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Role of Sb Substitution on Electrical Properties of Se-Te Glasses

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Abstract

Objectives: This paper reports the influence of Sb addition on the electrical properties on bulk samples of Se80-xTe20Sbx(x=0, 6, 12) glasses. **Methods/Statistical Analysis**: The amorphous samples have been prepared using melt quenching method and the pellets of the bulk sample of thickness 1mm and diameter 5mm has been formed using pelletiser. The I-V measurements of pellets have been recorded on Keithley electrometer in the temperature range 298-398 K. The data obtained is analysed to calculate activation energy of conduction and temperature dependent dc electrical conductivity. **Findings**: The observations suggest that conductivity decreases due to formation of highly crosslinked structure when Sb is added in less amount i.e. 6 at.wt% whereas on addition of Sb upto 12 at. wt. %, conductivity increases significantly due to increase in ring structure and decrease of steric hindrance in system. Earlier reports on the electrical properties of Se-Te-Sb system provided information upto 10 at. wt. % of Sb addition to Se-Te system, whereas in present study Sb is added upto 12 at. wt.% to get its influence on electrical properties of Se-Te system. **Applications/Improvements**: The higher content of Sb in Se-Te system increases the electrical conductivity which makes it a potential candidate for application in electronic devices.

Keywords: Activation Energy, Electrical Conductivity, Hopping Conduction, Hopping Energy, Se-Te-Sb Glasses

1. Introduction

Chalcogenide glasses are studied extensively in terms of optical, electrical, structural and phase transformational properties[1-5]. Electrical properties of chalcogenide glasses have been studied widely from the viewpoint of their applications in solid state electronic devices. Se-Te based glasses have attracted considerable interest due to their widespread applications in semiconductor devices. They are used as optical recording medium due to their good laser write sensitivity[6-10]. It has been reported that properties of these glasses are significantly affected by the presence of third impurity in the Se-Te matrix[11-13]. The addition of third impurity brings out configurational and structural changes in the host system that not only increases the stability of the system but also enlarges their domain of the application in various devices. The addition of antimony also increases the glass forming ability of Se-Te system[14-15].

The present communication, a detailed study has been carried out on the temperature dependent dc electrical properties of the system. The conduction activation energy has been studied in terms of hopping conduction and lastly, the effect of Sb concentration on Se-Te system has been discussed.

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2. Experimental Details

Amorphous samples of $Se_{80-x}Te_{20}Sb_x$(x=0,6,12) glassy alloys have been prepared using well established melt quenching method. Metal powders of Se, Te and Sb of high purity (99.999%) have been procured from Sigma Aldrich. The constituent materials were weighed and mixed using mortar and pestle. The mixture was then placed into a quartz ampoule and sealed at a vacuum of 10^{-5} Torr. The sealed ampoules was then placed in a muffle furnace where temperature increases gradually at rate of 2-3 °C/min to 950°C. The temperature of the furnace was maintained at 950°C for 12 hours. The ampoule was frequently rotated to facilitate the homogenization of the sample. The molten sample was then taken out from the furnace and was quickly plunged into ice-cooled water to get amorphous mixture. This amorphous mixture was then grinded gently to powder form using mortar and pestle. The pellets of the sample (thickness 1mm and diameter 5mm) were made using pelletizer. The samples were structurally characterized using X-ray diffractometer and the XRD patterns confirmed that samples are amorphous in nature. The I-V characteristics were recorded on Keithley electrometer in the temperature range 298-398 K.

3. Results and Discussion

The I-V curves recorded on Keithley electrometer were linearly fitted and slope of the curves was used to evaluate the dc electrical conductivity of the samples using the following relation[15-17]

$$\sigma_{dc} = \frac{L}{RA} \quad (1)$$

Where, R denotes resistance, L and A are thickness and cross sectional area of the pallet respectively. Figure 1 shows the plot of 1000/T versus ln s_{dc} for $Se_{80-x}Te_{20}Sb_x$(x=0, 6, 12) glassy alloys. The plots are fitted to straight line and slope of the straight line gives activation energy of conduction (ΔE) while the intercept gives pre-exponential factor (σ_o). The dc electrical conductivity (s_{dc}) obtained at 25°C is mentioned in Table 1 for all the samples under investigation.The dc electrical conductivity is dependent on temperature through the Arrhenius equation given by[18-19]

$$\sigma_{dc} = \sigma_o \exp\left[\frac{-\Delta E}{KT}\right] \quad (2)$$

Table 1. Values of Activation Energy of Conduction (ΔE), DC Electrical Conductivity (Δdc), pre-Exponential Factor (σo) and Density of Localized States at Fermi Level for for Se80-xTe20Sbx (x=0, 6, 12%) Glasses

Composition	ΔE (eV)	$\sigma_{dc(\Omega.cm)^{-1}}$ at 25 °C	$\sigma_{o(\Omega.cm)^{-1}}$	N(E)
$Se_{80}Te_{20}$	10.28×10^{-4}	2.07×10^{-10}	2.04×10^{6}	0.95×10^{40}
$Se_{74}Te_{20}Sb_6$	9.13×10^{-4}	0.57×10^{-10}	2.50×10^{4}	2.23×10^{40}
$Se_{68}Te_{20}Sb_{12}$	6.60×10^{-4}	4.98×10^{-9}	1.03×10^{2}	3.81×10^{40}

Where, ΔE is activation energy of conduction, σ_o represents the pre-exponential factor and K is Boltzman constant. Table 1 lists the value of ΔE and σ_o for both the samples.

Table 1 shows that dc electrical conductivity decreases with increase in Sb concentratio n upto 6 at.wt% and on further addition of Sb upto 12 at wt %, conductivity increases significantly. It is reported[3-4] that glasses containing Se in higher atomic weight percentage mostly contains about 40% of Se atoms in ring structure and 60% of Se atoms are bonded as polymeric chains. When Te is added, Se_6Te_2 rings are formed at the expense of Se_8 rings. Thus, the structure of Se-Te is mixed network of Se_6Te_2 rings, Se_8 rings and Se-Te copolymer chains. The rings are binded by strong covalent bonds while the chains have only Vander Wall bonds between them. The addition of Sb upto 6 at.wt% to Se–Te system causes its entry into the crosslink chains Se-Sb (214.20 kJ/mol) bonds of higher bond energy are formed replacing Te-Sb (205.8 kJ/mol) bonds of lower bond energy. This leads to a decrease in Se_8 rings which makes the system heavily cross linked. The steric hinderance increases which makes the system more rigid and results into decrease in dc electrical conductivity. On further increasing the concentration of Sb in Se-Te system, lower energy bonds (176.40 kJ/mol) of Sb-Sb bonds are favoured over Se-Sb bonds, which increases the Se_8 rings in the structure of Se-Te-Sb system. The increase in Se_8 rings makes the system more ordered by decreasing the degree of crosslinking and as a result increase in dc electrical conductivity is observed on addition 12 at.wt% of Sb to Se-Te system.

The I-V curves and the values of conduction activation energy indicate that conduction is mainly due to thermally assisted tunnelling of charge carriers. The conduction mechanism may be either hopping into localized states or in extended states over the mobility edge. In order to get clear distinction between the two conduction mechanism, Mott and Davis[20-21] proposed formulations to obtain hopping conductivity and other parameters related to hopping conduction mechanism. The is hop-

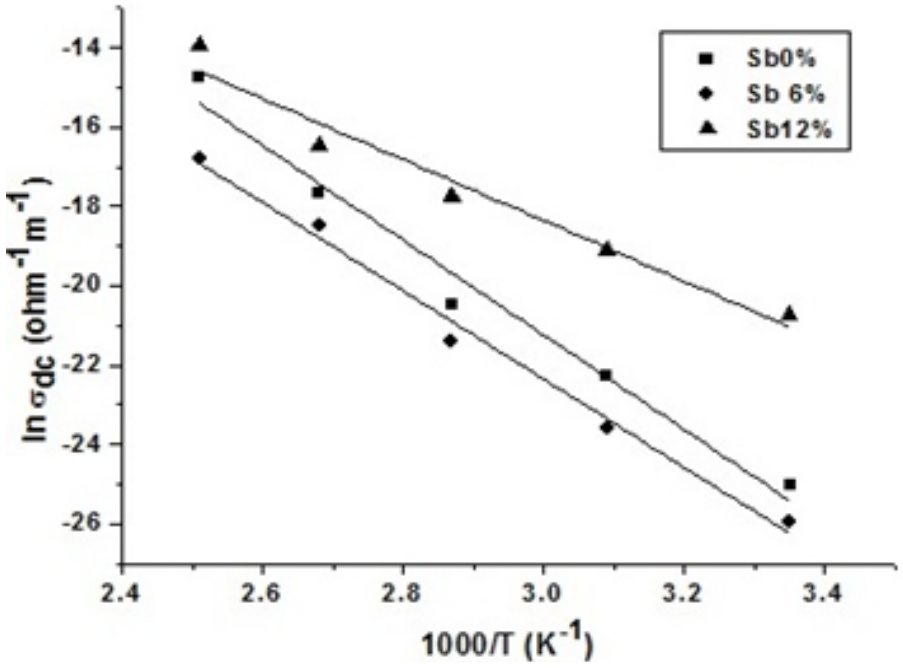

Figure 1. Temperature dependence of dc electrical conductivity for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glasses.

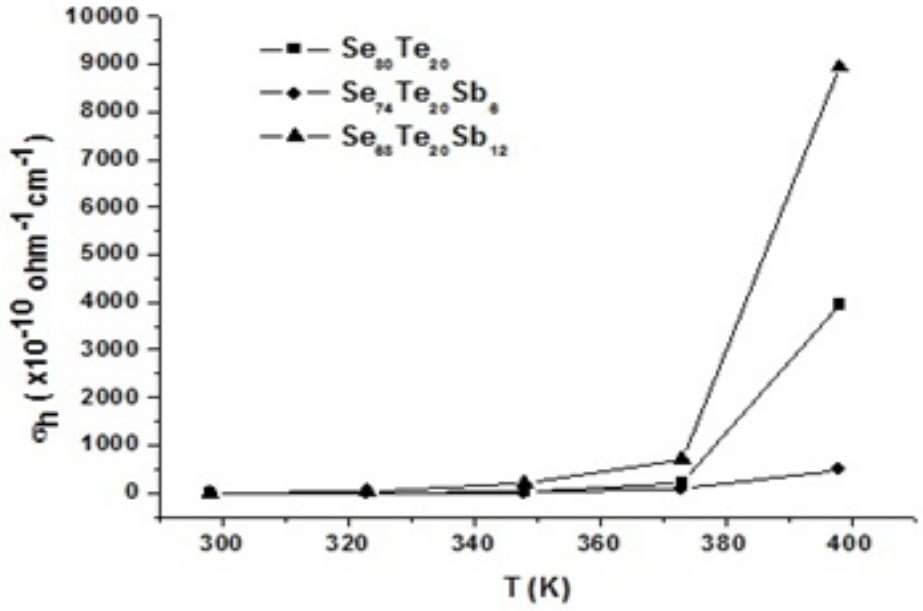

Figure 2. Plot of σ_h with temperature for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glassy alloys.

ping conductivity (σ_h) is dependent on temperature by the following relation:

$$\sigma_h \sqrt{T} = \sigma_o^{'} \exp\left[\frac{-B}{T^{1/4}}\right] \quad (3)$$

Where, $\sigma_o^{'}$ is pre-exponential factor and B is given by[22],

$$B = 2.1\left[\frac{\alpha^3}{KN(E)}\right]^{1/4} \quad (4)$$

Here, α describes the spatial extent of the localized wave function, which was assumed to be 0.1 $Å^{-1}$ and N(E) is the density of localized states at the fermi level.

Figure 1 is extrapolated to lowest temperature and value of σ_{low} is noted. The value of σ_h is then obtained by subtracting σ_{low} from $\sigma_{d.c.}$ obtained at different temperatures[23]. Figure 2 shows the plot of σ_h with temperature for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glassy alloys.

It is observed that hopping conductivity increases with increase in temperature and sudden increase is observed at 398 K. This is due to rise in activation energy of charge carriers (Figure 5) at high temperatures, which ease their hopping in various available states. The values of σ_h are also directly related to hopping distance (R_h) and hopping energy (E_h).

The values of R_h (Fig.4) depict that hopping distance decreases on increase of temperature which supports the rise in hopping conductivity with rise in temperature.

Figure 3 shows the the variation of $\ln \sigma_h \sqrt{T}$ with $T^{-1/4}$ for all the samples under investigation. These plots are linearly fitted and slope of these curves gives value of B given by eq. (4), which is further used to calculate density of localized states N(E). The value of N(E) for all the sam-

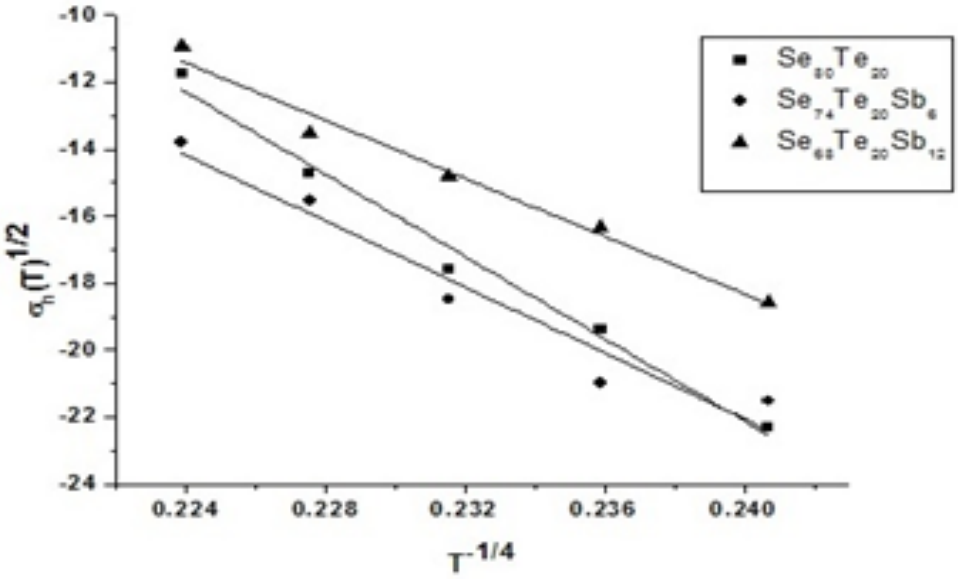

Figure 3. Plot of $\ln \sigma_h \sqrt{T}$ with $T^{-1/4}$ for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glassy alloys.

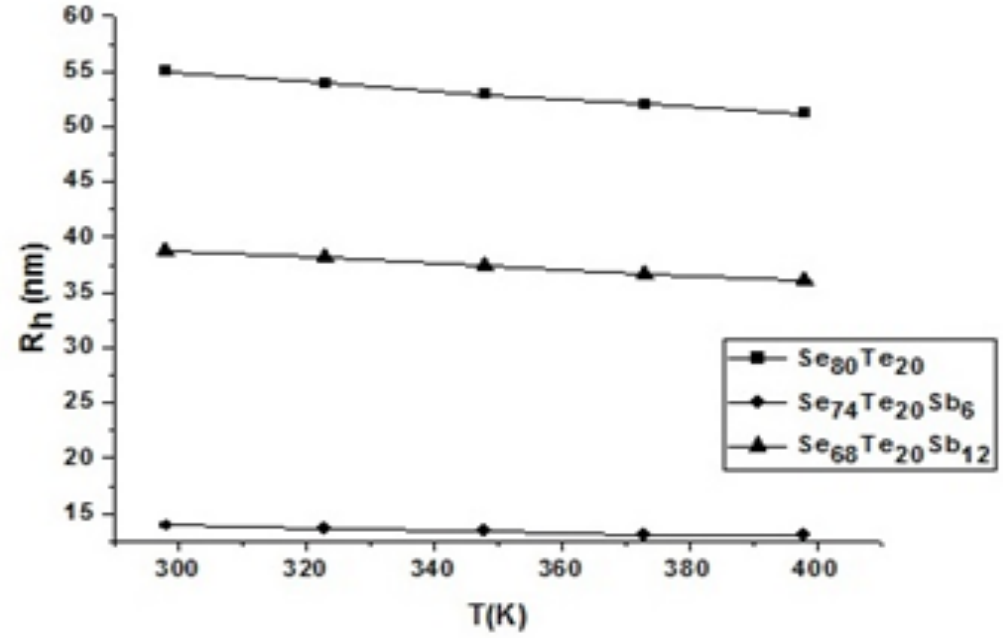

Figure 4. Variation of R_h with temperature for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glassy alloys.

ples under consideration are mentioned in Table 1. The values of N(E) indicate that density of localized states increases with increase in Sb content in the samples, which suggest that conduction in these glasses is also due to hopping into these localized states near Fermi level.

The value of N(E) is used to calculate hopping distance (R_h) and hopping energy (E_h) given by[24]

$$R_h = \left[\frac{9}{8\pi\alpha N(E)KT}\right]^{1/4} \quad (5)$$

$$E_h = \frac{3}{4\pi R_h^3 N(E)} = \left[\frac{2\alpha^3 K^3 T^3}{9\pi N(E)}\right]^{1/4} \quad (6)$$

Figure 4 shows the plot of R_h with temperature while Figure 5 shows the variation of E_h with temperature for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glassy alloys.

From Figure 4 and Figure 5, it is observed that hopping distance decreases while hopping energy increases with increase in temperature. This is due to availability of large number of localized states at higher temperature. It is noticed that R_h is highest for $Se_{80}Te_{20}$ glass, which suggest that due to large hopping distance, hopping conductivity is less for $Se_{80}Te_{20}$ glass. Further, it is noted that inspite of fact that R_h is lowest for $Se_{74}Te_{20}Sb_6$, σ_h is lowest for this sample. This may be attributed to high value of hopping energy (Fig. 5) needed for conduction in this sample. E_h is lowest for $Se_{68}Te_{20}Sb_{12}$ glasses, which indicate that very less energy is required for hopping conduction in this case. Moreover, a large number of localized states

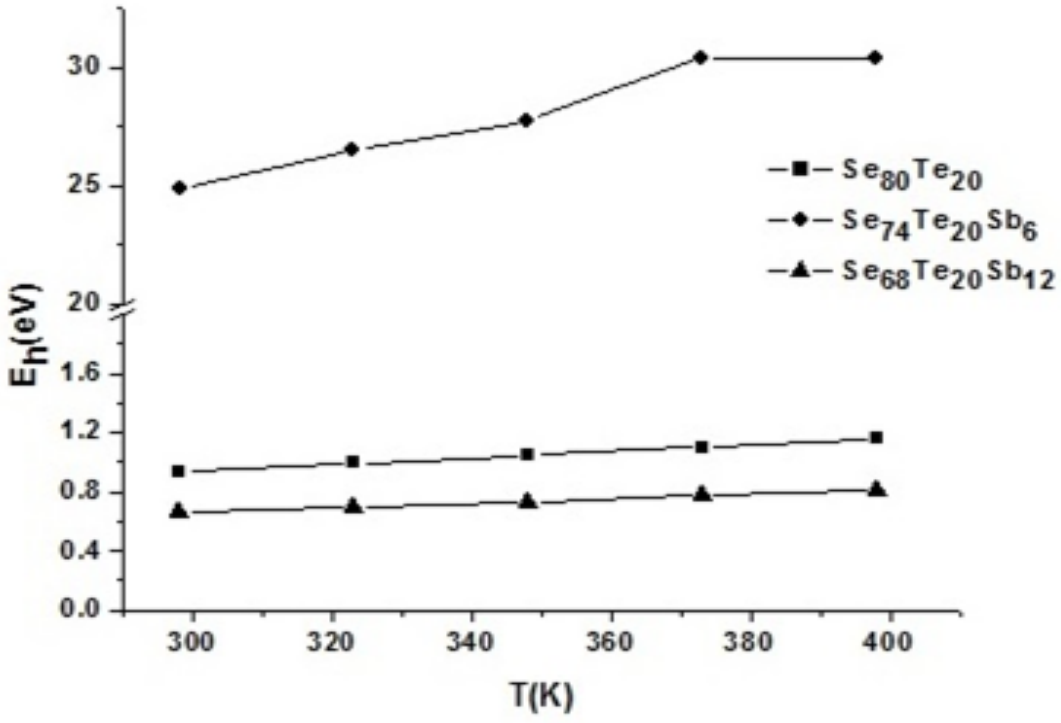

Figure 5. Variation of E_h with temperature for $Se_{80-x}Te_{20}Sb_x$ (x=0, 6, 12%) glassy alloys.

in $Se_{68}Te_{20}Sb_{12}$ glass favours the high value of electrical for this sample as observed in Table 1 and Fig.2.

4. Conclusions

The systematic study of dc electrical conductivity at various temeperatures reveals that on addition of antimony in less amount i.e. 6 at.wt.%, dc electrical conductivity (σ_{dc}) decreases due to steric hinderence created in structure while on further addition of Sb upto 12 at.wt.%, σ_{dc} increases due to more ordered structure. Moreover, it was also observed that hopping conductivity play a pivotal role in electrical conduction of Se-Te-Sb system. The values of hopping distance and hopping energy supports the above mentioned facts.

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