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DIELECTRIC DISPERSION IN $Te_9Se_{72}Ge_{19-x}Sb_x$ (x = 8, 9, 10, 11, 12) CHALCOGENIDE GLASSY ALLOY

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Dielectric dispersion and conductivity of $Te_9Se_{72}Ge_{19-x}Sb_x$ (x = 8, 9, 10, 11, 12) chalcogenide glassy alloy has been studied in frequency range 500 Hz to 1 M Hz and temperature range (300K-363K) below glass transformation region. Effect of temperature and frequency on dielectric response and conductivity of the material has been explained thoroughly using appropriate models. Activation energy of ac conductivity and other dielectric parameters have also been calculated to explore material's conduction mechanism and dielectric behaviour.

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1. Introduction

In diverse field of amorphous semiconductors, non-oxide glasses are in focus due to their extensive applications in technological fields. These glasses have low value of dielectric constant as compared to oxide glasses**.** Oxide glasses fall in the category of ceramics whereas the chalcogenide glasses fall in the category of semiconductors, as the electrical conductivity of these glasses ranges from 10^{-2}–10^{-16} $\Omega^{-1}cm^{-1}$ [1]. Chalcogenide glasses are successfully applied in photo electronics, thermoelectric devices, optical switching, ultra-fast telecommunication devices and phase change memory devices [2-9]. The dielectric behaviour of chalcogenide glasses reveals the structural information which is very useful for understanding the conduction mechanism. The dielectric relaxation in amorphous semiconductors is very useful to understand the origin of the dielectric losses and to determine the density of defect states in solids. Dielectric dispersion is the dependence of permittivity of a dielectric material on the frequency of an applied electric field. A lag between changes in polarization and changes in the electric field results in a dielectric loss. Electrical properties of chalcogenide glasses provides useful information about the band gap, density of states, mechanism of conduction, type of dominant charge carriers, and nature of defect states of the material [10,11]. The presence of defect states (D^+, D^-) in the mobility gap is the common feature of these glasses because of the absence of long range order in the atomic arrangements [12]. The density of defect states in mobility gap in the chalcogenide glasses controls the optical and electrical properties of the material. The frequency dependence of ac conductivity confirms the hopping conduction mechanism. Therefore, the measurement of ac conductivity of amorphous chalcogenide glasses can be used to obtain information about these states. The correlated barrier hopping (CBH) [13, 14] model of bipolarons (i.e., two-electron hopping charged defects D^+ and D^-) has been proposed to explain the ac conduction mechanism in glassy amorphous semiconductors. According to Guintini model [15], each pair of D^+ and D^- is assumed to form a dipole which creates a potential barrier over which the carrier can hop. This may lead to a decrease in the density of states due to the conversion of some bipolaron states (D^+, D^-) into a single polaron state (D^o) according to the relation $(D^+) + (D^-) \rightarrow 2(D^o)$. This theory successfully explained many features of the ac conductivity at low temperature particularly the parameter 's'. However, it does not explain the high temperature behaviour so well,

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particularly in the low frequency range. Shimakawa [16] suggested that the thermal excitation of D^+ and/or D^- states produces the D^o states and single polaron hopping (i.e. one-electron hopping between D^o and D^+ or D^-) is responsible at high temperature. Mostly selenium based glasses are preferred because of their commercial and technological applications. Pure Se is a good glass former, but its low glass transition temperature limits its applicability. Certain cross-linking agents like Ge, increase the thermal stability and glass forming ability by compromising on the optical and electrical properties by decreasing the defect states. Ge-Se glasses have been widely studied due to their extended amorphization range [17]. The addition of heavy metal impurities like Sb, enhances the optical properties due to creation of compositional and configurational disorder in the system. Sb behaves as a good tailoring agent to tune the optical and electrical properties in present Se-Te-Ge glassy system as it increases the density of defect states. Mehra et al. [18] have reported the creation of some new defect states with Sb contents in Se-Te binary glassy alloys during photoconductivity measurements. The present focus of this study is to report some electrical properties and determine the influence of antimony content on the electrical conductivity and dielectric behaviour of the Se-Te-Ge glassy system.

2. Experimental Details

The bulk samples of the $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \le x \le 12$) are prepared by melt quenching technique. High purity (99.999%) Se, Te and Sb (Alpha Aesar) and Ge (Acros Organics) are weighed according to their atomic percentages and are sealed in quartz ampoules at a pressure of ∽10^{-5} mbar. The sealed ampoules are kept inside a furnace, where the temperature is increased up to 1273^{o}K at a heating rate of 3–4^{o}K per min. These ampoules are frequently rocked for 15 h at the highest temperature to make the melt homogeneous. The quenching is done in ice cold water. The material is extracted by breaking the ampoules and is grinded to fine powder. The amorphous nature of all the samples at room temperature is ascertained by Panalytical X'Pert-Pro diffractometer (PW 3050/60) by using Cu target source (λ = 1.5483 Å) and diffractograms are shown in figure 1.

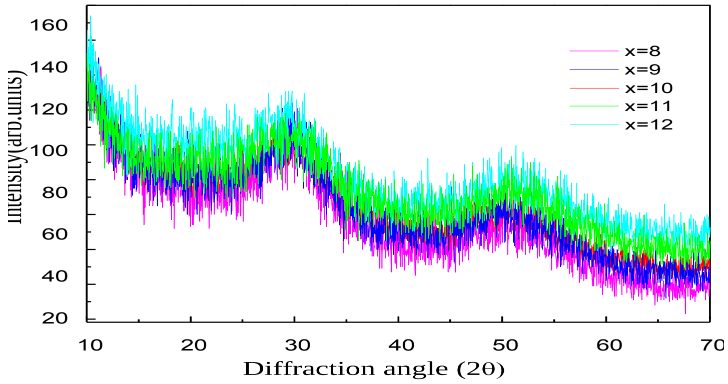

Fig. 1 X-ray diffractograms of the $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \le x \le 12$) glassy alloys

The glassy nature of the material is conformed using modulated differential scanning calorimetery. The glass transition temperature of the material varies from 371K to 362 K as Sb content changes in the composition [19] as shown in figure 2. To study the dielectric behaviour of the material, the pallets of diameter ∽10mm and thickness∽1.5 mm of the material are prepared by pressing the finely grinded powder in a steel die by using a hydraulic press under a load of 3-4 tons.

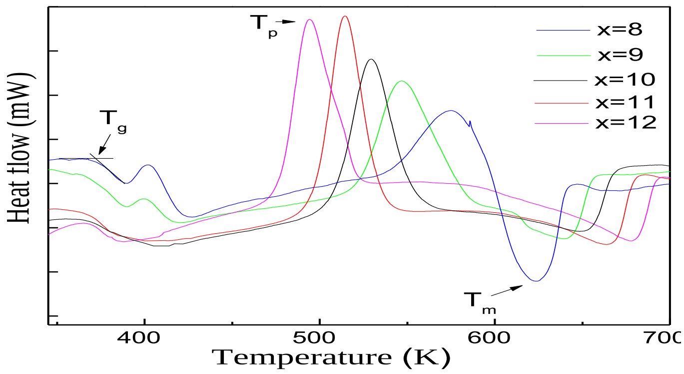

Fig. 2 DSC thermograms of $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \leq x \leq 12$) at heating rate 10K per minute

The pellets are coated with silver paste on both sides to ensure good electrical contact with the electrodes of the sample holder. The dielectric behaviour and ac conductivity of these pellets are analyzed with the impedance analyzer, Wayne Kerr 6500B. The response of the conduction mechanism and dielectric behaviour has been studied in the frequency range (500Hz -1MHz) and temperature range (300K to 363K) below the glass transformation region.

3 Results and Discussion

3.1 Frequency and Temperature Dependence of ac Conductivity:

3.1.1. ac Conductivity:

The conducting response of the material is studied in the frequency range 500 Hz to 1MHz at different temperature ranging from 300K to 363K. The total conductivity is given by [20]

$$\sigma(\omega) = \sigma_o + A\omega^s \tag{1}$$

here $\omega = 2\pi f$ is the angular frequency. A is pre factor and s is the frequency exponent. It has been found that both of these factors have the temperature dependence. The value of 's' approaches to 1 at room or low temperature. The term σ_o is low frequency (or dc) conductivity. The value of dc conductivity is very small as compared to ac conductivity. So σ_o can be neglected in the above equation and total conductivity is approximately equal to ac conductivity.

In semiconductors and some disordered materials, frequency dependence of ac conductivity is expected due to conduction in localized states [21]. The low temperature ac conduction can be explained by considering bipolaron hopping between D^+ and D^- states whereas the higher temperature behaviour is due to thermally activated single polaron hopping. At higher temperatures a number of thermally generated D^o states are produced with a temperature dependent concentration [14]. A common feature of all the amorphous semiconductors is that the ac conductivity $\sigma_{ac}(\omega)$ changes with frequency according to the following relation

$$\sigma_{ac}(\omega) = \mathrm{A}\omega^{\mathrm{s}} \tag{2}$$

In the above equation ($s \leq 1$) is the frequency exponent and is given by the following relation [14, 22]

$$s = \frac{d(\ln(\sigma_{ac}))}{d(\ln(\omega))} = 1 - \frac{6K_BT}{[W_m + k_BTln(\omega\tau_o)]} \tag{3}$$

here W_m is the maximum barrier height over which the electrons hop, T is the absolute temperature and k_B is the Boltzmann constant, ω is the angular frequency and τ is the relaxation time for the electrons to hop over the barrier height *W*. The variation of ac conductivity with frequency for the invested composition at different temperature has been shown in figure 3. The ac conductivity increases with increasing Sb concentration because the addition of Sb in the glassy matrix increases the density of charge carriers and band tailing.

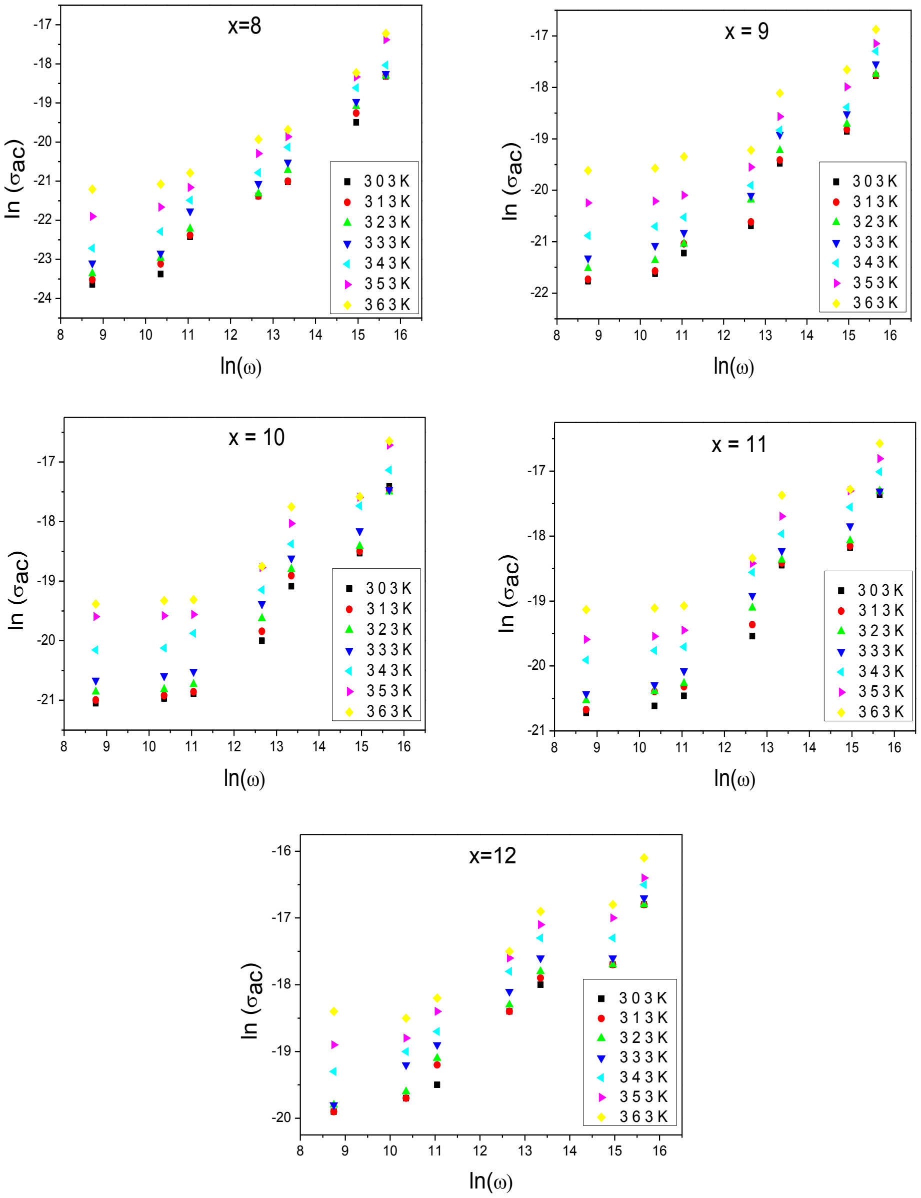

Fig. 3 Plots of $ln(\sigma_{ac})$ *versus* $ln(\omega)$ *at different temperature for* $Te_9Se_{72}Ge_{19\text{-}x}Sb_x$ ($8 \le x \le 12$) *glassy alloys*

The value of frequency exponent ‘s’ is calculated from the slopes of the plots of $ln(\sigma_{ac})$ versus $ln(\omega)$and its variation with temperature for the composition is shown in figure 4. The value of ‘s’ is found to be decreasing as the temperature increases and its value approaches to 1 as the temperature reduces. At a low temperature, ac conductivity is observed to be almost constant in the low frequency range and is found to be increasing exponentially in high frequency range. The behaviour of ‘s’ has been explained with correlated barrier hopping (CBH) model proposed by Elliot. According to this model, hopping takes place between close pairs of defect states that have a random distribution of the charged defect states and these pairs are called as non intimate valence alternation pairs (NVAP’s).Various authors have shown such observations [22-26].

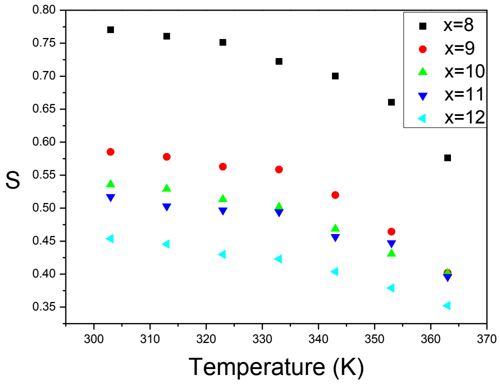

Fig. 4 Variation of frequency exponent 's' with temperature for $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \leq x \leq 12$) glassy alloys

The ac conductivity also increases as the temperature increases. The values of ac conductivity at different temperatures and frequencies for all glassy alloys are tabulated in table 1.

Table 1 *Values of ac conductivity of $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \leq x \leq 12$) glassy alloys at different temperatures and frequencies*

x	Frequency	ac conductivity(Ω^{-1} cm^{-1})			
		303K	313K	353K	363K
x=8	500KHz	3.41×10^{-9}	4.33×10^{-9}	1.09×10^{-8}	1.21×10^{-8}
	1MHz	1.10×10^{-8}	1.11×10^{-8}	2.82×10^{-8}	3.33×10^{-8}
x=9	500KHz	6.48×10^{-9}	6.71×10^{-9}	1.54×10^{-8}	2.16×10^{-8}
	1MHz	1.91×10^{-8}	1.93×10^{-8}	3.58×10^{-8}	4.71×10^{-8}
x=10	500KHz	7.48×10^{-9}	9.25×10^{-9}	2.29×10^{-8}	2.32×10^{-8}
	1MHz	2.16×10^{-8}	2.58×10^{-8}	5.50×10^{-8}	5.88×10^{-8}
x=11	500KHz	1.27×10^{-8}	1.31×10^{-8}	3.05×10^{-8}	3.13×10^{-8}
	1MHz	2.87×10^{-8}	3.03×10^{-8}	5.02×10^{-8}	6.35×10^{-8}
x=12	500KHz	2.10×10^{-8}	2.11×10^{-8}	3.96×10^{-8}	5.08×10^{-8}
	1MHz	4.98×10^{-8}	5.06×10^{-8}	7.92×10^{-8}	1.02×10^{-7}

The temperature dependence of the ac conductivity for $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \leq x \leq 12$) glassy alloys is studied in the temperature range (300K-363K) at different frequencies and is represented in figure 5.

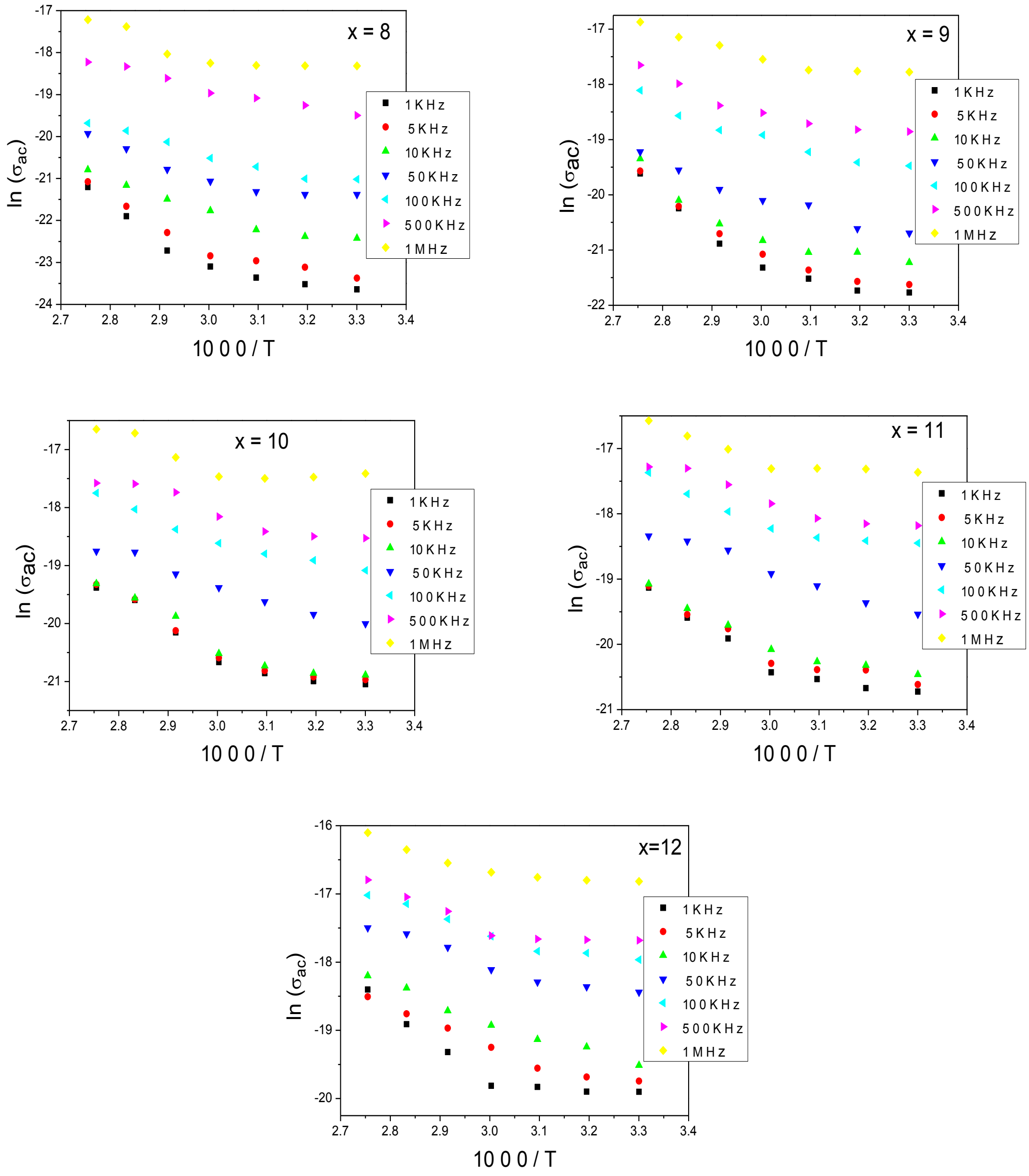

Fig.5 Plots of $ln(\sigma_{ac})$ versus 1000/T at different frequencies for $Te_9Se_{72}Ge_{19\text{-}x}Sb_x$ ($8 \le x \le 12$) glassy alloys

Figure 5 shows the variation of $ln(\sigma_{ac})$ versus 1000/T at different frequencies and approximately linear fits are found. It has been found that ac conductivity increases with an increase in the temperature as well as with frequency for all the samples of the composition. This shows that the ac conductivity in different localized states in the gap is a thermally activated process. In the intermediate temperature range the ac conductivity can be related with temperature [25] as follows

$$\sigma_{ac}(\omega) = \sigma_o e^{\left(\frac{-\Delta E_{ac}}{K_b T}\right)} \qquad (4)$$

here σ_o is constant. The activation energy $\Delta E_{ac}(\omega)$ of ac conduction is calculated at different frequencies using this equation and the variation of ΔE_{ac} with frequency for all the samples have been shown in figure 6.

It is clear that $\Delta E_{ac}(\omega)$ decreases with increasing frequency. This decrease may be attributed to the increase of applied field frequency which enhances the electronic jump between the localized states [22]. The results of the activation energy obtained for the samples support the hopping conduction mechanism.

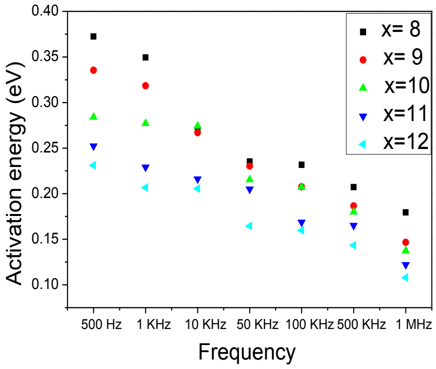

Fig. 6 Variation of ac activation energy versus frequencies for $Te_9Se_{72}Ge_{19-x}Sb_x$ *(*$8 \leq x \leq 12$*) glassy alloys*

3.2 Dielectric Dispersion and Behaviour with Temperature:

3.2.1 Dielectric constant:

The following equation has been used to calculate the values of dielectric constant ($\mathcal{E}'$)

$$\mathcal{E}' = \frac{Cd}{\mathcal{E}_o A} \tag{5}$$

In the above equation *C* is the capacitance of the material, *d* is thickness of the pellet, *A* is the area of the pellet and $\mathcal{E}_o$ is the permittivity of free space. The dielectric constant is found to remain constant with frequency at the room temperature as shown in the figure 7 (inset). Dipoles are unable to orient themselves at room temperature and the orientational polarization decreases with increase in the frequency. Hence dielectric constant remains constant at room temperature with increase in the frequency [23].

3.2.1 Dielectric Loss:

The dielectric loss is the imaginary part of the dielectric susceptibility and can be calculated using the relation [23]

$$\mathcal{E}'' = \mathcal{E}' tan(\delta) \tag{6}$$

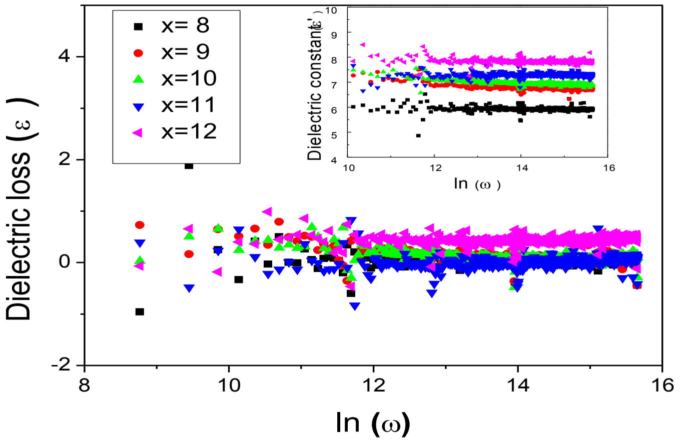

Fig. 7 Variation of dielectric constant and dielectric loss with frequency at room temperature

here $tan(\delta)$ is the dissipation factor *(D)* or the loss factor obtained by the impedance analyzer which symbolizes the inherent dissipation of electric energy in the dielectric material. It has been found that the dielectric loss decreases initially with frequency up to frequency 50 K Hz and after that remains constant. With an increase in the Sb content in the glassy matrix both the dielectric constant and dielectric loss increases as is clear from the figure 7.

The behaviour of dielectric constant and loss is studied in the frequency range 500 Hz to 1MHz with varying temperature below glass transition temperature for all the samples of the composition. The dielectric loss (ε'') obeys a power law with angular frequency (ω) for all the samples of the compositions [15, 27] and is given by

$$\varepsilon'' = A\omega^m, \quad m < 1 \tag{7}$$

here *A* is constant. Figure 8 shows the plots of dielectric loss (ε'') versus $ln(\omega)$ for the glassy alloys. The loss behaviour is also confirmed from the straight line graph between $ln(\varepsilon'')$ and $ln(\omega)$ shown in figure 9. The values of power 'm' calculated from the slope of these plots have been found negative for all the samples. The values of m are plotted as a function of temperature shown in figure 10. To explain the dielectric relaxation in chalcogenide glasses, Guintini [15, 27] have proposed a model based on Elliot's [13, 28] idea of hopping of charge carrier over the potential barrier formed due to columbic interaction between neighbouring sites forming dipoles between the charged defect states. The defect states D^+ and D^- which forms dipoles are also responsible for the transport properties of the material. The relaxation time of charge carriers depends upon the activation energy. As suggested by this model, the dielectric loss at a particular frequency and temperature range where the dielectric dispersion occurs is given by [15]

$$\varepsilon'' = (\varepsilon_o - \varepsilon_\infty)\, 2\pi^2 N \left(\frac{ne^2}{\varepsilon_o}\right)^3 \kappa_B T \tau_o^m W_m^{-4} \omega^m \tag{8}$$

where *n* is the number of electrons that hop, *N* is the concentration of localized sites, ε_o is static dielectric constant, ε_∞ dielectric constant at infinitely high frequencies. W_m is the maximum barrier height which can be calculated from the slopes of the straight lines obtained from plots between *'m'* and temperature are shown in figure 10**.** The value of *m* is related to temperature as follows

$$m = \frac{-4\kappa_B T}{W_m} \tag{9}$$

The relaxation time is also given by

$$\tau = \tau_o exp(\frac{W}{K_B T}) \tag{10}$$

It has been observed that there is decrease in the dielectric loss with increase in frequency and increases with rise in temperature which is clear from equation (8) also. The values of W_m are calculated by using equation (9) and these values are found as 0.1214eV, 0.1120eV, 0.1169eV, 0.06365eV and 0.0636eV for samples at x= 8, 9, 10, 11, 12 respectively.

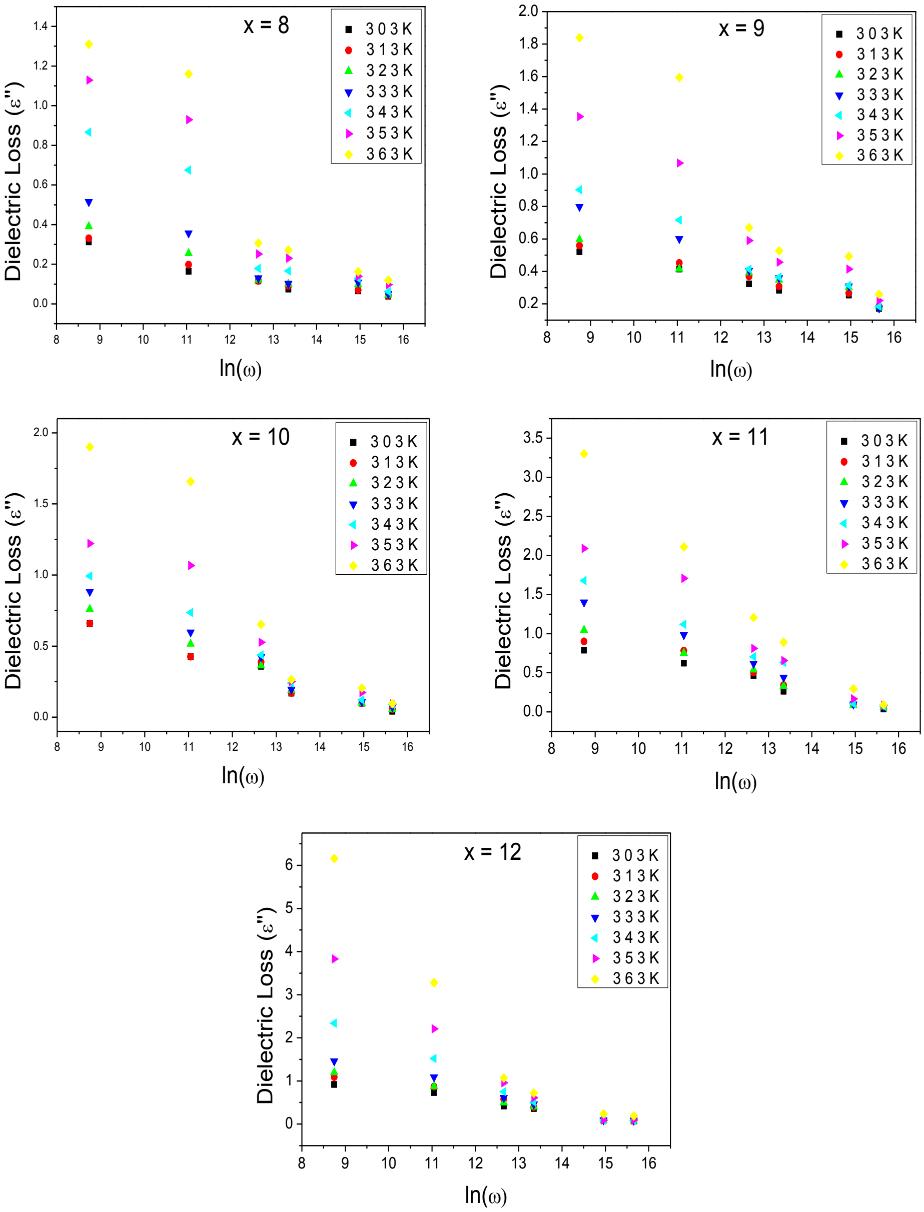

Fig. 8 Variation of ε'' *with* $\ln(\omega)$ *at different temperatures for* $Te_9Se_{72}Ge_{19-x}Sb_x$ *(*$8 \le x \le 12$*) glassy alloys*

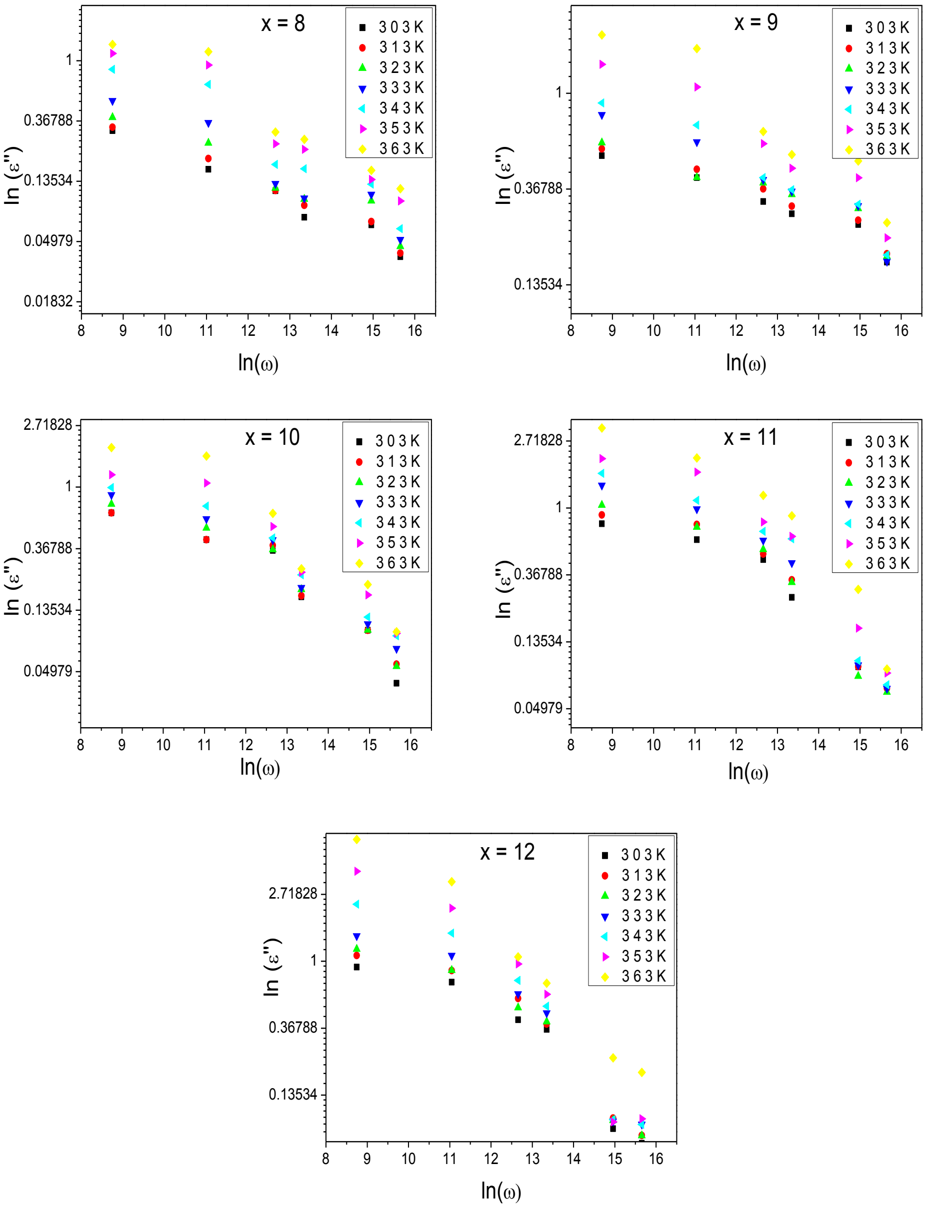

Fig.9 Variation of $ln(\varepsilon'')$ *with* $ln(\omega)$ *at different temperatures for* $Te_9Se_{72}Ge_{19-x}Sb_x(8\leq x \leq 12)$ *glassy alloys*

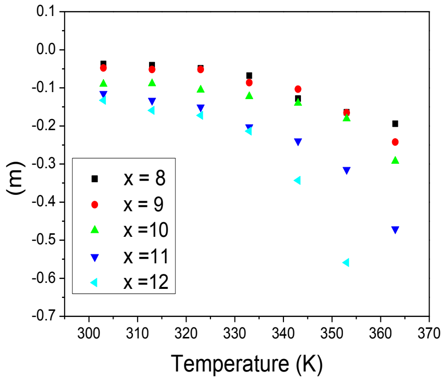

Fig. 10 Variation of parameter 'm' with temperature for $Te_9Se_{72}Ge_{19-x}Sb_x$ *(*$8 \leq x \leq 12$*) glassy alloys*

It has been observed that the dielectric loss increases as temperature increases and is having different values at different frequencies. The dielectric loss is also frequency dependent and it is large at low frequencies and at higher frequencies this loss decreases.

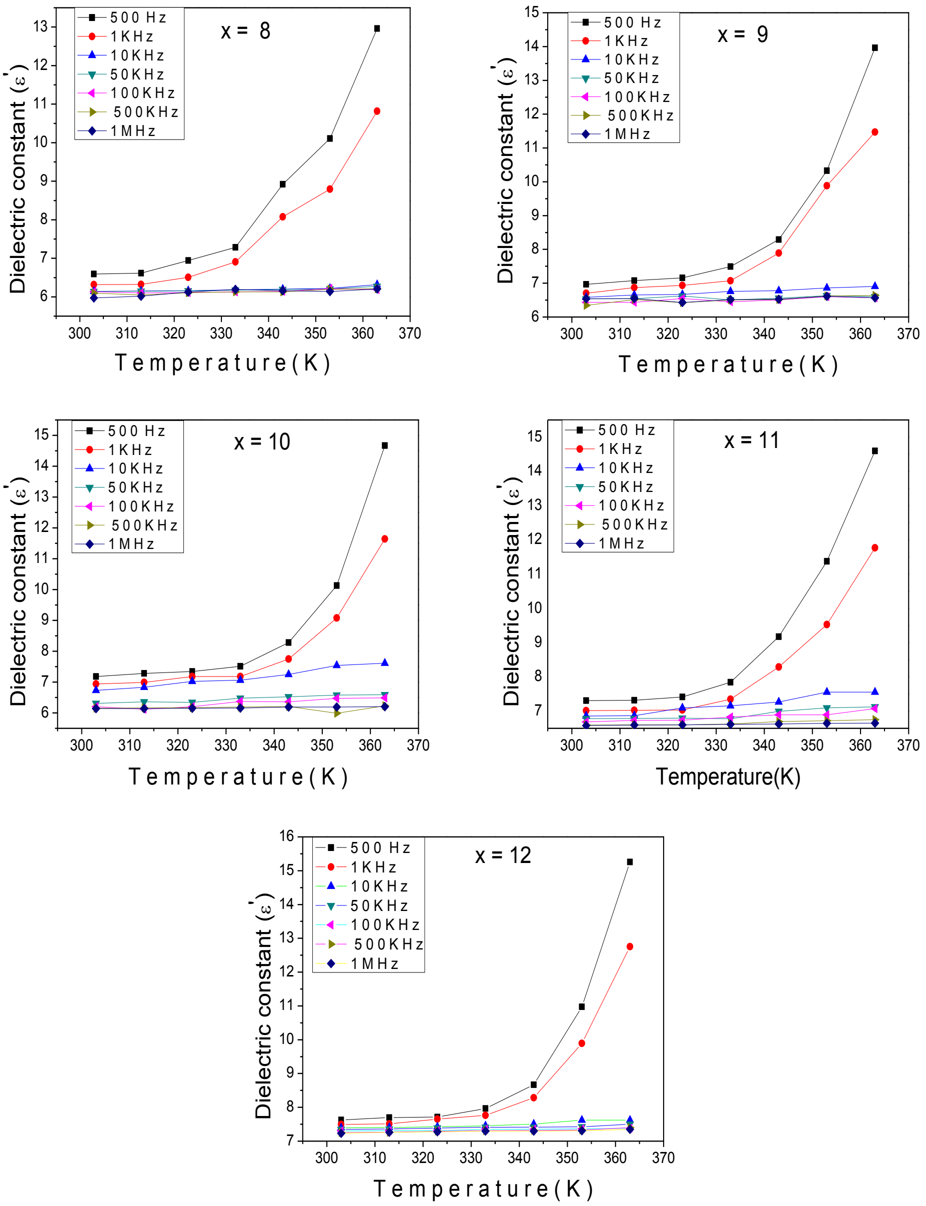

Fig. 11 Variation of dielectric constant (ε') with the temperature at different frequencies in $Te_9Se_{72}Ge_{19-x}Sb_x$ *(*$8 \leq x \leq 12$*) glassy alloys*

Figure 11 shows the temperature dependence of dielectric constant **(ε')** at different frequencies for $Te_9Se_{72}Ge_{19-x}Sb_x$ ($8 \leq x \leq 12$) glassy alloys. There is large variation in dielectric constant with increase in temperature at lower frequencies; dielectric constant at higher frequency above 10 KHz becomes approximately constant with temperature.

4. Discussion

The behaviour of dielectric constant with frequency and temperature depends upon various kinds of polarization such as ionic, electronic, and orientational and space charge polarization. The electronic polarization arises from the displacement of the valence electrons relative to the positive nucleus which can occur at frequencies upto10^{16}Hz. The ionic polarization is due to the displacement of negative and positive ions with respect to each other and the maximum frequency at which ionic polarization takes place is 10^{13} Hz. When molecules possess the permanent dipole moments which change their orientations on the application of electric field, it is called orientational polarization and this takes place at frequencies up to 10^{10} Hz. Space charge polarization is due to impedance mobile charge carriers by interfaces. The space charge polarization occurs at a frequency ranging from 1Hz to10^{3}Hz. The variation of dielectric constant with ac field frequency depends on all these polarizations [26]. When the applied field frequency is increased, the orientational polarization decreases as it takes more time than electronic polarization. The dipoles are not able to respond sufficiently to follow the field. Hence dielectric constant decreases with increasing frequency and approaches to a constant value at higher frequency.

On the other hand increase in dielectric constant with the rise in temperature over the whole frequency range can be attributed to the fact that orientational polarization depends upon the thermal motion of molecules. These dipoles remain frozen as they cannot orient themselves at low temperature. The facilitation of the orientation of the dipoles with increase in temperature increases the orientational polarization. Hence the dielectric constant increases as temperature increases. At higher temperature dipoles become rotationally free and the effect of molecular interaction energy reduces than thermal energy [29]. With increase in Sb concentration there is an increase in density of defect states in the material and hence the dielectric constant increases with the increase in Sb concentration. The different bonds present in the material possess the covalent character whose percentage can be calculated as;

Covalent character $= 100\% \exp(-0.25(\chi_A - \chi_B)^2)$, here χ_A and χ_B are the electronegativities of atoms A and B respectively. The percentages of covalent character for different bonds are calculated as Te-Se 95.06%, Sb-Se 93.44 % and Ge-Se 92.96%. Hence the ionic polarization does not have the significant effect in the total polarization, as material has the good percentage of covalent character [30].

In our composition, dielectric loss ($\boldsymbol{\varepsilon}''$) decreases with frequency and increases with an increase in the temperature. Dielectric loss ($\boldsymbol{\varepsilon}''$) also depends upon polarization. There is the frequency dependence of the dielectric loss and found to follow the $\varepsilon'' \propto \omega^m$ relation. The increase in dielectric loss ($\boldsymbol{\varepsilon}''$) with temperature can be explained according to Stevels [31]. The dielectric loss depends on conduction losses, dipole losses and vibration losses. The conduction losses involve the migration of ions over a large distance, when the ions jump over the highest potential. As these ions move they impart some of their energy to lattice in the form of heat and this heat lost per cycle of field is proportional to σ/ω. As the conductivity is small at low temperature, these losses are small. As the temperature increases, the conductivity increases, conduction losses also increase. So the dielectric loss increases with rise in the temperature and decreases with increasing frequency as the conduction losses are inversely proportional to frequency. At low frequency, the migrations of ions are the source of the dielectric loss. At moderate or low frequency range there is high value of dielectric loss due the conduction loss of the ions migration and ion polarization loss. At high field frequency dielectric loss (ε'') decreases as ions vibrations is the only source of dielectric loss.

5. Conclusion

The material has been prepared by the melt quenching technique. For the dielectric study of the material, pallets coated with silver are investigated. Dielectric dispersion has been studied below the glass transformation region in frequency range 500 Hz to 1 M Hz. Conductivity has also been studied and found to increase with rise in temperature which shows the semiconducting behaviour of the material along with frequency dependence showing hopping conduction mechanism in the material. Conductivity and dielectric response both have the temperature and frequency dependence exploring the material’s nature.

References

[1] K. Hulls, P. W. Mcmillan, J. Phys. D: Appl. Phys. **5,** 865 (1972).
[2] A.M. Andriesh, Semiconductors. **32,** 867 (1998).
[3] B. T. Kolomiets, V. M. Lyubin, Physica status solidi. **17,** 11 (1973).
[4] A. P. Gonc ¸ Alves, G. Delaizir, E.B. Lopes, L. M. Ferreira, O. Rouleau, C. Godart, Journal of ELECTRONIC MATERIALS. (2011).
[5] M. P. Slankamenac, S.R. Luki´c, M.B Živanov, Semicond. Sci. Technol. **24,** 085021 (2009).
[6] P. J. Walsh, R. Vogel, J. E. Edgar, Phys. Rev. **178,** 1274 (1969).
[7] S. Spalter , H. Y. Hwang , J. Zimmermann, T. Ktsufuji, T. Katsufuji, S.W. Cheong, M.E. Lines ,Optical Fiber Communication Conference. **3** (2000).
[8] S. Raoux, F. Xiong , M. Wuttig , E. Pop, Materials Research Society. **39** (2014)
[9] M. Nardone, M. Simon, I. V. Karpov, V.G. Karpov, J. Appl. Phys. **112,** 071101 (2012).
[10] J. M. Marshall, Rep. Proj. Phys. **46,** 1235. (1983)
[11] S. Gautam, A. Thakur, S. K. Tripathi, N. J Goyal, Non-Cryst. Solids. **353**, 1315 (2007).
[12] S. K. Tripathi, V. Sharma, A. Thakur, J. Sharma, G. S.S. Saini, N. Goyal, J. Non-Cryst. Solids. **351,** 2468 (2005).
[13] S.R. Elliott, Philos. Mag. B **36**, 1291 (1977).
[14] S.R. Elliott, Adv. Phys. **36,** 135 (1987).
[15] J.C. Guintini, J. V. Zanchetta, D. Jullen, R. Eholle, P. Hoenou, J. Non Cryst. Solids. **45,** 57 (1981)
[16] K. Shimakawa, Philos. Mag. B. **46,** 123 (1982).
[17] X. Fang, W. J. Bresser, P. Boolchand, Phys. Rev. Lett. **78,** 4423 (1997).
[18] R. M. Mehra, G. Kaur, P. C. Mathur, Phy. Solid State Commun.**85,** 29 (1993).
[19] A. V. Nidhi, V. Modgil, V. S. Rangra, J Therm Anal Calorim. **121,** 559 (2015).
[20] W. K. Lee, J.F. Liu, A. S. Nowick, Phys. Rev. Lett. **67,** 1559 (1991).
[21] J. Sharma, S. Kumar, Physica B **407**, 457 (2012)
[22] I. S. Yahia, N. A. Hegab, A. M. Shakra, A. M. Al-Ribaty, Physica B. **407,** 2476 (2012).
[23] N. A. Hegab, M. A. Afifi, H. E. Atyia, M. I. Ismael, Acta Phys. Polon A. **119,** 416 (2011).
[24] A. Sharma, N. Mehta, A. Kumar, J. Mater. Sci. **46,** 4509 (2011).
[25] N. Chandel, N. Mehta, A. Kumar, Curr. Appl. Phys. **12,** 405 (2012).
[26] N. A. Hegab, H. M. El-Mallah, Acta Phys.Polon.A. **116** (2009).
[27] J.C. Giuntini, J. V. Zancheha, J. Non Cryst. Solids. **34,** 419 (1979).
[28] S. R. Elliott, Solid State Commun. **27,** 749 (1978).
[29] D. K. Goel, C. P. Singh, R. K. Shukla, A. Kumar, J. Mater. Sci. **35,** 1017 (2000).
[30] L. J. Pauling, Nature of the Chemical Bond, Cornell University Press, New York, 1960.
[31] J. M. Stevels, H. Physik, in: Flugge (Ed) Springer, Berlin **350,** (1975)**.**