Experimental investigation on some electrical parameters of $\text{In}_{10-x}\text{Sn}_x\text{Se}_{90}$ ($x = 2, 4, 6,$ and $8$) chalcogenide glasses before and after $\gamma$-irradiation

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Abstract

Some electrical parameters of $\text{In}_{10-x}\text{Sn}_x\text{Se}_{90}$ ($x = 2, 4, 6,$ and $8$) chalcogenide glasses before and after $\gamma$-irradiation, at doses of $8, 16$ and $32$ kGy, are reported and discussed. $I$–$V$ measurements, for both unirradiated and gamma irradiated glasses, have been carried out in the temperature range of $170$–$290$ K for different electric fields. The recorded $I$–$V$ characteristics reveal that the studied glasses exhibit ohmic and non-ohmic behavior at low and high fields, respectively. Analysis of the experimental data, for unirradiated and gamma irradiated samples, in the high field range confirms the presence of space charge limited conduction (SCLC) in the glasses under investigation, while conduction mechanism in low temperature region occurs via variable range hopping (VRH) of charge carriers in the band tail of the localized states. The DC conductivity is found to increase with Sn concentration up to $6$ at.$\%$ of Sn, after which it decreases. The post gamma irradiation conductivity is, in general, improved at moderated doses i.e., $8$ and $16$ kGy. The density of localized states $N(E_F)$, before and after irradiation, is increased with incorporation of Sn up to $6$ at.$\%$, after which a drastic decrease is observed to occur. This is attributed to an increase in the average single bond energy of $\text{In}_2\text{Sn}_8\text{Se}_{90}$ which decreases the conductivity and the density of localized states for this particular composition. The hopping energy $W$ and hopping distance $R_{hop}$ have also been calculated and their values agree fairly well with the concept of (VRH) model and are in support of our results regarding the behavior of the electrical conductivity.

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1. Introduction

A survey of literature reveals that electrical measurements on some chalcogenide glasses were carried out since the sixties of the last century through the pioneer work of Ovshinsky [1]: The discovery of switching and memory effects in these glasses. Since then, these materials have drawn intensive attention from researchers and have reached now the stage of practical applications. Indeed, it is reported that [2–5] they have many applications in electronic and optoelectronic devices such as CD recording, optical memory, DVD-RAM, ovonic unified memory, and many other devices in which the memory effect is based on the rapid phase change produced under the action of electrical current pulses. It is evident that these electrical applications of chalcogenide glasses are based on the electron transport mechanism that is involved in these materials.

The nature of electron transport in chalcogenide glasses still remains under extensive investigation. However, studying the temperature and field dependence of electrical conductivity and analyzing the $I$–$V$ characteristics of these materials provide essential information about the nature of defect centers which are responsible for the type of conduction in amorphous glassy systems. The density of defect states in the mobility gap controls the electrical properties of chalcogenide glasses and can be calculated from the determination of the conduction mechanism. There are various models for the conduction mechanism from which the density of localized states can be determined. In the case of low temperatures, the non activated transport can be described by Mott’s variable range hopping (VRH) of charge carriers between localized states [6,7]. This model has been applied to a large number of materials among which are chalcogenide glasses [8–11]. The density of localized states $N(E_F)$ and other parameters can be obtained from the fit of the low temperature experimental data to Mott’s variable range hopping. In the case of high electric fields,
several models were proposed to explain the $I$–$V$ characteristics in these materials such as Poole–Frenkel effect [12–14] and space charge limited conduction [15].

In spite of their potential applications that have been mentioned earlier, yet chalcogenide glasses prepared by melt quenching technique lack stability [16,17] with temperature and time during their usage in certain applications. Hence, to avoid this instability, it is recommended that a glass must be heat-treated, stored for long period of time, and/or irradiated by external influences before their final use as suggested by many researchers [17–22]. According to the results of these studies, the structure of a glass is modified under the influence of irradiation, giving rise to changes in the various physical properties, such as a shift of the optical absorption edge, the refractive index, the activation enthalpy, relaxation time and the glass transition temperature without any atomic transmutation or surface damage. Although the studies concerning the effect of gamma irradiation on thermal and optical properties are few [23–30], but still less attention has been devoted to the effect of irradiation on the electrical properties of Se based ternary chalcogenide glasses.

The effect of gamma irradiation on thermal, optical and electrical properties of binary Se–Sn glasses has just recently been reported from this laboratory [18,19,23]. Besides, crystallization kinetics, thermal stability and glass forming ability of In$_2$Sn$_x$Se$_{200}$ ($x = 2, 4, 6,$ and $8$) glasses have also been studied by the group [31]. The effect of gamma irradiation on the optical behavior of the same compositions of Se–In–Sn glasses has recently been reported by us [32]. In this research paper, electrical properties of In$_{10}$.Sn$_x$.Se$_{90}$ ($x = 2, 4, 6,$ and $8$) chalcogenide glasses before and after gamma irradiation (at doses of $8, 16$ and $32$ kGy) will be covered through the measurements of $I$–$V$ characteristics and the temperature dependence of DC conductivity in the temperature range (170–290 K). In addition, the density of localized states $N(E_F)$, the hopping distance $R_{hop}$ and the hopping energy $W$ will be obtained using Mott’s variable range hopping model. It is found that the studied samples showed an ohmic behavior at low voltages (electric fields) while at higher voltages (field), a non-ohmic behavior is observed. The post $\gamma$-irradiation induced changes in the electrical properties of the studied glasses have also been discussed.

2. Experimental details

Homogeneous glassy alloys of In$_{10}$.Sn$_x$.Se$_{90}$ ($x = 2, 4, 6,$ and $8$) were prepared by the well-established melt quenching technique that is described elsewhere [31]. The glassy nature of the samples was confirmed by the DSC measurements. Samples in the form of small discs (diameter ~1.3 cm and thickness ~0.05 cm) were produced by grinding the alloys to a fine powder and then compressing them in a die under a load of ~6 Tons. The samples were irradiated at room temperature using Co$^{60}$ gamma ray sources (available at the Jordan Atomic Energy Commission). The duration of gamma irradiation is such that the overall accumulated doses were $8, 16$ and $32$ kGy. For measurement of DC electrical conductivity at different temperatures, a specially designed sample holder with two copper circular electrodes was used. In this arrangement the sample is sandwiched between the two electrodes and inserted in ARS closed cycle cryostat (model DE104) under a vacuum down to $10^{-4}$ mbar. The samples were coated with silver paint to ensure good electrical contact with the electrodes. $I$–$V$ characteristics were obtained at various temperatures ranging from $170$ to $290$ K by applying DC voltage ranging from $0$ to $20$ V across the samples and measuring the resultant current by Keithley 6430 digital electrometer. Samples’ temperature was monitored using LakeShore 331 temperature controller. The resistance of the samples at various temperatures was measured from the linear portions of the resultant $I$–$V$ curves.

3. Results and discussion

The typical $I$–$V$ characteristics in the voltage range (0–20 V) at different temperatures are presented in Fig. 1 for the non-irradiated In$_2$Sn$_x$Se$_{90}$ glass and Fig. 2 for the In$_8$Sn$_2$Se$_{90}$ glass irradiated at a $\gamma$-dose of $8$ kGy. Similar figures, but not shown, were also obtained for other compositions and doses. From these figures it is observed that, for both the cases before and after gamma irradiation and at all measuring temperatures, the studied samples exhibit an ohmic behavior at low voltage range (0–5 V) due to the presence of thermally generated carriers (electrons and/or holes) [14]. However, at higher voltage (higher electric field), the behavior deviates from the linearity, i.e., non-ohmic behavior is observed to occur.

In order to understand the whole mechanism in the studied samples, the issue could be divided into three parts. First one is related to the deviation of ohmic behavior towards non-ohmic behavior of the $I$–$V$ characteristics at higher applied voltage. The second is related to the composition and $\gamma$-dose dependence of conductivity, while the third is related to the variation of conductivity with temperature.

Non-ohmic behavior has been observed in many chalcogenide glasses [13,14,23–35]. Several models were used to interpret the nonlinearity of the $I$–$V$ characteristics of these glasses. One of these models is the space charge limited conduction (SCLC). In this model, the voltage applied across the sample causes an electrode to inject a non-equilibrium density of electronic charges. The injected charges are populated in the empty gap above Fermi level and will be larger than the thermally generated free carriers causing the $I$–$V$ characteristics to deviate from linearity [15,36]. According to the theory of space charge limited conduction model and in the case of a uniform distribution of localized states, the relation between the current and the voltage is given in good approximation by [15]:

$$I = \frac{2eA\mu n_0 V}{d} \exp(SV)$$  \hspace{1cm} (1)

where $e$ is the charge of electron, $A$ is the cross-sectional area of the sample, $\mu$ is the mobility, $n_0$ is the density of the thermally generated charge carriers, $d$ is the electrode spacing which is the sample thickness, and $S$ is given by:

$$S = \frac{2\varepsilon_0 \varepsilon_r}{eN(E_F)k_BTd^2}$$  \hspace{1cm} (2)

where $\varepsilon_r$ is the static value of the dielectric constant of the sample, $\varepsilon_0$ is the permittivity of free space, $N(E_F)$ is the density of traps (localized states) near the Fermi level and $k_B$ is the Boltzmann constant.
constant. It is evident from Eq. (1) that the plot of \( \ln(I/V) \) vs. \( V \) should be a straight line whose slope \( S \) should decrease with increasing temperature as clear from Eq. (2). Such a plot is shown in Fig. 3 for un-irradiated In\(_2\)Sn\(_8\)Se\(_90\) glass at different temperatures. Similarly, Fig. 4 depicts the same plot for In\(_6\)Sn\(_4\)Se\(_90\) glass irradiated at a dose of 8 kGy. It is found from this plot and similar plots that the DC conductivity increases with increasing temperature. In addition, the conductivity at a particular temperature and a given dose increased with increasing Sn content up to 6 at.% and then decreases for 8 at.% of Sn. This result can be explained on the basis of glass structure. Schottmiller et al. [37] have studied the structure of amorphous Se and the effect of some additives on its structure. They found that in the glassy Se about 60% of the atoms are bonded as polymeric chains, while rest of the atoms have a ring structure and that the additive element enters into the Se chains rather than into the ring structure. Hence, in Se–In glass In enters into the chains of Se and forms In\(_2\)Se\(_3\) structural units [32]. The addition of Sn to Se–In glass yields the required ternary, the structure of which contains SnSe\(_{4/2}\) structural units embedded in a matrix composed of In\(_2\)Se\(_3\). The formation of SnSe\(_{4/2}\) was confirmed by Wang et al. [38] in their FTIR study on the effect of Sn on the properties and structure of Se based chalcogenide glass. The addition of Sn causes the formation of Se–Sn (174.3 kJ/mol) bonds at the cost of Se–In (153.6 kJ/mol) bonds [32]. Because bonds are formed in the sequence of decreasing bond energy until the available valences of atoms are satisfied [39]. This decreases the average single bond energy of the glasses which in turn increases the cohesive energy and is responsible for the corresponding increase in the conductivity of the glasses up to 6 at.% of Sn. However, the decrease in the
conductivity at 8 at.% of Sn may be attributed to readjustment in the local environment which involves the formation of SnSe$_4^{2-}$ tetrahedral with coordination number of Sn = 4 [40,41] rather than 6. This makes the average coordination number of In$_2$Sn$_8$Se$_9$O equal to 2.22 rather than 2.38 [32] which increases the average single bond energy of this composition and hence decreases its conductivity. This further causes a decrease in the density of localized states as we will see. In addition, for In$_8$Sn$_2$Se$_9$O and similar compositions, the energy of this composition and hence decreases its conductivity at 8 at.% of Sn may be attributed to readjustment in the local environment which involves the formation of SnSe$_4^{2-}$ tetrahedral with coordination number of Sn = 4 [40,41] rather than 6. This makes the average coordination number of In$_2$Sn$_8$Se$_9$O equal to 2.22 rather than 2.38 [32] which increases the average single bond energy of this composition and hence decreases its conductivity. This further causes a decrease in the density of localized states as we will see. In addition, Fig. 6 for In$_8$Sn$_2$Se$_9$O and similar figures (not shown) for In$_n$Sn$_{2n}$Se$_{2n+8}$ and In$_2$Sn$_4$Se$_{16}$ show that the conductivity, in general, increases under the influence of $\gamma$-irradiation and attains its maximum value at 8 kGy $\gamma$-dose, while the maximum value of conductivity for In$_n$Sn$_{2n}$Se$_{2n+8}$ glass occurs at 16 kGy $\gamma$-dose as evident from Fig. 7.

The dependence of DC conductivity on temperature shows that two types of conduction channels are involved in the samples under consideration: The first is the one in the high-temperature region which arises from band conduction through the extended states. Meanwhile the other is in the low temperature region which is due to the excitations of the carriers into localized states at band edges that cause the variable range hopping conduction [6]. In the high-temperature region, the DC conductivity increases exponentially with temperature and can be expressed by the following Arrhenius formula [42]:

$$\sigma(T) = \sigma_o e^{-E_c/k_BT}.$$  

(4)

Here, $\sigma_o$ is the pre-exponential factor including the charge carrier mobility and the density of states. On the basis of $\sigma_o$ values one can decide whether the conduction mechanism took place in the extended states above the mobility edge or by hopping in the localized states. According to Mott [43] for conduction in the localized states, for which $\sigma_o$ is in the range $10^3$–$10^4\text{ }\Omega^{-1}\text{ cm}^{-1}$ [44,45], the value of the pre-exponential factor should be two or three orders smaller in magnitude than for conduction in the extended states and should become smaller for conduction in the localized states near Fermi level, $E_F$ is the corresponding activation energy, which for conduction in the extended states, represents the energy difference between mobility edge and Fermi level ($E_c - E_F$) or ($E_F - E_v$) for conduction in the localized states [33]. $E_o$ is usually approximately half of the optical band gap $E_o$, indicating that the Fermi level lies near the mid-gap ($E_o \approx 2E_o$) [41]. The values of $E_o$ for un-irradiated and irradiated samples can be calculated from the slopes of the linear fits of $\ln(\sigma)$ against $1/T$ in the high-temperature region (250–290 K). Fig. 8 shows such a plot for In$_2$Sn$_4$Se$_{16}$ glass for high and low temperature regions that are distinguished by the kink that occurs in the plot at the temperature where the vertical dashed line is drawn. The calculated values of $E_o$ are given in Table 1. It is evident that these values are smaller than the corresponding values of the half-width of the optical band gap, that are reported in our earlier communication ($E_o = 1.69$–2.12 eV) [32] for thin films of the same glasses. The obtained values of the pre-exponential $\sigma_o$ for un-irradiated glass are also given in the same table. Such small values of $E_o$ and $\sigma_o$ indicate that the conduction takes place because of thermally assisted charge carriers movement in the band tail of the localized states, which has also been observed for other chalcogenide glasses [44–46]. It is also clear from Table 1 that the value of the pre-exponential value increases with Sn concentration, with approximately two orders larger in magnitude for 8 at.% of Sn than that for other compositions. This is an indication that the mobility of charge carriers in the trap states increases with the addition of Sn.

$$\sigma(T) = \sigma_o e^{-E_c/k_BT}.$$  

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<table>
<thead>
<tr>
<th>Sample</th>
<th>Activation energy (meV)</th>
<th>The pre-exponential $\sigma_o(\Omega^{-1}\text{ cm}^{-1})$ un-irradiated</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se$<em>{90}$In$</em>{10}$Sn$_8$</td>
<td>88</td>
<td>40</td>
</tr>
<tr>
<td>Se$<em>{90}$In$</em>{10}$Sn$_4$</td>
<td>37</td>
<td>34</td>
</tr>
<tr>
<td>Se$<em>{90}$In$</em>{15}$Sn$_5$</td>
<td>34</td>
<td>15</td>
</tr>
<tr>
<td>Se$<em>{90}$In$</em>{15}$Sn$_4$</td>
<td>99</td>
<td>72</td>
</tr>
</tbody>
</table>
Table 2

<table>
<thead>
<tr>
<th>Sample</th>
<th>Density of localized state $N(E_F) \times 10^{27}$ (eV$^{-1}$ cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Dose (kGy)</td>
</tr>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>0.033</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>0.179</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>0.418</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>0.099</td>
</tr>
</tbody>
</table>

The conductivity in the low temperature region (170–250 K), increases slowly with increasing temperature, which suggests that the conduction occurs due to variable range hopping of charge carriers. This mechanism is characterized by Mott’s expression [6,7]:

$$\sigma(T) \sqrt{T} = B e_{\lambda}(T_0/T)^{\alpha}$$

where $B$ is a constant related to the density of localized states, $T_0$ is the degree of disorder $= \lambda e(\frac{k_B}{N(E_F)}$, $N(E_F)$ is the density of localized states at the Fermi level, $\lambda$ is a dimensionless constant ($=18$) and $\alpha$ represents the coefficient of exponential decay ($\exp(-\alpha R)$) of the localized states wave function ($\alpha \approx 0.124$ $\text{cm}^{-1}$) [47]. From Eq. (5), a plot of $\ln(\sigma T^{1/2})$ vs. $(T)^{-1/4}$ should give a straight line. The values of the density of localized state $N(E_F)$ have been obtained for all samples from the fit of the low temperature experimental data to Mott’s variable range hopping and tabulated in Table 2. Depending on this table, one can see that the density of localized states increases with the increase in Sn concentration in the system up to 6 at.% and then decreases at 8 at.%. The increase in the density of localized states reflects an increase in the conductivity and a corresponding decrease in the activation energy. This further agrees fairly well with our earlier result regarding the local environmental re-adjustment caused by the formation of SnSe$_{92}$ tetrahedral in In$_{4}$Sn$_{6}$Se$_{90}$ composition and leads to an increase in its average single bond energy and decreases its density of localized states. This is found to be associated with a shift in Fermi level with Sn incorporation [48]. In addition, it is also noticed that the density of localized states increases for all compositions with increasing γ-ray dose and attains its maximum value at 8 kGy, after this dose it decreases but still remains above that of un-irradiated glass. It is reported that [19,23,24,32,49,50] γ-irradiation of chalcogenide glasses causes some kind of chemical bonds rearrangement through breaking weak bonds and creation of new ones. According to the bonds distribution model which was suggested by Golovchak et al. [24] it is assumed that chalcogen located 1p-electrons as well as σ-electrons of covalent bonds are excited. However, these electronic disturbances are not strongly localized, but decay giving rise to significant atomic displacements within the whole glass network with a new distribution of bonds. It is more likely that the change takes place in the chains, that is, the bonds within the chains may be broken and built between chains. It is mentioned that Se-In bonds have bond energy less than that of Se-Sn bonds, so it is expected that Se-In bonds are more sensitive to gamma irradiation. Therefore, upon gamma irradiation some of Se-In bonds are broken leading to the formation of In-In homopolar bonds, because of their low energy of formation 100.32 kJ/mol [32] and the Se atoms re-built within the Se chains. This process allows the formation of defects which produce localized states that change the effective Fermi level due to an increase in carrier concentrations. This increase in carriers in localized states will lead to a decrease in the transition probabilities into the extended states, resulting in a reduction in the gap [29,32] with a corresponding increase in the conductivity.

Two other hopping parameters, the average hopping distance ($R_{\text{hop}}$) and the average hopping energy ($W$) can also be calculated. According to Mott [6,7] and Hill [51] these two parameters are given by:

$$R_{\text{hop}} = \left(\frac{9}{8\pi a k_B N(E_F)}\right)^{1/4}$$

Table 3

<table>
<thead>
<tr>
<th>Sample</th>
<th>Dose (kGy)</th>
<th>$R_{\text{hop}}$ ($10^{-7}$ cm)</th>
<th>$W$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0</td>
<td>8</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>2.6</td>
<td>43</td>
<td>1.1</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>1.7</td>
<td>28</td>
<td>0.8</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>1.4</td>
<td>22</td>
<td>0.74</td>
</tr>
<tr>
<td>SnSe$<em>{90}$In$</em>{4}$Sn$_{6}$</td>
<td>2.0</td>
<td>33</td>
<td>1.0</td>
</tr>
</tbody>
</table>
\[ W = \frac{3}{4\pi R_{\text{hop}}^2 N(E_F)} \tag{7} \]

The calculated values of \( R_{\text{hop}} \) and \( W \) at \( T = 230 \text{ K} \) are given in Table 3 for all samples and doses. From this table, it is clear that both of hopping energy and hopping distance decrease under the influence of \( \gamma \)-irradiation and they have their minimum values at gamma dose of 8 kGy. As the hopping energy and hopping distance decrease the conductivity due to variable range hopping model increases and this is in good agreement with our results mentioned above. In addition, the variations of the hopping energy and hopping distance with temperature for In\(_{10}\)Sn\(_4\)Se\(_90\) for the two cases before and after \( \gamma \)-irradiation are given in Fig. 9. This figure shows that the hopping distance decreases with increasing temperature, while the hopping energy increases with increasing temperature. These results are in complete agreement with the concept of variable range hopping which was developed by Mott [6,7].

4. Conclusions

Electrical parameters for In\(_{10-x}\)Sn\(_x\)Se\(_90\) (\( x = 2, 4, 6, \text{ and } 8 \)) chalcogenide glasses have been studied in the temperature range of 170–290 K before and after gamma irradiation at doses of 8, 16 and 32 kGy. The effect of composition and gamma irradiation on the electrical parameters of the present glasses can be summarized in the following:

- The DC conductivity increases with Sn concentration up to 6 at.% after which it decreases due to the formation of SnSe\(_4\) tetrahedral induced by local environmental readjustment which involves the change in the coordination number of Sn. It follows that it is not advisable to use the compositions of higher percentage of Sn in applications where good electrical conduction is required.
- The conduction in all the studied compositions occurs due to thermally assisted charge carriers movement in the band tail of the localized states, for both cases of un-irradiated and gamma irradiated samples. The obtained parameters of VRH model, \( \sigma_0 \), \( R_{\text{hop}} \), and \( W \) are in support of this argument.
- Irradiation at moderate doses of \( \gamma \)-ray seems to be good for improving the conductivity and the stability through increasing the chain length of Se.

Acknowledgements

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References