DC CONDUCTION AND MEYER-NELDEL RULE IN Se_{85-x}Te_{15} In_x

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In this paper, we report dc conduction in Se_{85-x}Te_{15} In_x (x= 0, 2, 6, 10, 15) chalcogenide glasses. The measurements have been made in the temperature range 213K to 293K. The ln(\sigma_{dc}) versus 1000/T plots are almost straight lines in whole studied temperature range, slopes of graphs indicate lowest activation energy for Se_{75}Te_{15}In_{10}. The dc conductivity (\sigma_{dc}) versus concentration of Indium graph shows that Se_{75}Te_{15}In_{10} is most conducting. It has been established that Meyer –Neldel (MN) rule is not obeyed for the complete series but MN rule is followed for the series if Se_{75}Te_{15}In_{10} is excluded.

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1. Introduction

Chalcogenide glasses have some interesting technological properties [1] due to which they have been extensively investigated in recent years. Selenium tellurium based chalcogenides have received particular attention because of their high potential for application in electronics [2-4]. Generally, undoped chalcogenide glasses show low electric conductivity which limits their technological application. Certain additives are used to improve their properties. The effect of an additive in an amorphous semiconductor may be widely different, depending on the conduction mechanism and the structure of material [5]. The addition of Indium as third element at different percentages in Se-Te binary chalcogenides produces stability [6] in these glasses. Keeping this in mind, we have studied the effect of Indium impurity on dc conduction in Se_{85}Te_{15}. This paper reports the dc conductivity behaviour of Se_{85-x}Te_{15}In_x (x=0, 2, 6, 10, 15) and the applicability of Meyer –Neldel (MN) rule in the studied glasses. The peculiar behaviour of Se_{75}Te_{15}In_{10} has been explained.

2. Experimental details

For the present work the glassy materials have been prepared by melt quench technique. The constituent elements (5N pure) in the required atomic weight percentages were sealed in properly cleaned quartz ampoules under a vacuum of the order of 10^{-5} mbar using diffusion pump. The sealed ampoules were heated upto 650\textdegree C in the rocking furnace initially. Later the temperature of the furnace was raised upto 700\textdegree C and maintained at this temperature for about 24hrs. Rocking has been done to ensure proper mixing and homogeneity of samples. Heated ampoules were immediately cooled in liquid nitrogen for the materials to go into glassy state. Samples were crushed, separated, ground and characterized for their amorphous nature using X-ray diffraction technique. Compressed pellets were prepared by grinding bulk-ingots into fine

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powder and compressing the powder in a die under a hydraulic press \((10^3 \text{kg/m}^2)\). A three terminal sample holder fabricated for the measurement of AC and DC conductivity of pellet- shaped samples has been used. A thermocouple inserted inside the sample holder and kept close to the sample measures correct temperature of the samples. A vacuum of the order of \(10^{-4}\) to \(10^{-5}\) Torr was achieved inside the sample holder using vacuum pump. For DC conductivity measurements, a constant voltage 20V was applied across all the samples using dc power supply (CROWN DC-regulated Power Supply 0-30V/2A) and the resulting current was measured with digital nanometer (Digital Nanometer DNM-121, Scientific equipment Roorkee) having least count of 0.1nA.

3. Results and discussion

The measurements of dc conductivity in the case of amorphous semiconductors have yielded valuable information about the transport mechanism in these materials. The majority of amorphous semiconductors (chalcogenides and tetrahedrals) show activated temperature dependence according to the relation (1),

\[
\sigma = \sigma_0 \exp(-\Delta \mathcal{E}/kT)
\]

where \(\Delta \mathcal{E}\) and \(\sigma_0\) are the activation energy and pre-exponential factor respectively. Figure 1 shows temperature dependence of dc conductivity for \(Se_{85-x}Te_{15}In_x\) \((x=0, 2, 6, 10, 15)\) in the temperature range of 213K-293K.

![Fig. 1. Temperature dependence of dc conductivity](image)

From the graphs, the activation energies for the \(Se_{85}Te_{15}In_2\), \(Se_{79}Te_{15}In_6\), \(Se_{75}Te_{15}In_{10}\), \(Se_{70}Te_{15}In_{15}\) come out to be 0.38eV, 0.43eV, 0.49eV, 0.29eV, 0.44eV respectively [7]. The activation energy is lowest for \(Se_{75}Te_{15}In_{10}\). The variation of dc conductivity with x at 293K is shown in Fig.2.
From figure 2, it is clear that dc conductivity of Se-Te glass increases with increase in Indium content at about room temperature and Se$_{75}$Te$_{15}$In$_{10}$ is most conducting as compared to others in the series. Similar results have been reported on dc conductivity in Se$_{100-x}$In$_x$ \[8\]. This behaviour can be explained by considering the structural changes due to introduction of more and more Indium atoms into the Se-Te system.

Se-Te system prepared by melt quench technique has structure consisting of Se$_8$ rings, Se$_6$Te$_2$ rings and Se-Te copolymer chains \[9\]. In the series under study, addition of Indium is at the cost of Selenium concentration, so Indium affects the Se-Se bonds the most. So the variation in electric conductivity can be explained on the basis of formation of Se-Se, Se-In and In-In bonds.

When we add Indium at the cost of Selenium, Indium makes ionic-covalent bonds with Selenium and is dissolved in Selenium chain. These bonds offer more conducting path in the system, so conductivity of Se-Te glasses increases with increase in Indium content. In the present case, when concentration of Indium is increased, some of Se-Se structural units are replaced by Se-In structural units in Se$_8$ rings which results in increase of chain length and hence increase in conductivity. For Se$_{75}$Te$_{15}$In$_{10}$, the system appears to possess maximum chain length heavily cross-linked with ring concentration which accounts for its maximum conductivity. This sudden increase in conductivity leads to fall in its activation energy. When the concentration of Indium is further increased, concentration of In-In bonds increases, the system becomes more disordered and density of localized states decreases which leads to decrease in conductivity of Se$_{75}$Te$_{15}$In$_{15}$ as compared to Se$_{75}$Te$_{15}$In$_{10}$.

The correlation between $\sigma_0$ and $\Delta\mathcal{E}$ is given by

$$\sigma_0 = \sigma_{00} \exp(\Delta\mathcal{E}/E_{MN})$$

where $\sigma_{00}$ and $E_{MN}$ are constants for the system. The above equation is known as Meyer – Neldel (MN) rule \[10\] which is applicable to various thermally activated phenomena. $E_{MN}$ is called Meyer – Neldel characteristic energy and $\sigma_{00}$ is MN pre exponential factor. Recently, Meyer-Neldel rule has been found to be applicable to thermally activated phenomenon like dc conduction and ac conduction in chalcogenide glasses \[11-17\].

From the plots in figure 1, the value of $\ln\sigma_0$ has been calculated for Se$_{85-x}$Te$_{15}$In$_x$. It has been observed that $\sigma_0$ is not constant for the series under study. The values of $\ln\sigma_0$ and $\Delta\mathcal{E}$ for the series under study are shown in Table 1.
Fig. 3. Plot of $\ln \sigma_0$ versus $\Delta E$ for Se$_{85-x}$ Te$_{15}$ In$_x$

From the results it is clear that $\ln \sigma_0$ increases with increase in $\Delta E$ as expected for chalcogenide glasses (Eqn.2) except for Se$_{75}$Te$_{15}$In$_{10}$. So we can say that MN rule is not obeyed for the whole series under study. This is shown in the plot between $\ln \sigma_0$ and $\Delta E$ (Figure 3). This deviation from MN rule is due to exceptional behavior of Se$_{75}$Te$_{15}$In$_{10}$.

If we exclude the Se$_{75}$Te$_{15}$In$_{10}$ from the series, plot between $\ln \sigma_0$ and $\Delta E$ is a straight line (Figure 3) indicating that $\sigma_0$ varies exponentially with $\Delta E$ according to the relation (2) and MN rule is obeyed.

The slope and intercept of straight line plot (Figure 3) yield Meyer–Neldel characteristic energy, $E_{MN} = 17.04$ meV and MN pre exponential factor, $\sigma_{00} = 1.678 \times 10^9$ Sm$^{-1}$. The values are close to the range suggested by Shimakawa and Abdel-Waheb [11] for chalcogenide glasses.

Using the values of $E_{MN}$ and $\sigma_{00}$, the expected $\sigma_0$ values have been calculated for all samples of Se$_{85-x}$Te$_{15}$In$_x$ (x=0, 2, 6, 15) (Table 3.3).

Table 3.3. Semiconducting Parameters for Se$_{85-x}$Te$_{15}$In$_x$

<table>
<thead>
<tr>
<th>x</th>
<th>$\Delta E$ (eV)</th>
<th>$\ln \sigma_0$ (Sm$^{-1}$)</th>
<th>$\ln \sigma_0 = \ln \sigma_{00} + \frac{\Delta E}{E_{MN}}$ (Sm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.38</td>
<td>1.3004</td>
<td>1.8725</td>
</tr>
<tr>
<td>2</td>
<td>1.05</td>
<td>5.2787</td>
<td>4.8904</td>
</tr>
<tr>
<td>6</td>
<td>1.06</td>
<td>7.5585</td>
<td>8.2761</td>
</tr>
<tr>
<td>15</td>
<td>0.99</td>
<td>6.5652</td>
<td>5.6734</td>
</tr>
</tbody>
</table>

A good agreement between the two values of $\ln \sigma_0$ confirms the validity of MN rule for Se$_{85-x}$Te$_{15}$In$_x$ (except x=10) glasses.

4. Conclusion

The study of dc conductivity shows that for the series of chalcogenide glasses under study, Se$_{75}$Te$_{15}$In$_{10}$ is most conducting which may be due to the creation of Se-In bonds and increase in chain length. MN rule is not as such followed for complete series but is obeyed if we exclude Se$_{75}$Te$_{15}$In$_{10}$ from the series. This is due to peculiar behavior of dc conduction in Se$_{75}$Te$_{15}$In$_{10}$.

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References