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Structural, Morphological and Optical Properties of Ag & Sb co-doped PbTe Thin films

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ABSTRACT - In this paper, we report the preparation of nanocrystalline Ag & Sb Co-doped PbTe thin films and the prepared films were analysed by different techniques. In addition to the microstructure, the optical properties including the transmission, absorption co efficient, refractive index, real and imaginary part of the dielectric constant and optical conductivity were investigated.

1. INTRODUCTION

Thermoelectric devices have attracted much interest because they can generate electrical power from temperature difference which can be easily obtained from the waste heat sources.1,2 The efficiencies of thermoelectric power generation as well as thermoelectric refrigeration in thermoelectric devices can be described in terms of the dimensionless figure of merit ZT which can be expressed as, $ZT = \frac{S^2 \pm T}{©}$ where S is the Seebeck coefficient, ρ is the electrical resistivity, κ is the thermal conductivity, and T is the absolute temperature [1]. Thus the good thermoelectric performance requires a large value for S and small value for ρ and κ. Theoretical calculations [3,4] and experimental observations [5,6] indicate that a significant improvement in TE efficiency can be achieved in nanostructured systems having both a high electronic density of states near the Fermi level and an enhanced rate of phonon scattering which reduces the lattice thermal conductivity. In 2004, Hsu et al.,[6] reported that the quaternary thermoelectric material AgPb18SbTe20 could attain a ZT value of about 2.2 at 800 K. This high ZT originates from the nanocrystals of Ag and Sb-rich phase embedded in a PbTe-rich matrix [6]. However the high ZT achieved by Hsu is difficult to replicate primarily due to the high electrical resistivity of the sample [7, 8] which yields a low power factor. Recently Li et al. [9, 10] studied this composition (AgPb18SbTe20) synthesized by combining mechanical alloying (MA) and spark plasma sintering (SPS) for its synthesis. They found that a large power factor (18-20 « W cm-1K-2) and a high figure of merit (1.2-1.5) could be obtained in compositions rich in Pb. Ag and Sb co-doped PbTe has the cubic NaCl-type structure [11]. In this paper, addition to the microstructure, the optical properties including the transmission, absorption co efficient, refractive index, real and imaginary part of the dielectric constant and optical conductivity were investigated.

2. EXPERIMENTAL DETAILS

Ag and Sb co-doped PbTe nanopowders prepared by the chemical method were taken as the source material for thermal evaporation. Using a conventional 12 A4D Hind Hivac Coating unit, Ag and Sb co-

doped PbTe nanopowders were evaporated from a molybdenum boat onto pre cleaned glass substrates. The vacuum chamber is pumped down to 2 $\times 10^{-6}$ mbar. The boat is gradually heated until the material started to evaporate. The vapor molecules leaving the source were deposited onto the substrate surface. The rate of deposition was maintained about 1Å / Sec and the thickness of the films is monitored by a conventional quartz crystal monitor. The thickness of the prepared nanocrystalline Ag and Sb co-doped PbTe thin films is 300 nm. The substrates were held at room temperature during the deposition process.

3. RESULTS & DISCUSSION

3.1 Structural Analysis

Fig.1 represents the X-ray diffraction pattern of nanocrystalline PbTe and Ag and Sb 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 indicates the purity of the films. Fig.1 shows that the diffraction peak of the (200) plane slightly shifts to lower angles due to doping of Ag and Sb into PbTe. This is attributed to the incorporation of Ag and Sb into the PbTe lattice. This incorporation 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.

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 1. The crystallite size of the prepared thin films was estimated and it is found to be 26.62 and 23.20 nm for undoped PbTe and doped PbTe respectively. The lattice constant obtained by refinement of XRD data for undoped PbTe thin film is 6.452 Å, which is in good agreement

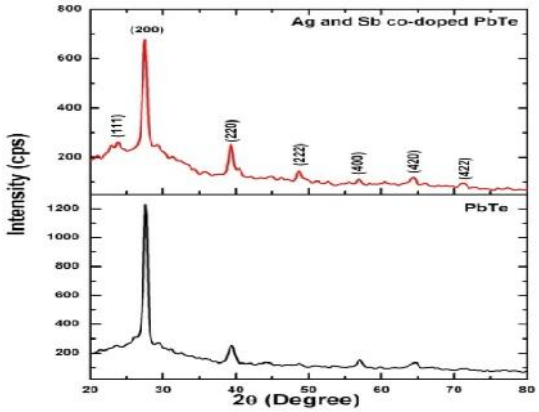

Figure.1 X-ray diffractogram of nanocrystalline PbTe and Ag &Sb co-doped PbTe thin films

Table 1. Structural parameters of PbTe and Ag & Sb co-doped PbTe nanoparticles

Sample name	Crystallite Size 'D'(nm)	Strain (£) 10^{-3}	Dislocation density(δ) x10^{15} lines / m^2	Number of crystallites / unit area	Lattice Constant 'a' (Å)	Volume of the cell 10^{-28} m^3
PbTe	26.620	1.359	1.19	2.648 x 10^{15}	6.452	2.685
Ag and Sb co-doped PbTe	23.201	1.600	1.67	2.73 x 10^{16}	6.479	2.690

with the standard value (a = 6.454 Å) of bulk fcc PbTe (JCPDS card No.78-1905; Fm3m). The lattice constant for Ag and Sb co-doped PbTe nanoparticles (6.479 Å) is higher than the standard value (6.454 Å). The increase 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 Ag and Sb 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.

3.2 Morphological and Compositional analysis

Fig.2 shows the surface morphology of nanocrystalline PbTe and Ag and Sb 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 Ag and Sb 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, Ag, Sb and Te elements and absence of impurities. The peak at less than 2 KeV corresponds to Silicon which is due to the glass substrate used for the preparation of thin film.

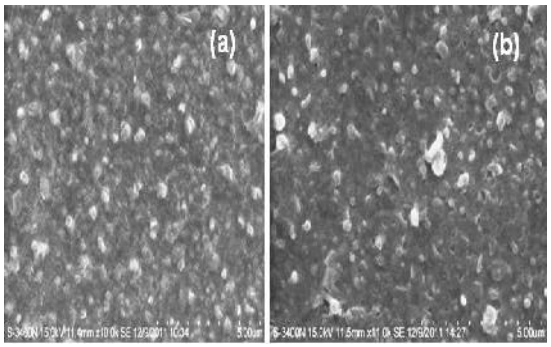

Figure.2 SEM image of nanocrystalline (a) PbTe and (b) Ag &Sb co-doped PbTe thin films

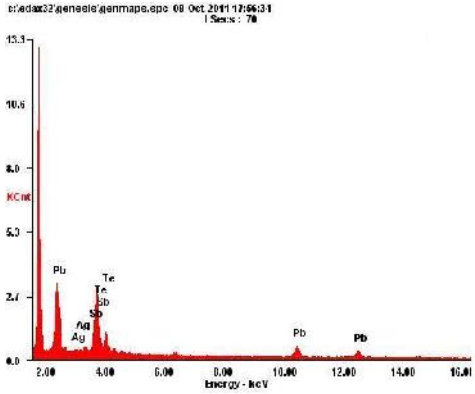

Figure.3 EDAX spectrum of nanocrystalline Ag & Sb co-doped PbTe thin films

3.3 Optical Analysis

Fig.4 shows a typical transmittance spectrum of nanocrystalline PbTe and Ag and Sb co-doped PbTe thin films. The value of transmittance increases with increase in wavelength. The transmittance of doped PbTe thin film is increased compared with undoped PbTe. In a polycrystalline material the nature of optical inter band transitions (direct or indirect) near the absorption edge can be determined by the relation between absorption coefficient (α) and the optical energy gap (Eg). Using the standard expression for direct transition between two parabolic bands $(\alpha h\nu)^2 = A(h\nu - E_g)$, the band gap of both undoped and doped PbTe thin films was estimated. The value of E_g is determined from an intercept on the energy axis of $(\alpha h\nu)^2$ versus hν plot as shown in Fig. 5 and the values are 1.153 and 1.342 eV. The band gap energy of doped PbTe is increased compared with undoped PbTe.

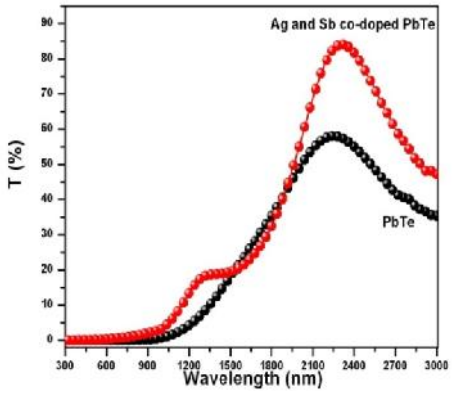

Figure.4 Transmittance spectra of the nanocrystalline Ag & Sb co-doped PbTe thin films

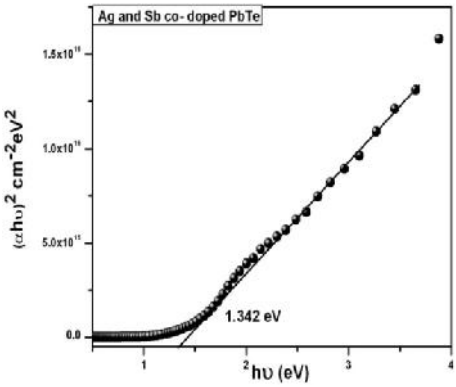

Figure.5 Tauc plot of (αhυ)2 versus hυ showing the estimation of direct band gap energy

The possible optical transitions in these films are found to be direct and allowed. It is worth pointing out that the observed optical band gap energy in nanocrystalline undoped and doped PbTe films is much larger than the bulk value of 0.32 eV due to quantum confinement effect. From XRD results, the

crystallite size of the film is around 26 and 23 nm for undoped and doped PbTe thin films, much smaller than the exciton Bohr radius of 150 nm in PbTe. Therefore, the prepared nanocrystallites in the films can lead to a strong quantum confinement effect, which may account for the optical band gap energy increase.

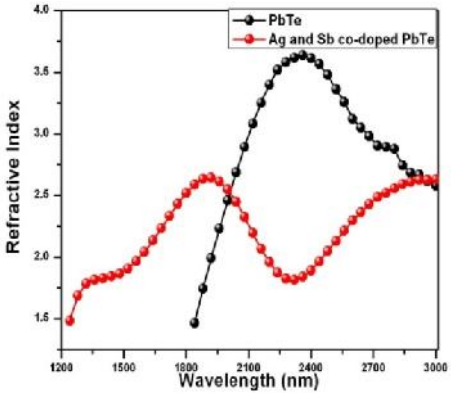

Figure.6 Spectral distribution of the refractive index of nanocrystalline PbTe and Ag & Sb co-doped PbTe thin films

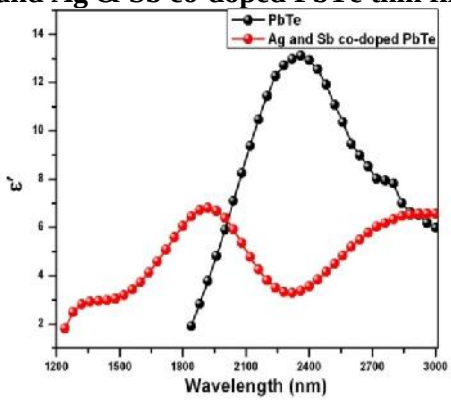

Figure.7 Wavelength dependence of real (ε') part of the dielectric constant

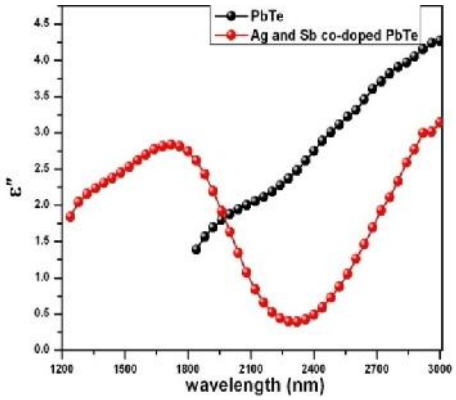

Figure.8 Wavelength dependence of imaginary (ε") part of the dielectric constant

The spectral distribution of the refractive index, real and imaginary part of the dielectric constant for the investigated films is presented in Fig. 6, 7 and 8. The variation of these parameters with wavelength exhibits an oscillatory nature which may due to the size effect in the prepared nanocrystalline thin films. The wavelength dependence of optical conductivity (σ_o) of nanocrystalline PbTe and Ag and Sb co-doped PbTe thin films is displayed in Fig. 9. Optical conductivity of doped PbTe thin films decreased by an order compared with the undoped PbTe. This may be due to the increase in transmittance of the doped PbTe.

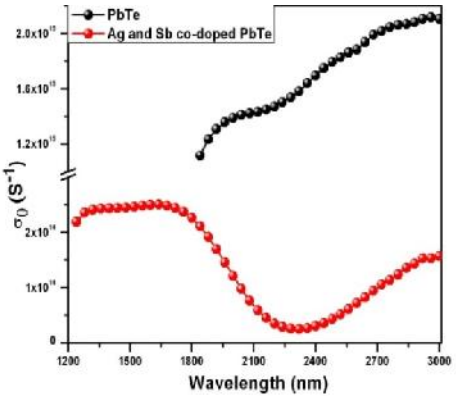

Figure.9 Variation of Optical Conductivity with photon energy

CONCLUSION

Nanocrystalline PbTe and Ag and Sb co-doped PbTe thin film was prepared by integrated physical-chemical approach. The method is simple, economical and highly efficient. The XRD analysis shows that the nanocrystalline PbTe and Ag and Sb co-doped PbTe thin film exhibits a pure NaCl-type structure. The possible optical transitions in these films are found to be direct and allowed. the observed optical band gap energy in nanocrystalline undoped and doped PbTe films is much larger than the bulk value of 0.32 eV due to quantum confinement effect.

REFERENCES

[1] L.D.Hicks, M.S.Dresselhaus, "Thermoelectric figure of merit of a one-dimensional conductor", Physical Review B, 47 (1993) 16631-16634.

[2] Y.M.Lin, M.S.Dresshaus, “Surface-state conduction through π-bonded chains”, Physical Review B, 68 (2003) 075304 (10 pages).
[3]T.C.Harman, T.J.Taylor, M.P.Walsh, B.E.Laforge, “Quantum Dot Superlattice Thermoelectric Materials and Devices”, Science, 297 (2002) 2229-2232.
[4]K.F.Hsu, S.Loo, F.Guo, W.Chen, “Cubic $AgPb_{(m)}SbTe_{(2+m)}$: bulk thermoelectric materials with high figure of merit”, Science, 303 (2004) 818-821.
[5] A.Kosuga, M.Uno, K.Kurosaki, S.Yamanaka, “Thermoelectric properties of $Ag_{1-x}Pb_{18}SbTe_{20}$ (x = 0, 0.1, 0.3)”, Journal of alloys and compounds, 387 (2005) 52-55.
[6] Zhou M, Li J F, Wang H 2007 Chin.Sci.Bull. 52 990-996
[7]M.Zhou, J.F.Li, T.Kita, “Nanostructured $AgPb_{(m)}SbTe_{(m+2)}$ system bulk materials with enhanced thermoelectric performance”, Journal of American Chemical Society, 130 (2008) 4527-4532.
[8]T.Su, H.Zhu, H.Li, X.Li, S.Li, “Composition-dependent transport properties of $Ag_{1-x}Pb_{18}SbTe_{20}$ prepared by high pressure”, Journal of alloys and compounds, 509 (2011) 2430-2433.
[9]W.Q.Ao, W.A.Sun, J.Q.Li, F.S.Liu, “Hydrothermal synthesis of nano-sized $AgPb_{18}SbTe_{20}$ thermoelectric powders”, Journal of alloys and compounds, 475 (2009) L22-L24.