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INFLUENCE OF ANNEALING ON THE STRUCTURAL AND OPTICAL CHARACTERISTICS OF INDIUM OXIDE THIN FILMS PREPARED USING THERMAL EVAPORATION IN A VACUUM METHOD

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ABSTRACT

In the work, the structures and the optical parameters were studied for In_2O_3 thin films Before and after annealing at different temperatures (400℃ and 500℃) thin films were prepared on glass substrate by by vacuum thermal evaporation. The X-Ray diffraction showed that In_2O_3, In_2O_3 Plasticizedthin films which were prepared to have a polycrystalline cubic structure. The most preferential orientation is along the (321) direction for all deposited In_2O_3 films. Some parameters of the films were calculated as the average grain size and the dislocation density. Optical properties measurement transmittance (T) of the film shows that they have high transmittance (82.7%) is obtained for at annealing 500℃. The optical properties such as absorption coefficient (α) using the transmittance measurement from an UV-vis spectrophotometer, with a range of wavelength (300-1100) nm. And also calculated energy gap for thin films. and also calculated energy gap for thin films, the result has been shown that the optical energy gap increasing with increasing annealing.

Keywords

thin film; structural properties; Indium oxide; band gap ;AFM

1. INTRODUCTION

Indium oxide is a transparent conductor, as well physically stable and chemically Inert. Usually, In_2O_3 crystallizes into a cubic bixbyite structure and has a melting point temperature of 1910°C and lattice constant 10.117 AO [1].It is an insulator in its stoichiometric form, where as in its non-stoichiometric form it behaves as a highly conducting semiconductor with a wide optical wide band-gap (3.7eV), providing reflectivity high in the infrared light range and high transparency in the visible light range [2]. Indium oxide (In_2O_3) is important transparent conducting oxide(TCO) materials that has application in flat panel display and optoelectronic devices due to high electrical conductivity and high optical transparency [3]. Indium oxide films can be prepared by several techniques, such as thermal evaporation, DC sputtering, chemical vapor deposition (CVD) and spray pyrolysis [4]. The vacuum thermal evaporation deposition technique consists in heating until Vaporization of the material to be deposited. The material steam finally condenses to form of thin layer on the cold substrate surface and on the vacuum chamber walls. Usually low pressures are used, about (10-6 - 10-5) Torr, to avoid a reaction between the vapor and atmosphere. At these low pressures, the mean free path of steam atoms is in the same order as the vacuum chamber dimensions, so these particles travel in straight lines from the evaporation source toward the substrate. Films produced by evaporation are relatively pure and thus are of interest from a theoretical standpoint . In this work effects of film annealed on the structural and optical properties of the indium oxide films prepared by vacuum thermal evaporation deposition technique have been studied.

2. EXPERIMENTAL DETAILS

2.1 Synthesis of Indium oxide (In_2O_3)

The glass substrate was cleaned with alcohol pure ethanol washed with deionized water ultrasonically cleaned in alcohol for about 15 minutes, washed with deionized

water. Indium thin films were deposited by thermal vacuum evaporation on a glass substrate at a vacuum pressure of 3.6x10-5 Torr. The thermal evaporator used was an EDWARD 306 to conduct thermal evaporation of indium process. Material of Indium pure was used and evaporated from a tnquestn, boat the vapor was condensed on cooled substrate. The oxidation experiment last for 1h in air atmosphere at temperature 400°C. and films finally leave to cool until it reaches room temperature.

2.2 Thicknesses Measurement

This method gives an approximate deposited film thickness using a sensitive electrical balance of type(Precise). The thin film substrates are weighted before and after deposition. The theoretical formula is given by [5].

$$t = \frac{m}{2\pi\rho r^2} \dots\dots\dots\dots (1)$$

where m is the mass of the material (In_2O_3), r is the distance between the substrate

and the boat, is the density of the material (In_2O_3).

Indium oxide thicknesses was measured by using optical interferometer method This method was based on interference of light beams reflected from thin film surface and substrate bottom He-Ne laser of wavelength (632nm) with incident angle 45^{O} was used and the thickness is determined using the formula[5].

$$t = \frac{\Delta x}{x} \times \frac{\lambda}{2} \cdots\cdots\cdots\cdots (2)$$

Where λ is the wavelength of laser light, Δx is the distance between two fringes and x is fringe width.

2.3 Characterization

The crystal structure of thin films (In_2O_3) was determined by X-ray diffraction using the Philips model, (PW/1710) diffract meter, with monochromatic CuKα radiation (λ = 1.541 A^{o} at 40 Kα and 30 mA) and 2θ ranges from 20° to 80°. The optical properties of indium oxide nanocomposites are measured by using UV/VIS Shimadzu 3101PC Spectrometer in range of wavelength (300 to 1100) nm . When the light of intensity (I_o) incident on the film of thickness (t) the transmitted intensity (I) can be obtained using the Lambert law [6].

$$I = I_o \exp(-\alpha t) \cdots\cdots\cdots\cdots (3)$$

I/I_o represents the transmittance (T), the absorption(A) [6].

$$A = \mathrm{Log}(\frac{1}{T}) \cdots\cdots\cdots\cdots (4)$$

Then absorption coefficient α is given by [7]

$$\alpha = 2.303(A/t) \cdots\cdots\cdots\cdots (5)$$

The energy gaps E_g of films were estimated using Tauc relation[8]

$$\alpha h\nu = B(h\nu - E_g)^r \cdots\cdots\cdots\cdots (6)$$

where B is the constant involving the properties of the bands, hv is the photon energy.

3. RESULTS AND DISCUSSION

3.1 XRD Analysis

The structure of (In_2O_3) thin films examined by X-Ray diffraction (XRD). before and after heat treatment at ca. (400- 550 °C) The installation of multi crystallization and type cubic , The growth of crystalline For the films crystalline directions (211) ,(222) ,(321), (400) and (642) reflections of the In_2O_3 phase as shown in Fig. 1. These data are compared with ASTM file No.00-006-0416 data.(321) is the predominant crystallographic plane indicating a perpendicular alignment of the c-axis of the grains.

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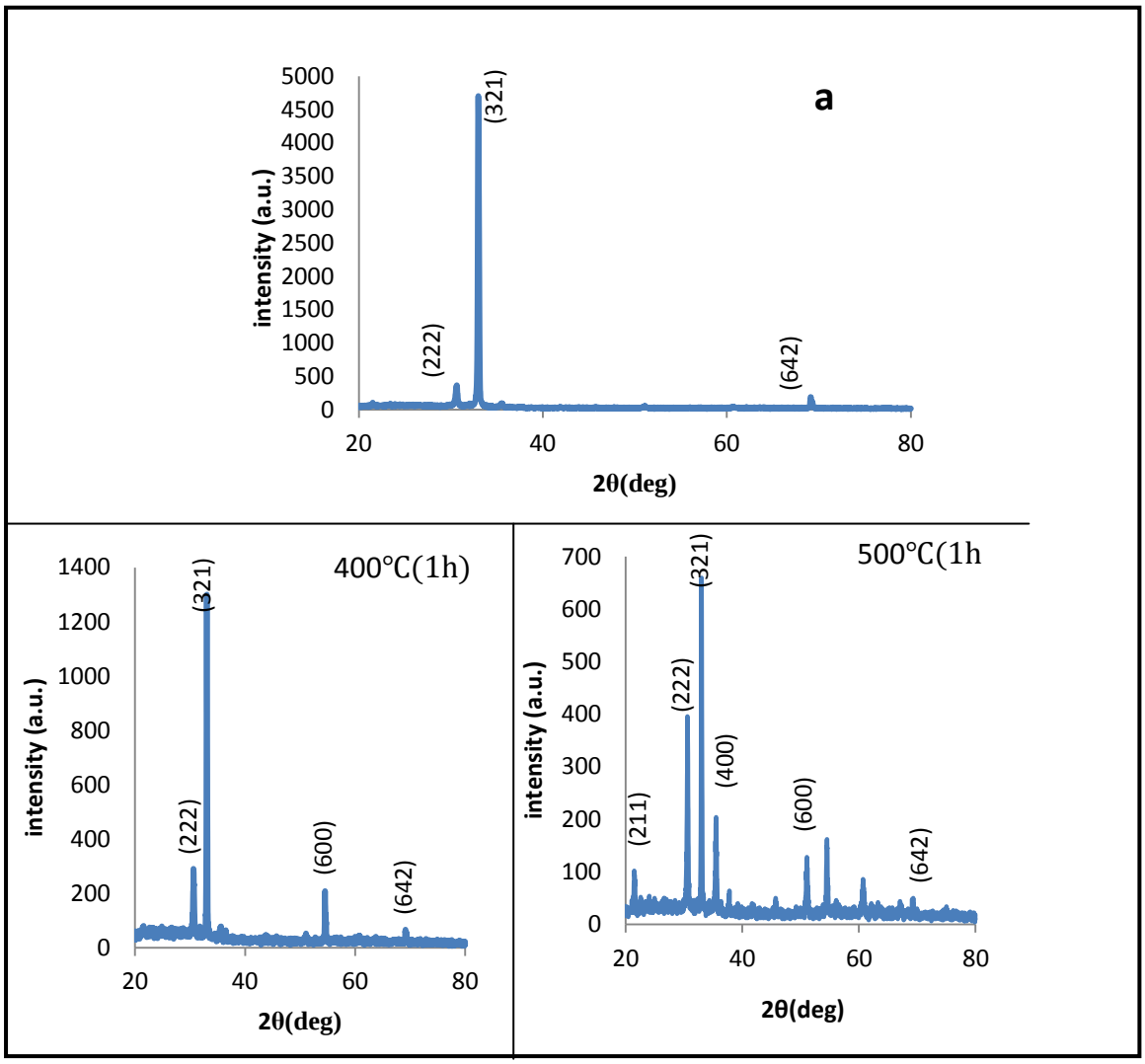

Fig. 1. The XRD pattern of In_2O_3 thin film prepared at different annealing temperatures: (a) as-deposited, (b) 400℃, (c) 500℃

The average crystallite size (D_{av}) was calculated using the Scherrer equation[9]:

$$D_{av} = \frac{K\lambda}{\beta COS\theta} \ldots\ldots\ldots\ldots (7)$$

where, k is the shape factor, λ = 0.154 nm is the wavelength of the X-ray, β is the full width half maximum FWHM of the diffraction peak and Θ is the angle of the diffraction peak , The values of average grain size listed in the Table 1, the values of the average grain size were increased At a temperature Annealing 500℃. This increase may be due to the That the effect of the degree of annealing temperatures led to the cancellation of some defects existing levels after obtaining the growth process of rearranging the crystalline granules.

The dislocation density (δ) defined as the length of dislocation lines per unit volume of crystallites was evaluated using equation (8)[10]

$$\delta = \frac{1}{D_{av}} \ldots\ldots\ldots\ldots (8)$$

3.2 AFM Measurements

A study of the surface morphology by AFM gives a 2D and 3D presentation of the grain arrangement. Fig. 2 shows the AFM picture of the In_2O_3 film prepared under the Before and after annealing. It can be seen that the film has better uniformity and crystallographic structure. The root mean square value of surface roughness (RMS) and roughness average (R) listed in the Table 1.

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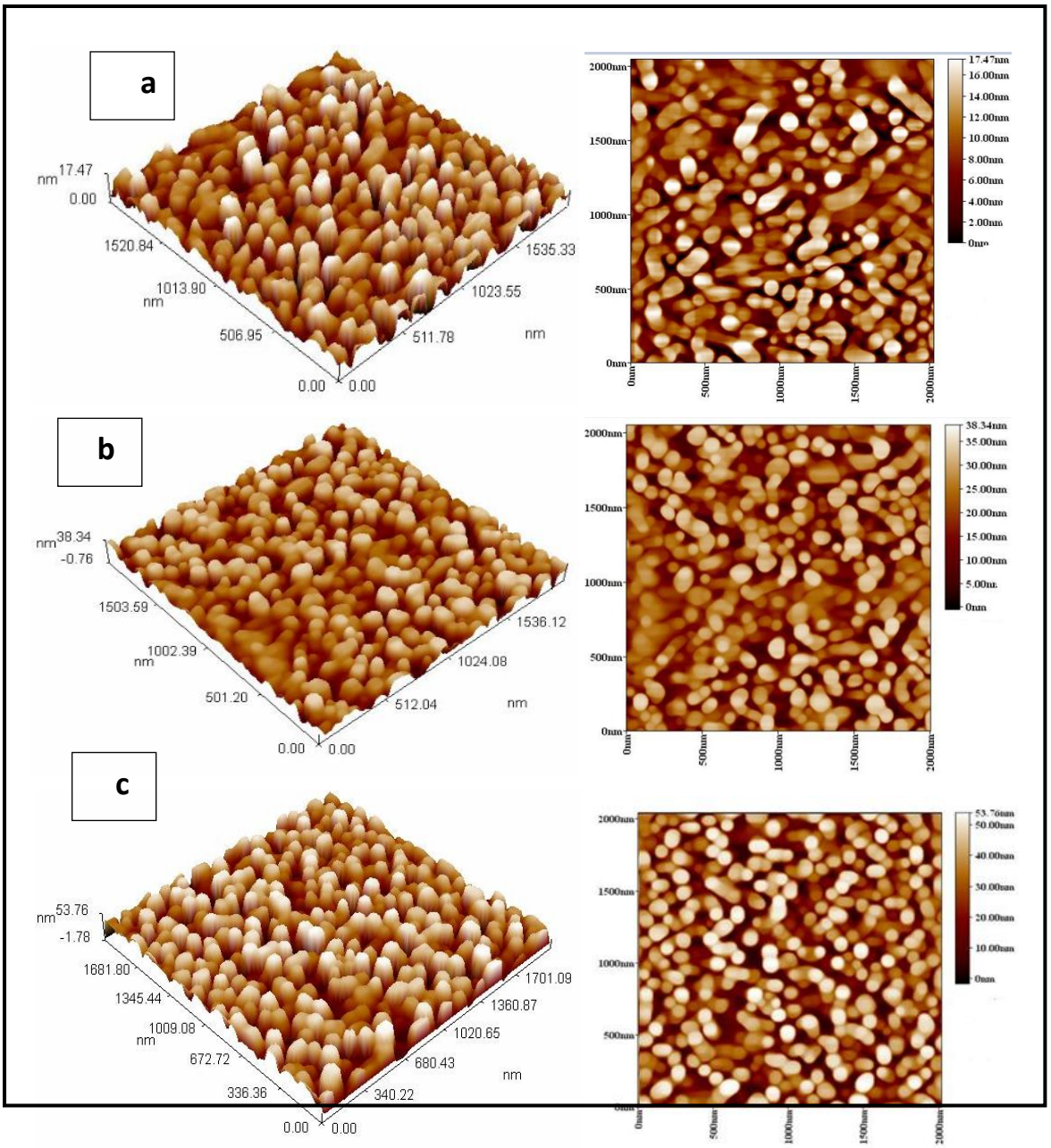

Fig.2. 2D-3D AFM topography of In_2O_3 film at (a)R.T (b)400℃ (c)500℃

Samples	Dav (nm)	R(nm)	RMS(nm)	σ x104(nm⁻²)
In2O3 at. R.T	56.8875	3.47	4.18	3.0901
In2O3 at. 400 °C	50.2995	6.62	7.9	3.8619
In2O3 at. 500 °C	68.5266	11.6	13.6	2.1295

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3.3 Optical Properties Calculations

The optical properties of In_2O_3 and Annealing In_2O_3 thin films were studied using UV–Vis spectrophotometer in the range 360–1100 nm at room temperature. The transmission (T) was recorded in the range of 360 nm to 1100 nm of the incident beam as shown in Fig. 3 The transmission in the visible region is found with increasing annealing .The average transmittance in the visible region is Almost (80%). The maximum value of the transmittance is greater than (82.7%) is obtained for at annealing 500 ℃ may be due to the structural and surface effects and increase in grain size.

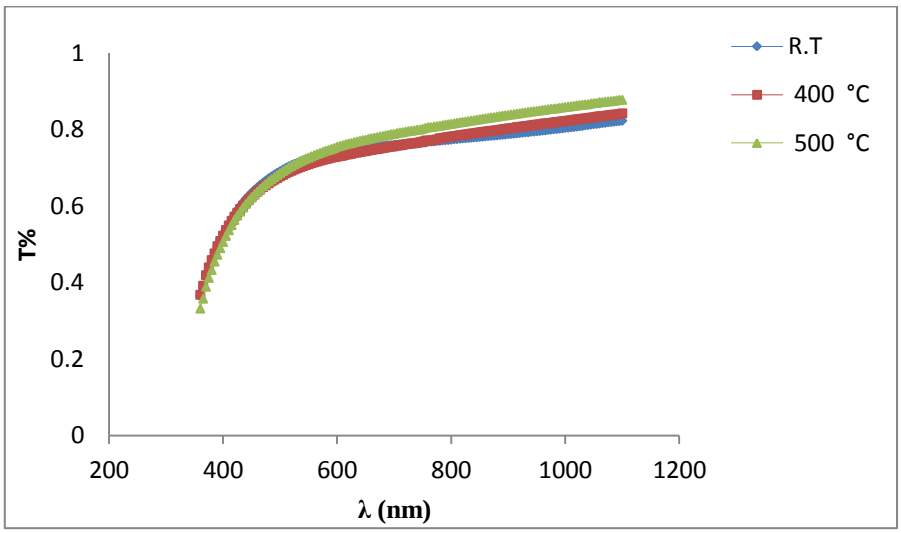
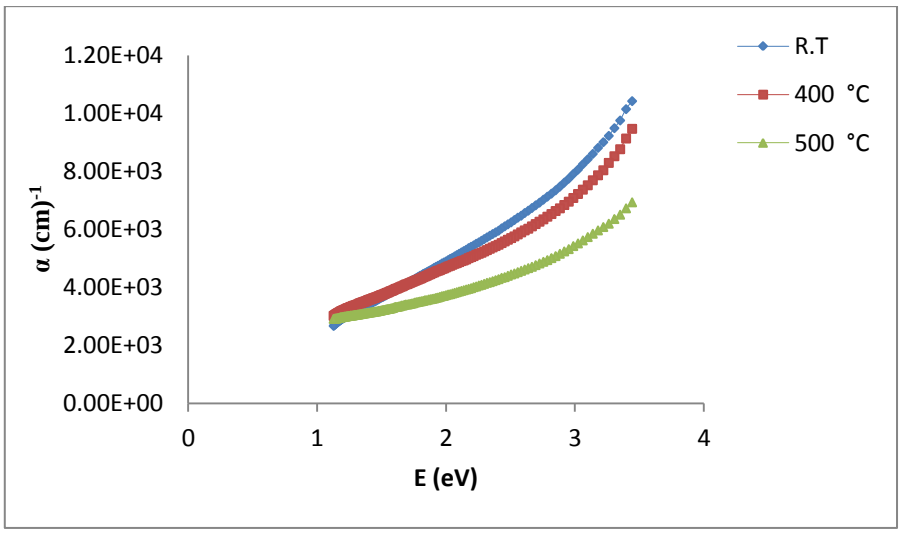


Fig. 3 shows Difference in absorption coefficient of In_2O_3 films prepared at different annealing ,We note that the absorption coefficient raise with increasing photon energy. The increase in the absorption coefficient With increasing annealing due to the small optical energy gap, and get the direct transfer, The absorption ($\alpha \geq 10^4$ cm^{-1}) is related to direct band transitions.



The band gap Eg depends on the type of transition .The values of absorption coefficient (α) plays an important role to limitation the type of transition .Here transitions are direct because (α) is more than (10^4 cm^{-1}), (when $\alpha < 10^4$ cm^{-1} the transition is indirect transition). These transitions are described by the relation (6). (r) takes the values (1/2, 3/2, 2) depending on the type of transition (r =1/2 for direct allowed transition , r = 3/2 for direct forbidden transition and r=3 for an indirect allowed transition). Fig. 4 shows the relation between $(\alpha hv)^2$ (hv) While Fig. 5 shows the relation between $(\alpha hv)^{1/3}$ (hv)

The values of the optical band gap (Eg) are calculated from the intercept of the extrapolation the straight line portion of the curves. accordingly, The Eg values for direct and indirect transition for (In_2O_3) at Room temperature (R.T), In_2O_3 at 400 ℃ and In_2O_3 at 500 ℃ thin films are summarized in Table 2. The energy gap was Increased with increasing annealing. This range is suitable to use this material for solar cell application. The calculated thickness of each deposited thin film is

(300±10) nm.

Table 2. The Eg values for direct and indirect transfers for films In_2O_3

Samples	Optical energy gab Eg (eV) (direct)	Optical energy gab Eg (eV) (indirect)
In_2O_3 at. R.T	3.1	2.9
In_2O_3 at. 400 ℃	3.2	2.85
In_2O_3 at. 500 ℃	3.4	2.7

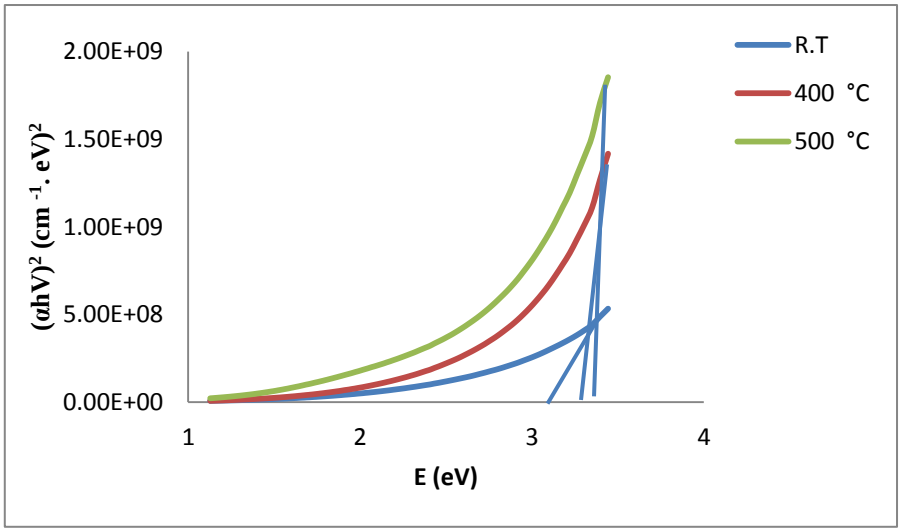


Fig. 4. Variation of $(\alpha hv)^2$ with photon energy (hv) of In_2O_3 films prepared at different concentration of annealing (R.T, 400 ℃and 500 ℃) direct transition

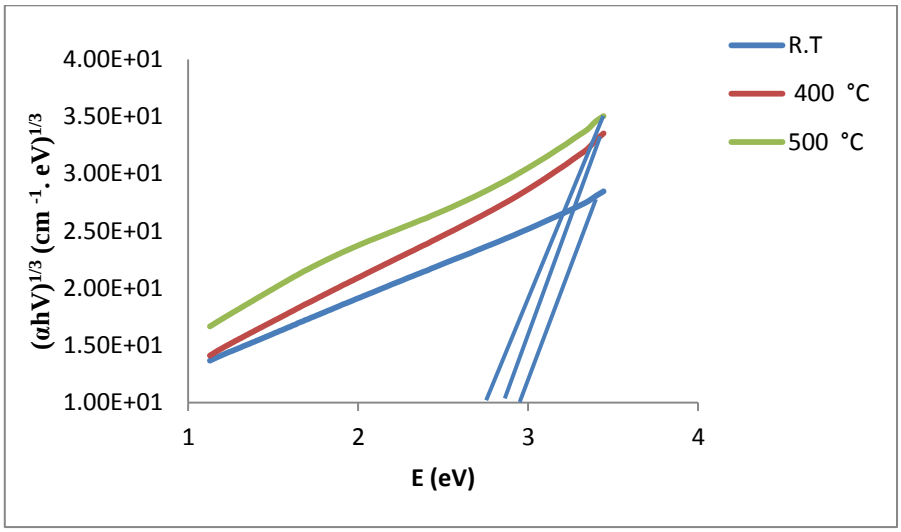


Fig. 5. Variation of $(\alpha hv)^{1/3}$ with photon energy (hv) of In_2O_3 films prepared at different concentration of Annealing (R.T, 400 ℃ and 500 ℃) indirect transition

4. CONCLUSION

This study elucidated the structural and optical properties of In_2O_3 and Different annealing films prepared via the vacuum thermal evaporation on glass substrates. The XRD spectrum shows that thin films are polycrystalline crystallized in the cubic phase. The values of the average grain size were increased with increasing annealing. The transmittance increases with increasing of annealing . The optical band gap was also found to vary from (3.1) to (3.4) eV . thus making the films suitable for solar cell applications Variations in optical properties with wavelength are found to be oscillatory in nature, which are attributed to the particular structure of the films and their Annealing. The precursor (In_2O_3 at:500℃) was found to be optimum parameters for the synthesis of zinc oxide thin films.

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