NANO AND MICRO INDIUM OXIDE STRUCTURE PREPARED USING LASER ABLATION METHOD

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Abstract

In the present work, effect of laser energy on optical and morphological properties of In_2O_3 trioxide thin film has been carried out using Reactive Pulsed Laser as a Deposition technique (RPLD). Q-switch Nd-YAG laser with (λ=1.06nm,t_b=7nsec) and different energy's has been use to ablated pure indium target and deposited on glass substrates and constant substrate temperature of (333K). The results films show that high transparency reached to about (80-95) % can be achieved with In_2O_3 film which itself decreases sharply with the decreasing of laser energy while the optical band gap is(3.6-3.8) eV at optimum laser energy (400mj). The optical properties of the prepared include optical microscopic measurement, optical transition measurement, surface uniformity measurement and FTIR.

الخلاصة :

في هذا العمل قد تم تنفيذ تأثير لطاقة الليزر على الخصائص البصرية والتركيبة للأغشية الرقيقة لثالث اوكسيد الانديوم In_2O_3 بأستخدام تقنية الترسيب بالليزر النبضي الفعال (RPLD) .Q -switch ND- YAG الليزر مع (λ =1.06nm) وامد نبضة (7nsec) حيث يعمل الليزر على ترسيب ذرات اوكسيد الانديوم المتبخرة بتأثير الليزر على قواعد من الزجاج . ودرجة الحرارة الفاعدة (333K) . تظهر النتائج التي تم تحقيقها شفافية عالية وصلت الى %(80-95) مع In_2O_3 حول الغشاء الذي يقل بشكل حاد مع انخفاض طاقة الليزر وكذلك تم الحصول على فجوة طاقة البصرية eV (3.6 – 3.9) في طاقة الليزر الامثل (400mj) . الخصائص البصرية للمعدة وتشمل قياس المجهرية الضوئية ، والنفاذية ، والانعكاسية ، وقياسا بالمجهر القوة الذرية وتحويلات فورير لمطيافية المنطقة تحت الحمراء .

Key word :PLD, In_2O_3 , surface morphology, optical properties, FTIR.

INTRODUCTION

The extensive and intensive investigation of indium oxide (In_2O_3) during the last two decades can be directly linked to its remarkable combination of electronic and optical properties [Bregman et al.1990, Grivas et al.1998]. Specifically, the high values for both electrical conductivity and transmission in the visible and near infrared make feasible the exploitation of In_2O_3 thin films in numerous optoelectronics applications, from solar cells to liquid crystal panel displays and switching devices [Grivas et al .1998]. Among these, indium oxide (In_2O_3) is an n-type degenerate semiconducting oxide which is finds optoelectronic applications such as flat panel displays, solar cells, photodiodes

[Mohan Babu et al.2004, Kaleemullaa et al.2009], etc. For the past three decades many researchers have reported on the physical properties of In_2O_3 films formed by different deposition methods such as flash evaporation [Peng et al. 2003, Maensirla et al. 2008], electron beam evaporation [Maensirla, et al. 2008], laser deposition [Manoja et al .2006] have been used for the fabrication of In_2O_3 thin films[Grivas et al. 1998]. In recent years much attention has been focused on reactive Pulsed Laser deposited In_2O_3 films because of their advantages such as deposited from metallic indium target in the presence of reactive gas of oxygen, high deposition rates, film uniformity on small area and precise control over the composition of the deposited film. The physical properties of In_2O_3 films prepared by this method mainly depend on the deposited parameters like oxygen partial pressure, substrate temperature, laser energy , number of laser pulses, angel of deposition and target to the substrate distance. The effect of laser energy on the target was reported earlier. In order to achieve optical transparency the substrate should be maintained at elevated temperatures during deposition of the films[Manoja et al .2006, Kaleemulla et al.2009]. In this investigation, we studied the influence of substrate bias on the crystallographic structure and optical properties of In_2O_3 films prepared by reactive pulsed laser deposition[Subrahmanyam.2006].

WORK

Undoped In_2O_3 thin films were deposited on cleaned glass substrates by using pulsed Nd:YAG laser deposition technique, Fig.(1) shows the schematic diagram of PLD system used in this study. Pulses of Q-switched Nd:YAG laser with(7ns) FWHM and (k =1.064 nm) were focused through (10cm) focal length of converging lens onto a high purity indium target (99.999% provided from Fluka com.) at 45_ angle of Incidence. The target rotated with frequency of(0.5Hz). The pulse energy density of laser at the target surface was maintained within the range (100-400 mj/cm^2). All films were produced using (100) laser shots and deposited at substrate temperature of 60C^o(this growth temperature was optimum) in background oxygen pressure (100 mbar) . The film thickness was measured by a stylus profilometer. The transmittance of the films was investigated in spectral range (200–800) nm using UV-VIS Shimatzu double beam spectrophotometer. 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.

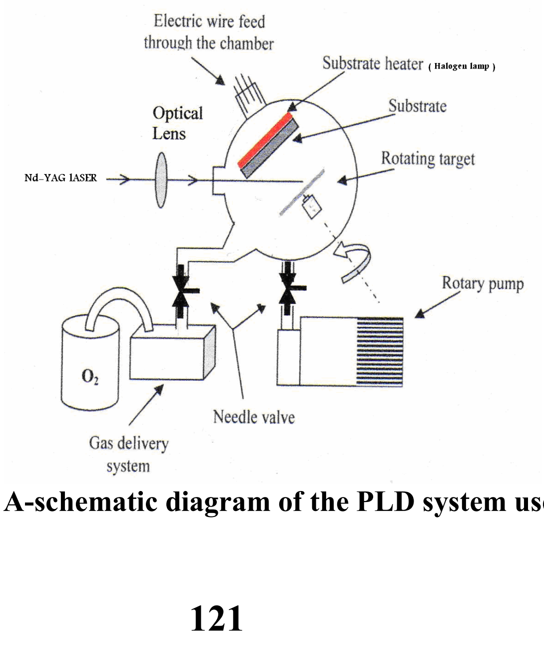
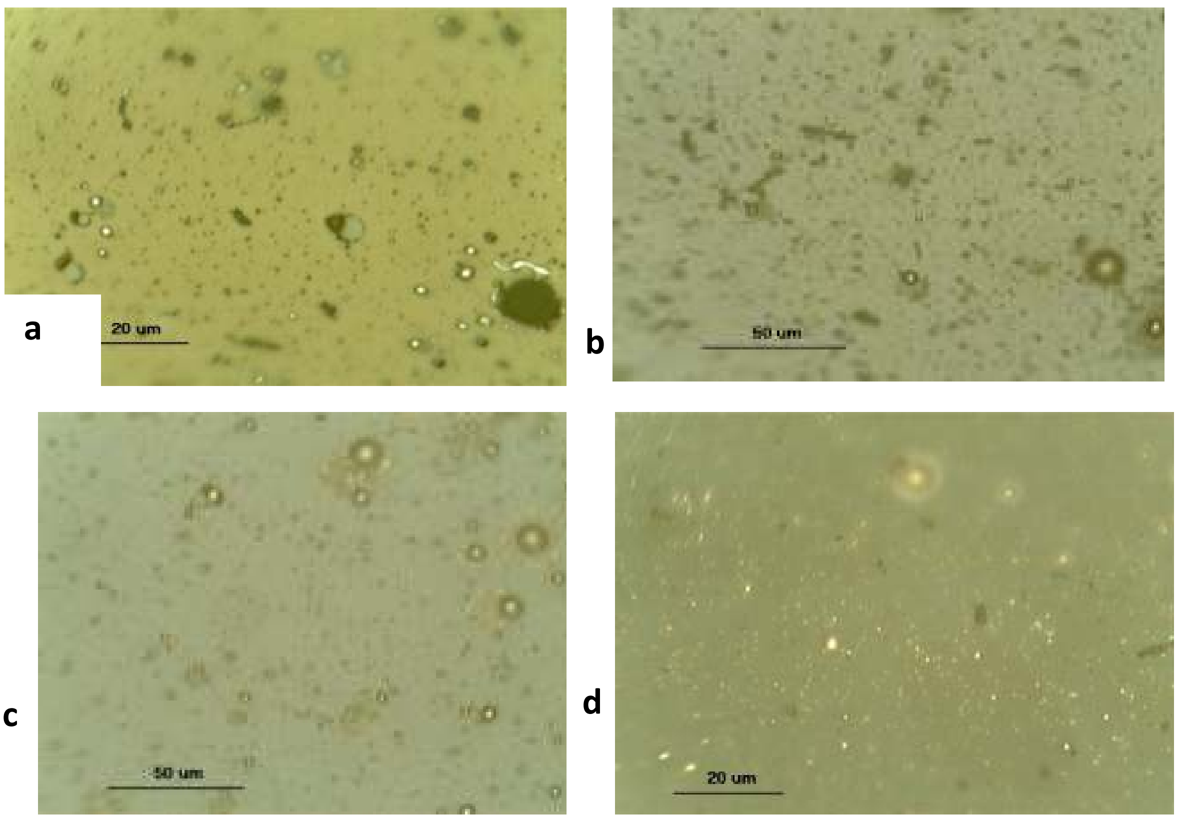


Figure (1) A-schematic diagram of the PLD system used.

RESULT AND DISCUSSION

Surface morphology of the laser produced transparent oxide (T.O) films. The optical micrograph of In_2O_3 thin films could be shown in Fig.(2), prepared at various growth conditions. It reveal that the film morphology can be easily recognized through the film homogeneity and color. Fig. (2), gives the optical micrographs of the films produced at room temperature in vacuum and (100mbar) oxygen pressure with different user energies, the color of films tends to be dark (nearly brown) which reflect the metallic nature of In_2 , which typically has dark brown color as well as the high reflectivity of the obtained film. It can be clearly noticed that the film color is the same as the physical color of the target metal, by increasing energy in Fig. (2-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 laser energy on indium target 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 energy of about (300 mj) shown in Fig.(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 (400mj) laser energy as shown in Fig.(d) and that color is a nature color of indium oxide that explain. In figure (e) the yellow particles of indium oxide and big metallic particles of pieces of indium that plowed (pulled) score effect of high energy of laser on indium target.



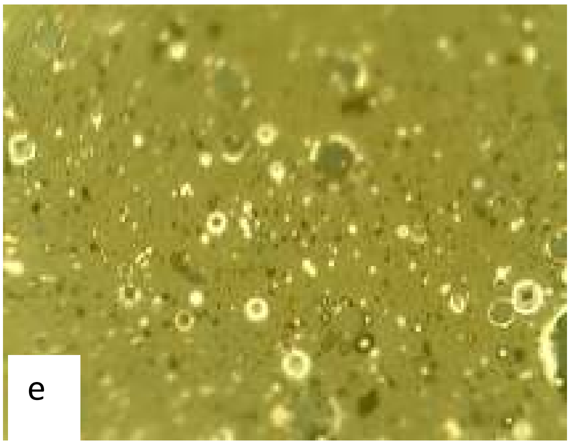


Figure (2) Surface morphology of In_2O_3 samples prepared at room tempreture and different laser energies (a- 100mj b- 200mj c- 300mj d- 400mj e- 500mj) and laser fluence of (89.17J /cm^2), x=1000 for all pictures.

The influence of laser power on the optical transmission spectra of In_2O_3 nanoparticle with constant number of pulses shown in Fig.(3), it was found that the transmission is increase with increase in the laser power density due to decrease the deposition efficiency and then the thickness of In_2O_3 film.

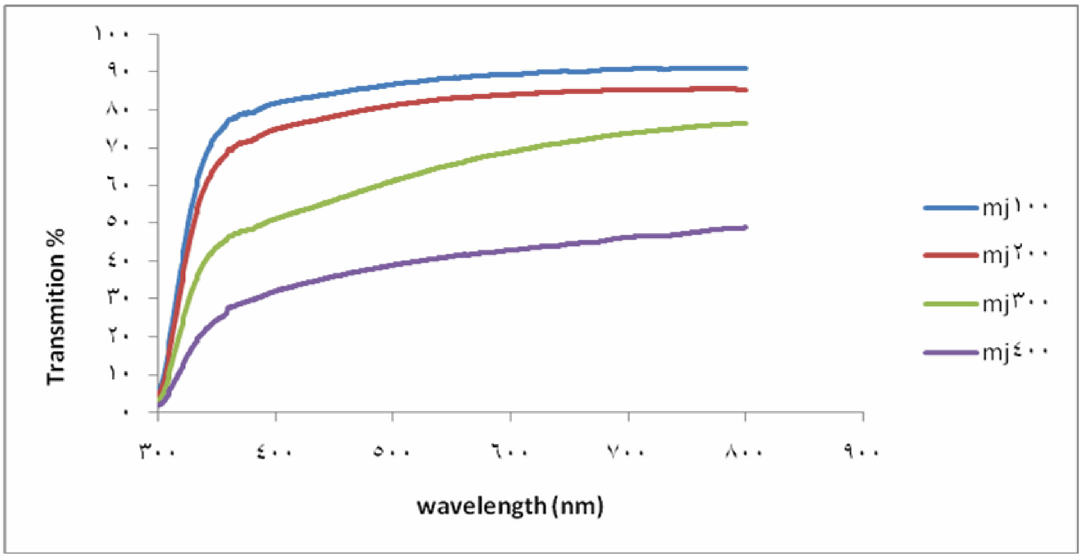


Figure (3) Optical transmission as a function of wavelength for In_2O_3 at different laser power density between(100-400) mj /pulse.

Figure (4) shows the optical absorption peaks, these peaks have as lightly red shift with the increase of laser power (increase in particle size). In fact, increasing the fluency means delivering more energy that implies ablating larger amount of material ,because of the plasma plume becomes more intense and the In_2O_3 particles cloud becomes bushy. 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 laser 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 fluency.

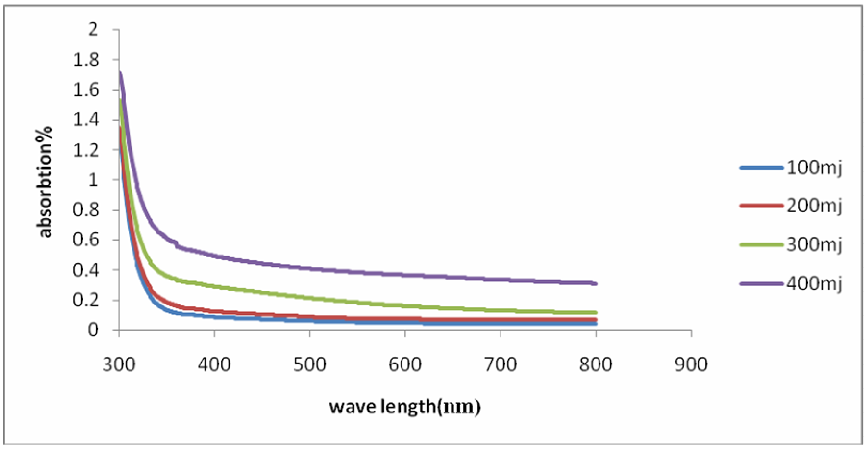
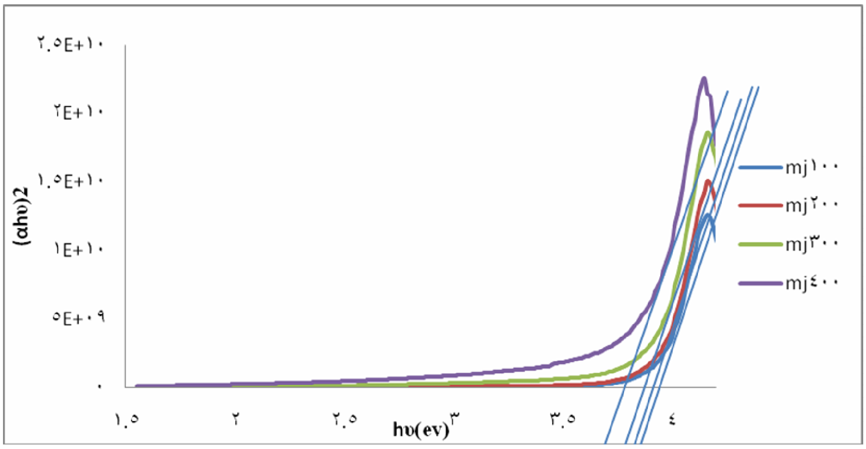


Figure (4) Optical absorbtion as a function of wavelength for In_2O_3 at different laser power density between(100-400) mj /pulse

the band gap energy Eg is found by plot (αhv) 2vs hν as shown in figure (5). The calculated band gap energies are (3.75, 3.8, 3.85, 3.9) corresponding to the (100,200,300,and 400 mj per pulse) are higher than the bulk In_2O_3, 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.



Figure(5) (α hυ)2 (eV/ cm)2 vs. hυ(ev) of In_2O_3 NPs at different laser power density.

Fig.(6) 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 acetyl acetone species [Guodong. 2011].Figure (6-a) give the FTIR result for film prepared at laser energy of (200mj), 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 MgO molecule could be recognize obviously by increasing the laser energy to (400) mj Fig(7-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 MgO molecule beside that ,the presences of the (987.49,918.05,817.7) cm^{-1} peak which related to Mg atom that still unoxide sized.

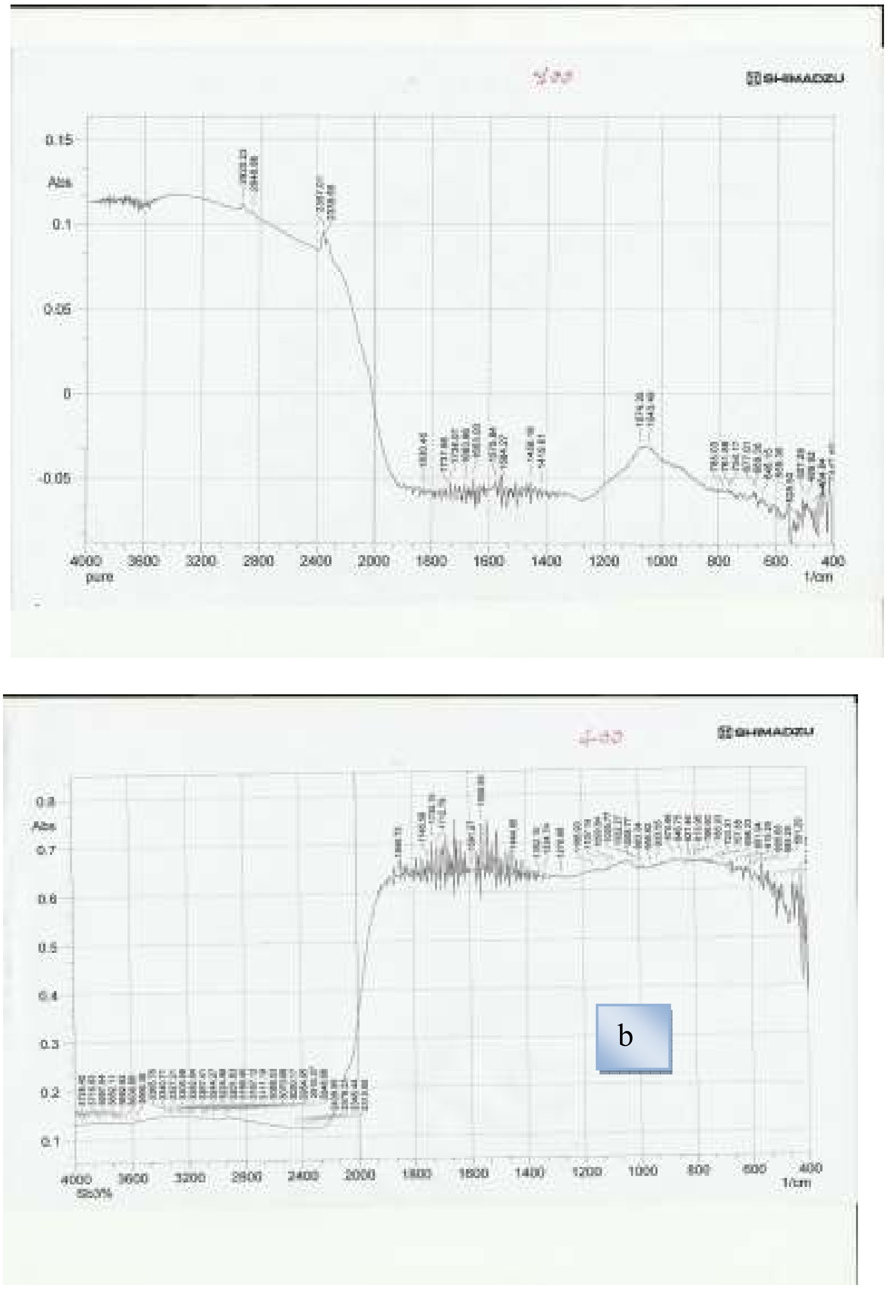


Figure (6) FTIR spectrum of In_2O_3 thin film deposited at a- 200mj,b-400mj.

The morphologies and grain sizes of the In_2O_3 thin films were examined by tapping-mode atomic force microscopy (AFM). Figure (7) show that the In_2O_3films fabricated on the different Laser energy preperation. The AFM micrographs of In_2O_3 thin films deposited at laser energy (100 mj) do not reveal any characteristic features and the films are amorphous. Whereas, it is remarkable to note that, the AFM data indicate nano-structured grain growth for In_2O_3 thin films deposited at energy of (200mj). The fine-microstructure can be seen in the image a of Fig.(7). 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. The grain size for In_2O_3 thin films deposited on these glass substrates increased from (30 to 370) nm with the increase in Laser energy from (100 to 200) mj. The image b of Fig. (8) represents the AFM micrograph of In_2O_3 films deposited at the highest energy of (400 mj). The root mean square value of surface roughness (rms) derived from the AFM image is (7.58 nm) and roughness average (5.97nm). It is clearly evident from the images shown in Fig.(8) that there is a change in the growth as well as arrangement of grains. The rms surface roughness for the films also increased from (15 to 155) nm with the increase in laser energy from (300 to 400) mj.

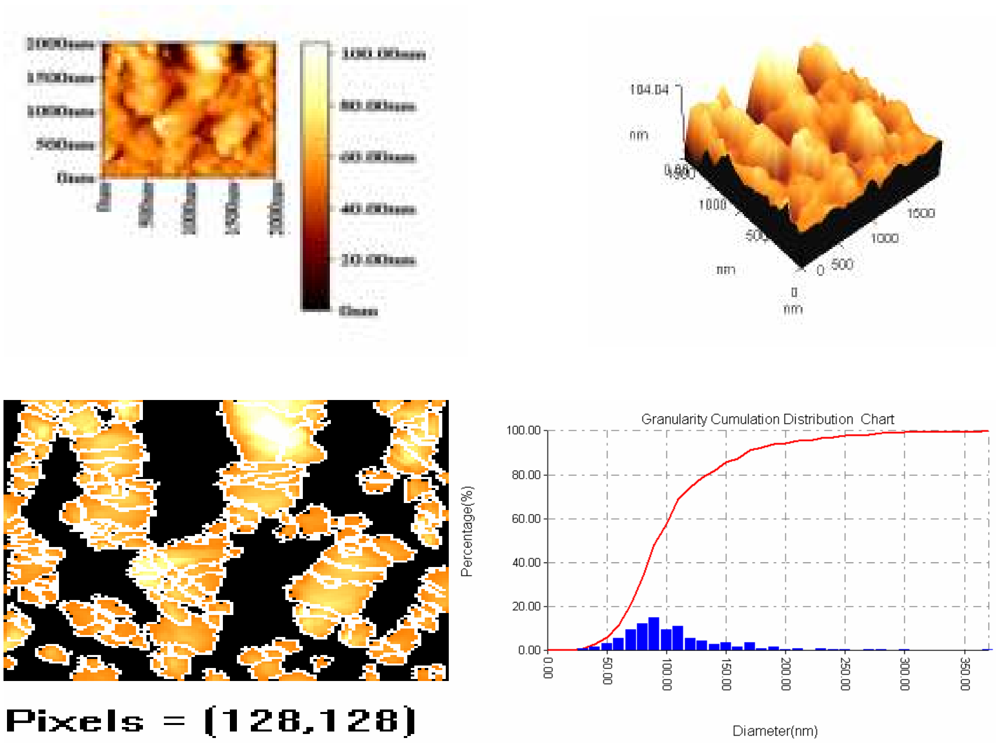


Figure (7) Show that the In_2O_3films fabricated on the lowest laser energy 100mj.

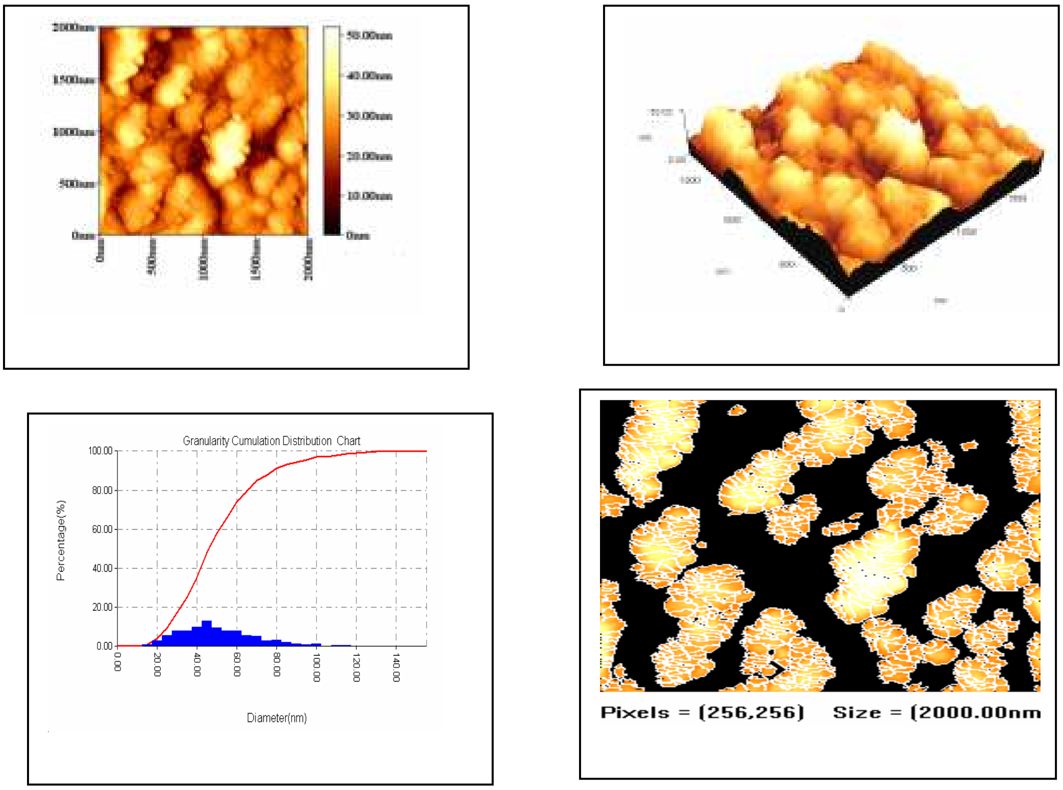


Figure (8) Represents the AFM micrograph of In_2O_3 films deposited at the highest energy of (400 mj).

CONCLUSION

We have successfully prepared high transparent In_2O_3 thin films using reactive pulsed laser deposition (RPLD). The optical and surface morphology results of indium oxide have been investigated as a function of laser energy. An average visible transmittance of (85%) and an optical band gap ranged between (3.6-3.9) eV was measured in 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). In the light of obtained results, these films can be used in optoelectronic and related high-technology applications.

REFERANCE

Bregman J. 1, Yoram Shapira1 and H. Aharoni2 .(1990)," Effects of oxygen partial pressure during deposition on the properties of ion- beam- sputtered indium-tin oxide thin films" J. Appl. Phys. 67, 3750; doi:10.1063/1.345017,(4 pages).

Grivas C., Gill D.S., Mailis S., Boutsikaris L. and Vainos N.A. (1998)," Indium oxide thin-film holographic recorders grown by excimer laser reactive sputtering" Appl. Phys. A 66, pp.(201–204).

Guodong Liu.(2011),"Synthesis,Characterization of In2O3 Nanocrystals and Their Photoluminescence Property" Int. J. Electrochem. Sci., 6 2162 – 2170.

Kaleemulla S., Sivasankar Reddy A. , Uthanna S. and Sreedhara Reddy P. (2009) ," Physical properties of In2O3 thin films prepared at various oxygen partial pressures" Mater. Lett Journal of Alloys and Compounds 479, pp.(589–593).

Maensira b* S. , Laokula P., Klinkaewnaronga J., Phokhaa S., Promarakc V. and Seraphin S. (2008)," Indium oxide (In2O3) nanoparticles using Aloe vera plant extract: Synthesis and optical properties " , Journal of optoelectronics and advanced materials Vol. 10, No. 3, March, pp. (161 – 165).

anoja P.K., Gopchandrana K.G., Peter Koshyb, Vaidyana V.K. and Benny Josephc. (2006), " Growth and characterization of indium oxide thin films prepared by spray pyrolysis" Optical Materials, Volume 28, Issue 12, September, pp.(1405–1411).

Mohan Babu P., Radhakrishnaa B , Venkata Rao G. , Sreedhara Reddy P. and Uthanna S.(2004)," Bias voltage dependence properties of dc reactive magnetron sputtered indium oxide films" Journal of Optoelectronics and Advanced Materials Vol. 6, No. 1, March, pp.(205– 210).

Peng X. S. ,. Meng * G. W., Zhang J. , Wang X. F., Wang C. Z. andZhang L. D.(2002), " Synthesis and photoluminescence of single-crystalline In2O3

Nanowires" J. Mater. Chem., , 12, pp.(1602–1605).

Subrahmanyam A., Barik U.K. (2006)," Electrical and optical properties of reactive dc magnetron sputtered silver-doped indium oxide thin films: role of oxygen " Applied Physics A: Materials Science & Processing Volume 84, Numbers 1-2, , 221-225, DOI: 10.1007/s00339-006, p.(3606).