American Journal of Materials Science and Engineering, 2013, Vol. 1, No. 3, 46-49
Available online at http://pubs.sciepub.com/ajmse/1/3/3

DOI:10.12691/ajmse-1-3-3

Structural and Optical Investigations of Amorphous $Se_{75-x}Te_{25}Sb_x$ Thin Films

D. K. Dwivedi[1,*], H. P. Pathak[1], R. K. Shukla[2], A. Kumar[2]

[1]Department of Physics, Amorphous Semiconductor Research Lab, M.M.M. Engineering College, Gorakhpur, India
[2]Department of Physics, H.B.T.I., Kanpur, India
*Corresponding author: dwivedidkphys@rediffmail.com

Received July 29, 2013; Revised September 02, 2013; Accepted September 04, 2013

Abstract Thin films of $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) glassy alloys have been deposited onto a chemically cleaned glass substrate by thermal evaporation technique under vacuum. Glassy nature of the films has been ascertained by X-ray diffraction pattern. The analysis by absorption spectra, measured at normal incidence, in the spectral range 400-1100 nm has been used for the optical characterization of thin films under investigation. The optical constants (absorption coefficient (α), extinction coefficient (k)) and optical band gap (E_g) have been studied. It has been found that extinction coefficient (k) decreases with increase in wavelength (λ). The absorption coefficient (α) is found to increase with incident photon energy. Optical band gap (E_g) has also been calculated and found to decrease with Sb content in $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) glassy system. The decrease of optical band gap (E_g) with Sb concentration has been explained on the basis of Mott and Davis model.

Keywords: *chalcogenide glasses, amorphous semiconductors, thin films, optical properties, optical band gap*

Cite This Article: D. K. Dwivedi, H. P. Pathak, R. K. Shukla, and A. Kumar, "Structural and Optical Investigations of Amorphous $Se_{75-x}Te_{25}Sb_x$ Thin Films." *American Journal of Materials Science and Engineering* 1, no. 3 (2013): 46-49. doi: 10.12691/ajmse-1-3-3.

1. Introduction

Chalcogenide glasses have recently attracted great deal of interest because of their potential applications in solid state devices [1-9]. These materials in particular Se glasses exhibit unique properties of reversible transformation [10]. This property makes these glasses very useful in optical memory devices [11,12,13]. Se based chalcogenide glasses have high transparency in the broad middle and for IR region and have strong nonlinear properties [14]. Apart from this application, amorphous selenium has been found to have tremendous potential in xeroxing applications and therefore a lot of attempts have been made to improve its properties by alloying [15,16]. Though amorphous selenium have got various device applications such as rectifiers, photocells, vidicons, xerography, switching and memory etc which make it attractive but pure selenium has disadvantages like short life time and low sensitivity. This problem can be overcome by alloying Se with some impurity atoms, which gives higher sensitivity, higher crystallization temperature and smaller aging effect [17,18,19]. In the present work Sb has been chosen as an additive element in amorphous Se-Te alloys. The third element behaves as chemical modifier as it is reported to expand the glass forming region and also creates compositional as well as configuratioanal disorder in the material with respect to binary alloys which will be useful in understanding the structural, electrical and optical properties of chalcogenide glasses [20-30]. The incorporation of a third element like Sb in Se-Te binary alloy is expected to change the optical and electrical properties of host alloy, which play a major role in device preparation. It has been reported that effect of alloying Sb with Se drastically improvers the thermal stability of Se [31]. The optical constants (absorption coefficient (α), extinction coefficient (k)) as well as band gap (E_g) are the most significant parameters in the amorphous semiconducting thin films. The optical behavior of a material is utilized to determine its optical constant. Therefore an accurate measurement of optical constants is extremely important. The aim of present paper is to study the effect of Sb incorporation on the optical properties of Se-Te matrix. The optical absorption spectra of the films of $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) are measured in the wavelength range 400-1100 nm, by double beam UV-VIS spectrophotometer. Optical parameters like absorption coefficient (α), extinction coefficient (k) and optical band gap (E_g) have been calculated for a-$Se_{75-x}Te_{25}Sb_x$ glassy system.

2. Experimental

Glassy alloys of $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) were prepared by applying melt quenching technique. The exact proportions of high purity (99.999%) Se, Te and Sb elements, in accordance with their atomic percentages, were weighed using an electronic balance (LIBROR, AEG-120) with the least count of 10^{-4} gm. The material was then sealed in evacuated (~10^{-5} Torr) quartz ampoule (length ~ 5cm and internal diameter ~ 8mm). The ampoule containing material was heated to 800°C and was held at that temperature for 12 hours. The temperature of the furnace was raised slowly at a rate of 3 - 4°C/minute. During heating, the ampoule was constantly rocked, by rotating a ceramic rod to which the ampoule was tucked away in the furnace. This was done to obtain homogeneous glassy alloy. After rocking for about 12 hours, the obtained melt was rapidly quenched in ice-cooled water. The quenched sample was then taken out by breaking the quartz ampoule. Thin films of glassy alloys of a $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) were prepared by vacuum evaporation technique, in which the substrate was kept at room temperature at a base pressure of 10^{-6} Torr using a molybdenum boat. The films were kept inside the deposition chamber for 24 hours to achieve the metastable equilibrium as suggested by Abkowitz [32]. The thickness of the film was measured using a single crystal thickness monitor. The XRD patterns of the films were recorded with the help of x-ray diffractometer (XPERT-PRO) using Cu-Kα radiation (λ=1.54Å).The tube was operated at 45 kV and 35 mA. The normal incidence absorption spectra of $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) thin films have been taken by a double beam UV-VIS-NIR computer controlled spectrophotometer (ECIL-Hyderabad, India, Model No.5704 SS) in the wave length range 400-1100nm.

3. Results and Discussion

3.1. Structural Property

The XRD pattern for $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) has been shown in Figure 1. The X-ray diffraction patterns indicate that the films are amorphous in nature.

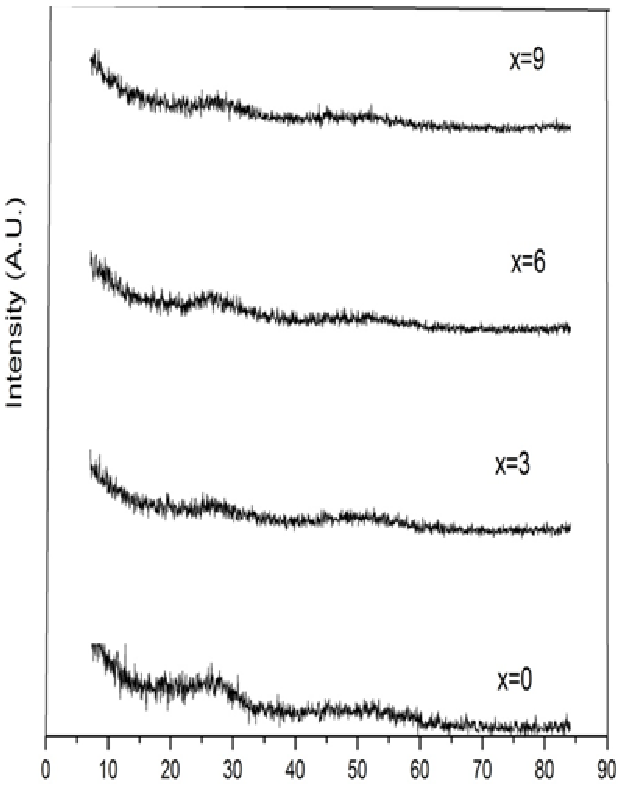

Figure 1. XRD pattern of Se75-xTe25Sbx (x = 0, 3, 6, & 9) thin films

3.2 Optical Properties

3.2.1. Absorption Coefficient (α) and Optical Band Gap (E_g)

The absorption coefficient (α) of the film was calculated using the equation (1). A plot of absorption coefficient (α) as a function of photon energy (hυ) is given in Figure 2, and values are given in Table 1. It has been observed that absorption coefficient (α) increases with increase in photon energy (hυ) as well as Sb concentration, for all the samples of a-$Se_{75-x}Te_{25}Sb_x$.

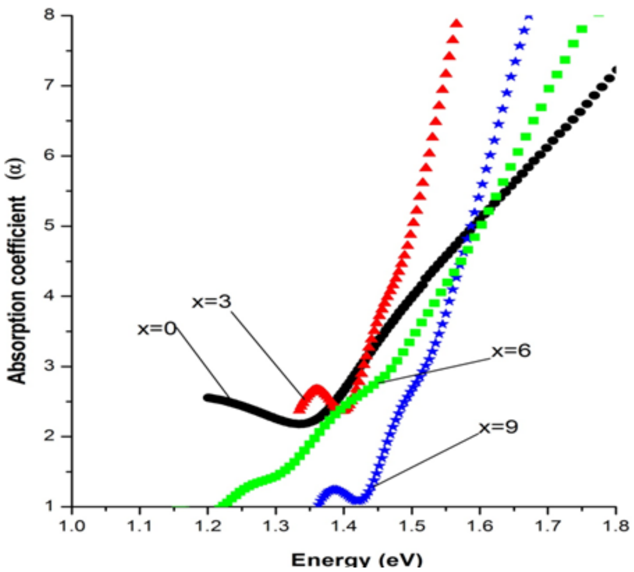

Figure 2. Variation of absorption coefficient (α) with photon energy (hυ) in $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) thin films

Table 1. Optical band gap (E_g), absorption coefficient (α) and extinction coefficient (k) for $Se_{75-x}Te_{25}Sb_x$ glassy system

S.No.	Sample	Optical Band Gap (E_g) in eV	Absorption Coefficient(α) (m^{-1})	Extinction Coefficient (k) 10^{-4}
1.	$Se_{75}Te_{25}$	1.41	1.99217	951.7
2.	$Se_{72}Te_{25}Sb_3$	1.21	8.19759	3616.0
3.	$Se_{69}Te_{25}Sb_6$	1.17	8.72369	4167.6
4.	$Se_{66}Te_{25}Sb_9$	1.11	8.86962	4237.1

The absorption coefficient (α) has been obtained directly from the absorbance against wavelength curves using the relation,

$$\alpha = OD/t \qquad (1)$$

Where OD is the optical density measured at a given layer thickness (t).

The analysis of the absorption coefficient has been carried out to obtain the optical band gap (E_g). The optical band gap has been determined from absorption coefficient data as a function of (hυ) by using Tauc relation [33,34,35].

$$(\alpha h\nu)^{1/2} = A\left(h\nu - E_g\right) \qquad (2)$$

Where A is the edge width parameter representing the film quality, which is calculated from the linear part of this relation and Eg is the optical band gap of the material. The variation of $(\alpha h\upsilon)^{1/2}$ with (hυ) for a-$Se_{75-x}Te_{25}Sb_x$ films are shown in Figure 3.

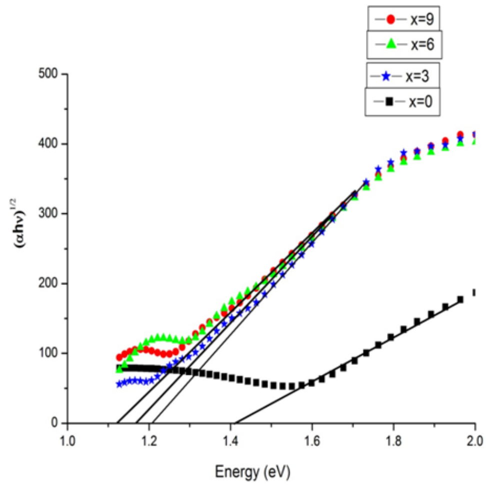

Figure 3. Variation of $(\alpha h\nu)^{1/2}$ with (hυ) in $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) thin films

The values of indirect band gap (E_g) have been calculated by taking the intercept on the x-axis. The calculated values of E_g for all glassy samples of a-$Se_{75-x}Te_{25}Sb_x$ are given in Table 1. It is evident from the Table 1 that the values of optical band gap (E_g) decreases with increasing Sb concentration.The decrease in optical band gap E_g indicates an increase in the density of defect states (DOS). The decrease in the optical band gap could also be discussed on the basis of density of state model proposed by Mott and Davis [35]. Chalcogenide thin films always contain a high concentration of unsaturated bonds or defects. These defects are responsible for the presence of localized states in the amorphous band gap.

The decrease in E_g along with the increase in the density of defect states may also be correlated with the electronegativity of the elements involved. The valence band in chalcogenide glasses is constituted by loan pair p-orbital contributed by the chalcogen atoms [36].These loan pair electrons will have a higher value of energies adjacent to electropositive atom than those of the electronegative atoms. Thus the addition of an electropositive element to the electronegative element may raise the energy of lone pair states, which is further responsible for the broadening of the valence band inside the forbidden gap. The electronegativities of Se, Te and Sb are 2.4, 2.1 and 2.05 respectively. Since Sb has lower electronegativity than Se, the substitution of Sb for Se may raise the energy of lone pair states, which may be further responsible for the broadening of the valence band. This leads to band tailing and hence shrinking of the band gap. Therefore, E_g decreases with Sb content.

Variation of (E_g) with Sb concentration in a-$Se_{75-x}Te_{25}Sb_x$ glassy sample is shown in Figure 4.

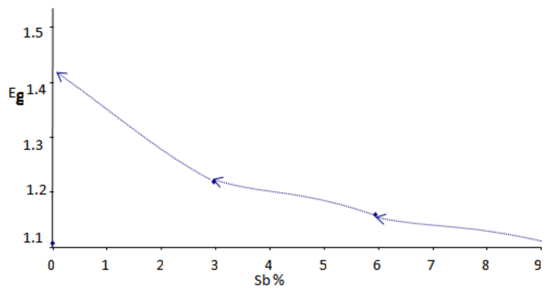

Figure 4. Variation of optical band gap (E_g) with Sb concentration in $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) thin films

3.2.2. Extinction Coefficient (k)

The optical behavior of the material has been utilized to determine its extinction coefficient (k). The extinction coefficient (k) has been calculated using the relation

$$K = \alpha\lambda/4\pi \qquad (3)$$

Where α = optical density /film thickness

Figure 5 shows the spectral dependence of k for $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) thin films. It is clear from the figure that k decreases linearly with an increase in λ for all the samples. This behavior is due to decrease in absorption coefficient with increase in λ. It is also evident from Table 1 that k increases with Sb concentration in $Se_{75-x}Te_{25}Sb_x$ thin films.

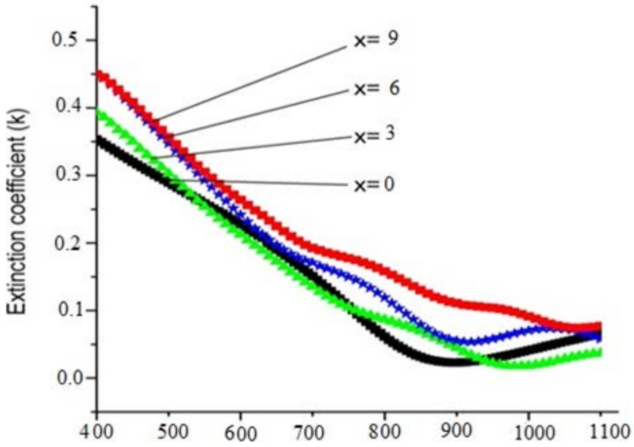

Figure 5. Variation of extinction coefficient (k) with wavelength (λ) in $Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) thin films

4. Conclusion

Thin films of a-$Se_{75-x}Te_{25}Sb_x$ (x = 0, 3, 6, & 9) glassy alloy have been deposited onto a chemically cleaned glass substrate by thermal evaporation technique. The glassy nature of the samples was ascertained by x-ray diffraction analysis The optical absorption spectra of a-$Se_{75-x}Te_{25}Sb_x$ glassy alloy has been studied in the wavelength range of 400-1100 nm by spectrophotometer. The optical parameters like absorption coefficient (α), extinction coefficient (k) and optical band gap (Eg) have been calculated for a-$Se_{75-x}Te_{25}Sb_x$ glassy system. It is observed that extinction coefficient (k) decreases with wavelength (λ) for all the samples. Absorption coefficient (α) is found to increase linearly with incident photon energy (hν) for all the samples. It has further been observed that both k and α increases with increasing Sb concentration in a-$Se_{75-x}Te_{25}Sb_x$ glassy alloy. It is also observed that optical band gap decreases with Sb content. The decrease in optical band gap with increase in Sb concentration may be due to the increase in the amount of disorder in the materials and increase in the density of defect states. The decrease in band gap could also be explained in terms of electronegativity difference between the elements involved in making the a-$Se_{75-x}Te_{25}Sb_x$ glassy system. Due to the large absorption coefficient and compositional dependence of absorption these materials may be suitable for optical memory devices.

Acknowledgement

The Authors are very much grateful to the University Grants Commission (UGC), New Delhi, India for providing financial assistance in form of major research project F.No. 39-452/2010(SR). The Authors also thanks Prof.O.N.Srivastava, Department of Physics, B.H.U., Varanasi and Prof. M.Zulfiquar, Department of Physics, Jamia Millia Islamia, New Delhi for providing the facility for XRD and optical measurements respectively.

References

[1] Seddon, A.B., “Chalcogenide glasses: a review of their preparation, properties and applications”, *J. of Non-crys Solids*, 184, 44-50, 1995.

[2] Kakiuchida, Hiroshi, Sekiya, Edson. H., Shimodaira, Noriaki, Saito, Kazuya, Ikushima and Akira. J., “Refractive index and density changes in silica glass by halogen doping”, *J. of Non-crys Solids*, 353, 568-572, 2007.

[3] Lucovsky, G., Baker, D.A., Paesler, M.A. and Phillips, J.C., “Spectroscopic and electrical detection of intermediate phases and chemical bonding self-organizations in (i) dielectric films for semiconductor devices, and (ii) chalcogenide alloys for optical memory devices”, *J. of Non-crys Solid*, 353, 1713-1722, 2007.

[4] Popescu, M., Sava, F., Lőrinczi, A., Socol, G., Mihăilescu, I.N., Tomescu, A. and Simion, C., “Structure, properties and gas sensing effect of SnSe2 films prepared by pulsed laser deposition method”, *J. of Non-crys Solids*, 353, 1865-1869, 2007.

[5] Sun, Jingjing, Akdogan, E. Koray, Klein, Lisa C. and Safari, Ahmad, “Characterization and optical properties of sol–gel processed PMMA/SiO_2 hybrid monoliths”, *J. of Non- crys, Solids*, 353,2807-2812, 2007.

[6] Voynarovych, I., Takach, V., Cheresnya, V., Pynzenik, V., Makaur, I. and Chernovich, S., *Phys and Chem. of Solid State*, 8 (4), 722, 2007.

[7] Singh, Abhay Kumar, Singh, Kedar and Saxena, N. S. "Effect of annealing on structures and effective thermal conductivity of $Se_{90}In_{10}$ chalcogenide glass”, *J. of Ovonic. Res.*, 4,107-111,2008.

[8] Dwivedi, D. K., Dayashankar and Dubey, M., “Effect of annealing on the structural and electrical properties of CdTe/ZnTe heterojunction thin”, *Chalcogenide Letters*, 6(2), 71-76, 2009.

[9] Prabhakar, S., Suryanarayanan, N., Rajshekhar, K. and Srikanth, S., “Lead selenide thin films from vacuum evaporation method-structural and optical properties”, *Chalcogenide Lett.*, 6(5), 203-211, 2009.

[10] Tanaka, K., “Structural phase transitions in Chalcogenide glasses”, *Phys. Rev. B*, 39, 1270-1279, 1989.

[11] Shimizu, A., “Data Detection using Pattern Recognition for Multi-level Optical Recording”, *Technical Digest of ISOM 2001*, 300-301, 2001.

[12] Akiyama, T., Uno, M., Kituara, H., Narumi, K., Nishiuchi, K. and Amada, N., “Rewritable Dual-Layer Phase-Change Optical Disk Utilizing a Blue-Violet Laser”, *Jpn J. appl. Phys.*, 40, 1598-1603, 2001.

[13] Ohta, T., “Phase-change optical memory promotes the DVD optical disk”, *J. Optoelectron. Adv. Mater.*, 3, 609-626, 2001.

[14] Harbold, J.M., Hilday, F.O., Wise, F.W. and Itkain, B.G., “Highly nonlinear Ge-As-Se and Ge-As-S-Se glasses for all-optical switching”, *IEEE Photonics Technology Letters*, 14(6), 822-824, 2002.

[15] Onozuka, A. and Oda, O., “Electrophotographic properties of SeTeSb Halogen alloy”, *J. Non-Cryst. Solids*, 103, 289-294, 1988.

[16] Srivastava, S.K., Dwivedi, P.K. and Kumar, A., “Steady state and transient photoconductivity in amorphous thin films of Se100-xInx”, *Physica B*, 183(4), 409-414, 1993.

[17] Shimakawa, K., “Residual photocurrent decay in amorphous Chalcogenides”, *J Non-Cryst Solids*, 77-78, Part 2, 1253-1256, 1985.

[18] Shim, Jae Yeob, Park, Sang Wook and Baik, Hong Koo, “Silicide formation in cobalt/amorphous silicon, amorphous Co_Si and bias-induced Co_Si films”, Thin Solid Films, 292 (1-2), 31-39, 1997.

[19] Saiter, J.M, Ledru, J., Hamou, A. and Saffarini, G., “Crystallization of As_xSe_{1-x} from the glassy state (0.005<x<0.03)”, *Physica B: Condensed Matter*, 245(3), 256-262, 1998.

[20] Khan, Zishan H., Zulfequar, M., Ilyas, M., Husain, M.and Begum, Kh. Selima, “Electrical and thermal properties of a-$(Se_{70}Te_{30})_{100-x}(Se_{98}Bi_2)_x$ (0 x 20) alloys”, *Current Applied Physics*, 2(2),167-174, 2002.

[21] Ahmed, A., Khan, S.A., Khan, Z.H., Julfiquar, M., Sinha, Kirti and Husain, M., “Differential Scanning Calorimetric study of Se_{80} Te_{20-x} Cu_x Chalcogenide glasses”, *Physica B* 382, 92-97,2006.

[22] León,M., Díaz, R., and Rueda,F., “Composition effects in ‘‘flash’’ evaporated $CuIn(Se_xTe_{1-x})_2$ films”, *J. Vac. Sci. Technol. A*, 12, 3082, 1994.

[23] Mehta, N., “Applications of Chalcogenide glasses in electronics and optoelectronics: A review”, *J. Scientific and Industrial Research*,65, 777-786, 2006.

[24] Roy, R.and Choudhary, V.S., *J.Opto-Electron. Adv. Mater.*, 8, 1352, 2006.

[25] Kumar, B. Rajesh and Rao, T. Subba, “Studies on structural and optical properties of vacuum evaporates In_2Te_3 thin films”, *Chalcogenide Lett.*, 8, 83-92, 2011.

[26] Sivamalar, S., Shanthi, J. and Kalugasalam, Pon, “Comparison of optical properties of PbPc and CuPc thin films”, *Chalcogenide Lett.*, 9, 275-297, 2012.

[27] Ghamdi, A.A., “Optical band gap and optical constants in amorphous Se_{96-x} Te_4Ag_x thin films”, *Vacuum*, 80(5), 400-405, 2006.

[28] Ali, Nisar, Ali, Zulfiqar, Akram, Rizwan, Aslam,S.M., Jabeen, Musarrat, Chaudhry, M. Nawaz, Iqbal, M.A. and Ahmad, N., “Study of $Sb_{28.47}Sn_{11.22}S_{60.32}$ compound as thin film for photovoltaic applications”, Chalcogenide Lett., 9,329-335, 2012.

[29] Zakery, A. and Elliott, S.R., “Optical properties and applications of chalcogenide glasses: a review”, *J.Non-Crys.Solids*, 330, 1-12, 2003.

[30] Shukla, S. and Kumar S., “Optical characterization of a-$Se_{85-x}Te_{15}Zn_x$ thin films”, *J. Pramana Physics*, 78,309-318, 2012.

[31] Pradeep, P., Saxena, N.S. and Kumar, A., “Crystallization and specific heat studies of Se_{100-x} Sb_x (x = 0, 2 and 4) glass”, *J. Phys Chem.Solids*, 58,385-389, 1996.

[32] Abkowitz, M., “Relaxation induced changes in electrical behavior of glassy chalcogenide semiconductors”, *Polymer Eng.Sci.*, 24(14), 1149-1154, 1984.

[33] Tauc, J., *Amorphous and Liquid Semiconductor*, Plenum Press, New York, 1979, 159.

[34] Urbach, F., *Phys Rev*, 92, 1324, 1953.

[35] Mott, N.F., Davis, E.A., *Electronics Processes in Non – Crys Mat.*, Clarendon, Oxford, 1979, 428.

[36] Kasrner, M., Adler, D. and Fritzsche, H., *Physics Rev.Lett.*, 37, 1504, 1976.