International Journal of
Applied Mathematics,
Electronics and Computers
IJAMEC
ISSN: 2147-8228 www.atscience.org/IJAMEC
Advanced Technology and Science
Original Research Paper

Influence of Thickness on Electrical and Optical Properties of Tellurium Thin Films Deposited by Chemical Spray Pyrolysis

Janan H. Saadie *[1], Abdulkhadhim A. Hasan[2]

Accepted 15th August 2014

Abstract: In this paper, Spray pyrolysis was applied to deposit Tellurium thin film on glass substrates at 498 ± 283 K in order to investigate the influence of thickness on the electrical and optical properties of the prepared thin films. The film thicknesses were measured *in situ* by weighting method. The thicknesses were from (150 to 800) nm. X-ray diffraction results showed that the Te thin films formed were polycrystalline with hexagonal structure. The effect of film thickness on the electrical properties of the films was investigated by Hall Effect measurement through the analysis of the I-V plots. The conductivity of the films and carrier concentration were highly increased when the films became thicker. Transmission and absorption spectrum of the prepared thin film had been recorded using UV-VIS-NIR spectrophotometer in the photon wavelength range of 300 - 2000 nm. The values of some important optical parameters of the studied films (absorption coefficient, and optical band gap energy) were determined using these spectra. The transmittance showed better results when thicknesses were being increased. In addition, the absorption coefficient is higher for thinner films and decreases for thicker films showing little dependence for absorption coefficient on thin film thickness in infra-red region of the wavelength. It was found from the optical properties studies that the type of transition of Te film is direct transition. Also, the optical energy band gap was evaluated for different thicknesses. The results have shown that the optical energy band gap was increased by the increase of thickness. Experiments and measurement results are presented.

Keywords: *Tellurium, Thin film, Electrical properties, Optical properties, Hall Effect, Spray pyrolysis*.

1. Introduction

Due to their structural, electrical and optical properties, Tellurium thin films are widely used in various technological areas, especially in microelectronic devices such as gas sensor [1-3], optical information storage [4] and other applications [5-7]. Recently researches have shown that Tellurium films may be used for detection of harmful gases at room temperature such as NO_2, CO, NH_3, and H_2S [8], selective surface devices among them [9], IR detectors, strain sensitive devices and thin film transistors. All these applications are due to remarkable physical properties of Te such as low band-gap and transparency in the infrared region [10-11].

The electrical conductivity studies are aimed at understanding the origin of the charge carrying species and the way in which they move through the thin films and its type. Also, knowledge of electrical properties of thin films is helpful in material study and characterization for device fabrication. Moreover, knowledge of optical constants of a material such as optical band gap, refractive index and extinction coefficient is quite essential to examine material's potential optoelectronic applications [12]. Further, the optical properties may also be closely related to the material's atomic structure and electronic band structure. On the other hand, the optical behaviour of materials is important to determine it's usage in optoelectronic devices. The electrical properties of semiconductors are strongly dependent upon the band gap [13]. Hence, accurate characterizations of electrical and optical properties are essential in thin films applications.

Various researches have characterized Tellurium films prepared by evaporation method [14-15]. Chemical spray deposition is a low cost and convenient method has recently been utilized to prepare thin polycrystalline films of a wide variety of compound semiconductors by a number of investigators among them [16][Recently, Tellurium thin film has been prepared by chemical spray-pyrolysis [17]. Also, both electrical and optical properties of the prepared thin film have been investigated [10]. However obtaining Te films with superior electrical and optical properties suitable for device applications is still a technological challenge. Many researchers have shown that the film thickness has a strong influence on the optical absorption among them [18]. In the present work, we report the effect of thickness on electrical and optical properties in the wavelength range from (300 to 2000) nm of Te thin films deposited on glass substrate by chemical spray pyrolysis. Optical process or absorption method was implied to determine the band gap energy. Also, electrical resistivity and conductivity were determined. The experiment and calculations results are presented and the most important factors are discussed.

2. Experimental and Measurement Techniques

2.1. Film Deposition Method

Spray pyrolysis is a processing technique being considered in research to prepare thin and thick films. Unlike many other film depositions, spray pyrolysis represents a very simple and relatively cost-effective method (especially with regard to equipment costs). It offers an extremely easy technique for preparing films of any

---

[1] *Material Engineering Department, College of Engineering, Kufa University, Al-Najaf /Iraq*
[2] *Electronics and Communications Engineering Department, College of Engineering, Kufa University, Al-Najaf /Iraq*
* *Corresponding Author: Email: jenan.saadi@uokufa.edu.iq*

Note: This paper has been presented at the International Conference on Advanced Technology & Sciences (ICAT'14) held in Antalya (Turkey), August 12-15, 2014.

composition. The method has been employed for the deposition of dense films, porous films, and for powder production. Even multilayered films can be easily prepared using this versatile technique. In this paper spray pyrolysis method is suggested to fabricate Tellurium thin films in order to investigate the effect of film thickness on both electrical and optical properties. Typical spray pyrolysis set up used in this work to prepare the thin films under study consists of glass nozzle, compressor carrier gas, substrate heater, and temperature controller. The general schematic of a spray pyrolysis deposition process is shown in Fig. 1.

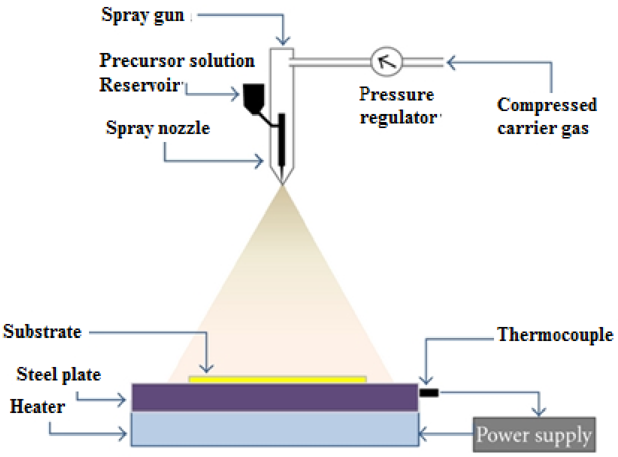

Figure 1. The schematic of the spray pyrolysis setup

A series of experiments were conducted to prepare samples of Tellurium thin films with different thicknesses using the schematic shown in Fig. 1. The thicknesses of Te thin films prepared for studying electrical and optical properties are in the range of (150-800) nm.

2.2. Thin Films Preparation

The thin films of Te with different thicknesses used for electrical and optical properties measurement are prepared by spray pyrolysis technique utilizing the set up shown in Fig. 1. The spray pyrolysis technique consists of spraying a solution containing a soluble salt of Tellurium onto a heated substrate. For Tellurium thin film preparation, A Tellurium dioxide TeO2 with molecular weight of 159.6 gm/mol is dissolved in Hydrochloric acid (HCl) with concentration of 37%. The acid was added and mixed with 0.399 gm from TeO_2 in order to prepare solution with 0.1 M after that distillation water was added to obtain solution of Tellurium salt according to the following chemical reaction equation [19]

$$TeO_2 + 4HCl = TeCl_4 + 2H_2O \qquad (1)$$

and

$$TeCl_4 = Te + 2Cl_2 \qquad \text{at } 498 \pm 283 \text{ K} \qquad (2)$$

This solution was sprayed onto a heated glass substrate using compressed air as a carrier gas. The glass substrate rested on steel surface which was heated indirectly by an electrical resistance heater. The steel surface temperature was monitored with thermocouple to provide a reference temperature. The exact temperature of the substrate surface was unknown and varied as a function of the heater temperature, the spray rate and the surface coverage. The substrate was Borosilicate with dimensions of 26 mm x 20 mm and thickness of 0.4 mm. The reason for using small thickness of substrate glass in our work is to prevent producing a big difference between the temperature of the upper and lower surfaces of the glass and providing a uniform temperature. The substrate temperature during deposition approximately 498± 283 K depending on spray rate. The time of one spray is 1-2 seconds and stop time for another spray is 30 seconds with gas flow rate of 35 L/min to yield thin films of thicknesses in the range of (150-800) nm. For measurement of Hall Effect, it is necessary to deposit four contacts in form of Ag strips made on the surface of the film by vacuum evaporation method. The strips dimensions are 4 mm in length, 4 mm in width, 0.4 µm in thickness and separated by 1 cm. The I-V measurements were taken by using a Keithley Source Meter.

Film thickness (t) in our study is an important parameter in the study of film electrical and optical properties. The thicknesses of prepared Te films were measured using weighting method which depends on the weight difference of the substrate before and after deposition processes of thin film. The weight difference is employed by a sensitive electronic microbalance, in this method the substrate was weighted before the deposition (m_1) and after the deposition (m_2) and the thickness of the films was obtained using (3)

$$t = (m_2 - m_1)/ \rho A \qquad (3)$$

where ρ is the density of the film material (g/cm^2) and (A) is the area of the film (in cm^2).

3. Results and Discussion

In order to get a better insight into the nature of the prepared film in the present work, X-ray diffraction study was made to determine its structure and to identify the components and phases. The X-ray diffraction pattern of sprayed Te is shown in Fig. 2. It is clear from the figure that the spectrum of Te exhibits sharp peaks at 2θ equal to 23^o, 27.5^o, 38.1^o, 40.4^o, 47^o, 49.6^o, 56.9^o which correspond to diffraction from (100), (101), (102), (110), (200), (201), and (202) planes of the hexagonal Te phase respectively . Both peaks highest and peaks position are in good agreement with ASTM X-ray powder file data for hexagonal Te (4-0554)[20]. Also, Fig. 2 shows that the film was polycrystalline in nature and only one phase was indicated in the film.

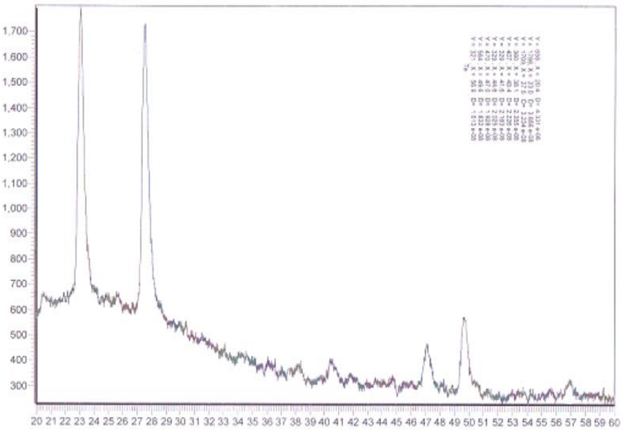

Figure 2. X-Ray diffraction of the prepared thin film.

The topographical properties of the film was investigated by optical reflection (Leitz-Metallux3) with magnification factor of 200 and shown in Fig. 3 (a, b, c) for thicknesses of 150 nm, 300 nm, and 500 nm respectively. The process of deposition of Te showed that the film was silver coloured, Also from Fig. 3, several droplets are shown including overlapping of the disk shapes on the surface as they are being deposited. The lighter surface of Te thin film as shown in Fig.3 (a) is for the thinner thin film (150 nm). This is due to that the droplet number density on the surface is not sufficient to ensure the formation of a thin film as compared to that shown in Fig. 3 (b) with thickness 300 nm, where it can be seen a good homogeneity and lower roughness. This is because that the individual droplets may spread uniformly on the surface of the substrate and form lamellae that may merge to form an integrated thin film. For Te thin film with thickness

(500 nm). a stacked-coin pattern with darker disks is created as shown in Fig 3 (c). Experimental results obtained here, however, suggest that Tellurium film was with good homogeneity and lower roughness for thin films with moderate thicknesses.

In order to study the electrical properties and evaluate the influence of Te thin film thickness variation on different electrical parameters, a series of experiments were conducted to achieve this purpose. Hall Effect measurements were also performed to determine the type of electrical conductivity of the prepared thin films and the variation of carrier concentration and conductivity with the film thickness. The variation in Hall voltage with the current is shown in Fig. 4 (a, b, c, and d) for thicknesses of 200, 400, 600, and 800 nm respectively. It is clear from the figure that the film has electrical conductivity of P type. Also

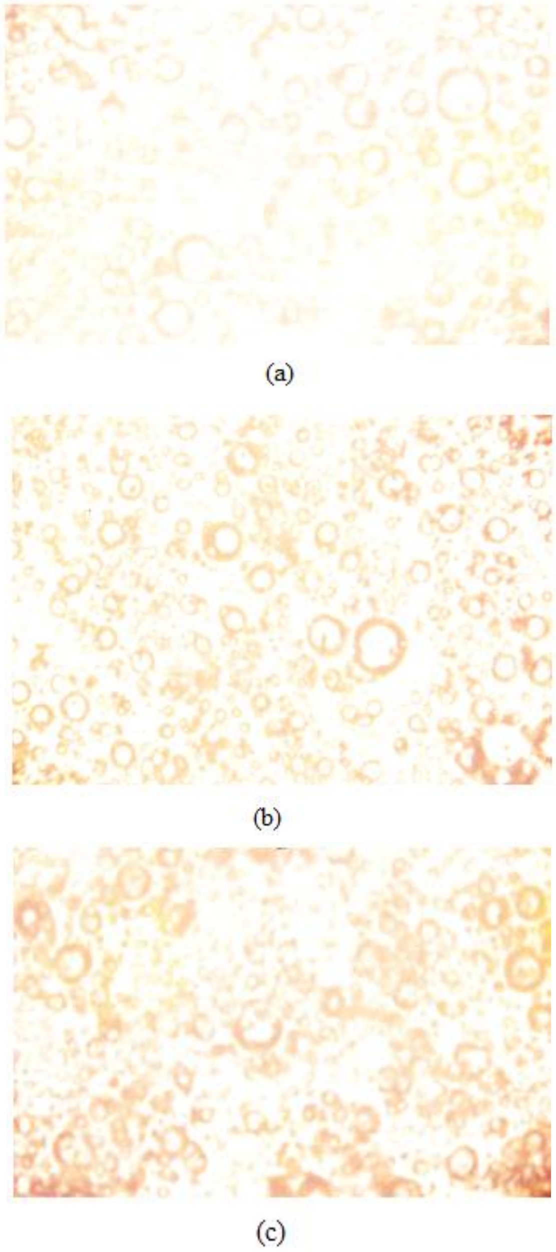

Figure 3. The topographical surface of the prepared thin films for different thicknesses (with magnification factor of 200).

(a) t = 150 nm (b) t = 300 nm (c) t = 500 nm

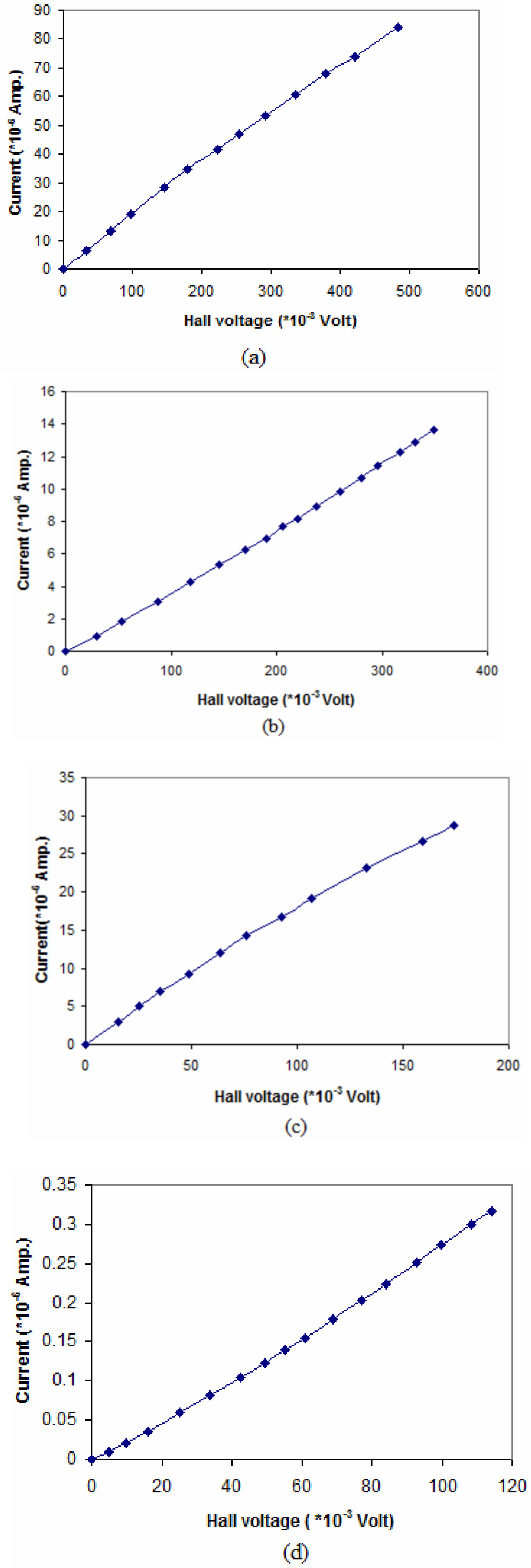

Figure 4. Variation in Hall voltage of Tellurium thin film with current for different thicknesses (a) t = 200 nm (b) t = 400 nm (c) t = 600 nm and (d) t = 800 nm

it can be observed from the figure that the increasing in the current passing through the film causes increasing in the induced voltage in the presence of normal magnetic field for the given thicknesses. Table I shows variations of carrier concentration, conductivity, and resistivity with thickness of the prepared films. It is clear from the table that the carrier concentration increases with increasing of film thickness. It can also be observed that there is a clear increasing of conductivity values with increasing film thickness. This change of the electrical conductivity values may be attributed mainly to three different contribution due to increase of the grain size and crystalline degree with increasing of thickness [21] as shown in Fig. 5 which shows variation of film conductivity with film thickness. It is clear that the conductivity slightly increases for thickness from (200-400) nm and it abruptly increases for large thicknesses from (400-800) nm. We believed that such difference in conductivity value of the film could be associated mainly with the change of carrier concentration as shown in Fig. 6. These results are in good agreement with those obtained by [18].

From the results of experiments and measurements, the optical properties of the prepared thin films were investigated and graphs for various thicknesses are plotted for this purpose. Fig. 7 shows the variation of the transmission (in percentage) with the wavelength. It is clear from the figure that the transmission in percentage increases with decreasing film thickness. The film with thickness of 150 nm displays a high transmittance value of 60 % in the infra-red region as compared to that displayed by the other two film thicknesses of 300 and 500 nm respectively. This is due to increasing of the scattering and attenuation effects with increasing thin film thickness. Fig. 8 shows plot of calculated values of absorption coefficient (α) versus the wavelength for different thicknesses of the prepared thin film. As shown in the figure, thin film has high absorption coefficient at low values of wavelength and decreases as the wavelength increases this is because of high transparency of thin film at these wavelengths. Also it can be seen from the figure that the absorption coefficient is higher for thinner films and decreases for thicker films showing little dependence for absorption coefficient on thin film thickness in infra-red region of the wavelength.

Table 1. Comparison of the measured electrical parameters for different thicknesses of the prepared thin films

Thickness (t) nm	Carrier concentration (P) cm^{-3}	Conductivity σ (Ω.cm)$^{-1}$	Resistivity ρ (Ω.cm)
200	2.57×10^{14}	6.2×10^{-3}	161
400	9.7×10^{15}	9.9×10^{-3}	101
600	3.6×10^{17}	4.2×10^{-2}	23.5
800	1.47×10^{19}	8×10^{-2}	12

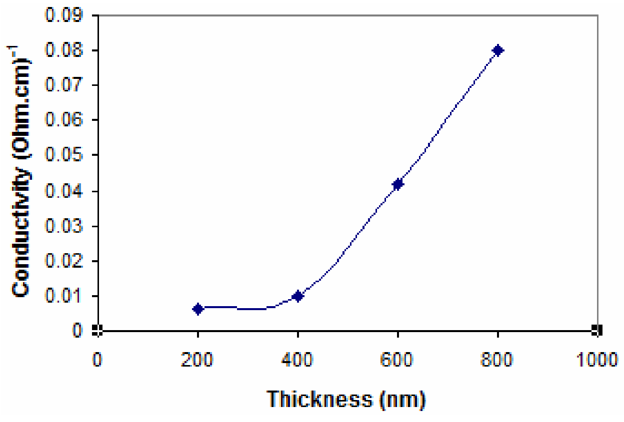

Figure 5. Variation of conductivity vs. thin film thickness.

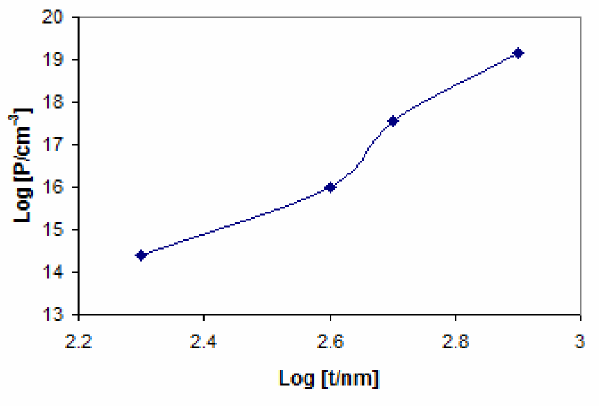

Figure 6. Variation of log of carrier concentration vs. log of thin film thicknesses

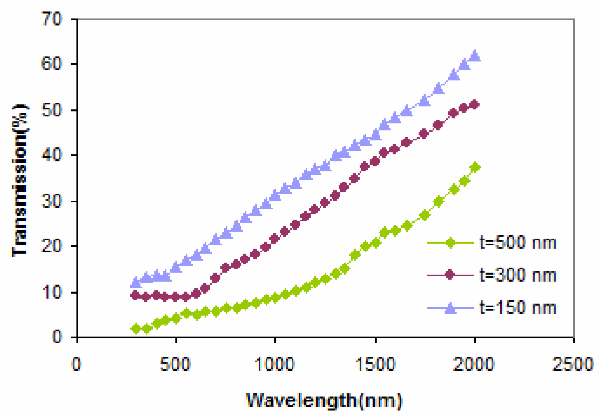

Figure 7. Variation of Transmission (%) vs. wavelength

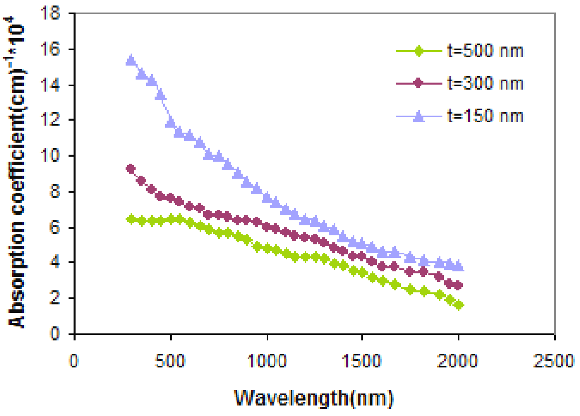

Figure 8. Variation of absorption coefficient vs. wavelength

In order to calculate the energy band gap of the prepared films, we plotted the values of $(\alpha h\nu)^2$ versus photon energy ($h\nu$) as shown in Fig. 9. It is clear from the figure that a linear variation at the short wavelengths (high photon energy) is obtained. This indicates obtaining a direct energy band gap by plotting extra straight line as illustrated in the figure. The energy band gaps for the prepared thin films are obtained from the intersection of the drawn lines of the indicated thicknesses with photon energy axis. These energy band gaps are found to be 0.68, 0.72, and 0.75 ev for thin film thicknesses of 150, 300, 500 nm respectively. From the figure, it is clearly observed that the value of energy band gap (Eg) increases with increasing thickness and have been attributed to the decrease of localized states in the band gap. The behaviour of variation of band gaps with thicknesses of the prepared thin films is similar to results reported by [13].

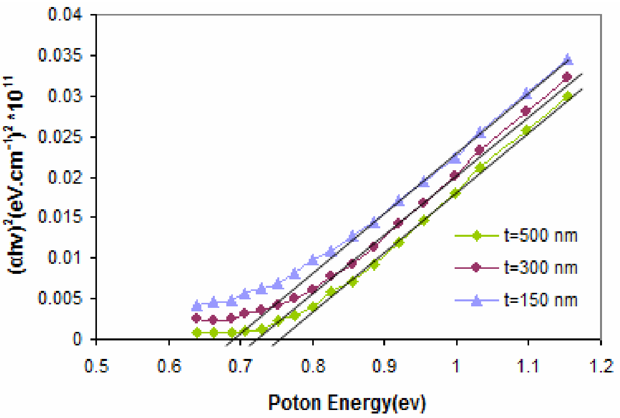

Figure 9. Variation of $(\alpha h\nu)^2$ vs. photon energy for different thicknesses

Conclusion

In this work, Tellurium thin film was prepared by using chemical spray pyrolysis to investigate the influence of variation of thin film thickness on the topographical surface of the prepared thin films and film characterization (both electrical and optical properties). Results of topographical surface have shown that films with moderate thickness resulting films with good homogeneity and lower roughness. With the aid of the X-Ray diffraction we observed that the prepared film has polycrystalline structure with peaks for Tellurium with hexagonal structure. Various parameters related to electrical and optical properties were calculated for different thicknesses of Te thin film. It is observed that the electrical conductivity values increase as the thickness of the films increases. This increase of the electrical conductivity with film thickness could be attributed to the lesser relative contribution of the carrier scattering at the film surface and grain boundaries. Also, the carrier concentration increases as thin films increases and Hall's effect showed that the prepared films were of P-type. Experiments results have shown that the transmission increases with decreasing thickness of thin film, which produces absorbance in opposite behaviour. In addition, the absorption coefficient increases with decreasing thin film thickness and the thickness of the thin has little influence on absorption coefficient in infrared region. It is found that the type of transition of Te is direct transition. Moreover, the band gap of the film increases with the increase of thickness. The obtained results give possibility to use the prepared film in manufacturing near IR detectors and selective surface devices. Also, such film may have application in solar energy collector.

References

[1] Tsiulyanu Tsiulyanu D, Marian S, Liess HD. Effect of annealing and temperature on the NO_2 sensing properties of tellurium based films, Sensors and Actuators B.100 (3): 2004, pp. 380-386

[2] Shashwati S, Muthe KP, Joshi N. Room temperature operating ammonia sensor based on tellurium thin films, Sensors and Actuators B. 98(2−3): 2004, pp. 154−159.

[3] Ramesh A .N. KambhampatiTsiulyanu D, Marian S, Miron V. High sensitive tellurium based NO_2 gas sensor. Sensors and Actuators” B.73 (1): 2001, pp. 35-42.

[4] Josef P, Dana P, Jaroslav B, et all. Nano-structured crystalline Te films by laser gas-phase pyrolysis of dimethyl tellurium, Anal Appl Pyrolysis. 71(2): 2004, pp. 739−746.

[5] Lozzi L, Santucci S, DI Nardo S. XPS, LEED and AFM investigation of the Si (100) surface after the deposition and annealing of tellurium thin films, Surface Science, 1996, pp. 352−354.

[6] Lozzi L, DI Nardo S, Passacantando M. Electron spectroscopy investigation of Te thin films deposited at room temperature on Si(100), Surface Science 569(1), 1995, pp. 331−333.

[7] Lozzi L, DI Nardo S, Passacantando M. Growth of Te thin films deposited at room temperature on the Si (100) 2×1 surface, J. of Electron Spectroscopy and Related Phen. 71(1), 1995, pp. 39-45.

[8] Tsiulyanu D. , Mocreac O. Impedance Spectroscopy of Sensitive to Harmful Gases Tellurium Thin Films, Journal of Non-Oxide Glasses, 3,2, 2011, pp. 43-50.

[9] Peterson M. J, Cocks F. H. Tellurium Selective Absorber Surfaces" Mater. Sci. & Engineering 41, 1979, pp. 143-147.

[10] Janan H. S., Optical Properties of Tellurium Thin Film Prepared by Chemical Spray Pyrolysis Method, Journal of Kufa- Physics 32, 2011, pp. 25-29.

[11] Mahdi H.S., Souad G.K. Marwa R. F. Structural and electrical properties of tellurium thin films prepared by vacuum thermal deposition, Iraqi Journal of Physics, 10, ,17, 2012, PP. 7-11.

[12] Chitra D. Jahanara B, Tahmina B. and Shamima C. Efeect of Thickness on the Optical Properties of GaAs Thin Films, Journal of Bangladesh Academy of Science, 37,1, 2013, pp. 83-91

[13] Dinesh C., Rajendra K. and Ram M. Influence of Thickness on Optical Properties of As_2Se_3 Thin Films” Turk J Phys., 30, 2006, pp. 519 – 527.

[14] Rusu M. On the Electrical and Optical Properties of the Stratified Tellurium Thin Film" Journal of Optoelectronics and Advanced Materials, 3, 4, 2001, pp. 867-872.

[15] Balasubram T. Narayandass K. Mangalaraj D. Electrical Properties of Thermally Evaporated Tellurium Thin Films, Bulletin of Materials Science. 20(1), 1997, pp 79-92.

[16] Celalettin Baykul M., Nilgun O. Band alignment of $Cd_{(1-x)}Zn_xS$ produced by spray pyrolysis method, Thin Solid Films 518, 2010, pp. 1925–1928.

[17] Janan H. S. Chemical Spray Deposition for Tellurium Thin Film” Journal of Babylon University-Engineering Sciences, 21, 3: 2013, pp. 981-986.

[18] Giraldi, T.R., Escote M.T. et. Effect of Thickness on the Electrical and Optical Properties of Sb Doped SnO_2 (ATO) Thin Films, Journal of Electroceramics, 13, 2004, pp.159–165.

[19] Encyclopedia of chemical technology Tellurium and

Tellurium Compounds, 22, pp. 658-673, John Wiley& Sons Inc., New York, 1980.

[20] Powder Diffraction File, Joint Committee on Powder Diffraction Standards, International Centre for Diffraction Data, Swarthmore, Pa., File (4-0554).

[21] Shadia J. Riyad N. Effect of Film thickness on the Electrical and Structural Properties of CdS: In Thin Films, American Journal of Applied Sciences 5(9), 2008, pp 1141-1143.

.