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PHYSICAL AND DIELECTRIC PROPERTIES OF Sn DOPED Se-Te GLASSY SYSTEM

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An attempt is made to study the effect of Sn on the physical and dielectric properties of tellurium rich chalcogenide glasses. The physical properties like, average coordination number, constraints, the fraction of floppy modes, lone pair electrons, heat of atomization, mean bond energy, cohesive energy, electronegativity and glass transition temperature, of $Se_{30}Te_{70-x}Sn_x$ system are studied for different values of x, i.e., for x= 0, 1.5, 2.5 and 4.5. The glass transition temperature and mean bond energy have been calculated by using Tichy Ticha approach, whereas cohesive energy is calculated by using chemical bond approach. The effect of Sn doping on the dielectric properties, i.e., dielectric constant and dielectric loss, of a bulk $Se_{30}Te_{70-x}Sn_x$ glassy alloy is also studied at room temperature in the frequency range 0.1 KHz to 3 MHz. It is found that the dielectric constant ε' (real permittivity) and dielectric loss ε'' decreases with increase in frequency and increases with tin content. The present results show that the system under consideration is a compact and stable glass former.

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Keywords: Chalcogenide glasses, Dielectric constant, Constraints, Heat of atomization, Fraction of floppy modes.

1. Introduction

The study of amorphous materials has received much attention in recent past due to their interesting electrical, optical and magnetic properties. Many workers [1-6] have reported the impurity effects on the various properties of chalcogenide glasses/semiconductors. It has been observed[7,8] that the effect of impurities strongly depend upon the composition of the glass, chemical nature of the impurity and method of doping. The impurity concentration is a critical factor and several physical properties are found to improve by the addition of certain impurities. In the recent past a lot of work has been done using Se as the base material owing to its high viscosity, yet it has short life time and low sensitivity[9]. The impurities like Ge, Te, Sb, Sn, As etc. are used to prepare alloys with Se which have more sensitivity, greater hardness, small aging effect and higher crystallization temperature[9,10]. The Se-Te binary alloys are widely studied in both thin films as well as in bulk form due to their electrophotographic applications like photoreceptors and laser printing. Generally, the binary alloys are covalent in nature and the addition of third element creates iono-crystals bounds that result in increased conductivity of the material. The addition of third element stabilizes the structure and makes the ternary system thermally more stable. Moreover, it helps in expanding the glass forming region and creates compositional and configurationally disorder in the glassy materials. In the present work we have studied the tellurium rich $Se_{30}Te_{70-x}Sn_x$ chalcogenide system with Sn as a dopant element. It is found[11] that the addition of Sn to Se-Te system results in structural changes in the material

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which in turn modifies the band structure and hence the properties of the material. Se-Te-Sn alloys show narrow glass forming region having high transmittance in the IR region due to its reduced optical band gap and low optical loss. Addition of Sn also increases the thermal stability of the material. Since, structure determination of glassy material is a challenging job because most of the experimental methods available are not applicable to them. Hence, it is better to study the compositional dependence of the various physical properties. The various physical properties, like average coordination number, constraints, lone pair electrons and heat of atomization, cohesive energy, etc., of $Se_{30}Te_{70-x}Sn_x$ alloy are studied using well established relations in the literature. It has been found that the values of lone pair electrons and average single bond energy decreases with increasing Sn content, whereas the average coordination number, constraints and heat of atomization, mean bond energy, glass transition temperature and cohesive energy increases with increase in the Sn content which is in agreement with the earlier results. The results of the present study show that $Se_{30}Te_{70-x}Sn_x$ alloy is a compact and stable glass former.

Dielectric relaxation studies are important to understand the nature and origin of dielectric losses, which is useful in determining the structure and defects in solids. The study of chalcogenide materials has shown that they exhibit dielectric dispersion at low frequencies. The addition of Sn to the Se-Te system is found to cause structural changes in the system and hence modifies the electrical properties of the material. It has been found[11] that the Sn concentration plays an important role in the variation of dielectric parameters with temperature. In this study the effect of Sn content on the dielectric properties of the bulk $Se_{30}Te_{70-x}Sn_x$ samples has been studied in the frequency range 0.1KHz to 3MHz. The dielectric parameters, i.e., dielectric constant ε' and the dielectric loss ε'' are determined using the Impedance Analyzer (Wayne Kerr 6500 B). It is found that the dielectric constant and dielectric loss decreases with increases in frequency. Whereas this decreases is large at low frequencies as compared to higher frequencies. Further, the present study shows that the dielectric parameters ε' and ε'' increases with the increase in Sn concentration. This increase in these parameters may be understood in terms of the increased defect states of Se-Te system.

The manuscript is organized as follows: the experimental detail is given in section 2, results and discussions are presented in section 3 and finally the manuscript is concluded in section 4.

2. Experimental details

The glassy alloy of $Se_{30}Te_{70-x}Sn_x$ system for x= 0, 1.5, 2.5, 4.5 in bulk form is prepared by melt quenching technique. High purity (99.99%) elemental substances were weighed according to their atomic percentages and were sealed in quartz ampoule under the vacuum of 10^{-5} Torr. The sealed ampoules were heated in a muffle furnace at an appropriate temperature of 800^{o}C for 15 hours by gradually increasing the temperature at a rate of 3-4^{o}C/ Min. During heating the ampoules were rocked frequently to ensure homogenization of the melt. The ampoules were then quenched in liquid nitrogen. The amorphous nature of the considered composition in powder form was confirmed by X-ray diffraction. The glassy alloy so prepared was grinded to a fine powder and pallets of diameter about 1cm were prepared by compressing the powder in a die under a pressure of 5 tons. The pallets were coated, both sides, with silver paste in order to obtain the good quality contact with the electrodes of the sample holder. The coated pellets were mounted between two steel electrodes inside a holder for dielectric measurements. Wayne Kerr 6500B Impedance Analyzer was used for measuring the capacitance (C) and dissipation factor (D), simultaneously. The dielectric constant ε' and dielectric loss ε'' for different compositions of $Se_{30}Te_{70-x}Sn_x$ system were determined from the parallel capacitance(C) and dissipation factor (D), measured at room temperature in the frequency range 0.1 KHz to 3 M Hz.

3. Results and discussion

3.1 Average Coordination number and number of Constraints

The coordination number is defined as the average atom coordination with its nearest neighbors of the constituents. It is useful in explaining the cross linking and testing the validity of topological concepts[12]. The average coordination number can be used to provide deeper understanding between the structural and material properties of a glassy alloy. The coordination number of covalent atom of chalcogenide glasses follows the 8-N rule [13], with N is the number of the outer most shell electrons in a given atom, and is given by the relation,

$$\langle r \rangle = \frac{((\alpha)N_{Se} + (\beta)N_{Te} + (\gamma)N_{Sn})}{\alpha + \beta + \gamma}, \quad (1)$$

here α, β and γ are the atomic percentage of Se, Te and Sn respectively and N_{se}, N_{Te} and N_{Sn} represent their respective coordination numbers. The calculated values of average coordination number, <r> are given in the table 1. From table 1 it is observed that the <r> increases with increase in the Sn content. It shows that the cross linking of chains between the atoms increases with increase in the Sn content. Since the minimum value of <r> obtained in our case is 2.0 hence we can say that the system under investigation is a good glass former.

Covalent networks in a glassy system are mechanically constrained by interatomic valence forces such as bond stretching and bond bending. In optimal glass formation the enumeration of mechanical constraints, i.e., bond stretching constraints and bond bending constraints are given[14] as N^{α}= <r> / 2 and N^{β}=(2<r>-3), respectively. The calculated values of average constraints i.e. ($N_{CON=}$ N^{α} + N^{β}), for $Se_{30}Te_{70-x}$ Sn_x system at different values of x are reported in table 1. The present results are consistent with the previous studies[15-17]. From table I it is observed that the value of average constraints increases with increase in Sn content. This increase in the average constraints results in the increase of the cross linking of chains between the atoms and hence leads to the compactness of the system as evidenced by <r>.

3.2 Lone Pair Electrons and Glass Forming Ability

In glassy alloys there are two types of interatomic forces. The first type of force is the short-range bonding interactions which are explained by valence force fields (VFF). The coordinates of the VFF are the bond vectors $\boldsymbol{R}_{ij}$ associated with directed s-p hybridized atomic orbitals connected to the nearest neighbors. These interactions are again of two types, i.e., two-body central forces 'α' associated with bond stretching, which depend on $\mathbf{R}_{ij}^2$, and three body non-central forces 'β' associated with bond bending, which depends on $\boldsymbol{R}_{ij} \bullet \boldsymbol{R}_{jk}$. The VFF has succeeded in representing the atomic interactions in large molecules[18] and covalent crystals [19, 20]. The second type of inter-atomic forces, which deserves special consideration, is the interaction between lone pair electrons. At short distances this force is repulsive, while at large distance it is attractive (Van der Waals force). Each chalcogen has pairs of lone pair electrons. The repulsive lone pair electron is a source of steric hindrance and has been used to define a minimum distance of approach. The attractive lone pair electrons are responsible for glass formation in chalcogenide alloys. It has been suggested [21] that, for a binary system the number of lone pair electrons must be greater than 2.6 and for ternary system the number must be greater than 1. The numbers of lone pair electrons, in the system under consideration, are calculated using the relation [21]

$$L = V - \langle r \rangle, \quad (2)$$

here L is the number of lone pair electrons and V is the number of valence electron of the whole system. The calculated value of lone pair electrons for $Se_{30}Te_{70-x}Sn_x$ at different values of x are

given in table 1. From table 1 it is evident that the minimum value of the number of lone pair electrons for the present system is 3.82, which shows that the system under study has a good glass forming ability. The system with large numbers of lone pair of electrons favors the glass formation as the strain energy of the system decreases with the increase in the lone pair electrons. The variation of lone pair electrons with Sn content is shown in fig.1. From the fig .1 it is clear that the number of lone pair electrons decreases with the increase in Sn content. The decrease in the lone pair electron might be due to the interaction between the Sn ion and lone pair electrons of bridging Se atoms.

Table 1. The values of various physical parameters, i.e., average coordination number <r>, average constraints N_{CON}, number of valence electron V, the number of lone pair electrons L, average heat of atomization $\overline{H}_S$ and average single bond energy $\overline{H}_S$/<r>, for $Se_{30}Te_{70-x}$ Sn_x system at different composition.

Composition	<r>	$N_{CON=}$ $N^{\alpha} + N^{\beta}$	V	L=V-<r>	$\overline{H}_S$ (kcal)	$\overline{H}_S$ /<r> (kcal/atom)
$Se_{30}Te_{70}$	2.00	2.00	6	4	47.02	23.51
$Se_{30}Te_{68.5}$ $Sn_{1.5}$	2.03	2.075	5.97	3.94	47.41	23.35
$Se_{30}Te_{67.5}$ $Sn_{2.5}$	2.05	2.125	5.95	3.90	47.67	23.25
$Se_{30}Te_{64.5}$ $Sn_{4.5}$	2.09	2.225	5.91	3.82	48.19	23.06

3.3 The Average Heat of Atomization

The heat of atomization of a ternary and higher order semiconductor compounds is a direct measure of the cohesive energy and the average bond strength for crystalline semiconductors [22] and is given by the relation

$$\overline{H}_S = \frac{\left((\alpha)H_S^{Se} + (\beta)H_S^{Te} + (\gamma)H_S^{Sn}\right)}{\alpha + \beta + \gamma}, \tag{3}$$

where α, β and γ are the atomic percentage of the elements Se, Te and Sn respectively and H_s^A is the heat of atomization of the element *A*. The average heat of atomization is calculated from equation (3), by using the values of heat of atomization of Se, Te and Sn as 49.4Kcal/atom, 46 Kcal/atom and 72 Kcal/atom, respectively from[18]. The calculated values of average heat of atomization $\overline{H}_S$ and average single bond energy $\overline{H}_S$/<r> are given in table 1. From table 1 one finds that with the increase in the Sn content or decrease in Te content the average single bond energy decreases. The variation of average single bond energy with Sn content is shown in fig. 2. From fig. 2 it is observed that the average single bond energy decreases with increase in Sn contents and is in agreement with the results[12] of Fouad et al.

3.4 Fraction of Floppy Modes

M.F. Thorpe [23] has suggested that there exist a finite fraction of zero frequency normal vibrational modes, called floppy modes. These floppy modes exists in the absence of weaker long range forces in the under coordinated networks. The fraction of floppy modes are related to the average coordination number by the relation

$$f = 2 - \frac{5}{6}\langle r \rangle. \tag{4}$$

The values of fraction of floppy modes are calculated from equation (4) and are reported in the table 2. From the table 2, it is clear that the fraction of floppy modes decreases with

increase in average coordination number. This shows that the system undergoes transition towards more rigidity with the increase in Sn content.

3.5 Mean Bond Energy and Glass Transition Temperature

The mean bond energy, <E>, is a function of average coordination number, the nature of the different bonds formed between atoms forming the glass and their energy. The mean bond energy for the $Te_xSe_ySn_z$ system is evaluated by Tichy Ticha approach [24], i.e.

$$\langle E \rangle = E_c + E_{rm} , \tag{5}$$

where E_c represents the energy contribution due to the heteropolar bonds and E_{rm} is the average bond energy per atom of the remaining matrix. The energy due to the heteropolar bonds, E_c, is defined as

$$E_C = P_r D_{hb} , \tag{6}$$

here P_r is the degree of cross linking which is given by

$$P_r = \frac{\alpha N_{Te} + \gamma N_{Sn}}{\alpha + \beta + \gamma} . \tag{7}$$

In equation (6) D_{hb} is the average heteropolar energy and is given by the relation[24]

$$D_{hb} = \frac{\alpha N_{Te} E_{Se-Te} + \gamma N_{Sn} E_{Se-Sn}}{\beta N_{Te} + \gamma N_{Sn}} , \tag{8}$$

The energy contribution due to the weaker bonds, E_{rm}, is given by the relation

$$E_{rm} = \frac{2[0.5\langle r \rangle - P_r] E_{Se-Se}}{\langle r \rangle} , \tag{9}$$

where $E_{A\text{-}A}$ is the contribution of homopolar bond energy and $E_{A\text{-}B}$ is the hetropolar bond energy contribution (values are not quoted here). In the above equations α, β, and γ are the atomic percentage of Se, Te, and Sn, respectively and N_{Se}, N_{Te} and N_{Sn} are their respective coordination numbers. The calculated values of the mean bond energy are listed in table 2 and are found to increase with the increasing Sn content.

The glass transition temperature of the present system is calculated from mean bond energy <E> using Tichy Ticha approach [24], i.e.

$$T_g = 311[\langle E \rangle - 0.9] , \tag{10}$$

and also from the E_g, optical energy gap, using the relation [25, 26]

$$T_g = T_0 + \delta(\langle r \rangle - 2) E_g / 32.2k , \tag{11}$$

where δ=0.55 and T_o=325±20 K are fitting constants[27]. In equation (11) E_g is the optical energy gap obtained from Shimakawa relation[28] and *k* is the Boltzmann constant. The values of glass transition temperature calculated by the two methods are reported in table 2. In table 2, T_{g1}

represents the transition temperature obtained from equation (10) and T_{g2} represents the results obtained from equation (11). The values obtained by two methods are comparable. From the table 2 it is inferred that the mean bond energy increases and hence the glass transition temperature increases due to the cross linking of the Se-Te chains with the Sn. The increase in the glass transition temperature with the addition of Sn may be due to more possibility of stronger bonds between Sn and Se. The variation of glass transition, Tg, temperature with mean bond energy in shown in fig. 3.

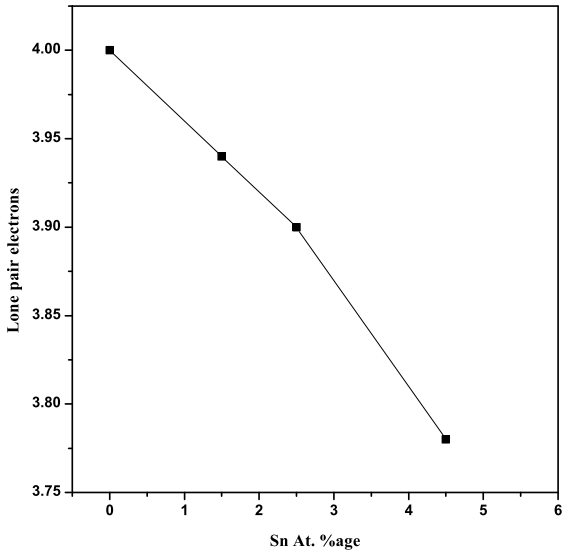


Fig. 1. Lone pair electrons versus Sn content in the $Se_{30}Te_{70-x}$ Sn_x system for different values of x, i.e., x= 0, 1.5, 2.5 and 4.5.

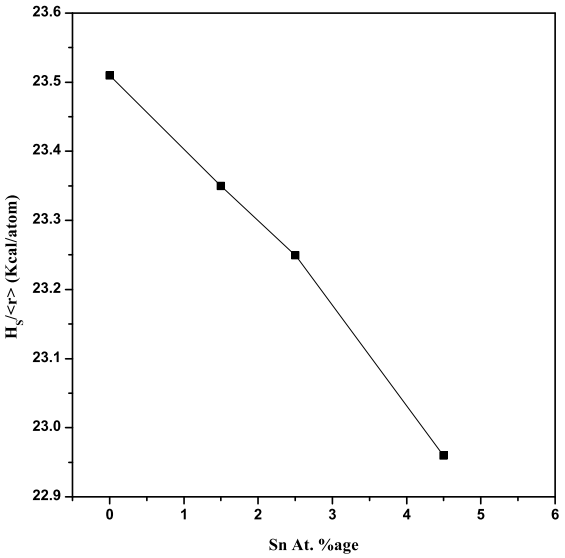


Fig. 2. Variation of average single bond energy $\overline{H}_S$ /<r> with Sn content in $Se_{30}Te_{70-x}Sn_x$ system for different compositions.

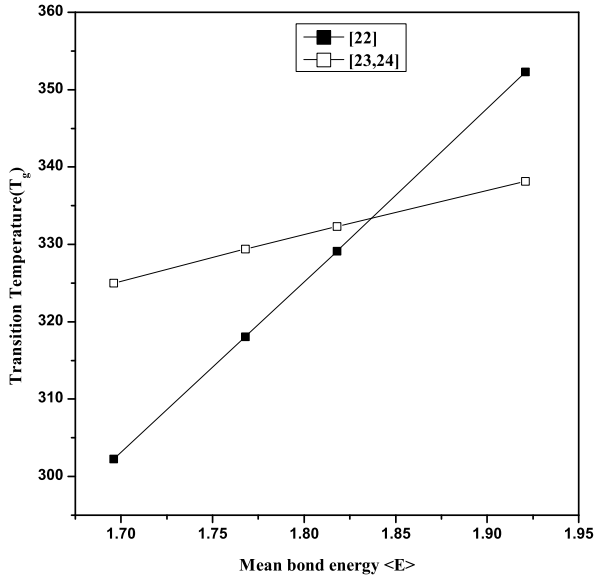


Fig. 3. Glass transition temperature T_g versus Mean bond energy <E> in the $Se_{30}Te_{70-x}Sn_x$ system for different concentration of Sn.

3.6 Cohesive Energy, deviation of Stoichiometry and Electronegativity

The cohesive energy (CE) is the measure of the stability of the material as it is the stabilization energy per atom of an infinitely large cluster of the material. The chemical bond approach (CBA) [29] is used to calculate the cohesive energy of the material under investigation. According to this approach the bond formation occurs in such a way that the bonds with higher energy are formed first and then the subsequent bonds with decreasing energy are formed, until the available vacancy of the atoms is fully occupied. In other words in the CBA the heteropolar bonds supercede the homopolar bonds. The cohesive energy is calculated by summing the bond energies of all the bonds expected in the material, i.e.,

$$CE = \sum (C_i D_i / 100) , \tag{12}$$

where C_i represent the number of expected chemical bonds and D_i is the energy of each bond respectively. The calculated values of cohesive energy for different compositions of $Se_{30}Te_{70-x}Sn_x$ are reported in the table 2. From table 2 it is observed that the cohesive energy increases with the addition of Sn, which means that average stabilization energy increases. This increase in the cohesive energy widens the gap between the bonding and antibonding orbitals and hence may leads to the increase in the optical energy gap[30]. The variation of cohesive energy with Sn content is shown in fig. 4. The electronegativity of the samples is calculated by Sanderson's principle[31]. According to this principle electronegativity of the alloy is the geometric mean of electronegativity of its constituent elements. The calculated values of the electronegativity for present system are listed in table 2. From the table 2 it is clear that the value of electronegativity goes on decreasing with the increase in Sn content.

The deviation of stoichiometry (R) is the ratio of the possible covalent bonds of chalcogen atoms to that of nonchalcogen atoms in the system. The values of R decide the nature of the material, i. e., for R > 1 the material is chalcogen rich and for R<1 the material is chalcogen poor. The value of R is obtained by the relation [24, 32]

$$R = \frac{XN_{Se} + YN_{Te}}{ZN_{Sn}}, \tag{13}$$

here X, Y, Z are the atomic fractions of Se, Te, Sn respectively. The calculated values of R are reported in table 2. From the table 2 it is observed that the minimum value of R is 21.22, which is much greater than 1 and hence confirms that the system under consideration is chalcogen rich system.

Table 2. The values of fraction of floppy modes f, mean bond energy <E>, glass transition temperature T_g, Electronegativity χ, cohesive energy and deviation of stoichiometry R for $Se_{30}Te_{70-x}Sn_x$ system at different compositions.

Composition	Fraction of floppy modes f	<E> (eV/atom)	Transition Temperature		χ	Cohesive energy(kcal/mol)	R
			T_{g1}	T_{g2}			
$Se_{30}Te_{70}$	0.333	1.696	302.28	325	2.115	37.35	∞
$Se_{30}Te_{68.5}$ $Sn_{1.5}$	0.308	1.768	318.07	329.41	2.110	37.63	65.66
$Se_{30}Te_{67.5}$ $Sn_{2.5}$	0.292	1.818	329.11	332.33	2.107	37.82	39
$Se_{30}Te_{64.5}$ $Sn_{4.5}$	0.258	1.921	352.28	338.13	2.101	38.23	21.22

3.7 Dielectric Properties

Dielectric analysis is important to understand the nature and origin of dielectric losses, which in turn may be useful in the determination of structure and defects in solids. Dielectric analysis measures the electric properties of a material as a function of frequency and temperature. This analysis may be used to determine the two fundamental electrical characteristics of the material, i.e. the ability to store the charge (capacitive nature) and ability to transfer the electronic charge (conductive nature). The frequency dependence of dielectric constant ε' and dielectric loss ε'' of $Se_{30}Te_{70-x}Sn_x$ system (pallets) is determined for different values of x, i.e., for x= 0, 1.5. 2.5 and 4.5. The values of dielectric constant ε' are calculated using the relation,

$$\varepsilon' = \frac{Cd}{\varepsilon_0 A}, \tag{14}$$

where C is the parallel capacitance of the pallet, A is the cross sectional area of the parallel surface of the pallets, d is the thickness of the pallet and ε_0 is the permittivity of free space. The variation of dielectric constant with frequency at room temperature is shown in fig. 5 (a). From the fig. 5 (a) it observed that the dielectric constant decreases with the frequency. This decrease is fast large at low frequencies as compared to higher frequencies that may be due to the electrode polarization effects[33-35]. The decrease in ε' with frequency at low frequencies may be explained in terms of the contribution of multicomponent polarizability, deformation and relaxation polarization.

The imaginary part of the dielectric constant ε'', i.e., dielectric loss, is determined from the relation,

$$\varepsilon'' = \varepsilon' \tan\delta, \tag{15}$$

where tan δ = tan(90-φ) is equivalent to the dissipation factor D obtained from the impedance analyzer. The variation of dielectric loss ε'' with frequency is plotted in fig. 5(b). From fig. 5(b) one can see that the dielectric loss ε'' follows the same trend as followed by the dielectric constant ε'. From, fig. 5 (a) and (b) it is noted that both the dielectric constant and dielectric loss increases with increase in the Sn concentration as earlier observed in the literature[11]. This increase in the dielectric parameters with Sn contents may be due to the increase of defect states in the band gap

near the Fermi level. This means that the hopping of the charge carriers in the defect states increases with increase in Sn concentration. From fig. 5 it is also observed that the ε' and ε'' decreases for x=4.5 this mean that there is a decrease in defect states in the band gap at this concentration. Further, the frequency dependence of the dielectric loss is found to follow the relation[36]

$$\varepsilon'' = C\omega^{m}, \tag{16}$$

where C is a constant. The ln (ε'') as a function of ln (ω) is plotted in fig. 6. The value of *m* can be calculated from the slope of the straight lines obtained in the fig. 6. The value of the exponent *m* so obtained is found to be negative which is consistent with earlier values of *m* obtained for the chalcogenide glasses[37]. According to Guintini equation[38] based on Elliott model for chalcogenide glasses the dielectric loss ε'' at a particular frequency is given by

$$\varepsilon'' = (\varepsilon_0 - \varepsilon_\infty)2\pi^2 N(ne/\varepsilon_0)^3 k_B T \omega^m \tau_0^m W_m^{-4}, \tag{17}$$

where $m = -4k_B T/W_m$, *n* is the no. of hopping electrons, ε_0 is the static dielectric constant, ε_∞ is the dielectric constant at high frequencies, N is the concentration of localized states, W_m is the maximum barrier height. For the present system the value of W_m is obtained as 0.035 eV which is comparable to the value of W_m for other chalcogenide materials [39,40]. The obtained value of W_m is consistent with the theory of hopping of charge carriers over a potential barrier between the defect states.

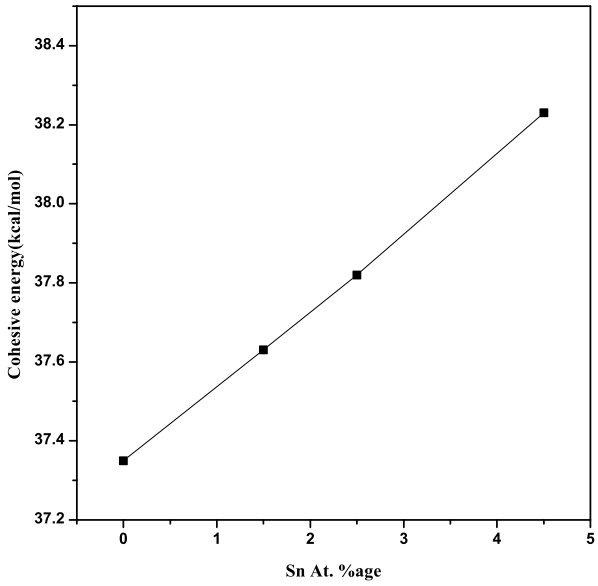
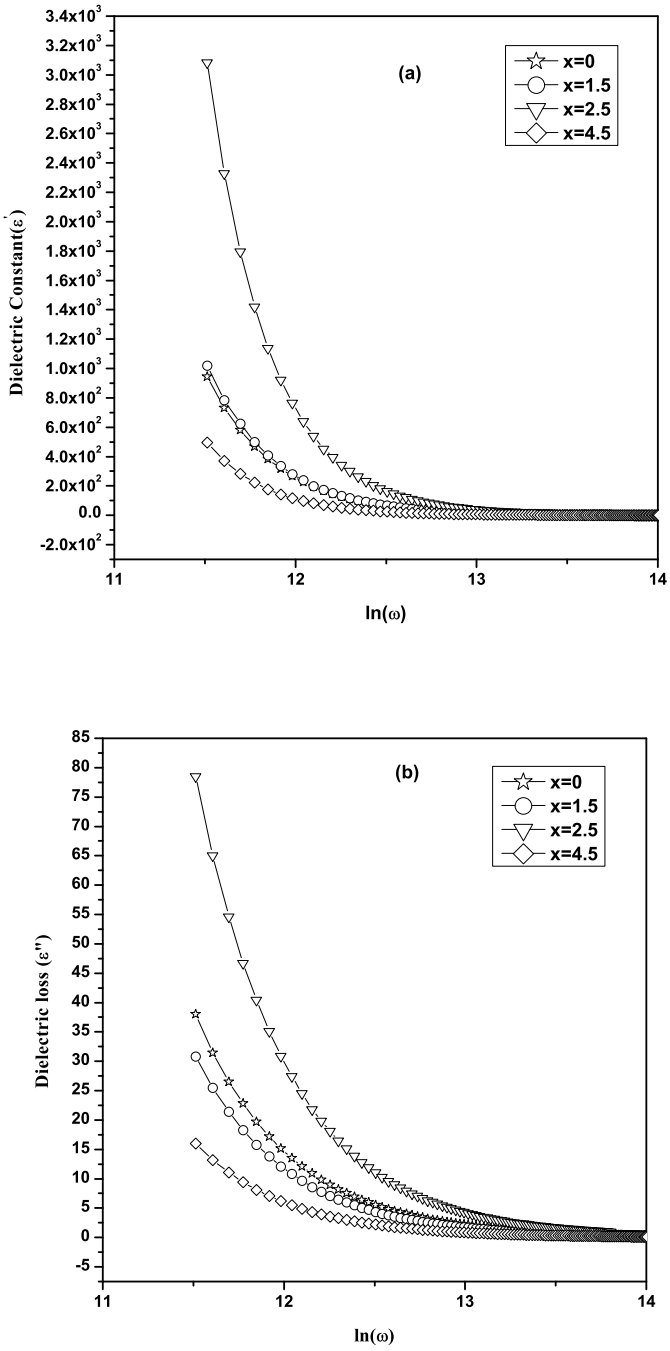


Fig. 4. Variation of Cohesive energy with Sn content in $Se_{30}Te_{70-x}Sn_x$ system for x= 0, 1.5, 2.5 and 4.5 .



F ig. 5. The frequency dependence of (a) dielectric constant ε′ and (b) dielectric loss ε″ at room temperature for different compositions of $Se_{30}Te_{70-x}Sn_x$ system.

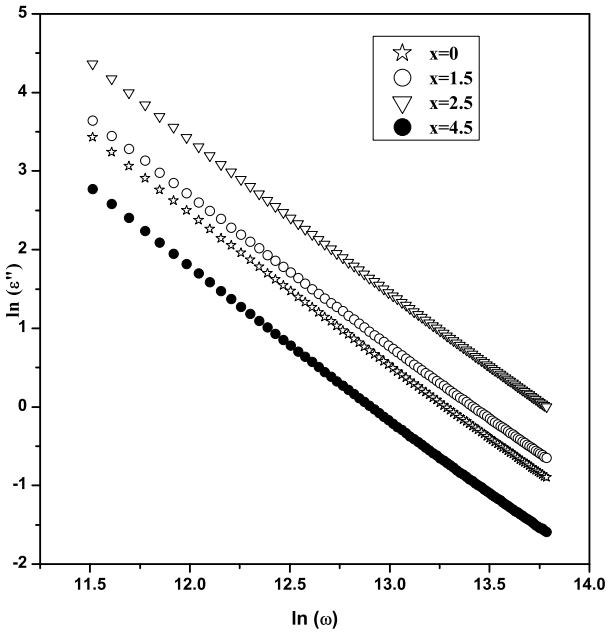


Fig. 6. ln ε″ versus ln(ω) for $Se_{30}Te_{70-x}Sn_x$ system at x= 0, 1.5, 2.5 and 4.5.

4. Conclusion

In the present work we have studied the effect of tin on the physical and dielectric properties of Se-Te glassy alloy. The material is prepared by the well known melt quench technique. The physical properties such as average coordination number, constraints, the number of lone pair electrons, the average heat of atomization, mean bond energy and glass transition temperature and cohesive energy are determined theoretically. The present study shows that $Se_{30}Te_{70-x}Sn_x$ is a good glasses former. Our predictions for the physical properties are consistent with the earlier predictions for chalcogenide glasses. From the lone pair and cohesive energy calculations one can say that the material is stable and good chalcogenide glass. For dielectric study the bulk material is converted into pallets and the pallets are coated with silver paste to make good contacts with electrodes. It is found that the dielectric constant and dielectric loss decreases with frequency. This decrease in the dielectric parameters with frequency can be attributed to the contribution of multicomponent polarization at low frequencies. Further, the increase in the dielectric parameters with Sn concentration shows that there is an increase in the defect states in the band gap at the Fermi level. The maximum value of barrier height (W_m) obtained from the dielectric loss is found in agreement with the earlier values obtained for chalcogenide glasses.

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