

## **OPTICAL AND ELECTRICAL PROPERTIES OF CADMIUM TIN OXIDE FILMS PREPARED BY VACUUM EVAPORATION TECHNIQUE**

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### **Abstract**

Preparation of highly conducting and transparent cadmium tin oxide ( $\text{Cd}_2\text{SnO}_4$ ) thin films by vacuum evaporation technique on glass substrate with thickness of (300, 600)nm is reported. The effect of thickness on structural, optical, and electrical properties of the films was investigated by using different techniques such as X-ray diffraction, atomic force microscopy, optical transmittance, and Hall measurement. The X-ray diffraction (XRD) studies revealed that the films are polycrystalline in nature with cubic structure and there are strong peaks at the direction (311), the intensity of the peaks increases with increasing thickness. The grain size varies between 19.1 and 21.4nm. The surface morphological was investigated by atomic force microscopy (AFM), which revealed the crystalline nature of the films. The optical parameters of the prepared films as transmittance, optical energy gap, refractive index, extinction coefficient, and the dielectric constants were found to be affected by varying the thickness. The resulting films show  $\geq 70\%$

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transmission in the visible region DC conductivity and Hall effect measurement as a function of thickness have been studied for these films DC conductivity measurement have been showed that the conductivity increases from 8.69 to  $11.62 (\Omega.cm)^{-1}$  when the film thickness increased. Hall Effect results showed that thin films were *n*-type. Also the charge carrier concentration increases with increasing thickness. The Hall mobility has been calculated, and it is decreased with increasing film thickness. The drift velocity of these films decreases with increasing film thickness.

## 1. Introduction

Transparent conducting oxides (TCOs) have been widely used in different areas due to their high optical transparency, low resistivity, and wide energy band gap. Hence, there has been great deal of work on investigating their preparation process and optimizing their properties [1]. Thin film of ternary oxides  $Cd_2SnO_4$  show extremely promising properties such as low, metal-like electrical resistivity ( $10^{-6}\Omega.m$ ), good transmissivity ( $\geq 80\%$ ) in the visible range of the light spectrum and high reflectivity in the IR range [2]. TCOs can perform a variety of important functions, including contacts, antireflection coatings, and chemical barriers. TCOs are becoming a more critical element in thin film photovoltaic devices. In the continued drive to increase solar cell efficiency and stability while reducing cost and optimizing performance, the optical, electrical, and materials properties of TCOs gain increasing importance. Among the various TCO materials available, cadmium stannate ( $Cd_2SnO_4$ ) is one of the potential candidates for solar cell application [1, 2].  $Cd_2SnO_4$  is an *n*-type semiconductor and have great technological interest due to their high quality electrical and optical properties superior to the conventional transparent conducting oxide materials [1-4]. In addition, these films have wide applications in photogalvanic cells, liquid crystal displays, heat mirrors, transparent electrode, and solar cells. Recently, it has been reported that,  $Cd_2SnO_4$  thin films have several significant advantages in the CdTe/CdS based

solar cells. Comparing with  $\text{SnO}_2$ , cadmium stannate films have better adhesion in the CdTe/CdS solar cells [5]. The reduced film thickness of  $\text{Cd}_2\text{SnO}_4$  films in the solar cell devices reduces the number of interconnects and reduces manufacturing costs. Thus preparing the low cost and quality cadmium stannate thin films is an important and useful task. Different ways of cadmium stannates obtaining are used depending on a purpose such as rf sputtering, DC reactive sputtering, metal organic MOCVD, radio frequency, pulsed laser deposition, dip coating, DC magnetron plasmatron sputtering, reactive magnetron sputtering atom beam sputtering, sol-gel deposition, electron beam evaporation, thermal treatment using the coprecipitation method, spray pyrolysis, and vacuum evaporation [5-21]. The optical and electrical properties of CTO coatings are, though, notoriously sensitive to process parameters and depend on control of film composition, structure, crystallinity, defect density, surface roughness, and dopant concentration [13]. In this paper, thin  $\text{Cd}_2\text{SnO}_4$  films have been prepared by thermal evaporation technique. The main task of this study is a search for studying the effect of the thicknesses on some structural morphological, optical and electrical properties.

## 2. Experimental

Thin  $\text{Cd}_2\text{SnO}_4$  films have been evaporated by using Edward (E306A) coating system under vacuum of about  $2 \times 10^{-5}$  mbar, from  $\text{Cd}_2\text{SnO}_4$  powder purity of about 99.999% manufacturer by Balzers Switzerland. These films deposited on 7059 corning glass slides with (300, 400)nm thickness at room temperature (*RT*). The prepared films have been annealed at 400K. The thin  $\text{Cd}_2\text{SnO}_4$  films structural analysis was examined by using Phillips X-Ray diffractometer system with  $\text{CuK}\alpha$  source. The surface morphology of the prepared films is investigated by means of atomic force microscopy (AFM). Optical transmittance spectrum

was recorded using UV-visible spectrometer in the range of 200-1100nm. The electrical resistance has been measured as a function of the temperature ( $T$ ). The measurements have been done by using sensitive electrometer type of Keithly Digital Electrometer (616) and vacuum electric oven. The resistivity and conductivity as a function of thickness can be calculated from these measurements.

Optical transmittance ( $T$ ) and absorptance ( $A$ ) spectra were performed over the wavelength ( $\lambda$ ) within the range of (300-1100)nm. These data were used to calculate the absorption coefficient ( $\alpha$ ), band gap energy ( $Eg$ ), and the optical constants (extinction coefficient, refractive index, and real and imaginary parts of dielectric constant).

The relation between the intensity of incident light ( $I_0$ ) and the transmitted intensity ( $I_T$ ) is represented by an exponential form [22, 23]:

$$I_T = I_0 \exp(-\alpha t), \quad (1)$$

where  $\alpha$  being the absorption coefficient and  $t$  is the film thickness. According to this equation, the optical absorption coefficient of thin films were evaluated from the transmittance data using the relation

$$\alpha = \ln[1 / T] / t, \quad (2)$$

where  $T = I_T / I_0$  is defined as the transmittance [22].

As a result of absorption coefficient data, the nature of transition (direct or indirect) is determined according to Tauc relation [23] given by:

$$\alpha E = B(E - Eg)^r, \quad (3)$$

where  $B$  is a constant,  $r$  is a constant whose value depend on the type of transition, where  $r$  is equal to 1/2 and 3/2 for allowed and forbidden direct transition, respectively, and  $r$  is equal to 2 and 3 for allowed and forbidden indirect transition, respectively. The term  $E$  in Equation (3) represent the photon energy, which can be calculated from the relation

$$E \text{ (eV)} = hv = 1.24 / \lambda(\mu\text{m}), \quad (4)$$

where  $h$  is Plank constant,  $v$  is the incident photon frequency, and  $\lambda$  is the photon wavelength.  $(\alpha E)^{1/r}$  of thin films is plotted against  $E$  to decide whether this material has allowed or forbidden direct or indirect band gap transition. Since the variation of  $(\alpha E)^2$  with  $E$ , at which the absorption coefficient  $\alpha \geq 10^4 \text{ cm}^{-1}$ , for thin films is a straight line indicating that the involved transition is allowed direct one. Extrapolation of the linear portion of the plot to the energy axis ( $\alpha E = 0$ ) yielded the direct optical band gap energy value of deposited thin film.

Transmittance and absorptance spectra were used to deduce the optical constants including the refractive index ( $n$ ), extinction coefficient ( $k_e$ ), and the real ( $\epsilon_r$ ) and imaginary ( $\epsilon_i$ ) parts of dielectric constant. The reflectance ( $R$ ) was evaluated from the relation [24]:

$$A + T + R = 1. \quad (5)$$

The index of refraction was estimated from the reflectance data using the equation–

$$n = \left( \frac{4R}{(R-1)^2} k_e^2 \right)^{\frac{1}{2}} - \frac{(R+1)}{(R-1)}, \quad (6)$$

where  $k_e$  is the extinction coefficient, which is determined from the formula [8]

$$k_e = \alpha\lambda / 4\pi. \quad (7)$$

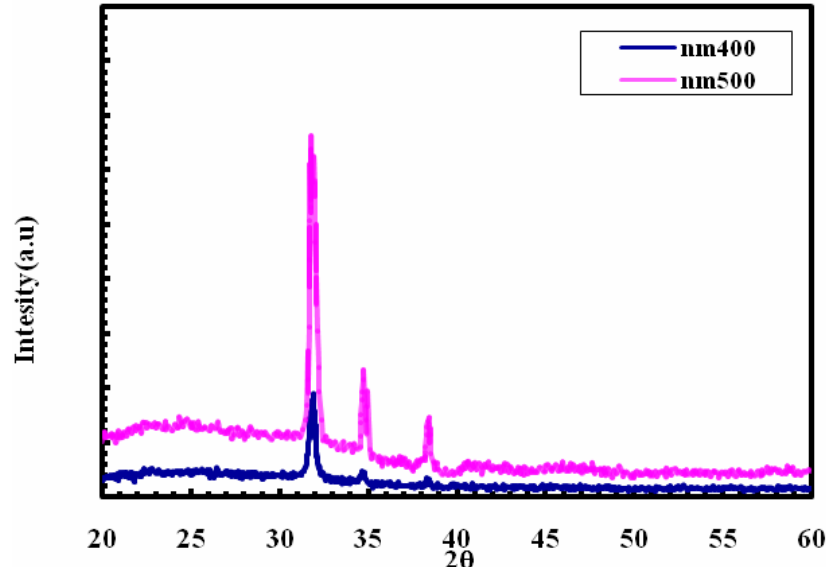
The real and imaginary parts of the dielectric constant of the films were calculated from the relations [25]

$$\epsilon_1 = n^2 - k_e^2, \quad (8)$$

$$\epsilon_2 = 2nk_e. \quad (9)$$

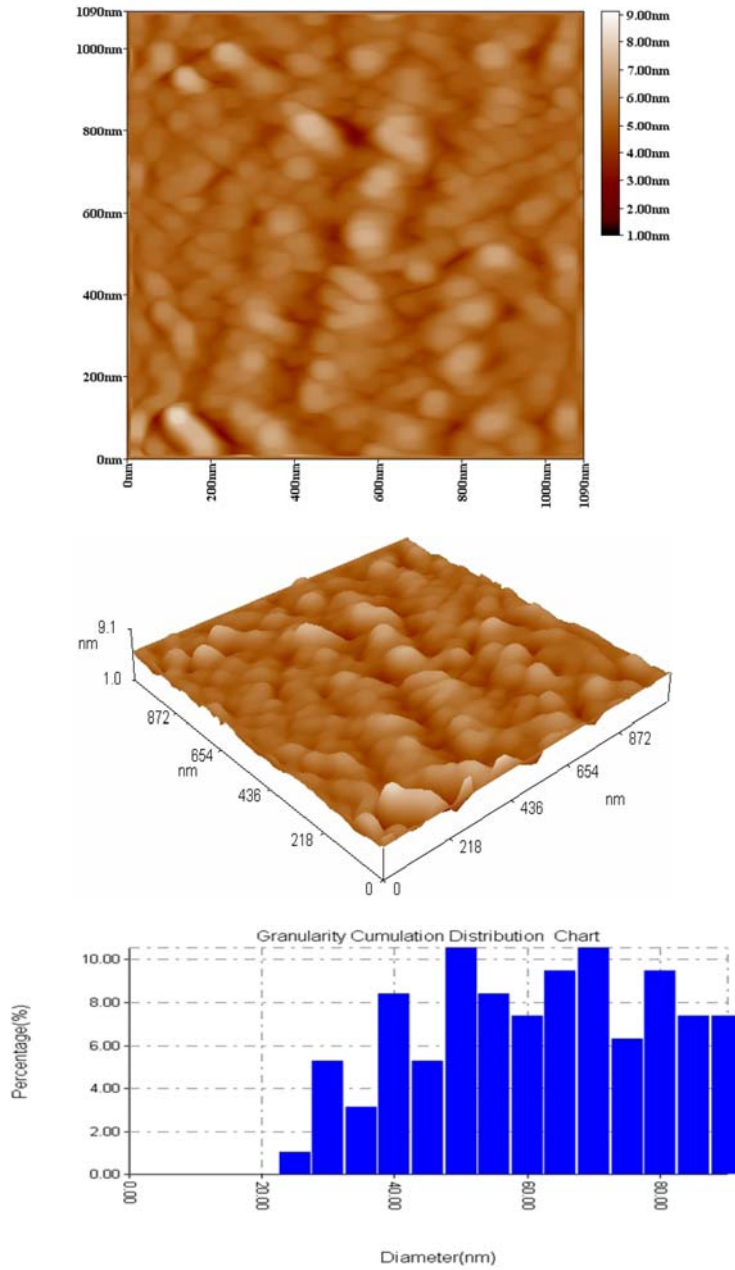
### 3. Results and Discussions

The structure of the  $\text{Cd}_2\text{SnO}_4$  thin films of thickness (400, 500)nm have been studied by X-ray diffraction, it is observed from this study that the structure for these films are polycrystalline of spinal cubic structure according to standard JCPDS, and a small amount of CdO may be responsible for the broad (400) reflection as shown in Figure 1, and the reflection surface (311), (222) have been appeared as shown in Figure 1, and this is in a good agreement with data for Eman et al. [14] and Jayaram et al. [15]. It is observed a preferred orientation growth along the (311), Xiaonan et al. [16] have found that the crystal structures is cubic and cubic spinal (CdO), also our results in agreement with Andrew et al. [17]. They found that the films are polycrystalline with cubic crystal structure, also it is observed that the intensity of the peaks increases with increasing film thickness, this is attributed to the increase in grain size of the films, for this, the grain size of the films as a function of thickness was determined by using Scherrer formula, and it is increases from 19.1 to 21.4nm with increasing film thickness from 300 to 600nm, respectively, and these values are agreement with Kumaravel et al. [12].



**Figure 1.** The XRD spectrum of thin  $\text{Cd}_2\text{SnO}_4$  films.

The surface morphology of  $\text{Cd}_2\text{SnO}_4$  thin films were studied by using AFM. Figure 2 shows the two and three-dimensional view of AFM micrographs of  $\text{Cd}_2\text{SnO}_4$  thin films at thickness of 600nm, respectively. The scanning is done over an area of  $1\mu\text{m} \times 1\mu\text{m}$ . The micrographs show that the surfaces of the  $\text{Cd}_2\text{SnO}_4$  thin films which consist of nanoscale particles are dense. Also, the studied reveal the uniform thickness of the films, and the presence of uniform grain growth in  $\text{Cd}_2\text{SnO}_4$  thin films. The maximum size of particles and the roughness of the surface of  $\text{Cd}_2\text{SnO}_4$  films of, 600nm thickness are calculated. The study reveals that films are smooth and uniform. The value of grain size is 59nm for thickness 600nm, and the films consisted of grains with size in the nanometer range which well matches with the results obtained from XRD, and these values are agreement with Jeyadheepan et al. [15, 18]. It is observed from roughness analysis that the root mean square value of surface roughness is 0.602nm at 600nm of thickness, the roughness is comparatively small. From the section analysis, it confirms the uniform grain growth in films. It has also been showed that the low surface roughness of the films is desired for solar cells and gas sensor applications. But, the low surface roughness is desired in solar cells, while rough surface is desired in gas sensor application in which rough surface has higher surface area. This is in a good agreement with data for Eman et al. [14, 15, 18]. Also Figure 2 shows the histogram of the percentage of  $\text{Cd}_2\text{SnO}_4$  films as a function of the grain size.



**Figure 2.** 2 and 3-D AFM images and the histogram of the percentage as a function of the grain size of  $\text{Cd}_2\text{SnO}_4$  films at thickness 600nm.



A basic reason why TCOs are of concern is that they can show transparency in a limited and well-defined range, normally encompassing visible light in the 400nm to 700nm wavelength interval. In the infrared (IR) region, their metallic property leads to reflectance and at sufficiently short wavelength, in the ultraviolet (UV), they become absorbing due to excitations across an energy gap [15]. Optical transmittance and absorption spectra of  $\text{Cd}_2\text{SnO}_4$  thin films, which are measured as a function of the wavelength of incident photons of two thicknesses are shown in Figures 3 and 4. In this spectrum, a distinct absorption peak at 398-400nm is found, it is quite clear that both thickness have excellent transparency in the visible range  $\geq 70\%$  transmission, the wavelength region from 450-1000nm shows very low absorption of incident light. We can see that increasing of thickness shifts the peak of transmittance spectrum to the red shift. The shift in the peak position films may be attributed to the crystallite of film structure by increasing the grain size, which is confirmed by XRD results. Also the spectrum of absorption has been studied as shown in the Figure 4, it is obviously that its behaviour is opposite to that of the transmittance spectrum. Also, it is observed that the transmittance decreases from 0.6 to 0.377 with increasing film thickness from 300 to 600nm, respectively, whereas the absorbance increases from 0.251 to 0.419 with increasing film thickness from 300 to 600nm, respectively, and the reflectance increases from 0.148 to 0.203 with increasing film thickness from 300 to 600nm, respectively. This is due to crystallization of film structure by increasing the grain size and slightly, the absorption edge shift to higher wavelength.

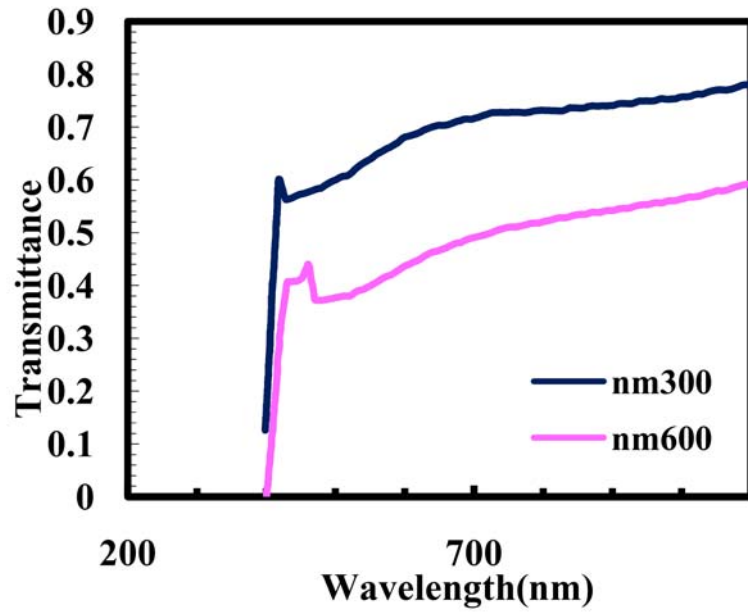


Figure 3. The transmittance spectrum of Cd<sub>2</sub>SnO<sub>4</sub> thin films.

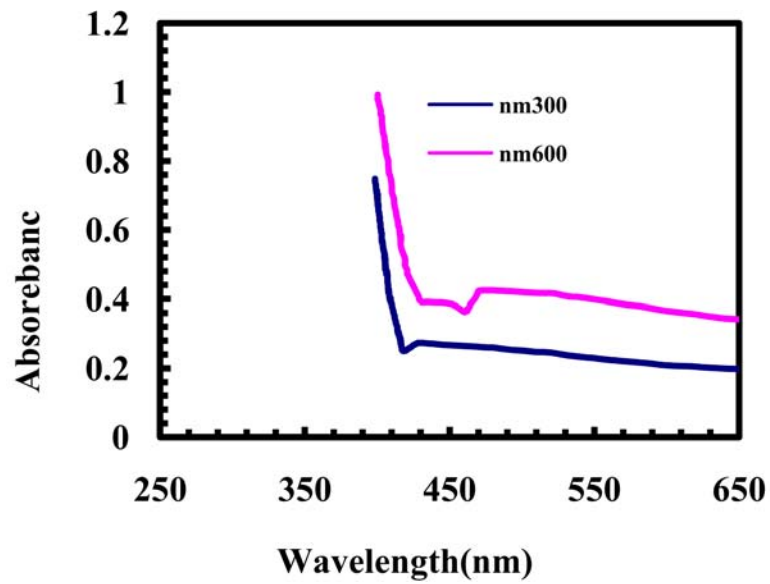
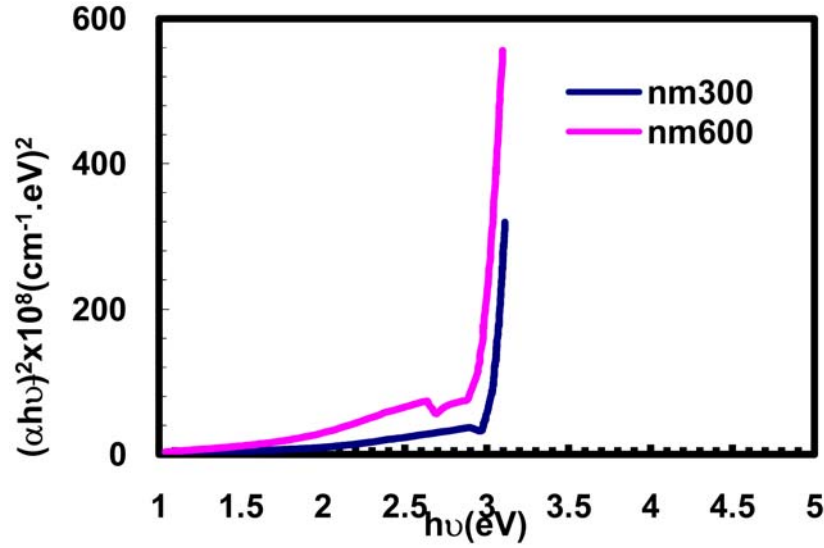


Figure 4. The absorption spectrum of Cd<sub>2</sub>SnO<sub>4</sub> thin films.

The optical constants of  $\text{Cd}_2\text{SnO}_4$  thin film at both thickness have been calculated, absorption coefficient ( $\alpha$ ) associated with the strong absorption region of the films was calculated from absorbance ( $A$ ) and the film thickness ( $t$ ),  $\alpha$  increases from  $(1.92 \text{ to } 3.22) \times 10^4 \text{ cm}^{-1}$  with increasing film thickness from 300 to 600nm, respectively. This is due to the decreasing value of energy gap with increasing of thickness.

From Figure 5, the energy gap is determined by plotting Tauc equation and taking the extrapolation of the linear portion, where ( $\alpha \geq 10^4 \text{ cm}^{-1}$ ) of the  $(\alpha h\nu)^2$  as a function of  $h\nu$  curve to  $\alpha = 0$ . We found that the last relation yielded a linear dependence, which describes allowed direct transitions. The direct energy gap value was found to be as shown in Figure 5, which nearly in agreement with the other literatures [10-18]. The value of optical energy gap decreases from 3.00 to 2.89eV with increasing film thickness from 300 to 600nm, respectively. The observed decrease in the band gap energy with increases in thickness is due to the changes in the barrier height to the size of the grain in crystalline film and large density of dislocation. This is in a good agreement with data for Jayaram et al. [15-19]. The optical behaviour of materials is generally utilized to determine its optical constants, for example, the refractive index ( $n$ ). The refractive index of  $\text{Cd}_2\text{SnO}_4$  films increases from 1.756 to 2.142 with increasing film thickness from 300 to 600nm, respectively. This behaviour is due to increase in the reflection which the refractive index depend on it. Measured refractive index is in good accordance with other author results ( $n = 1, 9, 2$ ) Dudonis et al. [13]. The extinction coefficient ( $k_e$ ) is directly related to the absorption of light. In the case of polycrystalline films, extra absorption of light occurs at the grain boundaries. This leads to non-zero value of ( $k_e$ ) for photon energies smaller than the fundamental absorption edge. It is increases from 0.076 to 0.128 with increasing film thickness from 300 to 600nm, respectively. This attributed to the same reason, which mention

previously in absorption coefficient. The values of real ( $\epsilon_1$ ) and imaginary ( $\epsilon_2$ ) dielectric constants with different thickness were calculated. The behaviour of  $\epsilon_1$  is similar to refractive index because the smaller value of  $k_e$  comparison of  $n$ , while  $\epsilon_2$  is mainly depends on the  $k_e$  values, which are related to the variation of absorption coefficient. It is found that  $\epsilon_1$  increases from 3.08 to 4.57 with increasing film thickness from 300 to 600nm, respectively, while  $\epsilon_2$  increases from 0.269 to 0.55 with increasing film thickness from 300 to 600nm, respectively.



**Figure 5.** The  $(\alpha h\nu)^2$  as a function of photon energy of  $\text{Cd}_2\text{SnO}_4$  thin films.

The major investigation on a transport conductive oxide material is its metal like resistivity and good electronic conduction. To investigate the electrical behaviour of TCOs, Hall measurements and DC conductivity have been measured. The resistivity and conductivity, activation energy and carrier concentration, mobility and drift velocity, are reported, of  $\text{Cd}_2\text{SnO}_4$  with increasing film thicknesses from 300 to 600nm, respectively.

The activation energy is calculated, and it decreases from 0.144eV to 0.106eV with increasing film thickness from 300 to 600nm, respectively, and these values are in agreement with Jayaram et al. [15]. Both films show increasing conductivity from 8.69 to 11.62(ohm.cm)<sup>-1</sup> with increasing film thicknesses from 300 to 600nm, respectively. Both films show an increase in conductivity with increasing temperatures which confirms the semi-conducting behaviour of both thin films, this is in a good agreement with Jayaram et al. [15]. Our Hall effect measurements showed that the films are *n*-type and low resistivity ( $8 \times 10^{-2}$ ohm.cm), and this value is in agreement with Hani et al. [21], the carrier concentration increases from  $0.140 \times 10^{19}$  to  $0.29 \times 10^{19}$ cm<sup>-3</sup> with increasing film thickness from 300 to 600nm, respectively, these values are in a good agreement with Dou et al. [3, 4]. Jeyadheepan et al. [18] and Hani et al. [21], noticed from these measurements that the mobility and drift velocity, respectively, decrease from 38.763 to 24.621cm<sup>2</sup>·V<sup>-1</sup>·sec<sup>-1</sup> with increasing film thickness from 300 to 600nm, respectively, Xiaonan et al. [16] and Krishnakumar et al. [1] found that the mobility was about 40 cm<sup>2</sup>·V<sup>-1</sup>·sec<sup>-1</sup> and the carrier concentration can be as high as  $9 \times 10^{19}$ cm<sup>-3</sup>. And Walter et al. [19] found that the mobility is 30 cm<sup>2</sup>·V<sup>-1</sup>·sec<sup>-1</sup> and carrier concentration is  $10^{20}$ cm<sup>-3</sup>, and Wu et al. [20] found that the mobility is 65 cm<sup>2</sup>·V<sup>-1</sup>·sec<sup>-1</sup> and carrier concentration is  $2 \times 10^{20}$ cm<sup>-3</sup>, also the drift velocity decreases from 387 to 246cm·sec<sup>-1</sup> with increasing film thickness from 300 to 600nm, respectively.

#### 4. Conclusion

Highly conductive and transparent Cd<sub>2</sub>SnO<sub>4</sub> conducting oxides thin films of two thicknesses have been prepared by thermal evaporation method. They are more-conductive, more transparent, have a lower surface roughness, are patternable, and are exceptionally stable. The

result of X-ray diffraction shows that the structure is polycrystalline with cubic structure, and the grain size increases with increasing film thickness. The surface morphological studies show that the sizes of the particles increase along with thickness of the film by the means of AFM and have low roughness. Crystallinity and grain size of the films Crystallinity and grain size of the films were found to increase with film thickness were found to increase with film thickness. The transmittance decreases with film thickness. On the basis of experimental results, we may conclude that band gap of thin films is also thickness dependent. The optical transition in  $\text{Cd}_2\text{SnO}_4$  film is direct transition and the value of optical energy gap decreases with increasing of thickness. The Hall effect measurement shows that for a given set of deposition conditions, the thicker films have better electrical properties The electrical resistivity and therefore activation energy are observed to be thickness dependent. The Hall effect measurements confirm the *n*-type nature of  $\text{Cd}_2\text{SnO}_4$  thin films. Also, the conductivity and charge carrier concentration increases with increasing thickness, whereas, the Hall mobility and drift velocity decreased with increasing of film thickness.

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