Dielectric properties of In-Se-Te glassy alloys

A S Maan* & D R Goyal
Physics Department, Maharshi Dayanand University, Rohtak 124 001, India

Received 4 November 2006; accepted 28 February 2008

In this paper, the dielectric properties of In$_{40}$Se$_x$Te$_{60-x}$ glassy alloys (x = 10, 20 & 30) have been reported. These measurements are carried out as a function of temperature and frequency in the range 120-350 K and 0.1-10 kHz respectively. The frequency and temperature dependence of dielectric constant $\varepsilon'$ and dielectric loss $\varepsilon''$ is almost similar in nature in all the alloys and the experimental results indicate that dielectric dispersion sets in at temperature exceeding 150 K and is more prominent at lower frequencies. $\varepsilon'$ and $\varepsilon''$ values as a function of temperature and frequency are maximum for x = 20 alloy, though similar behaviour is observed in all the samples. The experimental results seem to be explained in terms of the theory of dielectric dispersion based on two electrons hopping over a potential barrier. The frequency dependence of dielectric loss in present set of glasses at a fixed temperature is in good agreement with theory of dielectric dispersion.

Among amorphous semiconductors, chalcogenide glasses based on sulphides, selenide and telluride alloys have proved to be promising candidates for various optical and photonic applications. Chalcogenide glasses based on Se show high transparency in middle and infrared region and also have nonlinear properties and therefore could be useful for all-optical switching. Chalcogenide glasses are sensitive to the absorption of electromagnetic radiations and show a variety of photo-induced effects. Glassy alloys based on Se-Te are reported to be of commercial importance due to their use as optical recording media. In-Se glasses find application in optical and electronic communication, switching and memory device and photovoltaic applications. Various properties such as photoconductivity, charge conduction and optical absorption, solar cell applications have been reported in this system. Amorphous films of In-Se also exhibit anomalous feature such as negative seebeck coefficient in certain compositions, large fermi level shift, an anomalous rise of the photocurrent. In-Se-Te glassy systems have been relatively less investigated as compared to the In-Se System. Structural study of Se-Te-In alloy using X-rays reveal that selenium K-edge shift towards the lower energy side compared to Se-Te. Electrical conductivity and dielectric relaxation studies carried out on these glasses reveal that dc conduction and dielectric parameters ($\varepsilon$ and $\varepsilon''$) increase with increasing In% and are attributed to an increase in density of charged defects states. Study of photoconductivity and analysis of resulting recombination kinetics reveals that monomolecular recombination takes place in the In$_{40}$Se$_{30}$Te$_{30}$ glassy alloy.

Present work is related with the investigation of dielectric properties of In$_{40}$Se$_x$Te$_{60-x}$ alloys with Se to Te ratio being systematically varied. It is important to study the electrical properties as these are strongly dependent upon the density and distribution of gap states. An account of study of dielectric relaxation is presented. The observed dielectric loss has been analyzed in terms of existing theories of dielectric relaxation.

**Experimental Procedure**

Amorphous In$_{40}$Se$_x$Te$_{60-x}$ samples (x=10, 20 &30) were obtained in bulk form by the quenching technique using elements of 5 nines purity. For a given composition, elemental constituents weighed according to given stoichiometric ratios were sealed in quartz ampoules of 8 mm internal diameter under a vacuum of $\sim$10$^{-5}$ Torr. The ampoules were heated in a furnace up to the melting point of constituents and were kept at that temperature for 10 h. The temperature was raised at a heating rate of 3-4°C/min. The ampoule was constantly rocked to ensure homogenous nature of the glassy alloys. The heated
ampoules were then quenched in ice-cooled water. Bulk material as obtained was used for preparation of pallets for experimental use. Pellets of diameter 1.3 cm and thickness ~0.14 cm were prepared by compressing the finely ground powder in a die in a hydraulic press under a pressure ~ \(10^8\) kg/m\(^2\). Aquadecc electrodes were used for making contacts and the sample was then tightened between two steel electrodes in a cell. Reported measurements were carried out in a vacuum of \(10^{-3}\) Torr. A capacitance conductance bridge (Genrad model AP – 1620) was used for conductance and capacitance measurements. For connection purposes coaxial cables were used to ward off stray capacitances. Lead capacitance was subtracted from the measured capacitance while calculating the dielectric constant. Liquid nitrogen was used to cool down the sample and measurements were started from 100 K.

Results and Discussion

Dielectric studies have been carried out on the present set of glasses at different temperatures and frequencies. Figure 1 shows the variation of the dielectric constant (\(\varepsilon'\)) with temperature at four different frequencies in \(x = 30\) sample.

The dielectric constant remains fairly constant with temperature up to a particular temperature after which there is appreciable temperature dependence of \(\varepsilon'\) especially at low frequencies. Such behaviour may be attributed to the presence of frozen dipoles slowly attaining freedom of rotation with increasing temperature resulting in temperature dependent \(\varepsilon'\). A similar behavior of \(\varepsilon'\) with temperature has been found in other compositions. Figure 2 shows the temperature dependence of dielectric loss (\(\varepsilon''\)) in \(x = 30\) sample. As seen from the figure, \(\varepsilon''\) is almost temperature independent at \(T<150\) K and as
temperature increase \( \varepsilon'' \) becomes frequency dependent, the variation being more pronounced at low frequencies. Similar behaviour was observed in other compositions as well. To have a comparative view, variation of \( \varepsilon' \) and \( \varepsilon'' \) with Se content at a fixed temperature (296 K) and frequency (1.0 kHz) are shown in Figs 3a and 3b respectively. It is clear from the figures that \( \varepsilon' \) and \( \varepsilon'' \) do not show a linear dependence on Se content. As seen from the figures, \( \varepsilon' \) and \( \varepsilon'' \) are maximum for \( x = 20 \) composition. Similar discontinuity in other parameters related to photoconductivity with Se content has been reported for the present set of alloys.

To analyze the experimental results as in Figs 1 and 2, let us consider the model proposed by Guinini et. al.\textsuperscript{13} for dielectric dispersion in chalcogenide glasses. This model is based on the concept of charge carriers hopping over a potential barrier between charged defect states (\( D^+ \) and \( D^- \))\textsuperscript{14}. Each pair of sites (\( D^+ \) and \( D^- \)) is assumed to form a dipole with a relaxation time dependent on its energy\textsuperscript{15,16}; the latter being attributed to the existence of a potential barrier over which the carriers hop\textsuperscript{17}.

According to this model, corresponding to particular frequency and temperature where the dielectric dispersion happens, \( \varepsilon'' \) is given by

\[
\varepsilon'' = (\varepsilon_o - \varepsilon_{\infty})^2 \frac{2\pi^2 N(n e^2)}{\varepsilon_o} KT \tau_o^m W_m^{-4} \omega^m \quad \ldots (1)
\]

where,
\[
\begin{align*}
\tau_o & = -\frac{4kT}{W_m} \quad \ldots (2)
\end{align*}
\]

In accordance with Eq. (1), \( \varepsilon'' \) should follow a power law with frequency as

\[
\varepsilon'' = A\omega^m \quad \ldots (3)
\]

where, \( m \) must be negative and vary linearly with \( T \), as given in Eq. (2).

In present set of samples, it has been observed that \( \varepsilon'' \) has a power law dependence on frequency at temperatures where dielectric dispersion sets in. For this purpose, variation of \( \ln \varepsilon'' \) with \( \ln \omega \) is plotted in Fig. 4 at a number of temperatures.

Using these plots, values of \( m \) are computed and are found to be negative. Similar behaviour is present in other compositions of the present glassy system. The variation of \( m \) with temperature for \( x = 30 \) alloy is shown in Fig. 5 which clearly shows that \( m \) follows a linear relation with \( T \). Similar trend is observed in other compositions as well.

As discussed earlier, \( \varepsilon'' \) has a power law dependence on frequency at temperatures where dielectric dispersion occurs and the frequency exponent \( m \) also has negative values at different temperatures and varies linearly with \( T \).

This shows that the present results are in good agreement with Elliot’s theory of dielectric dispersion\textsuperscript{14} as mentioned earlier. A similar dependence of dielectric constant \( \varepsilon' \) and dielectric loss \( \varepsilon'' \) on frequency and temperature has been reported in Se-Te, Se\textsubscript{0.80}Te\textsubscript{0.10}M\textsubscript{0.10} (\( M = \text{cd, In and Sb} \))\textsuperscript{18} and Se\textsubscript{100-x}Sb\textsubscript{x} glassy alloys\textsuperscript{19} in the audio frequency range.
Also ε’ and ε’’ in these glasses are independent of temperature and frequency at low temperatures \( T < 150 \) K. Similar variation of ε’ and ε’’ with temperature and frequency has been reported in Se-Te-Sb and Se-Te-Ge glasses\(^{20,21}\). The results as mentioned above have also been explained on the theory of ac conductivity and dielectric relaxation by Guintini et al.\(^{13}\)

**Conclusions**

Temperature and frequency dependence of dielectric studies (ε’ and ε’’) has been studied in \( \text{In}_{40}\text{Se}_{x}\text{Te}_{60-x} \) glassy alloys (\( x = 10, 20 \) and \( 30 \)) in the range 120-350 K and 0.1–10 kHz respectively. The experimental results indicate that the dielectric dispersion exists in present set of alloys and the dispersion sets in at temperatures >150 K. The analysis based upon Elliot’s theory of two electrons hopping over a potential barrier seems to explain the behaviour in present set of glasses. At a particular temperature and frequency, values of ε’ and ε’’ are maximum at \( x = 20 \) and decrease as \( x \) is changed to 10 and 30. Such a discontinuity in ε’ and ε’’ with Se content (\( x \)) was also observed in case of different parameters obtained from photoconductivity studies on these samples.

**References**