



Original research article

Preparing and characterization of indium arsenide (InAs) thin films by chemical spray pyrolysis (CSP) technique

Ali K. Al-Mousoi, Mustafa K.A. Mohammed, Haider A. Khalaf*

Department of Applied Science, University of Technology, Baghdad, Iraq

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ABSTRACT

In this research, InAs thin films were prepared by chemical spray pyrolysis method at different substrate temperatures (250°C , 280°C , 310°C), with fixing the molarity of solution to 0.035 M and the deposition time to 30 min . X-ray diffraction shows that the structure of InAs films were polycrystalline and the most intensity planes approximately $25^{\circ}.2'$ and $29^{\circ}.1'$ corresponding to (111) and (200) orientation respectively. Surface analysis by atomic force microscopy (AFM) showed that the morphology of the deposited films is nanostructured. Optical measurements were characterized by UV-vis spectroscopy showed that the transmittance and the energy gap of InAs films decreased with increasing the substrate temperature due to the decreasing in the density of state by quantum confinement. Electrical characteristics showed that InAs film has been a negative Hall coefficient (R_H) value which confirm that InAs is *n*-type semiconductor and has carrier's concentration about $1.67 \times 10^{11} \text{ cm}^{-3}$.

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1. Introduction

Thin film is a material produced by the indiscriminate nucleation and accretion processes of separately condensation reacting atomic, ionic, or molecular sorts on a substrate. Thin films may consist of a numerous thickness range, it varies from a few nanometers to several micrometers and, thus they are best defined in terms of the preparation processes rather than by thickness [1]. Indium Arsenide (InAs) is an *n*-type important semiconductor material with a direct band gap of 0.36 eV and high electron mobility ($33,000\text{ cm}^2/\text{Vs}$) at room temperature. The band gap of InAs can be tuned from its bulk value which is (0.36 eV) to 2.6 eV that makes InAs suitable for heterojunction photovoltaic applications [2]. InAs has a cubic zinc blende structure and a lattice constant of about 6.0583 \AA with polycrystalline and the structure extremely affected by deposition parameters such as deposition time, substrate temperature and molarity of the precursor solution [3]. A lot of methods have been used for thin films production like chemical bath deposition (CBD), electrodepositing (ED), Chemical spray pyrolysis (CSP) and thermal evaporation technique. One of the varied methods, Chemical spray pyrolysis deposition, which used in this study, is an excellent technique because deposition of large surface with good homogeneity can be obtained at a low cost [4]. Spray pyrolysis is essentially a chemical reaction, which involves spraying a solution into a substrate fixed at high temperature, and finally the solution reacts to form the required thin film [5].

* Corresponding author.

E-mail address: haider14ak@gmail.com (H.A. Khalaf).

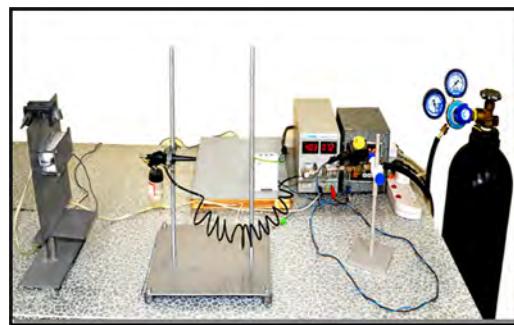


Fig. 1. Image of the chemical spray pyrolysis system.

2. Experimental work

2.1. Substrates and solutions preparation

Glass and Indium Tin Oxide coated glass (ITO) substrates were used for depositing thin films. First, the substrate was cut with suitable dimensions and after that the substrates were cleaned by hydrochloric acid (HCl), ethanol and distilled water (DIW), then dried by optical cleaning paper. The solution required for deposition of InAs was prepared by solving InCl_3 and AsO_3 in distilled water individually and mixing these solutions by using magnetic stirrer after that these two solutions were mixed together. The value of the volume of the distilled water and the mass of the materials were calculated to bring the molarity to 0.035 M by using the relationship [6].

$$M = (W_t/W_{wt}) \times (1000/V) \quad (1)$$

where M; the molarity of the solution.

W_t ; the mass of a material should be added to a certain volume of distilled water to obtain the required molarity.

W_{wt} ; molecular weight of the material in gm/mole.

V; the volume of the distilled water measured in (mL).

2.2. Deposition process

The chemical spray pyrolysis technique has been used in preparing the films on glass and ITO substrates (Fig. 1). The first step after solution preparation was putting the solution in the bottle of the nozzle and then the substrate was placed in the middle of the surface of the heater and left to reaches the required temperature (250°C , 280°C , 310°C). The pressure of the air compressor was in the range (3.5–5) bar. The distance between the nozzle aperture and the substrate was fixed to be 20 cm. After all these steps were done, then solution was sprayed in horizontal direction and make sure the droplets of solution is falling in regular manner on the substrate surface. The spraying time of the solution was controlled by electrical timer and electrical gas valve which make the time to be (2 s) on and (28 s) off. After finishing the deposition, the substrate left to cool to the ambient temperature. The X-ray diffraction measurement has been done and compared with Joint Committee on Powder Diffraction Standards (JCPDS), using Philips PW 1840 X-ray diffractometer of wavelength $\lambda = 1.5406 \text{ \AA}$ form Cu-K α . Atomic force microscopy (AFM) is used to measure size, granularity accumulation distribution and roughness average in nano-meter or even in sub nano level. In our project scanning probe microscope (CSPM-5000) was used as instrument to measure grain size and roughness of the films. Optical Measurement was done by a Cecile (CE 7200) Spectrophotometer supplied by Aquarius company was used to record the optical transmission for the films (375–900 nm). Finally, an Ohmic contact deposited via thermal evaporation technique was used to measure the electrical properties (DC measurement and Hall measurement). This process done under pressure of about 10^{-5} torr. Aluminum wires of 99.999 purity was used as an Ohmic contact material.

3. Results and discussion

3.1. Structural properties

Fig. 2A–C shows the X-ray patterns of the InAs thin films deposited at three different substrate temperatures (250°C , 280°C , 310°C) with fixing the molarity of solution to 0.035 M and the deposition time to 30 min. It is clear that all the films are polycrystalline with a cubic structure and the most intensity planes, which has the close value to the reference data (JCPDS) card no. 15-0869, are occurred at 2 θ values of approximately $25.2'$ and $29.1'$ corresponding to planes (111) and (200) respectively. At substrate temperature of 250°C the intensity of the peak (111) has a small intensity compared with (200) plane, and some minor peaks appears which belong to InCl_2 . This may be due to incomplete reaction of the solution,

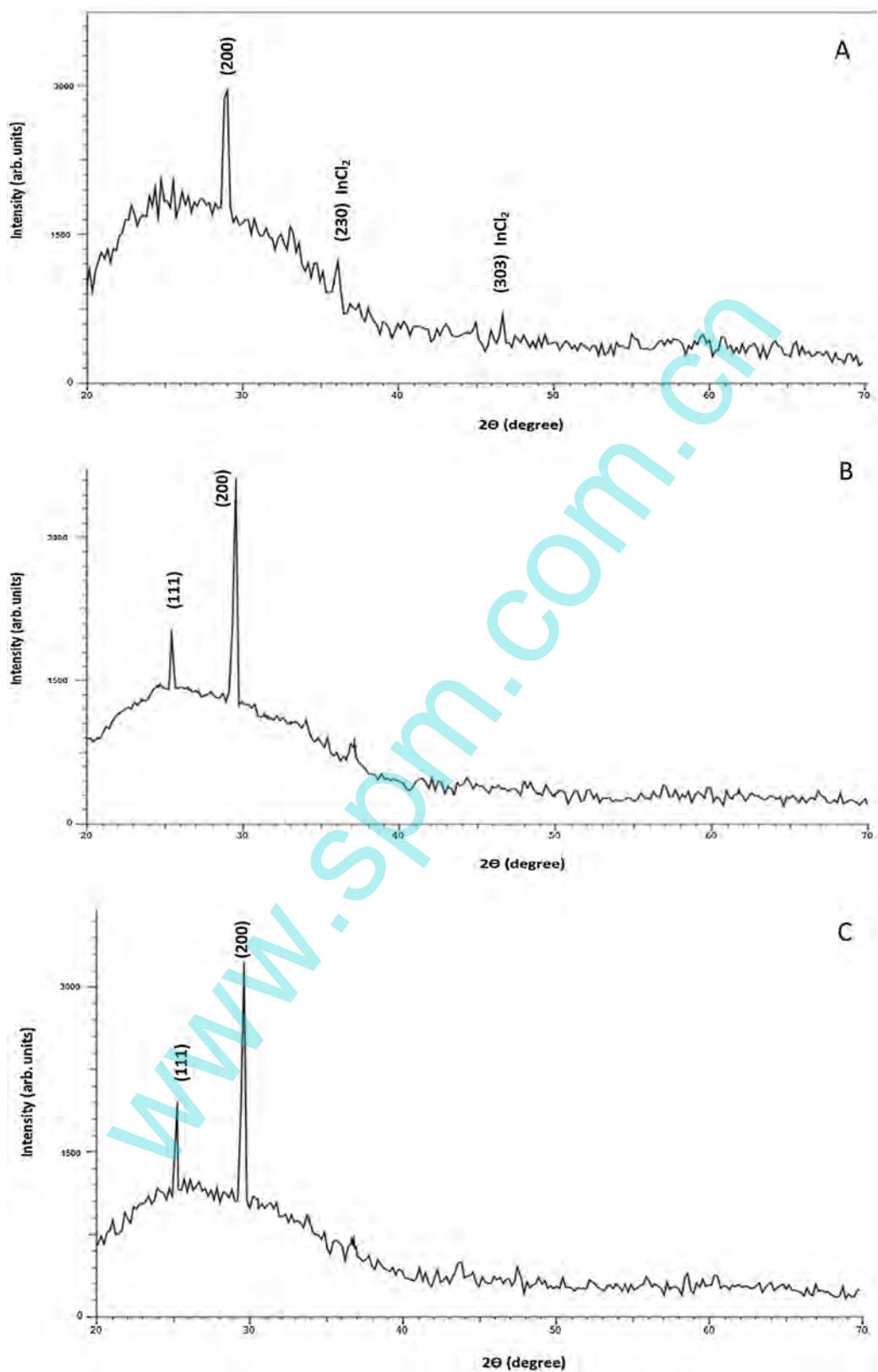


Fig. 2. XRD patterns of the InAs thin films deposited on glass substrate at substrate temperature: (A) 250 °C, (B) 280 °C and (C) 310 °C.

Table 1

The result data of XRD of the InAs thin films deposited on glass substrate at different substrate temperatures.

Substrate temperature (°C)	FWHM (°) at (200)	Grain size (nm)	a (Å)	Strain (%)
250	0.5297	15.49	6.151	1.53
280	0.5125	16.01	6.11	0.8533
310	0.4997	16.43	6.08	0.358

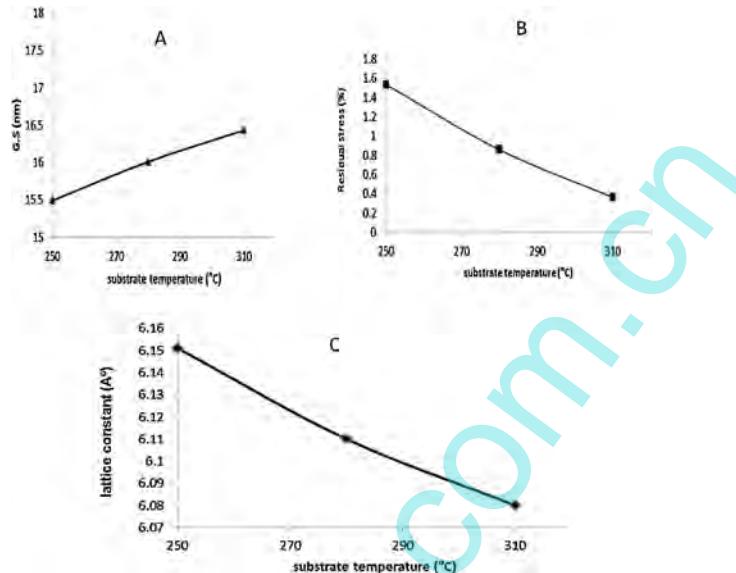


Fig. 3. Showing the effects of the substrate temperature of the deposited InAs film on (A) grain size, (B) residual stress, and (C) lattice constant.

and these InCl_2 peaks are disappeared in higher substrate temperature. All peaks are shifted slightly to lower 2θ value (higher value of the lattice constant) which means there is a strain in the lattice due to the deposition on amorphous substrate and the difference of thermal expansion between the substrate and the film. The full width at half maximum (FWHM) is an important property of the film which indicates to the degree of crystallite growth that occurs in the film. From (Table 1) it is obvious that when increasing the substrate temperature, the (FWHM) decreased and that leads to an increase in the grain size. Fig. 3A–C shows the dependence of structural properties of the film such as the grain size, lattice constant and the micro strain upon the temperature of the substrate, the micro strain is a structural defect occurs in film during the deposition process, and it is reached the minimum value as the highest temperature in case of this study (310°C) as well as the closer lattice constant to the standard value.

3.2. Morphological properties

The surface morphology of films has been investigated using AFM images, which produces topological images of surfaces at very high magnification. The AFM images of the InAs thin film in both 2D and 3D form are shown in Fig. 4A and B, the deposition parameters of the InAs film were (time of the spraying is 30 min, substrate temperature is 310°C and molarity of the solution is 0.035 M), the roughness of the film is 1.43 nm and the average particles diameter is 78.37 nm, this can be seen from Fig. 4C, where the high particles diameter value (40%) is about of 75 nm. The result of the particle size, that calculated by using Scherrer-Debye formula, is smaller than that estimated from AFM measurement indicating that grains are probably an aggregate of many crystallites.

3.3. Optical properties

Fig. 5 shows the optical transmittance of InAs thin films prepared at different substrate temperature, less transmittance found with high substrate temperature. In spite of the effect of temperature on the transmittance curves but there is a distinguish difference in the shape of the curves, this may be due to the effect of impurities which act as levels between conduction band and valence band.

Absorption coefficient (α) is calculated by using Eq. (1).

$$\alpha = 2.303 \left(\ln \frac{1}{T} \right) \frac{1}{t} \quad (1)$$

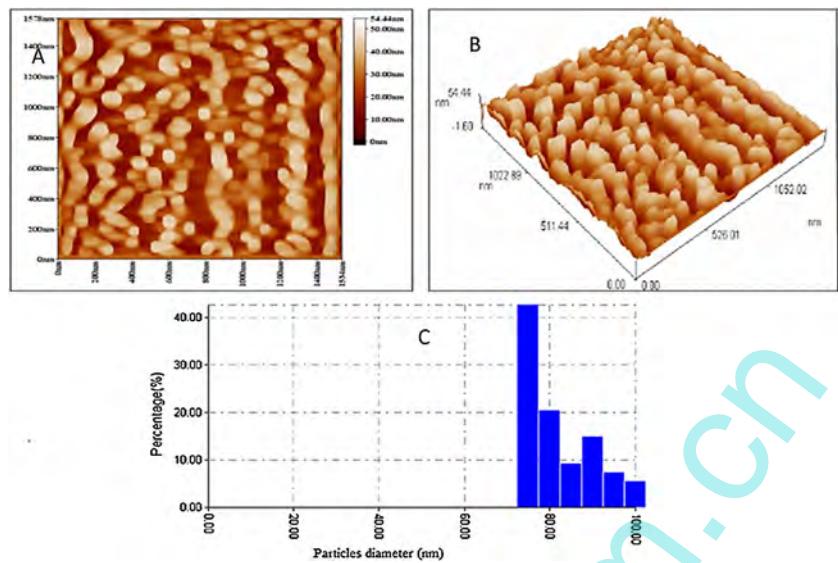


Fig. 4. (A) 2D, (B) 3D AFM images and (C) granularity cumulation chart for InAs film prepared by spray pyrolysis technique.

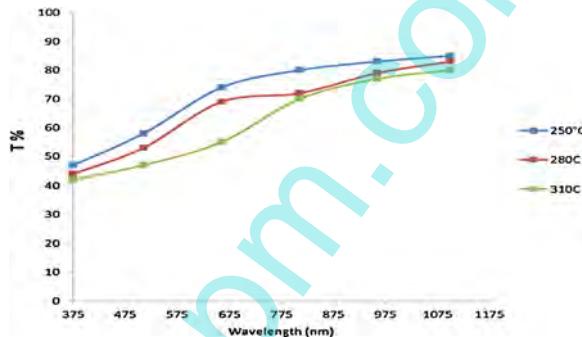


Fig. 5. Transmittance as a function of wavelength for InAs thin films deposited at different substrate temperatures.

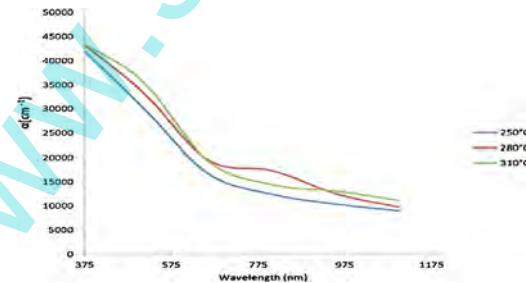


Fig. 6. Absorption coefficient as a function of wavelength for InAs thin films prepared at different substrate temperature.

where t is the thickness of thin film, and T is the transmission. In the direct band gap structure or direct transition semiconductors (present case), the absorption coefficient and optical band gap (E_g) are related by [7].

$$\alpha = A(h\nu - E_g)^{\frac{1}{2}} \quad (2)$$

where A is a constant, h is Plank's constant and ν is the frequency of the incident photon. Fig. 6 shows the effect of substrate temperature on the spectral absorption coefficient. All films have high absorption coefficient at short wave lengths (400 nm), for longer wave length a sharp decreasing occurs, and for $\lambda \geq 650$ nm the curves show a saturation behavior. The sharp decreasing and saturation are an indication to the presence of nano films.

Different energy gap values, of InAs thin films deposited at different temperatures, are shown in Fig. 7, these values are much bigger than the bulk value (0.34 eV) due to the decreasing in the density of state which caused by quantum confinement.

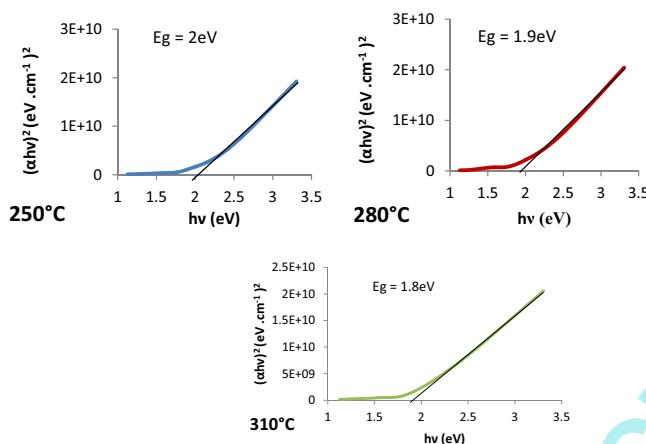


Fig. 7. Shows the estimated energy gap of InAs films deposited at different substrate temperature.

Table 2

Hall effect data of the deposited film according to the optimum condition.

Film type at 310 °C	$R_H (\text{cm}^3/\text{C})$	Majority carriers	Carrier concentration (cm^{-3})	$\rho (\Omega \text{ cm})$	$\mu_H (\text{cm}^2/\text{V s})$
InAs	-3.74×10^7	n	1.67×10^{11}	2.05×10^5	180

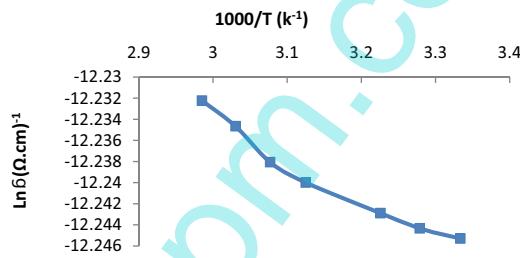


Fig. 8. Plots of $\ln(\sigma)$ vs. $1000/T$ for InAs thin films deposited at substrate temperature 310 °C.

Table 3

Values of the activation energy for the deposited film.

Film type	$E_{A1} (\text{eV})$	$E_{A1} (\text{eV})$
InAs	0.34	–

4. Electrical properties

4.1. Hall measurements

Hall measurements were performed so as to investigate the electrical properties of InAs thin films such as resistivity, mobility, carrier concentration and semiconductor type. The results of Hall measurements show that InAs has a negative Hall coefficient (R_H) values which confirm that InAs is *n*-type semiconductor, on the other hand. All data related to the Hall measurements are listed in Table 2.

4.2. Dc conductivity measurements

Unlike metals, semiconductors conductivity increased as the ambient temperature increased [8]. In order to estimate the activation energy (EA) which can be defined as the energy required to make the carrier charge moving from the Fermi level to the conduction band (if the material is *n*-type) or from the Fermi level to the valence band if the material is *p*-type, the conductivity of the film was studied as a function of $1000/T$ as shown in Fig. 8, the EA values are listed in Table 3.

5. Conclusions

The InAs thin film prepared by chemical spray pyrolysis technique at different substrate temperatures, it's a reliable, cheap and simple method has been used. The substrate temperature has an important influence on some properties of the deposited films especially the structural and optical properties.

References

- [1] K.L. Chopra, P.D. Paulson, V. Dutta, Thin-film solar cells: an overview, *Prog. Photovoltaics Res. Appl.* 12 (2004) 69–92.
- [2] Samuel L. Liao, *Microwave Devices and Circuits*, third edition, Prentice Hall, Englewood Cliffs, New Jersey, 1996, 533p.
- [3] F.S. Bahabri, R.H. Orainy, Sara Al Balwi, Study of some structural and transport properties of evaporated indium arsenide (InAs), *Thin Films* (2011).
- [4] Mohammad Ghaffar Faraj, Effect of thickness on the structural and electrical properties of spray pyrolysed lead sulfide thin films, *Am.J. Condens. Matter Phys.* 5 (2) (2015) 51–55.
- [5] Mustafa Öztaş, Metin Bedir, Thickness dependence of structural, electrical and optical properties of sprayed ZnO:Cu films, *Thin Solid Films* 516 (2008) 1703–1709.
- [6] R. Thiagarajan, M. Mahaboob Beevi, M. Anusuya, T. Ramesh, Influence of reactant concentration on nano crystalline PbS thin films prepared by chemical spray pyrolysis, *Optoelectronics Adv. Mater.: Rapid Commun.* 6 (1–2) (2012) 132–135.
- [7] S. Dimitrijev, *Understanding Semiconductor Devices*, Copyright by Oxford University Press, 2000.
- [8] V. Khranovskyy, A. Ulyashin, G. Lashkarev, B. Svensson, R. Yakimova, Morphology, electrical and optical properties of undoped layers deposited on silicon substrate by PEMOCVD, *Thin Solid Films* 516 (2008) 1396–1400.