic force microscopy (AFM). The fine-microstructure can be seen in Figure. (5) the film composed of spherical particles varying in size and the average grain size is about 49.89 nm. The root mean square value of surface roughness (rms) derived from the AFM image is 14.4nm and roughness average 10.4 nm . The increase in grain size was associated with a change in shape and distribution of grains making up films with the increase in laser energy at the laser Gaussian mode.

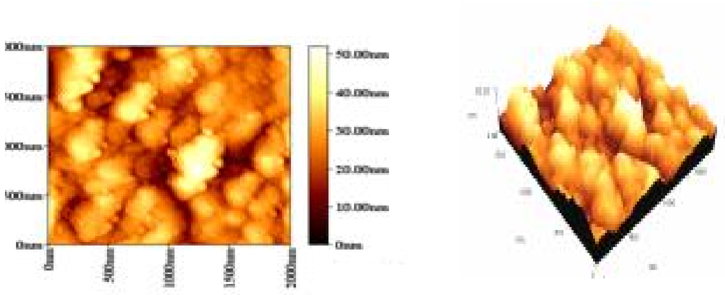
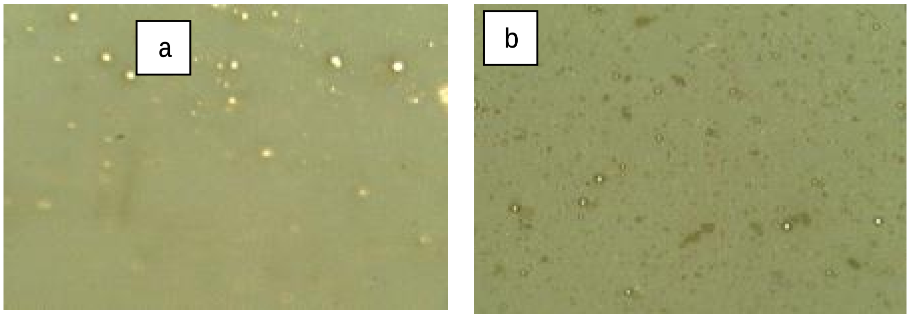


Figure (5) represents the AFM micrograph of In_2O_3 films deposited at 50C

The surface micrograph of the In_2O_3 thin films prepared at various substrate temperatures could be shown in Figure (7), film morphology can be easily recognized through the film homogeneity and color. The results show the surface morphology at different substrate temperatures. The color of films tends to be dark (nearly brown) which reflect the metallic nature of In, which typically has dark brown color as well as the high reflectivity of the obtained film as shown in fig (7-a). It can be clearly noticed that the film color is the same as the physical color of the target metal, by increasing energy in Figure (7-b) we can recognize the change in the deposited film color from dark brown to light brown nearly yellow, which may be attributed to the fact that effect of heating of substrate and interacting with oxygen atoms have been fired rather than reacted with In atoms to form In_2O_3. Droplets and particulates of submicron sizes have been observed over the film surface at low energy and they are sprayed randomly as dark regions on the film surface which explain its homogeneity. At laser heat of about (60 C^o) shown in figure (7-c) where oxides particulates begin to form the metal atoms that are still available in the film structure reflect. The incident light appears as dark yellow dots in the microscope picture, while the oxides appear as light yellow particulates for In_2O_3 molecules at ($70C^o$) temperature as shown in Figure (7-d), and that color is a nature color of indium oxide that explain and the yellow particles of indium oxide and big metallic particles of pieces of indium that plowed(pulled) score effect of heat transfer on indium target[19].



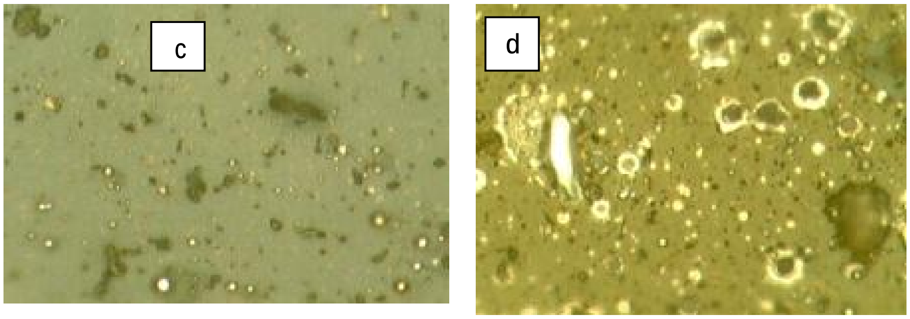


Figure (7) Surface morphology of In_2O_3 samples prepared at laser energy of 400 mJ and different tepreture of substrate (a- 40C^o b-50C^o c-60C^o d-70C^o) and laser fluence of (400)J /cm^2 ,x=1000 for all pictures.

CONCLUSION

In_2O_3 Nano structure thin film was successfully prepared using reactive pulsed laser deposition (PLD) method . Optical transmition of prepared at deferent substrate temperature show an average visible transmittance of 85% and an optical band gap ranged between 3.6-3.82 eV with the absence of any post deposition heat treatment. The RMS value reveled the preparation of high quality thin film with average roughness of about 5.97 nm. The microscope measurement it was been found the optimum condition as a light yellow particulates for In2O3 molecules at (50C^o) substrate temperature . The FTIR results shown An increasing the formation ability of the In_2O_3 molecule could be recognize obviously by decreasing heating of substrate to(50C^o) at bonds (632.6,860.7,1615.3,1640,172697,1740)cm^{-1} . In the light of obtained results, these films can be used in optoelectronic and related high-technology applications.

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