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Dielectric Properties and AC Conductivity Measurements of Amorphous $Ge_{15}Se_{85}$ Glass

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In the present study, investigations of dielectric parameters viz dielectric constant $\varepsilon'(\omega)$, dielectric loss $\varepsilon''(\omega)$ and *AC* conductivity measurements have been made for bulk chalcogenide $Ge_{15}Se_{85}$ glass in the frequency range 10 to 500 kHz within the temperature range from 300 to 390 K. The variation of dielectric constant and dielectric loss with frequency at room temperature is reported and discussed in the investigated glassy binary alloy.

Keywords: Chalcogenide semiconductors, Dielectric constant, Dielectric loss, *AC* conductivity.

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1. INTRODUCTION

Great consideration has been given to chalcogenide glasses in last two decades because of their potential applications in various solid state devices both in scientific and technological fields. Among the amorphous chalcogenide alloys, mostly selenium (Se) based materials are preferred due to its commercial use and technological importance. But in pure state it has disadvantages because of its short life time and low sensitivity [1]. The problem can be overcome by alloying selenium with some impurities such as Ge, Te etc which in terms gives high sensitivity, greater hardness, high crystallization temperature and small ageing effects as compared to pure Se glass [2].

Ge-Se glasses have been widely studied due to their extended amorphization composition range [3]. The fact that bulk glasses are easily obtained, makes Ge-Se system an ideal one to investigate a great variety of properties and their correlation with structure and composition. This system is extensively studied and glass formation in this system occurs predominantly in alloys enriched with Se and containing 0 to 25 at. % of Ge [4]. Furthermore, addition of Ge into the polymeric Se matrix produces a cross linking of selenium chain, mediated by the formation of $Ge(Se_{1/2})_4$ tetrahedral units [5]. At low doping ($x < 15$ at. %), the tetrahedral units are sparsely distributed in the background matrix, with rather flexible interconnections. By $x = 15$ at. %, the amount of Ge becomes sufficient to join some of the tetrahedral pairs by corner sharing [6].

Chalcogenide glasses in Ge-Se system are used as switching, memory elements and optoelectronic devices and are interesting material for infrared optics too. It has been observed that the addition of the third element to Ge-Se system effectively controls its electrical, optical, thermal, dielectric and physical properties. Moreover, Ge-Se system has drawn more attention as some metallic additives have been found [7-9] to change the conduction type from p to n in these glasses.

Structural studies of these glasses are very important for better understanding of transport mechanisms. The study of dielectric properties of chalcogenide glasses is predictable to reveal structural information, which in fact, can be useful for the understanding of conduction mechanism in these materials. In addition, study of temperature and frequency dependence of dielectric parameters, particularly in the range of high frequency where the dielectric dispersion occurs can be of utmost importance for the understanding of the nature as well as the origin of the various losses happening in these materials.

In the present work, we have reported and discussed the temperature and frequency dependence of dielectric properties as well as *AC* conductivity measurements of glassy $Ge_{15}Se_{85}$ alloy. Studies of *AC* conductivity and dielectric properties of Ge-Se system are rare and have not attracted much interest. Therefore, the aim of this work is to analyze the frequency and temperature dependence of the *AC* conductivity $\sigma_{ac}(\omega)$, the dielectric constant $\varepsilon'(\omega)$ and the dielectric loss $\varepsilon''(\omega)$ for the amorphous $Ge_{15}Se_{85}$ chalcogenide glass.

2. EXPERIMENTAL DETAILS

Glassy alloy of $Ge_{15}Se_{85}$ has been prepared using the melt quenching technique. 5N highly pure materials (99.999 %) are weighed according to their atomic percentages and sealed in a quartz ampoule (length ~ 5 cm, diameter ~ 12 mm) under a vacuum of ~ 2×10^{-5} mbar. The sealed ampoule has been kept inside a furnace where the temperature is raised to 1000 °C at a rate of 3-4 °C/min. The ampoules are rocked frequently for 24 h at maximum temperature to make the melt homogenous. The quenching is done in the ice cooled water very rapidly to prevent crystallization. The amorphous nature of the glassy alloy has been verified by X-ray diffraction. Fig. 1 shows the XRD pattern for $Ge_{15}Se_{85}$.

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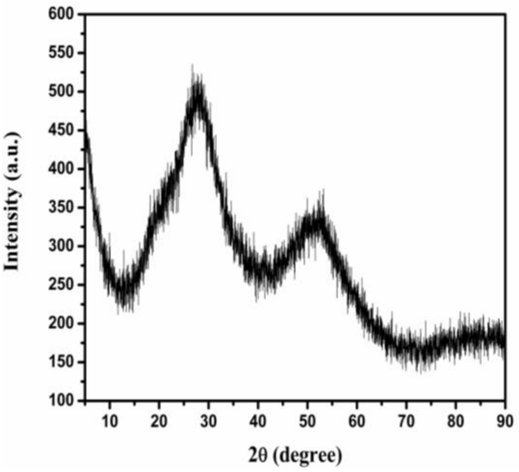

Fig. 1 – XRD pattern of $Ge_{15}Se_{85}$ glassy alloy

For the dielectric measurements, the pallets of powder are prepared by a die using a hydraulic machine at a load of 5 tons. The pallets are made of different thicknesses; 1.622 mm, 1.673 mm and 1.677 mm and diameter 9.957 mm, 10.058 mm and 10.042 mm respectively, to study the conduction process in the investigated system. The reproducibility of the results has been checked by making many runs at different times over the entire temperature and frequency ranges for the three thicknesses. However, only the results of the thickness 1.622 mm are reported here. The dielectric parameters viz dielectric constant ε' and the dielectric loss ε'' are determined using the Impedance Analyzer (Wayne Kerr 6500 B). The dielectric constant ε' (the real part of the dielectric constant) is calculated using the relation:

$$\varepsilon' = \frac{Cd}{\varepsilon_0 A} \quad (1)$$

where C is the capacitance of the sample, d is the pallet thickness, A is the cross-sectional area and ε_0 is the free space permittivity. The dielectric loss ε'' (imaginary part of the dielectric constant) is calculated from the equation:

$$\varepsilon'' = \varepsilon' \tan\delta \quad (2)$$

where (δ = 90-Φ) and Φ is the phase angle. Moreover, $\tan\delta$ is known as the dissipation factor or loss factor obtained from the impedance analyzer.

3. RESULTS AND DISCUSSION

The frequency and temperature dependence of the dielectric constant ε', the dielectric loss ε'' and *AC* conductivity $\sigma_{ac}(\omega)$, are investigated for bulk sample of the glassy $Ge_{15}Se_{85}$ alloy in the frequency range 10 to 500 kHz and in the temperature range 300-390 K, to study the dielectric behavior in the investigated system.

3.1 Dielectric Properties of $Ge_{15}Se_{85}$ Glass

3.1.1 Temperature and Frequency Dependence of the Dielectric Constant (ε')

The variation of dielectric constant (ε') with frequency (ω) at room temperature is shown in Fig. 2. It is observed that the dielectric constant decreases with the frequency. This decrease is fast at low frequency as compared to higher frequency that may be due to the electrode polarization effects [10, 11]. The decrease of ε' with frequency can be attributed to the fact that at low frequency, ε' for polar material is explained by the contribution of multi-component of polarization viz electronic, ionic, dipolar or orientation and space charge [12]. Thus, the total polarization (P) of the dielectric material can be represented as the sum of these four polarizations [13]:

$$P = P_e + P_i + P_d + P_s \quad (3)$$

Dielectric materials exhibit at least one of these polarization types. This depends on the bonding and structure of the material and frequency [14].

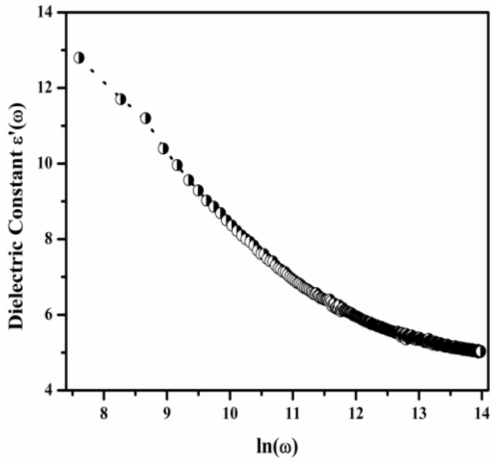

Fig. 2 – Frequency dependence of dielectric constant at room temperature for $Ge_{15}Se_{85}$ chalcogenide glass.

Fig. 3 shows the temperature dependence of the dielectric constant $\varepsilon'(\omega)$ at different frequencies. It indicates that there is an increase in the dielectric constant with temperature. The increase in the dielectric constant of the sample may be attributed to the electric field which is accompanied by the applied frequencies. Such field will cause some ordering inside the sample as well as the formation of an electric moment in the entire volume of the dielectric and in each separate polarizing molecule. The molecular dipoles in polar material cannot orient themselves at low temperature. When the temperature rises, the dipoles orientation is facilitated and the dielectric constant increases. As the frequency is increased, the variation in the field become too rapid for the molecular dipoles to follow, so that their contribution to polarization becomes less with a measurable lag because of internal frictional forces [15].

3.1.2 Temperature and Frequency Dependence of the Dielectric Loss (ε'')

The variation of dielectric loss $\varepsilon''(\omega)$ with frequency at room temperature is shown in Fig. 4. It is obvious from Fig. 4 that the dielectric loss also follows the same trend as followed by the dielectric constant $\varepsilon'(\omega)$. The decrease in the dielectric loss with frequency in the investigated system may be attributed to the fact that the migration of the ions in the glass is the main source of the dielectric loss at low frequencies. Therefore, dielectric loss at low and moderate frequencies is characterized by the higher values of $\varepsilon''(\omega)$ due to the contribution of ion jump and conduction loss of ion migration in addition to the ion polarization. However, at higher frequencies, the ion vibrations may be the only source of dielectric loss. So, $\varepsilon''(\omega)$ decreases at higher values of frequency [16].

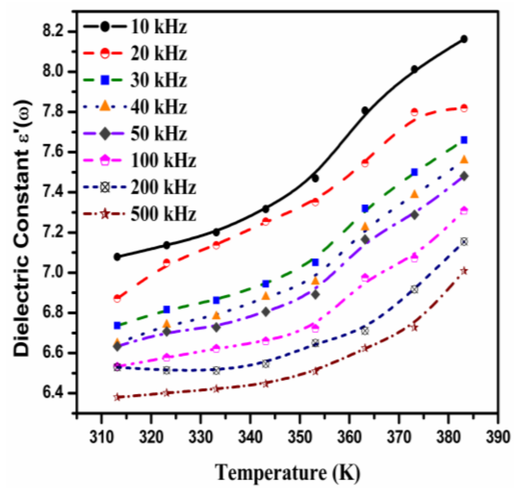

Fig. 3 – Variation of the dielectric constant $\varepsilon'(\omega)$ with temperature at different frequencies for $Ge_{15}Se_{85}$

The variation in the dielectric loss $\varepsilon''(\omega)$ as a function of temperature at different frequencies is also shown in Fig. 4 which illustrates that $\varepsilon''(\omega)$ exhibits temperature dependence and is found to be more at low frequencies. The origins of dielectric loss are conduction losses, dipole losses and vibration losses [17]. As the temperature increases, the electrical conduction losses increase with further increasing of the dielectric loss $\varepsilon''(\omega)$.

The dielectric loss is also given by the following empirical relation [16]:

$$\varepsilon'' = B\omega^m \quad (4)$$

where B is a constant and m is a power of angular frequency. The plot between $\ln(\varepsilon'')$ and $\ln(\omega)$ should be a straight line and the slope of the plot enables us to obtain the value of m (Fig. 6). The value of the exponent m obtained from the negative slope of Fig. 6 is found to be negative ($m = 0.55691$ at $T = 298$ K) in the studied sample which is consistent with earlier values of m obtained for the other chalcogenide glasses [18].

According to Guintini et al. [19] model for dielectric dispersion in chalcogenide glasses based on the Elliot's idea [20], the dielectric loss at a particular frequency is given by:

$$m = -\frac{4kT}{W_m} \quad (6)$$

where W_m is the maximum barrier height i.e. the energy required to move the electron from a site to infinity and is found equal to 0.18 eV for the present investigated $Ge_{15}Se_{85}$ glassy alloy.

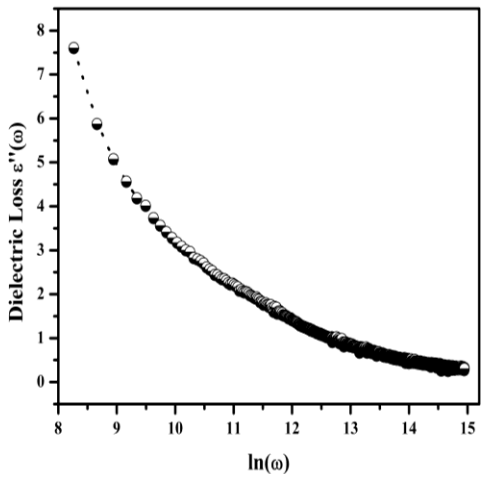

Fig. 4 – Frequency dependence of dielectric loss at room temperature for $Ge_{15}Se_{85}$ chalcogenide glass

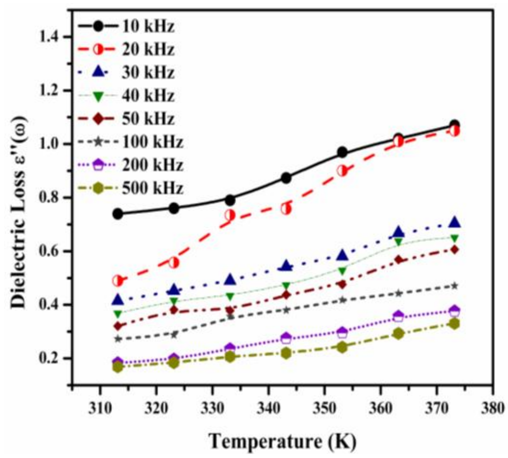

Fig. 5 – Variation of the dielectric loss $\varepsilon''(\omega)$ with temperature at different frequencies for $Ge_{15}Se_{85}$

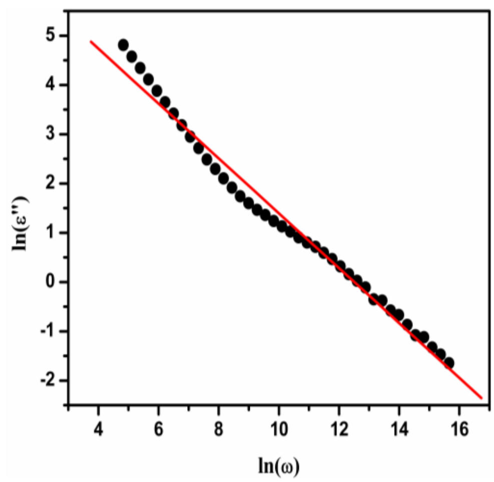

Fig. 6 – Plot of $\ln(\varepsilon'')$ versus $\ln(\omega)$ for chalcogenide $Ge_{15}Se_{85}$ glass at room temperature

3.2 Temperature and Frequency Dependence of *AC* Conductivity

The *AC* conductivity of the studied sample has been deduced from the data of dielectric constant $\varepsilon'(\omega)$ and loss factor $\tan\delta$ using the following relation:

$$\sigma_{ac} = \varepsilon' \varepsilon_o \omega \tan \delta \,, \quad (7)$$

where symbols have their usual meaning.

A common feature to all the amorphous semiconductors is that *AC* conductivity $\sigma_{ac}(\omega)$ changes with frequency according to the following relation [21]:

$$\sigma_{ac}(\omega) = A_o \omega^s \,, \quad (8)$$

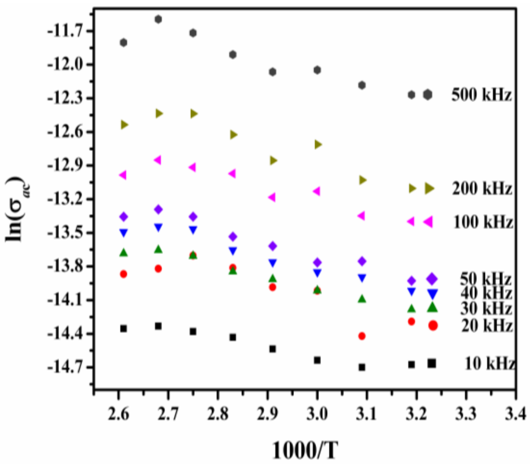

Fig. 7 – Variation of AC conductivity $\sigma_{ac}(\omega)$ with temperature at different frequencies for chalcogenide $Ge_{15}Se_{85}$ glass

where A_o is a constant dependent on temperature, ω is the angular frequency and the exponent *s* denotes the frequency dependence of σ_{ac} and it is generally less than or equal to unity and is found equal to 0.45 in the studied glass. Fig. 7 shows the frequency dependence of $\sigma_{ac}(\omega)$.

Fig. 7 shows the variation of *AC* conductivity as a function of reciprocal temperature at different frequencies. This figure depicts the temperature independence and the frequency dependence of $\sigma_{ac}(\omega)$ in the investigated temperature and frequencies ranges for the studied chalcogenide alloy. It indicates that $\sigma_{ac}(\omega)$ is not thermally activated in this range of temperature.

4. CONCLUSION

Dielectric properties and conductivity measurements have been made for $Ge_{15}Se_{85}$ glassy alloy at different frequencies and temperature. Dielectric constant and dielectric loss are found to decrease with frequency at room temperatures in the studied system. The AC conductivity measurements shows that $\sigma_{ac}(\omega)$ is independent of temperature. However, it is found to vary with different frequencies in the temperature and frequency range of the investigated binary alloy.

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