

Electrical Conduction Properties of Sb & Se Co-Doped PbTe Thin Films

L. Kungumadevi¹ and R. Sathyamoorthy²

¹PG and Research Department of Physics, Kongunadu Arts and Science College, Coimbatore, Tamil Nadu, India

²Department of Physics, Mother Teresa Women's University, Kodaikanal, Tamil Nadu, India

E-Mail:sivarivudevi@gmail.com

Abstract- ThenanocrystallineSb& Se co-doped PbTe thin films were prepared by using an integrated physical-chemical approach by evaporating chemically synthesized Sb& Se co-doped PbTenanopowders on glass substrates. The prepared films were analysed by XRD, SEM, EDAX and their results were discussed in detail. The DC electrical conduction mechanism in Al/SbPbTeSe/Al thin film sandwich system at different temperatures (298–423 K) was studied and was found to be Poole-Frenkel type.

Keywords: Thin Films, Crystal Structure, Electrical Properties,Metal–Semiconductor–Metal, Poole-FrenkelType

I. INTRODUCTION

There have been several studies on the growth and characterization of Sb& Se co-doped PbTenanopowders prepared by different techniques [1-4], there is still a lack of understanding of the electron conduction mechanism of Sb& Se co-doped PbTe thin films under dc field, which strongly influence the device performance. In general, dc electrical conduction through a polycrystalline semiconductor in a metal–semiconductor–metal (MSM) sandwich structure has been a complicated process and involves several conduction mechanisms. The thorough literature survey revealed that no researchers have studied the conduction mechanism of nanocrystallineSb& Se co-doped PbTe materials in thin film form. Hence, an attempt has been made to analyze electrical conduction of the nanocrystallineSb& Se co-doped PbTe thin films prepared by integrated physical and chemical approach by evaporating chemically synthesized Sb& Se co-doped PbTenanopowders on glass substrates. In this paper, we have prepared the nanocrystallineSb& Se co-doped PbTe thin films and studied their properties under dc field.

II. EXPERIMENTAL DETAILS

Sb& Se co-doped PbTenanopowders prepared by the aqueous chemical method at 110° C for 4 h were taken as the source material for thermal evaporation. Using a conventional 12 A4D Hind Hivac Coating unit, pure aluminium (99.999%, Aldrich Chemicals Company, India) was evaporated from a tungsten filament onto pre cleaned glass substrates through suitable masks to form the base electrode. Sb& Se co-doped PbTenanopowders were then evaporated from a molybdenum boat under a pressure of 2×10^{-6} mbar to form a dielectric layer. An aluminium counter electrode was evaporated onto the dielectric to form Al/ AgSbPbTe/Al (MSM) structure.

The X-ray diffraction measurements were performed on a Shimadzu XRD 6000 X-ray diffractometer using $\text{CuK}\alpha$ ($\lambda = 1.54056 \text{ \AA}$) radiation. A Quanta 200 FEG scanning electron microscope (SEM) equipped with Energy Dispersive Analysis by X-rays (EDAX) and S-3400N, Hitachi makescanning electron microscope are employed to analyze the surface morphology and composition of nanocrystallinePbTe thin films. A dc regulated power supply was used in dc conduction studies keeping the Al/PbTe/Al structure in vacuum at different temperatures 303 to 483 K. Digital voltmeter and a digital milliammeter were used for voltage and current measurements.

III. RESULTS AND DISCUSSION

A. Structural Analysis

Fig.1 represents the X-ray diffraction pattern of nanocrystallinePbTe and Sb& Se co-doped PbTe thin film. The existence of sharp peaks in the diffractogram suggests the polycrystalline nature of the films. The observed d-spacing and hkl planes are in good agreement with the JCPDS (78-1905) X-ray file data of Cubic PbTe, confirm the rock salt (NaCl) structure of the prepared thin films. From the XRD, it can be inferred that the thin films are grown with a preferential orientation along the (200) plane. The absence of any diffraction peaks related to elemental Te or Pb indicated the quality of the films. Fig.1 shows that the diffraction peak of the (200) plane shifts to slightly to higher angles due to doping of Sb and Se into PbTe. This is attributed to the assimilation of Sb and Se into the PbTe lattice. This assimilation will lead to an increase in the strain in the lattice which in turn will lead to a decrease in the intensity of the diffraction peaks.

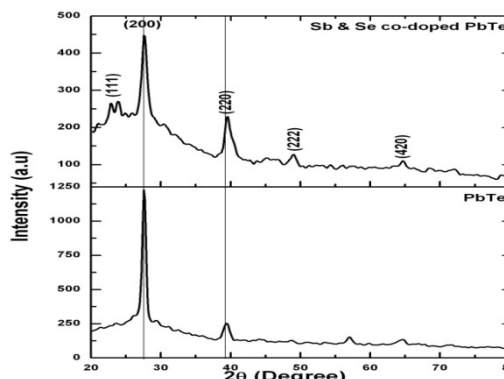


Fig.1 X-ray diffractogram of nanocrystallinePbTe and Sb& Se co-doped PbTe thin films

The crystallite size, strain, dislocation density, number of crystallites, volume of the cell and lattice constant were calculated and their values are presented in Table I. The crystallite size of the prepared thin films was estimated and it is found to be 26.62 and 8.92 nm for undoped PbTe and doped PbTe respectively. The lattice constant obtained by refinement of XRD data for undoped PbTe thin film is 0.6452 nm, which is in good agreement with the standard value ($a = 0.6454$ nm) of bulk fcc PbTe (JCPDS card No.78-1905; Fm3m).

TABLE I STRUCTURAL PARAMETERS OF NANOCRYSTALLINE PbTe AND Sb & Se CO-DOPED PbTe THIN FILMS

Sample name	Crystallite Size 'D' (nm)	Strain (ϵ) 10^{-3}	Dislocation density (δ) lines / m^2	Number of crystallites/unit area	Lattice Constant 'a' (Å)	Volume of the cell $10^{-28} m^3$
PbTe	26.62	1.359	1.19×10^{15}	2.648×10^{16}	6.452	2.685
Sb and Se co-doped PbTe	8.921	4.083	1.67×10^{16}	3.555×10^{17}	6.444	2.670

The lattice constant for Sb and Se co-doped PbTe thin film (0.6444 nm) is lower than the standard value (0.6454 nm). The decrease in lattice parameter is the evidence for build-up of the distortion in PbTe crystal structure. The appearance of this distortion can be caused probably by the Sb and Se ions occupied in the PbTe lattice. The strain, dislocation density, number of crystallites per unit area and the volume of the cell of the doped PbTe are increased compared with the undoped PbTe.

TABLE II EXPERIMENTAL AND THEORETICAL VALUES OF β

T (K)	Experimental field lowering coefficient (β) $10^{-5}(\text{mV})^{1/2}$		Theoretical field lowering coefficient (β) $10^{-5}(\text{mV})^{1/2}$	
	PbTe	Sb and Se co-doped PbTe	β_{PF} (a=1)	β_{SC} (a=4)
298	6.134	1.436	1.231	0.615
323	7.595	1.576		
348	7.156	1.781		
373	7.953	1.820		
398	9.056	2.068		
423	9.412	2.248		

B. Morphological and Compositional Analysis

Fig.2 shows the surface morphology of nanocrystalline PbTe and Sb & Se co-doped PbTe thin films. The micrograph shows that the substrate is well covered with a large number

of densely packed nanocrystalline grains. The absence of cracks and pinholes in the micrograph reveals that the prepared thin films are smooth and have uniform surface. Elemental analysis of nanocrystalline PbTe and Sb & Se co-doped PbTe thin film is carried out by EDAX. Fig.3 represents the EDAX spectrum of the prepared nanocrystalline thin films. The spectrum shows the presence of only Pb, Sb, Te and Se elements and the peak at less than 2 KeV corresponds to Silicon which is due to the glass substrate used for the preparation of thin film.

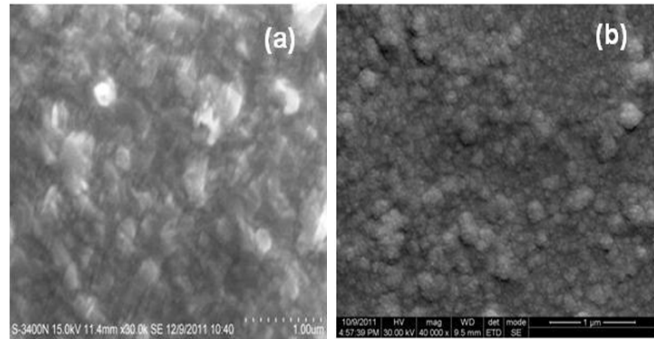


Fig.2 SEM image of nanocrystalline (a) PbTe and (b) Sb & Se co-doped PbTe thin Films

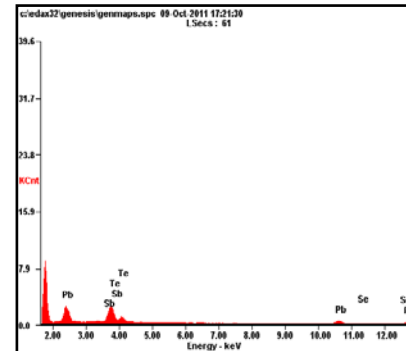


Fig.3 EDAX spectrum of nanocrystalline Sb and Se co-doped PbTe thin films

C. DC Conduction Studies

The variation of current as a function of voltage at different temperatures on undoped and doped PbTe thin films is shown in Fig. 4. It is observed that the current exhibits a voltage dependence of the form $I \propto V^n$, where n depends on the field and temperature. From the figure, it is observed that the current (I) exhibits linear dependence with applied voltage, which leads to ohmic nature. The field dependence of current for the prepared films at different temperatures is shown in Fig. 5. It is seen that the plots become linear which indicate that the conduction mechanism may be of either Richardson-Schottky (RS) or of the Poole-Frenkel (PF) type [5]. Both the mechanisms involve a relation of the form

$$I \propto \exp\left(\frac{eF^{1/2}\beta}{K_B T}\right)$$

Where I is the applied current, F the applied field, β the field lowering coefficient and K_B the Boltzmann constant.

Further to know the exact conduction mechanism prevailing in the

PbTe thin films, the theoretical field lowering coefficient (β_{theory}) was calculated, which is then compared with that of experimentally determined ($\beta_{experimental}$) given by [6],

$$\beta_{theory} = \left(\frac{e}{a\pi\epsilon\epsilon_0} \right)^{1/2} \quad (5)$$

Where, e is electronic charge, ϵ_0 is the permittivity of free space and ϵ is the effective dielectric constant, $a = 4$ for Richardson Schottky and $a = 1$ for Poole-Frenkel emission.

The dielectric constant of PbTe is 38 [7]. The values thus obtained for Schottky, β_{SC} and Poole Frenkel β_{PF} are $1.231 \times 10^{-5} \text{ eV} \text{ V}^{-1/2} \text{ cm}^{1/2}$ and $0.615 \times 10^{-5} \text{ eV} \text{ V}^{-1/2} \text{ cm}^{1/2}$ respectively. The experimental β value was determined from the plot of Log I vs. $F^{1/2}$ and is presented in Fig. 5. The β value calculated from the slope of Fig. 5 coincides with the theoretical β_{PF} value. The experimental β_{PF} values at various temperatures for nanocrystalline PbTe and Sb & Se co-doped PbTe thin films are presented in Table III. From Table III it is clear that the experimental β value for doped PbTe thin film is closer to the calculated β_{PF} than β_{SC} .

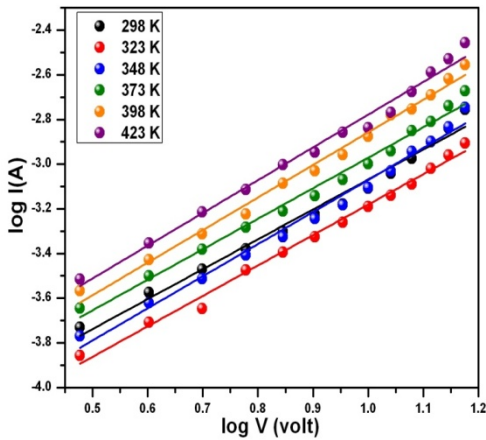


Fig.4 Variation of current with voltage at different temperature

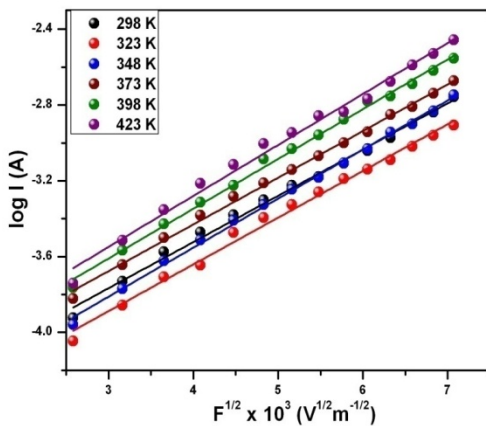


Fig.5 Dependence of current with square root of the field

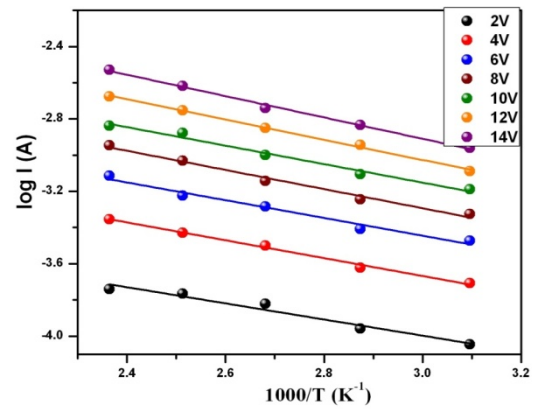


Fig.6 Plot of log I versus 1000/T

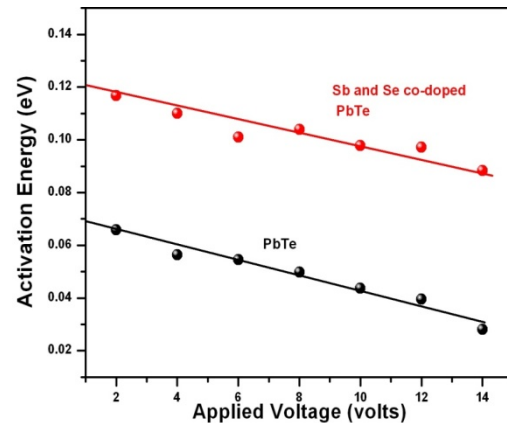


Fig.7 Activation energy as a function of applied voltage

Hence it can be proposed that the dominating conduction mechanism for doped PbTe thin films may be of the Poole - Frenkel type. The discrepancy observed between the theoretical and experimental values of β_{PF} can be attributed to the accumulation of electronic charge close to the injecting electrode, which reduces the effective field [8].

The experimental values of β for undoped PbTe film are more than the theoretical value by a factor in the range 4.90 – 8.18. Hence, the conduction mechanism cannot be explained by the classical Poole-Frenkel equation. Therefore, a modified Poole-Frenkel equation is applied to explain the conduction mechanism in PbTe films, in which the electrons produced by thermal ionization of donor –like centers hop between the localized states as a result of thermal activation. Mohammed *et al.* [9] have reported similar observation for CdTe films.

The activation energy has been determined at different constant applied voltages and is given in Fig.6. The estimated values of the activation energies are given in Table III. It is seen that the activation energy decreases with increase of applied voltage, indicating that the potential barrier has been lowered in the presence of an external electric field. Fig.7 depicts the activation energy versus the applied field. The zero activation energy is calculated from the graph and presented in Table III.

TABLE III FIELD DEPENDENCE OF ACTIVATION ENERGY

Voltage (V)	Activation energy (eV)	
	PbTe	Sb & Se co-doped PbTe
2	0.0658	0.1168
4	0.0564	0.1106
6	0.0545	0.1017
8	0.0498	0.1047
10	0.0437	0.0978
12	0.0395	0.0972
14	0.0281	0.0884
Zero activation energy	0.0695	0.1206

IV. CONCLUSION

Nanocrystalline PbTe and Sb & Se co-doped PbTe thin film was prepared by integrated physical-chemical approach. The XRD analysis shows that the nanocrystalline undoped and doped PbTe thin film exhibits a pure NaCl-type structure. The micrograph shows that the substrate is well covered with a large number of densely packed nanocrystalline grains. The dominating conduction mechanism for Sb & Se co-doped PbTe thin films may be of the Poole - Frenkel type.

REFERENCES

- [1] P. F. P. Poudeu, J. D'Angelo, H. J. Kong, A. Downey, J. L. Short, R. Pcionek, T. P. Hogan, C. Uher and M. G. Kanatzidis, "Nanostructures versus Solid Solutions: Low Lattice Thermal Conductivity and Enhanced Thermoelectric Figure of Merit in $\text{Pb}_{9.6}\text{Sb}_{0.2}\text{Te}_{10-x}\text{Se}_x$ Bulk Materials", *Journal of American Chemical Society*, Vol. 128, pp. 14347-14355, 2006.
- [2] J. C. Caylor, K. Coonley, J. Stuart, T. Colpitts and R. Venkatasubramanian, "Enhanced thermoelectric performance in PbTe-based superlattice structures from reduction of lattice thermal conductivity", *Applied Physics Letters*, Vol. 87, pp. 23105, 2005.
- [3] T. C. Harman, P. J. Taylor, M. P. Walsh and B. E. LaForge, "Quantum Dot Superlattice Thermoelectric Materials and Devices", *Science*, Vol. 297, pp. 2229-2232, 2002.
- [4] K. F. Hsu, S. Loo, F. Guo, W. Chen J. S. Dyck, C. Uher, T. Hogan, E. K. Polychroniadis and M. G. Kanatzidis, "Cubic $\text{AgPb}_m\text{SbTe}_{2+m}$: Bulk Thermoelectric Materials with High Figure of Merit", *Science*, Vol. 303, pp. 818-821, 2004.
- [5] R.M. AbadelLatif, "DC Electrical measurements on evaporated thin films of vanadium pentoxide", *Physica B: Condensed Matter*, Vol. 254, pp. 273-276, 1998.
- [6] S. Gogoi and K. Barua, "D.C. electrical properties of vacuum-deposited CdTe films", *Thin Solid Films*, Vol. 92, pp. 227-230, 1982.
- [7] Z.H. Dughaish, "Lead telluride as a thermoelectric material for thermoelectric power generation", *Physica B: Condensed Matter*, Vol. 322, pp. 205-223, 2002.
- [8] A. C. Rastogi and K.L. Chopra, "Structure-sensitive optical scattering by thin polyvinylchloride (PVC) films", *Thin Solid Films*, Vol. 34, pp. 377-380, 1976.
- [9] W.F. Mohammed, Ma'an A. S. Yousif, "The electrical properties of post-deposition annealed and as-deposited In-doped CdTe thin films", *Renewable Energy*, Vol. 26, pp. 285-294, 2002.