Physical characterization of ternary *In-Se-Te* chalcogenide glasses

A. M. Abousehly[1], N. Afify[2], K. A. Aly[1,3,] and A. M. Abd Elnaeim[1]*

[1]Department of Physics, Faculty of Science, Al-Azahar University, 71542 Assiut, Egypt.
[2]Department of Physics, Faculty of Science, Assuit University, 71515 Assuit, Egypt.
[3]Physics Department, Faculty of Science and Arts, King Abdulaziz University (KAU), Khulais, Saudi Arabia.

Abstract: Different compositions of the $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at.%) chalcogenide system were prepared by the usual melt quench technique. The results of the Differential Scanning Calorimeter (DSC) under on-isothermal conditions were reported in order to study the effect of replacement of In atoms by Se or *Te* atoms on the glass transition, crystallization kinetics and the thermal stability for $In_x(Se_{75}Te_{25})_{100-x}$ glasses. The glass transition temperature was found to increases with the increase in In content. The thermal stability of the studied glasses has been evaluated using various thermal stability criteria (ΔT, Hg and S), based on the characteristic temperatures such as the glass transition temperature (T_g),the temperature at which crystallization begins (T_c), the temperature corresponding to the maximum crystallization rate (T_p), A comparison of various simple quantitative methods to assess the level of stability for $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at.%) glasses is presented. The obtained results were discussed in terms of the glass density, molar volume,compactness and chemical bond approach.

Keywords: Thermal stability; amorphous glasses; semiconductors.

1 Introduction

Glasses of chalcogen elements were the initial object of study because of their interesting semiconducting properties [1,2] that makes it to use for infrared optical elements, infrared optical fibers, and for the transfer of information [3,4]. They have also used in many technological applications, as xerography, switching and memory devices, photolithographic process, and in the fabrication of inexpensive solar cells, and more recently as reversible phase change optical recorders and more recent importance in optical recording [5,6,7,8,9,10,11]. Recording materials must be stable in the amorphous state at low temperature and have a short crystallization time. Promising materials with these characteristics have been recently studied [12,13]. Therefore, it is very important to know the glass stability of these types of materials. Different simple quantitative methods have been suggested in order to evaluate the level of stability of the glassy alloys. Most of them as Dietzel and Hruby [14, 15] are based on the characteristic temperatures (the glass transition temperature (T_g), the temperature at which

crystallization begins (T_c), the temperature corresponding to the maximum crystallization rate (T_p), and the melting temperature (T_m). Some of the suggested methods [16, 17] are based on the crystallization activation energy. The characteristic temperatures (T_g, T_c and T_p) are easily and accurately obtained by the differential scanning calorimetry [18] during the heating processes of the glass sample. Dietzel [14] introduced the first glass criterion, $\Delta T = T_c$-T_g (T_c is the temperature at which crystallization begins), which is often an important parameter to evaluate the glass forming ability of the glasses. Saad and Poulain [19] obtained two other criteria, weighted thermal stability $H_g = \Delta T/T_g$ and $S=(T_p$-$T_0)$ $\Delta T/T_g$ criterion where T_o is the initial temperature. In the present work, the above-mentioned criteria have been applied to the $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at.%) glasses. It is found that, the parameters ΔT, Hg and S decrease with increasing the In content. Bearing in mind that, the values of these parameters increase with increasing stability, it is possible to suggest that, the free *Te* content glass, the greater thermal stability. In addition, the glass transition temperature were found to increase with the increase of In

* Corresponding author e-mail: kamalaly2001@gmail.com

content this behavior were discussed in terms of the glass density, compactness, molar volume, fragility and the chemical pond approach.

2 Theoretical background

The theoretical basis for interpreting kinetic data is provided by the formal theory of transformation kinetics. This theory describes the evolution with time (t) of the volume fraction crystallized (x) by Johnson, *Mehl* and Avrami equation [20]:

$$x = 1 - exp(-(Kt)^n) \qquad (1)$$

where n is an integer or half integer depends on the mechanism of growth and the dimensionality of the crystal, K is the effective (overall) reaction rate constant, which obeys an Arrhenius expression for the absolute temperature:

$$K(T) = K_0 exp\frac{-E}{RT} \qquad (2)$$

where K_0 is the frequency factor, T is the absolute temperature and E is the effective activation energy describing the overall crystallization process. The rate constant K in a non-isothermal *DSC* experiment was found to changes continually with time due to the change in the temperature, therefore $Eq.1$ can be generalized to:

$$x(t) = 1 - exp[-(\int_0^t K[T(t')]dt')^n] = 1 - exp(-I^n) \qquad (3)$$

where $K[T(t')]$ is still given by $Eq.2$, and $T(t')$ is the temperature at time t' [21]. The crystallized volume fraction depends on time (t) through the temperature, $T(t)$, and the same is true for the integral I. The time integral in $Eq.3$ is transformed to temperature integral, yielding:

$$I(T) = \frac{K_0}{\alpha} \int_{T_0}^{T} exp(\frac{-E}{RT'})dT' \qquad (4)$$

which represented by several approximate analytical expressions [22]. By using the substitution $y' = E/RT'$, the above integral has been represented by the sum of the alternating series:

$$S(y') = -\frac{e^{-y'}}{y'^2} \sum_{k=0}^{k=\infty} \frac{(-1)^k(k+1)!`}{y'^k} \qquad (5)$$

Considering that, in this type of series the error produced is less than the first term neglected and bearing in mind that in most crystallization reactions $y' = E/RT' >> 1$. Therefore, it is possible to use only the two first terms of this series and the error introduced is not greater than 1%. By assuming that, $T^2(1-2RT/E)exp(-E/RT) >>$

$T_0^2(1 - 2RT_0/E)exp(-E/RT_0)$, where T_o is initial temperature, then equation 4 can be rewritten in the form of:

$$I = K_0 E(\alpha R)^{-1} e^{-y} y^{-2}(1 - 2y^{-1}) \qquad (6)$$

The maximum crystallization rate in a non-isothermal process which occurs at the peak of the exotherm at time t_p and temperature T_p [20] is found by putting d^2x/dt^2, thus obtaining the relationship:

$$d^2x/dt^2 = nK_p(I)_p - (n-1)K_p - \frac{\alpha E(I)_p}{RT_p^2} = 0 \qquad (7)$$

$nK_p(I)_p = (n-1)K_p - \frac{\alpha E(I)_p}{RT_p^2}$

Substituting for $(y = E/RT) and (K = K_0 exp(-E/RT))$ into Eq. 6, one obtains :

$$I = RT^2 K(\alpha E)^{-1}(1 - 2RT/E) \qquad (8)$$

Substituting the last expression for I into $Eq.79$, one obtains the relationship:

$$I_p = (1 - 2RT_p/nE)^{1/n} \qquad (9)$$

When this relationship is equated to $Eq.$ 8 this gives: $RT_p^2 K(\alpha E)^{-1}K_0(-E/RT_p) = (1 - 2RT_p/nE)^{1/n}$ $\times (1 - 2RT_p/E)^{-1}$ or in a logarithmic form

$$ln(T_p^2/\alpha) + ln(K_0 R/E) - E/RT_p \approx (2RT_p/E)(1 - 1/n^2) \qquad (10)$$

where the function $ln(1 - z)$ with $z = 2RT_p/nE$ or $z = 2RT_p/E$ is expanded as a series and only the first term has been taken. Note that $Eq.10$ reduces to the Kissinger expression for the $n = 1$ case as one might have anticipated since this corresponds to the homogeneous reaction case. Thus, it can be seen that, the Kissinger method is appropriate for the analysis not only of homogeneous reactions, but also for the analysis of heterogeneous reactions which are described by the *JMA* equation in isothermal experiments [20]. The right-hand side (RHS) of $Eq.10$ is generally negligible in comparison to the individual terms on the left hand side for $\alpha \leq 100 K min^{-1}$. This approximation in $Eq.10(RHS)$ implies:

$$ln(T_p^2/\alpha) = E_c/RT_p - ln(k_0 R/E) \qquad (11)$$

where $E_c = E$ (the activation energy for crystallization) and the quoted approximation might introduce a 3% error in the value of E_c/R in the worst cases.

3 Experimental details

Different compositions of bulk $In_8 Se_{92-x} Te_x$ (x= 0, 5, 10, 15, 20 and 25 at. %) chalcogenide glasses were prepared

starting by *In*, *Se* and *Te* elements with high purity (99.999%) by the usual melt quench technique.

The elements were heated together in an evacuated (10^{-3} *Pa*) silica ampoule up to 1250 *K*, then the ampoule temperature kept constant for about 24 h. During the heating process the ampoules were shaken several times to maintain their homogeneity, then the ampoule was quenched in ice cooled water to avoid the crystallization process. The amorphous state of the materials was checked using x-ray (Philips type 1710 with *Cu* as a target and *Ni* as a filter, $\lambda = 1.5418$ *A*) diffractometer. The absence of the crystalline peaks confirms the amorphous state of the prepared samples. The elemental compositions of the investigated specimens were checked using the energy dispersive $x-ray$ (Link Analytical *Edx*) spectroscopy.

The compositions so determined agreed with those of the starting materials. The thermal behavior was investigated using calibrated *Shimadzu* 50 differential scanning calorimeter. About 15 *mg* of each sample in powdered form was sealed in standard aluminum pan and scanned over a temperature range from room temperature to about 770*K* at different uniform heating rates ($\alpha = 2.5$, 5, 10, 15, 20 and 30 *K/min*.).

Density measurements of the considered samples were made by applying Archimedes method using the hydrostatic weighting in toluene. A single crystal of germanium was used as a reference material for determining the toluene density, ρtol. The samples density (ρs) was determined from the relation;

$$\rho_s = \frac{W_{air}}{W_{air} - W_{tol}} \rho_{tol} \tag{12}$$

where W is the weight of the sample. For each composition, the experiment was repeated five times to get the average density of the sample (ρ_s) with a precision of .3%. Knowing the density helps us to determine the molar volume, V_m, through the following equation:

$$V_m = \sum n_t . M_t / \rho_s \tag{13}$$

where n_i and M_i are the molar fraction and molar weight of a component i, respectively. The compactness d was calculated by the formula [23, **?**, 25]

$$\delta = \frac{\sum_i c_i . A_i / \rho_i - \sum_i c_i . A_i / \rho}{\sum_i c_i . A_i / \rho} \tag{14}$$

where c_i, A_i and ρ^i are the atomic fraction, the atomic weight and the atomic density of the *ith* element of the glass and is the measured density of the glass. Thus, δ is a measure of the normalized change of the mean atomic volume due to chemical interactions of the elements forming the network of a given solid [26, 27]. Consequently, it is more sensitive to changes in the structure of the glass network as compared to the mean atomic volume.

4 Results and discussion

Fig. 1 shows the $x-ray$ diffraction patterns for the $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses. The absence of the diffraction lines in the $x-ray$ patterns indicates that the glasses have amorphous structures. *Fig.* 2(*a*) shows the *DSC* thermograms for the amorphous the $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses recorded at heating rate 10 K/min. As shown in this figure, there is a very small single endothermic peak. This peak is attributed to the glass transition temperature range as shown in *Fig.* 2(*b*) which represents the strength or rigidity of the glass structure. Also there is an exothermic peak originating from the amorphous-crystalline transformation.

The exothermal -peak has two characteristic points: the first is the onset temperature of crystallization (T_c) and the second is the temperature corresponding to the maximum crystallization rate (T_p). As shown in this figure the characteristic temperatures are found to affect by the addition of Indium content. *Fig.* 3 represents the density,ρ_s, and the molar volume, V_m, as a function of In content. It's well known that the density and molar volume changes are related to the change in the atomic weight and the atomic volume of the elements constituting the system. The atomic weights of the *In*, *Se* and *Te* are 114.82, 78.7 and 127.6 respectively [28].

This behavior was expected because the density of In is the highest one (see *Table* 1). *Fig.* 4 shows the glass transition temperature, T_g, and the molar volume V_m, as a function of In content, from this figure we can notice that, the glass transition temperature increases while the V_m decreases with increasing the In content. This behavior can be ascribed to where the molar volume decrease leads to the decrease in bond lengths consequentially the glass rigidity increase and therefore the T_g increases. In other words the glass transition temperature is known to depend on several independent parameters such as the average coordination number [29,30]. The average coordination number (Nr) for the $In_x(Se_{75}Te_{25})_{100-x}$ glasses can be written as [31]:

$$N_r = 3X_{In} + 2X_{Se} + 2X_{Te} \tag{15}$$

where X is the mole fraction, by using the values of Nr for *In*, *Se*, and *Te* as 3, 2 and 2 respectively [32,33], the values of N_r for the $In_x(Se_{75}Te_{25})_{100-x}$ glasses are obtained. Values of N_r for the $In_x(Se_{75}Te_{25})_{100-x}$ glasses are listed in *Table* 2. It can be seen that N_r increases with increasing In content. The increase in the glass transition temperature which is accompanied by an increase in the coordination number can be ascribed to the increase of the rigidity (strength) of the system with increasing the In content.

The above results can also be discussed on the basis of a parameter called fragility (F), which characterizes and quantifies the anomalous non-Arrhenius transport

Table 1: The characteristics temperatures (T_g, T_c, and T_p), the activation energy of crystallization (E_c), the frequency factor (K_o) and the ΔT, Hg, S, criteria for $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10 at.\%$) glasses.

Composition	α (K/min)	T_g (K)	T_p (K)	E_c Kcal/mol	K_0 (s^{-1})	ΔT (K)	H_g	S (K)
$(Se_{0.75}Te_{0.25})_{100}$	2.5	304.70	365.58			54.24	0.178	1.182
	5	308.49	374.31			58.41	0.189	1.403
	10	312.35	384.38	20.10	3.051×10^{10}	65.25	0.209	1.416
	20	317.12	394.05			69.91	0.221	1.548
	40	321.98	402.60			73.59	0.229	1.607
$In_2(Se_{0.75}Te_{0.25})_{98}$	2.5	311.06	371.84			51.79	0.167	1.497
	5	314.51	380.54			55.71	0.177	1.828
	10	318.50	388.86	23.52	2.156×10^{11}	60.72	0.191	1.838
	2.5	322.28	397.63			66.36	0.206	1.851
	5	326.02	404.06			69.78	0.214	1.884
$In_4(Se_{0.75}Te_{0.25})_{96}$	2.5	315.05	375.38			49.38	0.157	1.716
	5	318.65	382.55			53.02	0.166	1.810
	10	322.13	390.31	25.12	1.498×10^{12}	57.71	0.179	1.878
	2.5	325.44	398.88			63.22	0.194	1.985
	5	329.06	406.68			67.42	0.205	2.090
$In_6(Se_{0.75}Te_{0.25})_{94}$	2.5	321.91	378.90			46.68	0.145	1.495
	5	325.04	385.48			50.64	0.156	1.527
	10	327.80	393.08	26.93	1.192×10^{13}	54.91	0.168	1.737
	20	331.19	400.91			59.22	0.179	1.878
	40	335.09	408.64			62.96	0.188	1.990
$In_8(Se_{0.75}Te_{0.25})_{92}$	2.5	324.61	381.05			45.06	0.139	1.580
	5	327.73	388.42			48.67	0.149	1.785
	10	330.98	395.29	27.85	3.518×10^{13}	53.08	0.160	1.801
	20	334.00	402.63			57.47	0.172	1.920
	40	337.09	410.61			61.65	0.183	2.171
$In_{10}(Se_{0.75}Te_{0.25})_{90}$	2.5	326.53	384.15			44.58	0.137	1.780
	5	329.39	390.43			47.90	0.145	1.911
	10	332.91	397.85	29.07	1.371×10^{14}	51.81	0.156	2.043
	20	336.77	404.71			55.29	0.164	2.077
	40	341.14	412.64			59.00	0.173	2.162

Table 2: The compactness (δ), the fragility (F), average coordination number (N_r), The number of expected *Se-Te*, *Se-Se* and *In-Se* bonds and the calculated cohesive energy for $In_x(Se_{0.75}Te_{0.25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses according to the chemical bond approach

Composition	δ	E_g (kj mol^{-1})	F	N_r	Se-Te	In-Se	Se-Se	CE(kcal. Mol^{-1})
$(Se_{0.75}Te_{0.25})_{100}$	-0.134	128.17	22.986	2	50	0	50	44.12
$In_2(Se_{0.75}Te_{0.25})_{98}$	-0.096	152.61	26.841	2.02	49	6	46	45.17
$In_4(Se_{0.75}Te_{0.25})_{96}$	-0.066	169.18	29.417	2.04	48	12	42	46.23
$In_6(Se_{0.75}Te_{0.25})_{94}$	-0.037	188.19	32.158	2.06	47	18	38	47.29
$In_8(Se_{0.75}Te_{0.25})_{92}$	-0.016	199.45	33.755	2.08	460	24	34	48.34
$In_{10}(Se_{0.75}Te_{0.25})_{90}$	0.004754	210.12	35.355	2.1	45	30	30	49.4

behavior of glassy materials near the erogidicity breaking glass transition region [34,35,36]. Fragile glasses are substances with non-directional *interatomic/intermolecular* bonds. Strong glasses are those which show resistance to structural degradation and usually associated with a small C_p. Fragility (F) is calculated by using the following relation [37]:

$$F = \frac{E_g}{T_g.R.ln(1.009)} \quad (16)$$

where all the symbols have their usual meaning [28]. The value of (F) is found to increase with N_r as stated in *Table* 2. This behavior indicates that the glasses become more fragile and their tendency to structural rearrangement increases with increasing non-directional interatomic bonds. The bond energies $D(A - B)$ for heteronuclear bonds have been calculated by using the empirical relation:

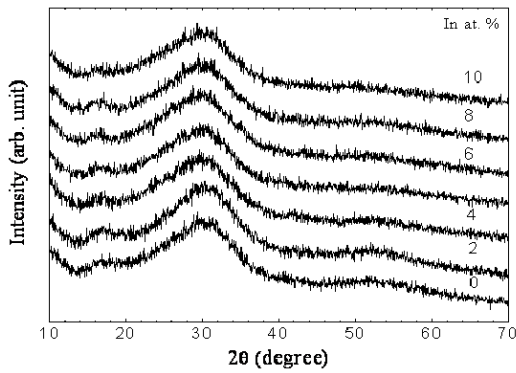$$D(A-B) = [D(A-A).D(B-B)]^{1/2} + 30(x_A - x_B)^2 \quad (17)$$

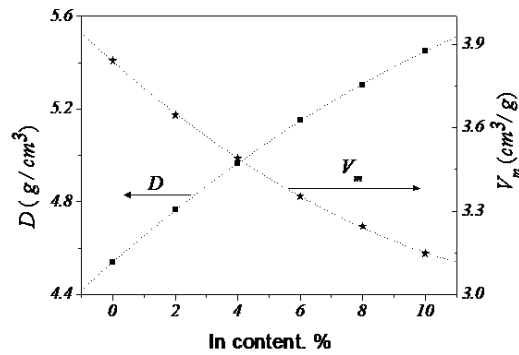**Figure 1:** X-ray diffraction patterns of the $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses
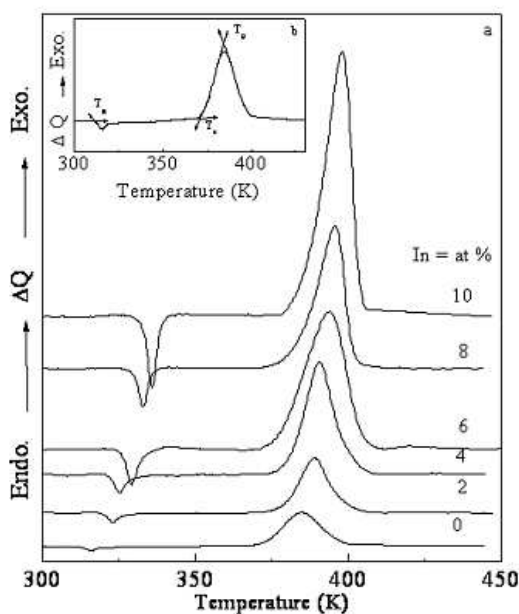

Figure 2: (a)The *DSC* thermograms for the amorphous the $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses recorded at heating rate 10 *K/min*. (b) The characteristic temperatures of the *DSC* thermogram for $Se_{75}Te_{25}$ glass recorded at heating rate $10K/min$.

Figure 3: The density and the molar volume as a function of In content for $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses.

Figure 4: The plots of $\ln(T_p^2/\alpha)$ versus $In_x(Se_{75}Te_{25})_{100-x}$ ($0 \leq x \leq 10$ at. %) glasses.

proposed by Pauling [38], where *[D(A-A)* and *[D(B-B)* are the energies of the homonuclear bonds ($in units kcal/mol.$) [39], x_A and x_B are the electronegativity values for the involved atoms [40]. Bonds are formed in the sequence of decreasing bond energy until the available valence of atoms is satisfied [41].

In the present compositions, the *In-Se* bonds with the highest possible energy ($54.321 kcal mol^{-1}$) are expected to occur first followed by the *Te-Se* ($44.197 kcal mol^{-1}$) to saturate all available valence of *Se*. There are still
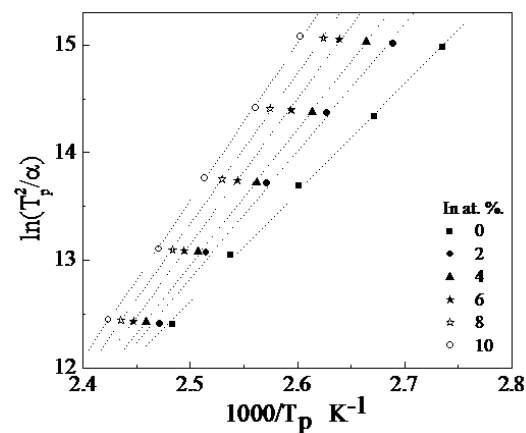
unsatisfied as which must be satisfied by *Se-Se* defect homopolar bonds that decreases with the increase of In content. Based on the chemical bond approach, the bond energies are assumed to be additive. Thus, the cohesive energies were estimated by summing the bond energies over all the bonds expected in the material. Calculated values of the cohesive energies for all compositions are presented in *Table* 2. These results indicate that, the cohesive energies of these glasses show an increase with increasing *In* content. Therefore, it can be concluded that the increase of T_g with increasing In content is most probably due to the replacement of *Se-Te* and *Se-Se* bonds by the strongest In-Se bonds. It should be mentioned that the approach of the chemical bond neglects dangling bond and other valence defects as a first approximation.

Also van *der* Walls interactions are neglected, which can provide a means for further stabilization by the formation of much weaker links than regular covalent bonds. The linear relation of versus

$1/T_p In_x(Se_{75}Te_{25})_{100-x}$ glasses are plotted in *Fig*.4 to obtain the activation energy of crystallization (E_c) and the frequency factor K_0. The obtained values of the activation energy of crystallization and the frequency factor are listed in *Table* 2. It is found that E_c increases with increasing In content, this increase is a result of the increase of T_p with increasing In content. The thermal stability for the $In_x(Se_{75}Te_{25})_{100-x}$ glasses can be estimated by using the characteristic temperatures (T_g, T_c and T_p).

The stability criterion parameters (ΔT, Hg and S) based on the characteristic temperatures are calculated for the $In_x(Se_{75}Te_{25})_{100-x}$ glasses and listed in *Table* 1. The stability criterion parameters allow the prediction of the glass forming ability of a material. The larger their values, the greater the glass thermal stability should be. *Table* 1 show that (ΔT, Hg and S) decrease with the increase of In content, so we can say that the $Se_{75}Te_{25}$ glass is the most stable one.

5 Conclusions

The addition of *In* at the expense of *Te* or *Se* atoms in $In_x(Se_{75}Te_{25})_{100-x}$ glasses results in an apparent increase in the characteristic temperatures (T_g, T_c and T_p), the activation energy for glass transition and the activation energy for crystallization. The thermal stability for the $In_x(Se_{75}Te_{25})_{100-x}$ glasses has been evaluated by using various criteria. The obtained results of the ΔT, Hg and S criteria indicates that, the thermal stability of $In_x(Se_{75}Te_{25})_{100-x}$ glasses decreases with the increase of In content. The obtained results were discussed in terms of the glass density, molar volume, compactness, fragility and the chemical bond approach.

Acknowledgement

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