



Compositional dependence of thermal stability, glass-forming ability and fragility index in some Se–Te–Sn glasses

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ABSTRACT

Non-isothermal DSC thermograms were obtained for the ternary $\text{Se}_{90}\text{Te}_{10-x}\text{Sn}_x$ ($x=2, 4, 6$ and 8) chalcogenide glasses in order to determine the melting temperature T_m , glass transition temperature T_g , onset T_c and peak T_p temperatures of crystallization. These temperatures were utilized to investigate the thermal stability through the calculations of temperature difference ($T_c - T_g$), the glass transition activation energy E_t , the parameter S and the average value of crystallization rate factor (K_p). In addition, the glass forming ability was estimated by the criteria of reduced glass transition temperature, T_{rg} and Hruby parameter H_R . The fragility index m for the present glasses was determined in order to see whether these materials are obtained from strong or fragile glass-forming liquid. Results reveal that, both thermal stability and glass forming ability exhibit a maximum at $x=4$ at.% of Sn. Meanwhile, the prepared glasses were obtained from strong glass-forming liquid as evident from the fragility index calculations. The compositional dependence of the above parameters was discussed on the basis of Philips and Thorpe topological model and the critical composition occurs at an average coordination number (r) = 2.16 but not 2.40. This is due to the formation of iono-covalent bonds when the glass doped with heavy elements like Sn.

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1. Introduction

In the last 40 years, the interesting properties of chalcogenide glasses make them considered for several applications like digital X-ray imaging [1], solar cells [2], bio-sensing [3], lithography [4], optical elements (lenses, waveguides, gratings, fibres etc.) [5–7] and switching devices [8,9]. In addition, the phase change between crystalline (high conductive) and amorphous (low conductive) states in these materials can be produced by either short electrical pulse or short laser pulse that heats the material above the crystallization or melting temperature, this property make them used in the latest and attractive applications of the phase-change rewritable optical disk, which has a large capacity and is easily handled like the floppy disk and the digital versatile disk (DVD) [10].

Thermal stability and glass-forming ability (GFA) are related but independent properties for a given glass. Glass-forming ability of a glassy alloy is related to the ease by which melt can be cooled with the avoidance of crystal formation [11]. On the other hand, thermal stability represents the resistance to devitrification of glassy alloy through the nucleation and growth processes [12].

Efforts [13–18] have been made to study and to relate thermal stability and glass-forming ability of chalcogenide glasses with their composition because of its importance in determining the utility of chalcogenide alloys as recording materials or in other practical applications. Further, the composition dependence of thermal stability, glass-forming ability and other thermal parameters yields valuable information about the evolution of network connectivity and the network topological thresholds.

Amorphous selenium (*a*-Se) is one of the most widely used amorphous semiconductors. The structure of *a*-Se consists of randomly mixed long polymeric Se_n chains in which various portions of a chain have ring fragments in contrast to a structure which is a mixture of Se_n chains and Se_8 rings [19]. Strong covalent bonds exist between the atoms in the chains and rings, whereas in-between the chains it is weak bonds of Van der Waals type. Se is an excellent glass former, but pure Se has disadvantages such as short lifetime, thermal instability, low crystallization temperature and low sensitivity. To improve these properties, Te is added to Se. Se–Te glassy alloys have advantage over pure *a*-Se because of their higher photosensitivity, greater hardness, higher crystallization temperature and smaller ageing effect [20]. Substitution of Te by Se in Se–Te glasses breaks up the Se_8 ring structure and slightly increases the chain fraction but reduces the chain length [21]. The increase in T_g with Te addition can be assumed to be due to the small fraction of Te atoms that are triply bonded so that they cross-link chains [22]. However,

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Se–Te glassy alloys still have poor thermo-mechanical properties. In order to overcome this problem, to enhance the thermal stability and to enlarge the domain of applications, it is necessary to add a third element (like Sn) as a chemical modifier which expands the glass forming area and also creates compositional and configurational disorder as comparing with the binary Se–Te glassy alloys. The substitution of some Te atoms by Sn atoms causes variation in the physical properties of the primary binary compound [23].

Differential scanning calorimeter (DSC) is a very useful tool that usually used to determine the transformations temperatures, such as glass transition T_g , crystallization T_c and T_p and melting T_m temperatures and to evaluate, using these temperatures, thermal stability and GFA of chalcogenide glasses. In the present work, thermal stability in $\text{Se}_{90}\text{Te}_{10-x}\text{Sn}_x$ ($x=2, 4, 6$ and 8) samples has been studied through the calculations of the temperature difference ($T_c - T_g$), the thermal stability parameter S , the glass transition activation energy E_t , and the average value of crystallization rate factor (K_p). On the other hand, GFA has been investigated through the calculations of the reduced glass transition temperature T_{rg} and Hruby parameter H_R . Meanwhile, to ascertain that the prepared glasses were obtained from strong glass-forming liquids, the fragility index m has also been obtained. An attempt has also been made to apply the topological threshold model on the present glassy system.

2. Experimental details

Glassy alloys of $\text{Se}_{90}\text{Te}_{10-x}\text{Sn}_x$ ($x=2, 4, 6$ and 8) were prepared by melt quenching technique that described elsewhere [15]. About (10 ± 0.5) mg of the powder samples were capsulated in aluminum pan and subjected to the differential scanning calorimeter (Perkin Elmer DSC-7). Heating rates of 5, 10, 15, 20 and 25 K/min were applied in the temperature range from room temperature to about 500°C . The temperature precision of this equipment is ± 0.1 K with an average standard error of about 1 K in the measured values. The DSC equipment is calibrated prior to measurements, using high purity standards Pb, Sn and In with well-known melting points. The operation of a differential scanning calorimeter is based on measurement of the thermal response of an unknown specimen as compared with a standard when the two are heated uniformly. A typical differential scanning calorimeter consists of two sealed pans; a sample pan and a reference pan (which is generally an empty sample pan). These pans are often covered by lids that act as a radiation shield. The two pans are heated, or cooled, uniformly while the heat flow difference between the two is monitored. This can be done at a constant temperature (isothermally), but is more commonly done by changing the temperature at a constant rate (non-isothermally) as in this work.

3. Results and discussion

3.1. Thermal stability

The non-isothermal DSC thermograms at five different heating rates 5, 10, 15, 20 and 25 K/min for $\text{Se}_{90}\text{Te}_6\text{Sn}_4$ alloy are shown in Fig. 1, as an example. Similar thermograms were obtained for other compositions. Four characteristic temperatures, namely the glass transition temperature T_g , the crystallization temperatures T_c and T_p and the melting temperature T_m , are observed in the all resultant curves. Both T_g and T_c have been defined as the temperatures which correspond to the intersection of two linear portions adjoining the transition elbow of the DSC traces in the endothermic and exothermic directions, respectively [17]. The characteristic temperatures T_g , T_c , T_p and T_m are the factors that used to determine the stability of the glass and its GFA by using different simple quantitative methods.

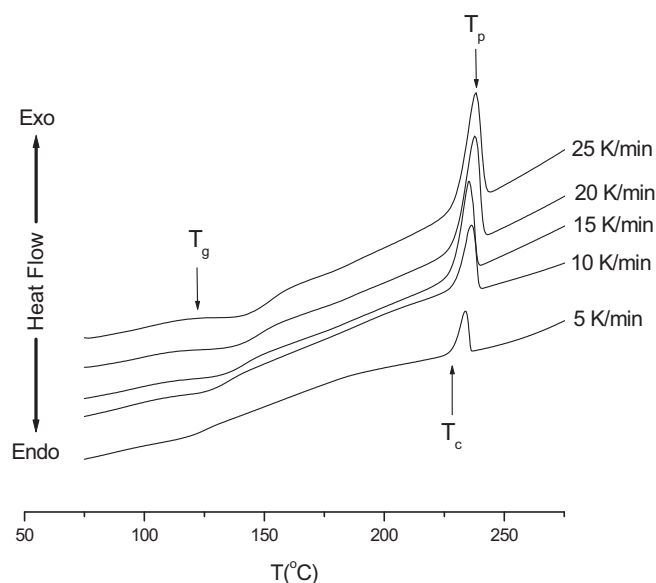


Fig. 1. DSC non-isothermal thermograms at different heating rates for $\text{Se}_{90}\text{Te}_6\text{Sn}_4$.

The glass transition temperature T_g represents the strength or rigidity of the glassy structure of the alloy, but it is alone does not give any information on the thermal stability of the glass [24]. However, Dietzel [25] suggested that the difference between the crystallization and the glass transition temperatures ($T_c - T_g$) is a good indication of thermal stability. Usually, in a stable glass, the crystallization peak is closed to the melting temperature while in an unstable glass; the crystallization peak is closed to the glass transition temperature. Therefore, the higher the value of ($T_c - T_g$), the more is the delay in the nucleation process, the greater is the thermal stability, and the easier is the glass formation [12]. The values of ($T_c - T_g$) for the different compositions of the studied glasses a heating rate of 5 K/min is given in Table 1. Saad and Poulin [26] used this difference to introduce another thermal stability parameter S , given by:

$$S = \frac{(T_c - T_g)(T_p - T_c)}{T_g} \quad (1)$$

where $T_p - T_c$ is related to the rate of devitrification transformation of the glassy phases and S reflects the resistance to devitrification of the glassy phase, and hence the glass of higher values of S -parameter has higher stability. Values of S -parameter for all compositions have been calculated for all heating rates and plotted in Fig. 2. It is clear from this figure and from Table 1 that $\text{Se}_{90}\text{Te}_6\text{Sn}_4$ glass is the most stable in the composition range of investigation.

The glass transition activation energy E_t is another indicator of thermal stability. It can be calculated from the variation of T_g with the heating rate β , using the procedures described by Moynihan

Table 1

The temperature difference ($T_c - T_g$) at heating rate of 5 K/min, the average value of crystallization rate factor (K_p) and the reduced glass transition temperature T_{rg} at different heating rates of the studied samples.

Composition	$T_c - T_g$ (K)	$\langle K_p \rangle$ (min) ⁻¹	T_{rg}				
			Heating rate (K/min)				
			5	10	15	20	25
$\text{Se}_{90}\text{Te}_8\text{Sn}_2$	127	5.36×10^{-3}	0.49	0.50	0.51	0.51	0.52
$\text{Se}_{90}\text{Te}_6\text{Sn}_4$	129	3.51×10^{-3}	0.48	0.50	0.50	0.51	0.51
$\text{Se}_{90}\text{Te}_4\text{Sn}_6$	123	5.87×10^{-3}	0.49	0.50	0.51	0.51	0.52
$\text{Se}_{90}\text{Te}_2\text{Sn}_8$	125	6.76×10^{-3}	0.49	0.50	0.50	0.51	0.51

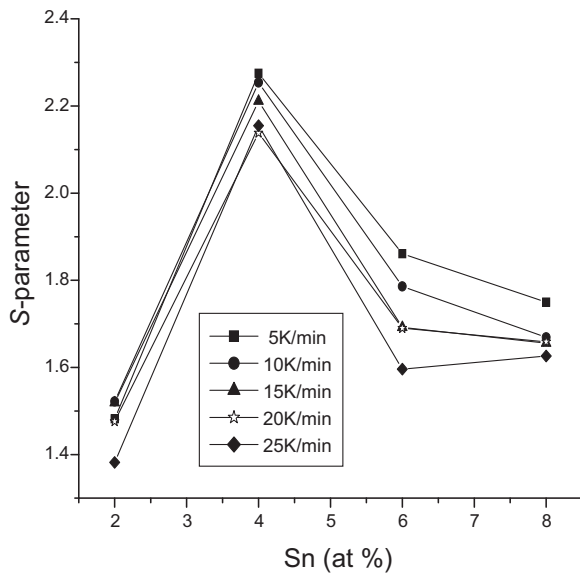


Fig. 2. The dependence of S -parameter on Sn concentration.

et al. [27], through the following equation:

$$\frac{d(\ln \beta)}{d(1/T_g)} = \frac{-E_t}{R} \quad (2)$$

where R is the universal gas constant ($=8.314 \text{ J K}^{-1} \text{ mol}^{-1}$). Fig. 3 shows plots of $\ln \beta$ vs. $1000/T_g$ for all compositions. The values of the activation energy E_t are calculated from the slopes of the resulting straight lines and plotted in Fig. 4 as a function of Sn content. The glass transition activation energy is the amount of energy, which is absorbed by a group of atoms in the glassy region so that a jump from one metastable state to another is possible. This means, this activation energy is involved in the molecular motion and rearrangement of atoms around the glass transition temperature. When the sample is reheated in the DSC furnace, the atoms undergo infrequent transitions between the local potential minima separated by different energy barriers in the configuration space where each local minimum represents a different structure. The most stable local minimum in the glassy region has lower internal

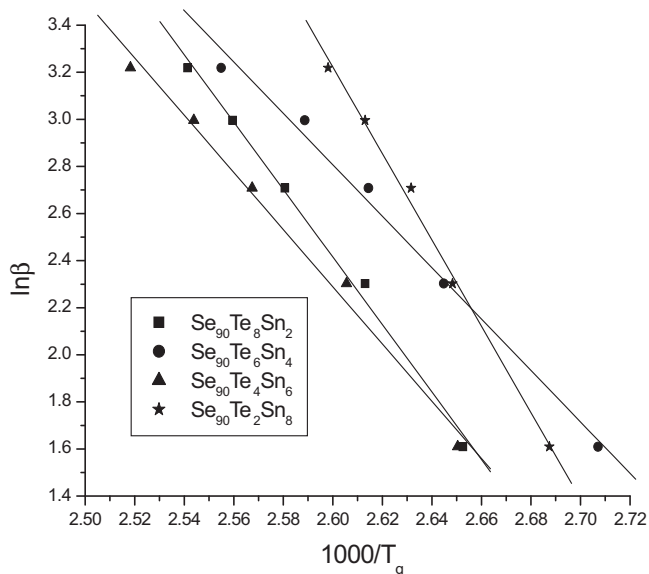


Fig. 3. Plot of $\ln \beta$ vs. $1000/T_g$ for $\text{Se}_{90}\text{Te}_{x-10}\text{Sn}_x$ ($x = 2, 4, 6$ and 8) glasses.

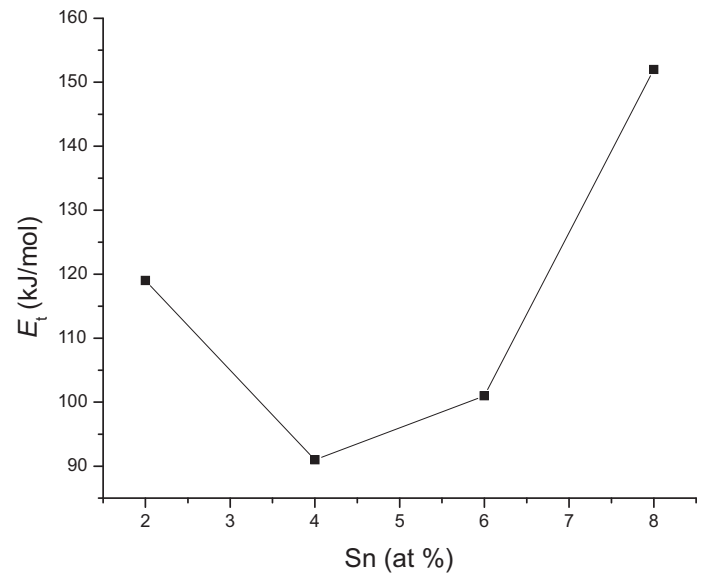


Fig. 4. The dependence of glass transition activation energy (E_t) on Sn concentration.

energy. Accordingly, the atoms in a glass having minimum activation energy have higher probability to jump to the metastable (or local minimum) state of lower internal energy and hence are the most stable [28,29]. Fig. 4 shows that a minimum in E_t occurs at $x = 4$ at.% of Sn, this indicates that this particular glass ($\text{Se}_{90}\text{Te}_6\text{Sn}_4$) has the larger probability to jump to a state of lower configurational energy and hence this glass has the higher stability in the investigated glassy region. This result is in agreement with the above conclusion that drawn from the values of $(T_c - T_g)$ and S -parameter.

For further confirmation of the above glass stability measurements, the crystallization rate factor K_p corresponding to the temperature at which the crystallization rate is maximum, have been calculated from the analysis of DSC curves using the following condition [30]:

$$\frac{\beta E_c}{R K_p T_p^2} = 1 \quad (3)$$

where the symbols carry their usual meanings used throughout the text and E_c is the activation energy of crystallization calculated in accordance with Ozawa equation [31]. The average values of crystallization rate factor (K_p), calculated by averaging the values of K_p found for each exothermic peak at different heating rates, are listed in Table 1 for all compositions. It is reported [32] that the glass with maximum stability will have minimum rate of crystallization. It is obvious that the minimum value of $\langle K_p \rangle$ occurs at 4 at.% of Sn which means that this composition requires longest time to be fully crystallized and hence it has the greatest stability against devitrification.

3.2. Glass-forming ability

GFA is a measure of the ease by which the glass can be formed. The first parameter that introduced to estimate the GFA is the reduced glass transition temperature T_{rg} given as [33]:

$$T_{rg} = \frac{T_g}{T_m} \quad (4)$$

For glass forming systems, T_{rg} has the value in the range $1/2 \leq T_{rg} \leq 2/3$ [34]. Turnbull [35] proposed that for a given composition the glass forming ability should decrease with the reduced glass transition temperature T_{rg} . The calculated values of T_{rg} for the present $\text{Se}_{90}\text{Te}_{10-x}\text{Sn}_x$ ($x = 2, 4, 6$ and 8) glasses at different heat-

Table 2

The values of H_R at different heating rates, the fragility index m and the average coordination number (r) of the studied samples.

Composition	H_R					m	$\langle r \rangle$
	Heating rate (K/min)						
	5	10	15	20	25		
Se ₉₀ Te ₈ Sn ₂	0.48	0.47	0.47	0.46	0.44	23.3	2.08
Se ₉₀ Te ₆ Sn ₄	0.49	0.48	0.47	0.47	0.46	17.9	2.16
Se ₉₀ Te ₄ Sn ₆	0.47	0.45	0.44	0.43	0.42	19.7	2.24
Se ₉₀ Te ₂ Sn ₈	0.47	0.46	0.45	0.44	0.44	30.2	2.32

ing rates are given in Table 1. All these values are closed to 0.5 and lie in the last accepted range. In addition to the present work, several works [12,13,36,37] showed that T_{fg} is almost constant value (mainly = 2/3) for all glassy alloys. Hence it does not reflect any difference in the GFA among different compositions of the studied glasses. Therefore, it is important to look for another parameter of GFA that differentiate between different compositions. Hruby [38] has introduced a parameter H_R given by:

$$H_R = \frac{T_c - T_g}{T_m - T_c} \quad (5)$$

This parameter H_R , in fact, combines the nucleation and growth aspects of phase transformation where higher values of $T_c - T_g$ delay the nucleation process and the small values of $T_m - T_c$ retard the growth process of the nucleated crystals. The H_R values for all heating rates are calculated and listed in Table 2. According to Hruby, glasses with $H_R \leq 0.1$ are difficult to form and require higher cooling rates, whereas glasses with $H_R \geq 0.4$ can be easily formed with moderate quenching rates [39]. It is clear from Table 2 that the maximum value of H_R , for the same heating rate, occurs at 4 at.% of Sn and all values of H_R are greater than 0.4. This further means that Se₉₀Te_{10-x}Sn_x ($x = 2, 4, 6, \text{ and } 8$) alloys are good glass former and the composition with $x = 4$ at.% is the best in the group. The glass transition of chalcogenide glasses, during which the glassy state turns into supercooled liquid state, on heating in the DSC furnace, is a process of diffusion. The more difficult is the atomic diffusion, the more is the difficulty in the nuclei growth. This means the more time the glass transition needs, the higher is the value of H_R and hence the better is the GFA [15].

3.3. Fragility index

Angell [40,41] introduced the concept of fragility, which is defined as the increasing rate of the viscosity of a supercooled liquid at the glass transition temperature during the process of cooling process. According to Angell, glass-forming liquids can be classified into strong and fragile liquids, depending on their fragility. The labels 'strong' and 'fragile' refer to the stability of the intermediate range order in the liquid as the temperature increases from that of the glass transition towards the melting point [38,42,43]. The viscosity for strong glass-forming liquids follows Arrhenius temperature dependence with almost constant apparent activation energy for viscous flow. In contrast, the viscosity for fragile glass-forming liquids shows highly non-Arrhenius temperature dependence and exhibits steep changes in the apparent activation energy for viscous flow from a very low value above the melting temperature to a very high value when approaching the glass transition [44]. The fragility of a given glass can be quantified by the fragility index m which is a measure of the rate at which the relaxation time decreases with increasing temperature around T_g and is given by [45]:

$$m = \frac{E_t}{RT_g \ln 10} \quad (6)$$

where E_t is the glass transition activation energy. This fragility index is usually referred to as the kinetic fragility index. The upper and lower limits of parameter m are theoretically estimated between 16 for 'strong' systems and 200 for 'fragile' systems. The 'strong' glass-forming liquids are characterized by covalent directional bonds that form a spatial network. On the other hand, 'fragile' liquids are composed from molecular units connected by isotropic bonds of Van der Waals type. The fragile structures are more susceptible to the thermal degradation in vicinity of the glass transition. Most of chalcogenide undercooled melts lie in the middle region between these two extremes [46,47]. The fragility index has been obtained for all samples at all heating rates and listed in Table 2 for a heating rate of 5 K/min, as an example. It is clear that these values of m for Se₉₀Te_{10-x}Sn_x ($x = 2, 4, 6, \text{ and } 8$) glassy alloys are near the lower limit ($m \approx 16$) which gives an indication that all the prepared glassy alloys in this study are obtained from strong glass forming liquids.

3.4. Determination of the average coordination number

The concept of average coordination number (r) is useful in describing the crosslinking in a covalently bonded solid. The coordination number of covalently bonded atoms in a glass is given by $8 - N$ rule, where N is the outer shell electrons (the column in the periodic table to which the element belongs). However, when metals atoms are added to a glassy material, this rule is violated and the metal atoms are found to be in higher coordination state [48]. For a multicomponent chain forming chalcogenide glassy system, the average coordination number is defined simply as the atoms averaged covalent coordination of the constituents. For the present glassy system Se_zTe_ySn_x, the average coordination number (r) is calculated using the following equation [49]:

$$\langle r \rangle = 8 - \frac{6z + 6y + 2x}{100} \quad (7)$$

where 6, 6 and 2 are the numbers of valence electrons in Se, Te and Sn, respectively, z , y and x their respective concentration in the glassy composition. Using this equation, the values of (r) for the present Se–Te–Sn glassy samples are obtained and listed in Table 2.

It is well known that [50,51] physical and chemical properties of chalcogenides are strongly depend on composition, so that; they appear to vary according to the average coordination number (r). Compositional-dependent studies on the physical properties of binary and ternary chalcogenide glasses give evidence for the existence of mechanical and chemical thresholds at a certain composition in these materials. According to Phillips and Thorpe rigidity theory [52] it is suggested that many properties exhibit an extremum at (r) = 2.40 at which there is a transformation from underconstrained floppy network to overconstrained rigid network. At this average coordination number it is assumed that the network attains its maximum stability [14]. However, Tanaka [53] suggested that anomalies in physical properties (including T_g) of chalcogenide glasses near (r) = 2.67 may result from a change of network dimensionality from a two-dimensional (layered) structure to a three-dimensional network. When the glass-forming liquids in any Se-rich glassy system are traversed on an average coordination number scale from (r) = 2.40 to (r) = 2.80, three distinct types of liquids can be identified based on Angell's strong and fragile classification: (i) underconstrained liquids (number of degrees of freedom exceed the number of constraints in the system) in which an increase in (r) from 2.00 to 2.40 makes them both thermodynamically and kinetically strong; (ii) overconstrained liquids (the number of constraints exceeds the number of degrees of freedom in the system) in which an increase in (r) from 2.40 to 2.67 makes them thermodynamically fragile but kinetically strong; and (iii) Se-deficient liquids ($r > 2.67$) in which an increase in (r) above 2.67 makes them thermodynamically strong but kinetically fragile [43].

In the present study, the calculated values of $\langle r \rangle$ (Table 2) indicate that the glass of maximum stability and maximum glass forming ability occurs at $\langle r \rangle = 2.16$. The obtained value is less than 2.40 and may be attributed to an important limitation of the topological model. In this model, Phillips and Thorpe have assumed that the interaction between atoms is purely covalent while obtaining the balance condition. This assumption may be valid for glasses showing electronic conduction but is not necessarily valid for glasses containing heavy elements like Sn. It seems that Sn doped chalcogenide glasses show ionic conduction arising from ionic or ionic-covalent behavior of bonds for host materials with Sn [15]. In addition, the values of $\langle r \rangle$ for all compositions are in the range of underconstrained liquids, which means that the present glasses are obtained from thermodynamically and kinetically strong glass forming liquids. This result agrees with the result obtained from the fragility index calculations.

The effect of Sn content on the glass transition activation energy, thermal stability, and glass forming ability of $\text{Se}_{90}\text{Te}_{10-x}\text{Sn}_x$ ($x = 2, 4, 6, \text{ and } 8$) chalcogenide glasses has already been reported from this laboratory [15]. According to this study, it is found that $\text{Se}_{90}\text{In}_4\text{Sn}_6$ glass is thermally most stable with higher glass forming ability and less activation energy for glass transition process. Besides, the above mentioned glassy composition, which occurs at an average coordination number of 2.36, has the lower rate of crystallization. Indeed, application of chemical bond approach to glass transition temperature and crystallization kinetics of the same glassy series as carried out by the group [54] reveal that the crystallization activation energy has its lower value at 6 atomic percentage of Sn. This was attributed to the formation of $\text{SeSn}_{4/2}$ structural units dissolved in Se chains. However, for the present study it is reported [55] that the structure of Se–Te glass consists of Se_8 member rings, Se_3Te mixed rings and Se–Te chains. It is mentioned earlier that the bonds between atoms in the rings are strong covalent, while weak van der Waals forces exist between the chains. Therefore, addition of Sn content to Se–Te glasses leads to the formation of Se–Sn and Te–Sn bonds of energy 174.3 and 161.23 kJ/mol, respectively, calculated as described elsewhere [55]. The bond energy of Te–Sn in $\text{Se}_{90}\text{Te}_{10-x}\text{Sn}_x$ is higher than that of Se–In [56] in $\text{Se}_{90}\text{In}_{10-x}\text{Sn}_x$ series, so the cohesive energy of the sample under investigation is greater than that of Se–In–Sn. This makes the Se–Te–Sn glasses heavily cross-linked which is why T_{gs} for them are higher than those of Se–In–Sn glasses. However, thermal stability indicators $T_c - T_g$, S-parameter and E_f reveal that Se–In–Sn alloys are more stable than Se–Te–Sn. The behavior of GFA parameters is in support of this argument.

4. Conclusions

Based on the obtained values of the characteristic temperatures T_g , T_c , T_p and T_m , thermal stability, GFA and fragility index of $\text{Se}_{90}\text{Te}_x\text{Sn}_{10-x}$ ($x = 2, 4, 6 \text{ and } 8$) glasses were estimated. Results indicate that there is a strong relationship between thermal stability and GFA of the studied glasses. The most stable glass $\text{Se}_{90}\text{Te}_6\text{Sn}_4$ is found to be the best glass former and the prepared glasses were obtained from a strong glass forming liquids, as evident from the fragility index calculations. The calculations of the average coordination number $\langle r \rangle$ of the four compositions reveal that the maximum stability occurs at an average coordination number of $\langle r \rangle = 2.16$ but not that of 2.40 as suggested by Phillips–Thorpe model.

This shift in the value of the threshold coordination number is due to the formation of ionic-covalent bonds when Se rich chalcogenide glasses doped with heavy elements like Sn, while Phillips–Thorpe model takes into consideration the case of glasses with purely covalent bonds.

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