Indian Journal of Pure & Applied Physics
Vol. 41, June 2003, pp. 466-469

# Pressure dependence of thermal conductivity and thermal diffusivity of Se-Te -In chalcogenide glasses

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Received 3 February 2003; revised 7 April 2003; accepted 25 April 2003

Measurements of effective thermal conductivity ($\lambda_e$) and effective thermal diffusivity ($\chi_e$) of twin pellets of $Se_{80}Te_{20-X}In_X$ ($x$= 2, 4, 6 and 10) chalcogenide glasses, prepared under pressure (load) of 5, 7 and 9 tons, have been made at room temperature, using transient plane source (TPS) technique. The results indicate that, both the values of $\lambda_e$ and $\chi_e$ increase with the increase of In concentration. Besides, $\lambda_e$ and $\chi_e$ also depend upon the load at which the pellets have been formed. The composition and pressure (load) dependent behaviour of $\lambda_e$ and $\chi_e$ is explained in terms of the ion-covalent type of bond which In makes with Se as it is incorporated in the Se-Te glasses.

[Keywords: Thermal conductivity, Thermal diffusivity, Se-Te-In chalcogenide glasses]

## 1 Introduction

Great attention has been given to chalcogenide glasses, in recent years, mainly due to their wide range applications as solid-state devices both in scientific and technological field[1]. These glasses exhibit unique IR-transmission and electrical properties that make them useful for several applications such as threshold switching, memory switching, inorganic photo-receptors, IR transmission and detection through lenses and optical wave guides e.g. in welding and surgery[2]. Especially selenium alloys exhibit a unique property of reversible transformation. This property makes these systems very useful in optical memory, X-ray imaging and photonics. The addition of indium as third element in different percentage in Se-Te binary chalcogenide glasses produces stability of these glasses[3]. The effects of element as an additive to binary glasses have been extensively studied by other workers[4]. In the present work, an investigation has been undertaken to study the variation of $\lambda_e$ and $\chi_e$ of these samples with composition at different pressure using transient plane source (TPS) technique[5].

## 2 Transient Plane Source (TPS) Theory

The TPS method consists of an electrically conducting pattern (Fig. 1) in the form of a bifilar spiral,

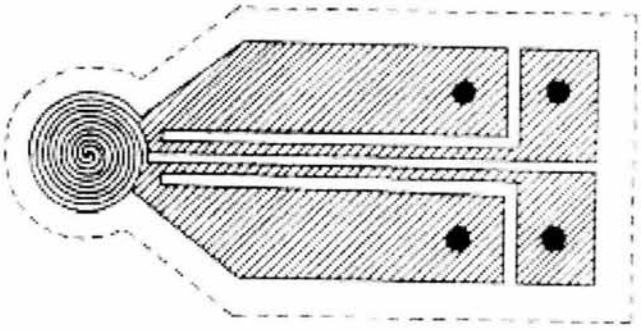

Fig. 1 — Schematic diagram of TPS sensor

which also serves as a sensor of the temperature increase in the sample. Assuming the conducting pattern to be in the $y$-$z$ plane of a coordinate system, placed in an infinite solid material, the rise in the temperature at a point $y$-$z$ at time, ($t$), due to an output power through the spiral per unit area $Q$ is given by[5]:

$$\Delta T(y,z,\tau)=\frac{1}{4\pi^{3/2}a\lambda}\int_0^\tau\frac{d\sigma}{\sigma^2}\int_A dy',dz'Q\left(y',z',t\frac{\sigma^2a^2}{\chi}\right)\exp\left[\frac{-(y-y')^2-(z-z')^2}{4\sigma^2a^2}\right] \quad \ldots(1)$$

where $\chi(t\text{-}t') = s^2 a^2$, ? = $a^2/\chi$

and t = $[t/\theta]^{1/2}$

$a$ is the radius of the hot disc (source and the sensor) which gives a measurement of the overall size of resistive pattern and $\theta$ is known as the characteristic time. $\sigma$ is a constant variable, $\lambda$ is the thermal conductivity in units of W/mK and $\chi$ is the thermal diffusivity in unit of $m^2/s$ of the sample. The temperature increase $\Delta T\ (y, z, \tau)$ because of flow of current through the sensor gives rise to a change in the electrical resistance $\Delta R(t)$ which is given as:

$$\Delta R(t)=\alpha R_0\ \overline{\Delta T(\tau)} \quad \ldots(2)$$

where $R_o$ is resistance of TPS element before the transient recording has been initiated, at room temperature, a the temperature coefficient of resistance (TCR) and $\overline{\Delta T(\tau)}$ the properly calculated mean value of the time-dependent temperature increase of the TPS element. $\overline{\Delta T(\tau)}$ is calculated by averaging the increase in temperature of TPS element over the sampling time, because the concentric ring sources in TPS element have different radii and are placed at different temperatures during the transient recording. During the transient event, $\overline{\Delta T(\tau)}$ can be considered to be a function of time only, whereas, in general, it will depend on such parameters, as the output power in TPS element, the design parameters of the resistive pattern, and the thermal conductivity and thermal diffusivity of surroundings.

It is possible to write down an exact solution for the hot disc, if it is assumed that, the disc contains a number ?$m$' of concentric rings as sources. From the ring source solution we immediately get:

$$\overline{\Delta T(\tau)} = \frac{P_0}{\pi^{3/2} a\lambda} D_s(\tau) \quad \ldots(3)$$

where

$$D_s(\tau) = [m(m+1)]^{-2}$$

$$\int_0^{\tau} \frac{d\sigma}{\sigma^2}\left[\sum_{l=1}^{m} l \left\{\sum_{k=1}^{n} k \ \exp\frac{-(l^2+k^2)}{4\sigma^2 m^2} L_0\left(\frac{lk}{2\sigma^2 m^2}\right)\right\}\right] \quad \ldots(4)$$

In Eq. (4), $P_o$ is the total output power, $L_o$ the modified Bessel function and $l$, $k$ the dimensions of the resistive pattern. To record the potential difference variations,

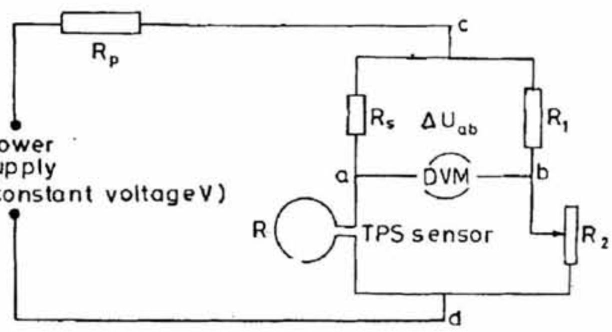


Fig. 2 — Schematic diagram of electrical circuit used for simultaneous measurement of effective thermal conductivity and effective thermal diffusivity

which normally are of the order of a few millivolts during the transient recording, a simple bridge arrangement as shown in Fig. 2 has been used. If it is assumed that, the resistance increase will cause a potential difference variation $\Delta U(t)$ measured by the voltmeter in the bridge, the analysis of the bridge indicates that:

$$\Delta E(t) = \frac{R_s}{R_s + R_0} I_0 \Delta R(t) = \frac{R_s}{R_s + R_0} \frac{I_0 \alpha R_0 P_0}{\pi^{3/2} a\lambda} D_s(\tau) \quad \ldots(5)$$

where

$$\Delta E(t)=\Delta U(t)[1-C.\Delta U(t)]^{-1} \quad \ldots(6)$$

and

$$C = \frac{1}{R_s I_0 \left[1 + \frac{\gamma R_p}{\gamma(R_s + R_0) + R_p}\right]} \quad \ldots(7)$$

The definition of various resistances is found in Fig. 2.

$R_p$ is the lead resistance, $R_s$ a standard resistance with a current rating that is much higher than $I_o$, which is the initial heating current through the arm of the bridge containing the TPS-element. ? is the ratio of the resistances in two ratio arms of the bridge circuit, which is taken to be 100 in the present case.

### 3 Experimental Details

High purity (99.999%) Se, Te and In in appropriate atomic percentages were weighed into a quartz glass ampoule (length 5 cm and internal diameter 8 mm). The contents of the ampoule (5 g) were sealed into a vacuum of $10^{-6}$ Torr and heated in a furnace where temperature was raised at a rate of 3-4 °C per min up to 925 K, and kept around that temperature

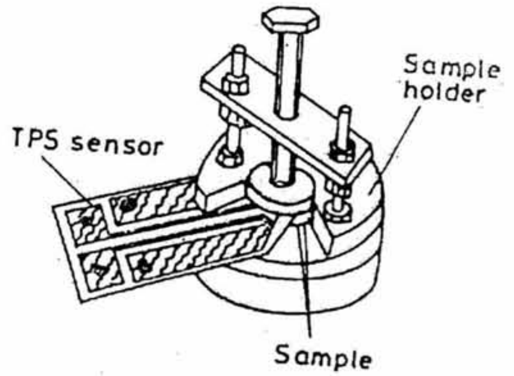


Fig. 3 — Sample holder diagram with TPS sensor

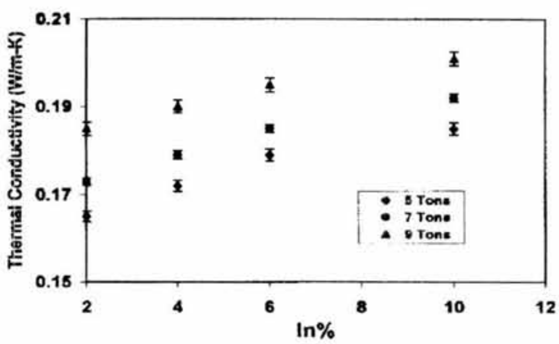


Fig. 4 — Thermal Conductivity with In percentage

for 7-8 hrs, to ensure the homogeneity of the samples. The molten samples were then rapidly quenched in ice-cooled water. Samples obtained by quenching were in the form of glasses. The glassy nature has been confirmed through X-ray diffraction. These bulk glasses were then crushed to fine powders by grinding process. Pellets of thickness 1 mm and diameter 12 mm were prepared by a pressure machine at different pressures.

The samples are in the form of pellets of 12 mm diameter and 1 mm thickness, and the surfaces of these pellets are smooth so as to ensure perfect thermal contact between the samples and the heating elements, as the TPS sensor is sandwiched between the two pellets of sample material in the sample holder [Fig. 3].

The change in the voltage was recorded with a digital voltmeter, which was online to the personal computer.

The power output to the sample was adjusted according to the nature of the sample material and was, in most cases, in the range $6\times10^{-6}$-$16\times10^{-6}$ W/m.[2]

The measurements reported in this paper were performed with a TPS element. It is made of a 10 μm -thick nickel foil (having a resistance of about 3.26 Ω and a TCR around $4.6\times10^{-3}$ $k^{-1}$) with an insulating layer made of 50 μm-thick kapton, on each side of the metal pattern. Evaluation of these measurements was performed in a way that was outlined by Gustafsson[5]. In experiments with insulating layers of such thickness, it is necessary to ignore the voltage recorded during the first few seconds because of the influence of the insulating layers. However, owing to the size of the heated area of the TPS element, the characteristic time of the experiment is so long that, it is possible to ignore a few second of recorded potential difference values and still obtain very good result.

An important aspect of the design of any TPS element is that, the pattern should be such that, as large a part of the "hot" area as possible should be covered by the electrically conducting pattern, as long as there is insulation between the different parts of the pattern. This is particularly important when insulating layers are covering the conduction pattern and the surface(s) of the sample. It should be noted that, the temperature difference across the insulating layer can, after a short initial transient, be considered constant.

## 4 Results and Discussion

Simultaneous measurement of effective thermal conductivity and effective thermal diffusivity of pellets of $Se_{80}Te_{20-x}In_x$ ($x$ =2,4,6 and 10) glasses, compacted under loads of 5, 7 and 9 tons, were carried out, at room temperature, using TPS technique. Variation of effective thermal conductivity ($\lambda_e$) and effective thermal diffusivity ($\chi_e$) with the composition ($x$) of indium, at different pressure has been plotted in Figs 4 and 5, respectively.

It can be observed from Figs 4 and 5, that the effective thermal conductivity and effective thermal diffusivity of the glasses increased slightly with the increase of composition of indium in the Se-Te-In glass at different pressure. Slight increase in the effective thermal conductivity and effective thermal diffusivity could be explained by considering the structural changes due to the introduction of more and more indium atoms.

The structure of the Se-Te system prepared by melt quenching is regarded[6] as a mixture of $Se_8$ rings, $Se_6Te_2$ rings and Se-Te copolymer chains[7]. In the present case, the addition of In is at the cost of Te concentration. Indium makes iono-covalent bonds with Se and is probably dissolved in the Se- chain, making the system more and more thermally stable with the increase of

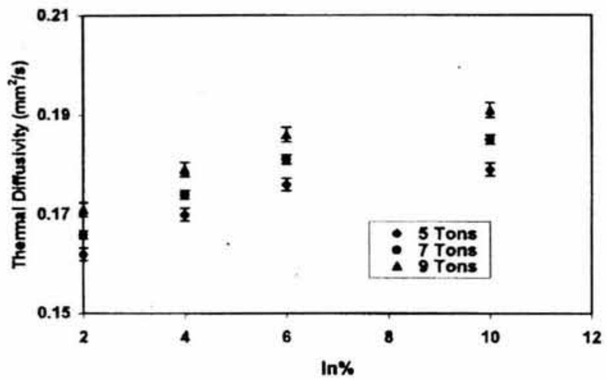

Fig. 5 — Thermal diffusivity with In percentage

indium in the binary Se-Te system. Thermal stability of the chalcogenide glasses is directly related to the thermal conductivity and thermal diffusivity[8]. These iono-covalent bonds having high dissociation energy offer more conductive path in the system and hence provide an easier heat flow from one point to another in the alloy. The effect of formation of iono-covalent bond in these glasses is also reflected in several kinetic parameters[9] during their thermal analysis using differential scanning calorimetry (DSC).

## 5 Conclusion

Formation of iono-covalent bond in Se-Te system with the increased concentration of In seems to be responsible for the slight increase in effective thermal conductivity and diffusivity.

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