Journal of Non-Oxide Glasses Vol. 8, No 2, 2016, p. 47 - 57

DEPENDENCE OF DIELECTRIC PARAMETERS AND A.C. CONDUCTIVITY ON FREQUENCY AND TEMPERATURE IN BULK $Se_{90}Cd_8In_2$ GLASSY ALLOY

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In the present paper the effect of frequency and temperature on dielectric parameter and a.c. conductivity of $Se_{90}Cd_8In_2$ glassy alloy has been reported.$Se_{90}Cd_8In_2$ chalcogenide semiconducting glassy alloy has been prepared by melt quench technique. The prepared glassy alloy has been characterized by techniques such as scanning electron microscopy (SEM) and energy dispersive x-ray (EDAX). The effect of frequency and temperature on dielectric parameters and a.c. conductivity have been investigated in the frequency range 5×10^2Hz - 1×10^5Hz and in temperature range 308-333K. It is observed that a.c. conductivity $\sigma_{ac}(\omega)$, dielectric constant (ε') and dielectric loss factor (ε'') depends on frequency and temperature. The frequency dependence of $\sigma_{ac}(\omega)$is found to be linear and obey the power law ω^s where $s\leq1$.A strong dependence of $\sigma_{ac}(\omega)$ and exponent s on temperature has been interpreted in terms of correlated barrier hopping (CBH) model. The dielectric loss data has been utilized to determine the barrier height W_m. The value of W_m has been recorded in accordance with the Elliott's theory of hoping of charge carriers over potential barrier between charge defect states in case of chalcogenide glasses. Frequency dependence of activation energy of a.c. conduction has also been studied.

(Received May 10, 2016; Accepted June 21, 2016)

Keywords: Chalcogenide glasses, Melt quench technique, a.c. conductivity, Dielectric properties, SEM, EDAX

1. Introduction

Non crystalline chalcogenide alloys and compounds have been subject of extensive investigations in the last few decades because they possess unique, interesting properties as well as possibilities of tailoring their properties [1]. The ability to control and modify the properties of chalcogenide glassy alloys, even after their fabrication just by exposing them to external effects such as γ-rays, x-rays, laser beams and thermal treatment have drawn attention of researchers and scientists to these materials[2-5]. Such materials are widely used in optical digital versatile discs (DVD) and phase change random accesses memory devices [6-8], in fabrication of optical fibers in low cost solar cells [9-11]. Since such materials are highly photo sensitive, IR transparent and have high refractive index, therefore they are suitable candidates to be used for advanced optoelectronic applications [5-7]. Such glasses are considered as promising semiconducting materials because they possess largest nonlinear susceptibility among inorganic glassy materials [12-14].

Among chalcogenide glasses Se-based glassy alloys have got wide technological applications in optoelectronic devices, such as photoreceptor in vidicon tubes [15], xerography machine and x-ray imaging [16-17]. Se based glassy semiconductors exhibit high resistivity, low sensitivity, short life time and thermal instability which impose some limitations on their applications [18-19].

Several attempts have been made to overcome the limitations of short life time and thermal instability by adding third element as an additive. The effect of various additives on

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structural, electrical and optical properties of Se-based chalcogenide glasses have been reported in the literature [20-25].

Dielectric materials have been the subject of special attention. These materials are being employed in various industrial devices such as dynamic access memory, microwave filter, voltage controlled oscillators and other telecommunication technologies [26-27].

Among VI-II-III group ternary compound Se-Cd-In has found applications in optoelectronics and solar cells due to its higher value of absorption coefficient [28]. The study of dielectric behavior of chalcogenide glasses is expected to reveal structural informations, which infact may be useful for the understanding of conduction mechanism in these glasses. In addition, a study of temperature dependence of dielectric properties specially in the range of frequencies where dielectric dispersion occurs can be of great importance to understand the nature and origin of losses occurring in these materials [29]. Since these materials are co-valently bonded solids, therefore dispersion is not expected at low frequencies. However, recently it has been reported [26,30-31] that dielectric dispersion loss does exists in these glasses even at very low frequency. Therefore origin and nature of dielectric loss in such materials has become a matter of renewed attention and it is interesting to study the behavior of these materials in ac fields which gives the important information about the transport process in localized state in forbidden gap [32].

Measurements of frequency and temperature dependent dielectric parameters and a.c. conductivity of different chalcogenide semiconductors have been subject of many researchers and different models have been proposed to explain the experimental results [33-34].

Literature survey on the dielectric measurements [35-38] shows that dielectric losses are dipolar in nature in these materials and can be under stood in terms of hopping of charge carriers over a potential barrier as suggested by Guintini [39].

In view of above, in the series of a systematic study of dielectric relaxation and a.c. conductivity in Se-based chalcogenide glass in our laboratory[21,26,40-41] we have made dielectric measurements in glassy $Se_{90}Cd_8In_2$ alloy to study the effect of frequency and temperature on dielectric properties and a.c. conductivity.

The present paper reports the dielectric relaxation and a.c. conductivity measurements in glassy$Se_{90}Cd_8In_2$ alloy. SEM and EDAX have been measured to analyses the surface morphology and elemental compositions of $Se_{90}Cd_8In_2$ glassy alloy. Dielectric parameters and a.c. conductivity measurement of $Se_{90}Cd_8In_2$ glassy alloy has been carried out in the frequency range5×10^2Hz - 1×10^5Hz and in temperature range 308-333K.

2. Experimental details

The bulk sample of $Se_{90}Cd_8In_2$ternary chalcogenide glass was prepared from stoichiometric mixture of highly pure (99.999%) Se, Cd and In elements by the melt quench technique. The exact amounts of constituent elements were weighed according to their atomic weight percentage using an electronic balance (LIBROR, AEG-120) with the least count of 10^{-4}gm and placed into ultra-cleaned quartz ampoules(length ≈ 5cm and internal diameter ≈ 8mm). The ampoules were evacuated and sealed under a vacuum of 10^{-5} Torr to avoid reaction of alloying elements with oxygen at a higher temperature. The sealed ampoules were heated in a furnace at rate of 4–5 K/min, the temperature raised up to 800°C and kept at that temperature for 12 h. During the heating process, the ampoules were constantly rocked by rotating ceramic rod to ensure the homogeneity of alloying materials. The ampoules with molten materials were rapidly quenched into ice-cooled water. The ingots of glassy materials were taken out from ampoules by breaking them and grinded into fine powder with the help of mortar. Surface morphology was studied using the JEOL, Japan JSM-6510 Model SEM. The magnification used was 5000×. The compositional analysis of the prepared alloy was studied by EDAX attachment to the above mentioned SEM model. Dielectric and electrical conductivity measurements have been done with Wayne Kerr Electronics, UK; model: 4255 in the frequency range from 5×10^2Hz - 1×10^5Hz and in temperature range 308-333K. For this, glassy samples were pressed into cylindrical pellet forms having diameter 10mm and thickness about 0.82mm under uniform load of 5 tons using hydraulic press. A pellet was sandwiched between two circular silver discs in order to ensure good electrical contact between sample and electrodes of the LCR meter. This whole assembly of sample and discs is placed between the electrodes of the LCR meter. The temperature measurement was facilitated by a copper constantan thermocouple close to sample. A vacuum of the order of 10^{-3}Torr was maintained over the entire temperature range. For determination of a.c. conductivity, the dissipation factor and capacitance were measured. All the measurements were carried out under dark condition in the cryostat. The signal voltage level was kept at 0.02 Volts.

The dielectric constant ε' was calculated using the relation:

$$\varepsilon' = Cd/A\varepsilon_o \qquad (1)$$

where, C is the capacitance of the sample, d is thickness of the pellet, A is area of the pellet and ε_o is the free space permittivity.

The dielectric loss ε" was calculated from the relation:

$$\varepsilon'' = \varepsilon' \mathrm{Tan}\delta$$

where, δ=90º-θ and θ is the phase angle.

If ω is the frequency of applied signal and ϵ_o is the permittivity of free space, then a.c. conductivity is given as

$$\sigma_{ac}(\omega)=\omega\varepsilon_o\varepsilon'' \qquad (2)$$

$$\sigma_{ac}(\omega)=2\pi f\varepsilon_o\varepsilon' \mathrm{Tan}\delta \qquad (3)$$

3. Results and Discussion

3.1 Surface morphological analysis

SEM is a promising technique for the topographic analysis, which gives important information regarding to growth mechanism, shape and size of the sample. Scanning electron micrograph of the studied sample is shown in fig.1. From SEM micrograph it is evident that image of the sample is uniform and without any pin holes or cracks and there is formation of conchoidal contours, which shows the presence of some micro-crystallites embeded in the glass matrix of the synthesized material.

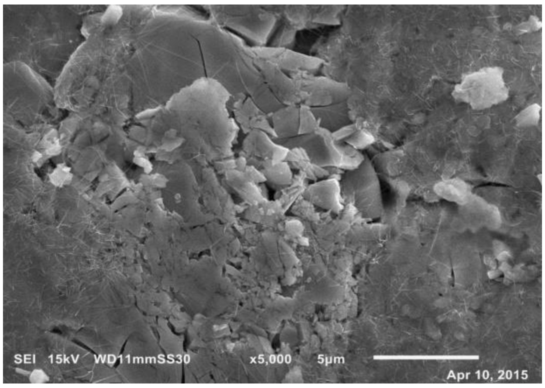

Fig.1. SEM images of $Se_{90}Cd_8In_2$ glassy alloy at 5000× magnification.

The elemental compositions of $Se_{90}Cd_8In_2$ glassy alloy were checked by energy despersive X-ray analysis (EDAX). The obtained percentage of the composition is $Se_{91.2}Cd_{5.8}In_3$ as shown in Fig.2. EDAX analysis indicates that there is absence of impurity elements in the studied coposition.

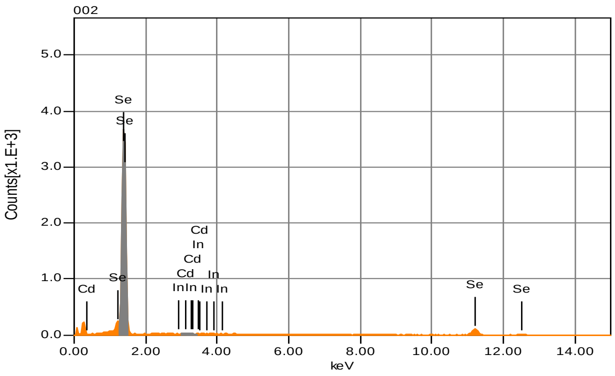

Fig.2: Energy dispersive X-ray analysis (EDAX) for $Se_{90}Cd_8In_2$ glassy alloy.

3.2 Dielectric properties

The dielectric investigations specify the dielectric properties of a material as a function of frequency and temperature. Two fundamental electrical characteristics of materials can be examined from the dielectric analysis.

(i) The capacitive insulating nature, which represents its ability to store electrical charge

(ii) The conduction nature, which represents its ability to transfer electric charge. Therefore, the dielectric analysis leads to provide the dielectric constant (ε') and dielectric loss (ε") of a material.

In the succeeding section proper attention has been paid to investigate the dielectric properties of $Se_{90}Cd_8In_2$ glassy alloy in the frequency range 5×10^2Hz - 1×10^5Hz and in temperature range 308-333K. As a general features of the obtained results, the frequency and temperature dependence of dielectric constant (ε') and dielectric loss (ε") are investigated.

3.2.1Frequency dependence of dielectric constant (ε') and dielectric loss(ε")

Fig. 3 shows the frequency dependence of dielectric constant (ε') of $Se_{90}Cd_8In_2$ glass alloy at different temperature.

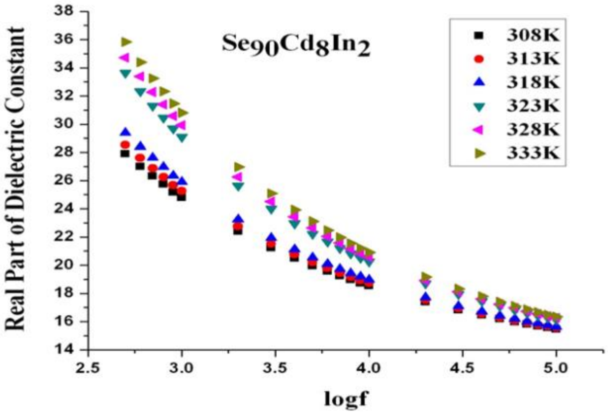

Fig.3. Variation of real part of dielectric constant with logf for $Se_{90}Cd_8In_2$ at different temperature.

It has been observed that ε' decreases with frequency and it attains a constant value at higher frequencies. The decrease of ε' with frequency can be attributed to the fact that at low frequencies ε' for polar material is explained by contribution of multi component of polarizability viz deformational (electronic and ionic) and relaxation (orientational and interfacial) polarization. The sum of above four type of polarization gives the total polarization of dielectric materials [42].

First of all electronic polarization arises due to displacement of valence electrons with respect to positive nucleus. Such type of polarization appears at frequencies upto 10^{16}Hz. The second type is the ionic polarization which appears due to displacement of positive and negative ions with respect to each other. Maximum frequency for ionic polarization is 10^{13} Hz. The third type is the dipolar polarization which appears in the materials having molecules with permanent electric dipole moments capable of changing orientation into the direction of applied electric field. Such polarization appears at frequencies up to 10^{10} Hz. The last one is the space charge polarization which appears due to the impedance mobile charge carriers by interfaces. Such type of polarization typically occurs in the frequency range 1-10^3 Hz. The ionic polarization does not play an important role in the total polarization. The orientational polarization decreases with increase in frequency because it takes more time then electronic and ionic polarization. This decreases the value of dielectric constant (ε') with increase in frequency which ultimately reaches a constant value at higher frequency range, which correspond to interfacial polarization.

The variation of dielectric loss (ε") with frequency at different temperature for $Se_{90}Cd_8In_2$ glassy alloy is shown in fig.4. From fig.4 it is observed that ε" is also found to be decreasing with increase in frequency.

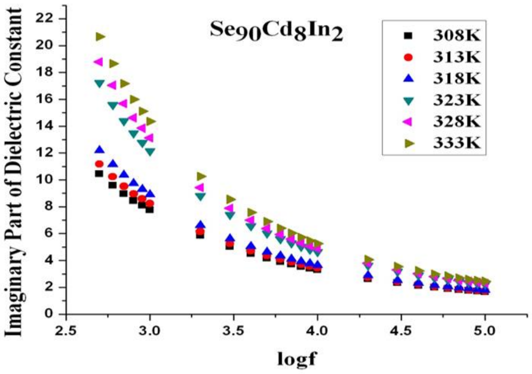

Fig.4: Variation of imaginary part of dielectric constant with logf for $Se_{90}Cd_8In_2$ at different temperature.

The decrease of ε" with frequency can be attributed to the fact that, at low frequencies, the value of ε" is due to migration of ions in the material. At moderate frequencies ε" is due to the contribution of ion jumps, conduction loss of ion migration and ion polarization losses. At high frequencies ion vibration may be the only source of dielectric loss and therefore ε" has the minimum value [43].

According to Guintini theory dielectric loss at a particular frequency and temperature can be expressed as [39]

$$\varepsilon'' = (\varepsilon_o - \varepsilon_\infty) 2\pi^2 N (ne^2/\varepsilon_o)^3 kT\tau_o^m W_m^{-4} \omega^m \qquad (4)$$

where, n is the number of electron that hop, N is the localized state density, ε_o and ε_∞ are the static and optical dielectric constants, ω is the angular frequency (ω= 2πf) and W_m is the maximum barrier height which is related to the band gap.

Dielectric loss ε" is found to increase with increase in temperature according to power law with frequency:

$$\varepsilon'' = A\omega^m \qquad (5)$$

Where m is a power of angular frequency and can be expressed as

$$m = -4kT/W_m \qquad (6)$$

Using the plots of fig.4 we have plotted logε" versus logf plots in the region where the variation of ε" with frequencies is appreciable (Fig.5). The values of power m have been calculated from the slopes of straight lines obtained from such plots. The evaluated values of m have been plotted as a function of temperature in fig.6. It is found that m has negative values and its magnitude decreases linearly with temperature. This is in good agreement with eq.(4), which clearly indicates that the Guintini [39] theory of dielectric relaxation based on the hopping of charge carriers over a potential barrier as suggested by Elliott [44] is applicable in the case of present ternary glasses.

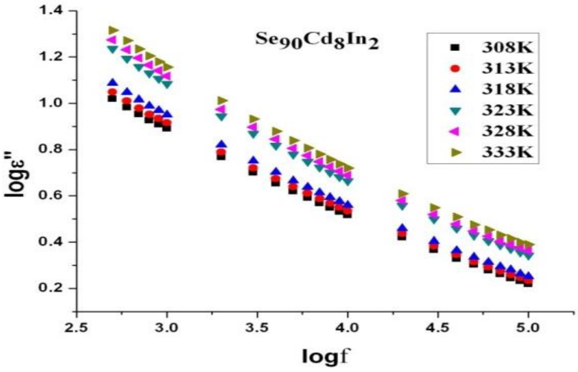

Fig.5: Frequency dependence of logε" at deferent temperatures forSe$_{90}$Cd$_8$In$_2$glassy alloy.

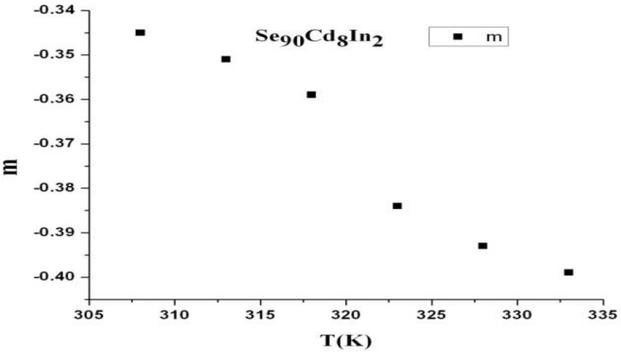

Fig.6: Variation of m versus temperature for Se$_{90}$Cd$_8$In$_2$ glassy alloy.

3.2.2Temperature dependence of dielectric constant (ε′) and dielectric loss(ε")

Variation of dielectric parameters ε' and ε" with temperature at different frequencies are shown in fig.7 and 8 respectively.

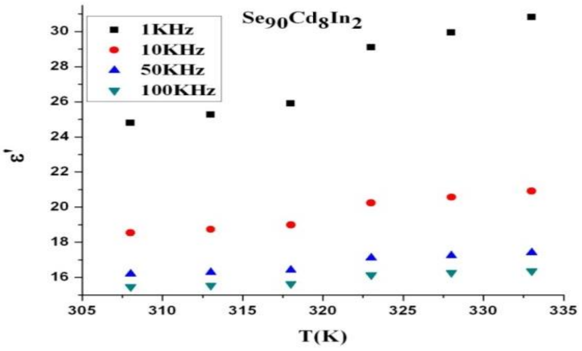

Fig.7: Temperature dependence of dielectric constant (ε') for Se$_{90}$Cd$_8$In$_2$ glassy alloy.

From fig.7 it is evident that ε' increases with increasing temperature. This can be attributed to the fact that orientational polarization is connected to the thermal motion of the molecules, so dipoles can not orient themselves with varying electric field at low temperatures. At low temperatures it is found almost constant. The orientation of dipoles is facilitated with increasing temperature; which causes increase in the value of orientational polarization, which ultimately results into increase of dielectric constant ε' with temperature.

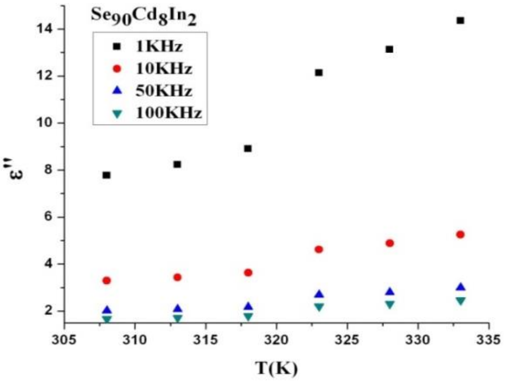

Fig.8: Temperature dependence of dielectric loss (ε'') for $Se_{90}Cd_8In_2$ glassy alloy.

Figure 8 shows the temperature dependence of dielectric loss ε'' of $Se_{90}Cd_8In_2$ glassy alloy at different frequencies. It has been observed that ε'' increases with increase in temperature.

The dielectric loss can be divided into three parts i.e. conduction losses, dipole losses and vibrational losses [45]. The conduction losses have minimum value at low temperatures because it is proportional to $\sigma_{ac}(\omega)$. The conduction loss is owing to migration of ions over large distances. When the ions move they give some of their energy to the lattice as heat. Since $\sigma_{ac}(\omega)$ increases with increasing temperature, which causes the increase in conduction loss. Therefore ε'' increases with increasing temperature. The characteristic of low dielectric constant and dielectric loss with high frequency for given sample suggest that the sample possess enhanced optical quality with lesser defects and this parameter is of vital importance for various nonlinear optical materials and their applications in devices.

3.3Frequency dependence of AC conductivity

Total conductivity of the material is the sum of a.c. and d.c. conductivities. Therefore a.c. conductivity $\sigma_{ac}(\omega)$ is expressed as[46-47]

$$\sigma_{ac}(\omega)= \sigma_t(\omega)-\sigma_{dc} \qquad (7)$$

where,$\sigma_t(\omega)$ is the total conductivity and σ_{dc} represents d.c. conductivity. Since d.c. component σ_{dc} is negligibly small as compared to $\sigma_t(\omega)$ in the studied sample, therefore $\sigma_t(\omega)$ is considered to be $\sigma_{ac}(\omega)$. The variation of $\log\sigma_{ac}(\omega)$with frequency in the range 5×10^2Hz - 1×10^5Hz has been investigated for the temperature range 308K-333K. Variation of $\sigma_{ac}(\omega)$ with frequency at constant working temperature has been plotted in fig.9. From fig.9 it is evident that $\sigma_{ac}(\omega)$ increases linearly with frequency at a constant working temperature.

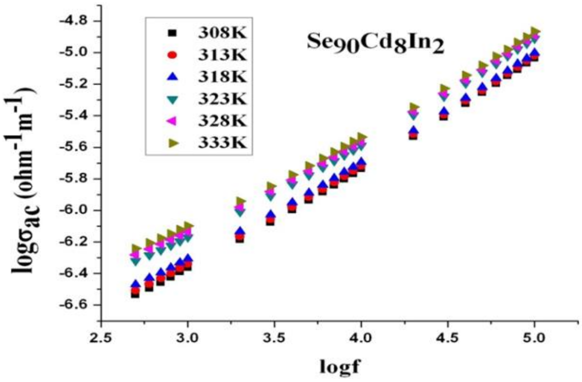

Fig.9: Frequency dependence of $\sigma_{ac}(\omega)$ for $Se_{90}Cd_8In_2$ glassy alloy at different temperature.

Generally a.c. conductivity in semiconductors is given as[47,48]

$$\sigma_{ac}(\omega)=A\omega^s \qquad (8)$$

where, A is a constant, ω is the angular frequency and s is the frequency exponent. Value of frequency exponent s is obtained from the slopes of a.c. conductivity versus frequency plots. The dependence of frequency exponent s with temperature for the studied composition is shown in fig.10. It has been observed that s has values in the range 0.66 to 0.60, which is less then unity and decreases with increase of temperatures.

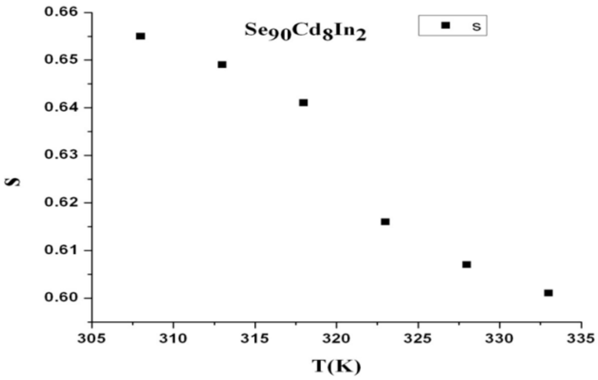

Fig.10: Temperature dependence of the frequency exponent s for $Se_{90}Cd_8In_2$ glassy alloy.

The observed behavior of s are in agreement with the correlated barrior hopping model (CBH) [48-50], such behavior have also been reported by other workers [26, 36, 51-54].

According to CBH model, the conduction occurs via bipolaron hopping process where two polaron simultaneously hop over the potential barrier between two charged defect states D^+ and D^- and barrier height is correlated with the inter-site separation via a columbic interaction. Shimakava [55] further proposed that at higher temperature D^o states are produced by thermal excitation of D^+ and D^- states and a single polaron hopping (which includes one electron hopping between D^o and D^+ and hole between D^o and D^-) becomes dominant.

3.4Temperaturedependence of a.c. conductivity

Fig.11 shows the variation of $\sigma_{ac}(\omega)$ versus 1000/T for studied composition at different frequency values.

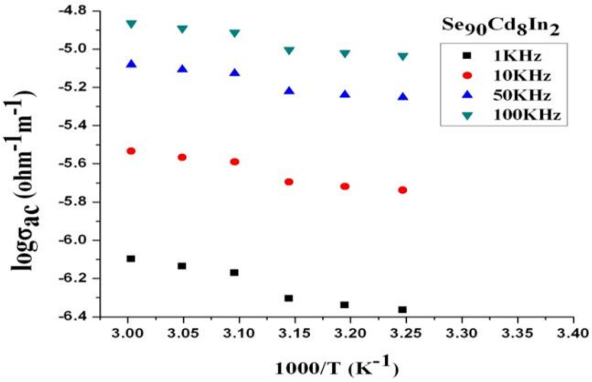

Fig.11: Temperature dependence of $\sigma_{ac}(\omega)$ for $Se_{90}Cd_8In_2$ glassy alloy.

It is evident that a.c. conductivity decreases non-linearly with reciprocal of temperature, which suggest that $\sigma_{ac}(\omega)$ is thermally activated process with single activation energy from various localized state in the band gap. The activation energy of a.c. conduction has been calculated at different audio frequencies using the Arrhenius temperature dependence of$\sigma_{ac}(\omega)$ [56].

$$\sigma_{ac} =(\sigma_o)_{ac}\exp[-\Delta E_{ac}/kT] \qquad (9)$$

Where, $\sigma_{ac}(\omega)$ is called the activation energy for thermally activated a.c. conduction and$(\sigma_o)_{ac}$ is well known pre-exponential factor.

The activation energy ΔE_{ac} and pre-exponential factor $(\sigma_o)_{ac}$ have been determined using equation(9) at different frequencies for each glassy alloys.

The frequency dependence of activation energy has been plotted in fig. 12. It is evident from the figure that activation energy ΔE_{ac} decreases with frequency which may be attributed to increase of field frequency which is responsible for electronic jump between localized states. Our present results are in good agreement with recently reported results by various authors in amorphous semiconductors [26,43, 57-60].

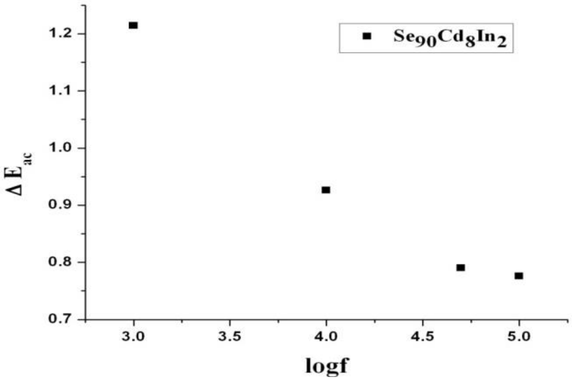

Fig.12: Frequency dependence of a.c. activation energy for$Se_{90}Cd_8In_2$ glassy alloy.

4. Conclusion

$Se_{90}Cd_8In_2$ chalcogenide glassy alloy has been prepared by melt quench technique. The prepared sample has been characterized by SEM, EDAX and impedance spectroscopic technique. Both dielectric constant ε' and dielectric loss ε'' are found to have decreasing trend with increase in frequency and increasing trend with increase in temperature. It is found that both ε' and ε'' show frequency dispersion at low frequency and show low values at high frequencies. The temperature and frequency dependence of a.c. conductivity $\sigma_{ac}(\omega)$ are studied in the frequency range 5×10^2Hz -

1×10^5Hz and temperature range 303-328K. A.C. conductivity has been found to obey the power law ω^s, where $s\leq1$. $\sigma_{ac}(\omega)$ increases with increase of frequency in the measured temperature range while s decreases with increase of temperature. These results are in good agreement with the correlated barrier hopping (CBH) model. Value of maximum barrier height was estimated from the data of dielectric loss, which is in good agreement with the theory of hopping of charge carriers over a potential barrier between charge defect states. These calculations are performed according to Guintini equation based on Elliot model of chalcogenide glasses. The low value of ε' and ε" at high frequencies suggest that the prepared alloy possess enhanced optical quality with lesser defect and is therefore, suitable for nonlinear optical materials applications.

Acknowledgement

The authors are very much grateful to the University Grants Commission (UGC), New Delhi, India for providing financial assistance in form of major research project F.No. 39-452/2010 (SR). The authors also thank Dr. Neeraj Mehta, Department of Physics, B.H.U. Varanasi for providing the facility of conductivity measurement.

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