ESTIMATION OF GLASS-FORMING ABILITY AND GLASS STABILITY OF 
Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ GLASSES BY USING THE THERMAL PROPERTIES

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Glass forming ability parameters defined by $T_{rg} = T_g/T_{f1}$ and $K_h = (T_{c1}-T_g)/(T_{f1}-T_{c1})$, glass stability parameters such as $K_w = (T_{c1}-T_g)/T_{f1}$, $\Delta T = T_{c1}-T_g$ and $K_l = T_{c1}/(T_g+T_{f1})$ and the degree of undercooling $\Delta T_r = (T_{f1}-T_{c1})/T_{f1}$ are analysed for glasses of the ternary system Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ as function of the As$_2$Se$_3$ concentration. These parameters are formulated by different combinations of the following characteristic Differential Scanning Calorimetry (DSC) temperatures: the glass transition temperature ($T_g$), the first crystallization temperature ($T_{c1}$) and the first melting temperature ($T_{f1}$). Variations of the above parameters indicate that the studied glasses can vitrify easily and become increasingly stable when the concentration of As$_2$Se$_3$ increases. Good correlations between $\Delta T_r$ and $K_h$ and between $\Delta T_r$ and parameters ($K_w$, $\Delta T$ and $K_l$), are found implying a low frequency of homogeneous nucleation in the thermally stable glasses. The degree of undercooling $\Delta T_r$ is an important parameter for the glass forming ability and the glass stability of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ glasses.

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Keywords: Glass transition temperature, First crystallization temperature, First melting temperature, Glass forming ability, Glass stability, Degree of undercooling

1. Introduction

The ability of substances to vitrify on cooling from the melt is known as glass-forming ability (GFA). Glass formation of materials containing one or more elements sulphur (S), selenium (Se) or tellurium (Te) in combination with elements from IVth and Vth group of the periodic table is relatively easy. Many kinds of these materials have been prepared by means of melt quenching method (Saffarini and Saiter, 2006; Repkova et al, 2006; Singh et al, 2006; Soliman and El-Den, 2007; El-Mokhtar, 2007)[1-5]. Several parameters or criteria have been proposed to reflect the relative GFA among bulk glasses on the basis of different calculation methods (Lu and Liu, 2002, 2003)[6, 7]. One is the reduced glass transition temperature $T_{rg} = T_g/T_{f1}$ which is the ratio between the glass transition temperature $T_g$ and the first melting temperature $T_{f1}$ of the corresponding glass-forming system (Kumar et al, 2006; Chol-Lyong et al, 2006)[8,9]. Another parameter $K_h = (T_{c1}-T_g)/(T_{f1}-T_{c1})$, where $T_{c1}$ is the first crystallization temperature, is also used as a measure of the glass-forming tendency of materials by (Farid, 2002)[10], (Aljihmani et al, 2003) [11] and (Nikhil Sur et al, 2006) [12].

Once a glass is made for instance by fast quenching a melt, its stability can be easily investigated. Thus the supercooled liquid range $\Delta T = T_{c1}-T_g$ (Kumar et al, 2006) [8], the parameter $K_w = (T_{c1}-T_g)/T_{f1}$ (Avramov et al, 2003; Nasciemento et al, 2005) [13, 14] and a new criterion $K_l = T_{c1}/(T_g+T_{f1})$ of (Lu and Liu 2002, 2003) [6, 7] are used to evaluate the glass stability against crystallization on heating.

The thermal stability of the metastable supercooled liquid obtained at temperatures between $T_g$ and $T_{f1}$ or liquids temperature $T_l$ can be discussed from the kinetics aspect because it would be informative in supercooled liquids, the frequency of homogeneous nucleation depends

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on the degree of undercooling $\Delta T_r = (T_f - T_c)/T_f$ as predicted by the classical theory of homogeneous nucleation.

The aim of this study is to calculate the values and interpret the variations of the glass-forming parameters ($T_{rg}$ and $K_h$), the glass stability parameters ($\Delta T$, $K_s$, and $K_l$) of the ternary system $Sb_2Se_3-As_2Se_3-Sb_2Te_3$ as function of $As_2Se_3$ concentration. Correlation between these parameters will be made in order to show the importance of the degree of undercooling for glass forming ability and glass stability and a possible relationship between glass forming ability and glass stability for glasses of the above system will be verified.

2. Materials and method

Glasses of $Sb_2Se_3-As_2Se_3-Sb_2Te_3$ system were prepared by our vitreous semiconductors group in the Laboratoire de Chimie des Matériaux Inorganiques by union of arsenic, selenium, tellurium and antimony of guaranteed purity (99.999%), introduced in stoechiometric proportions into sealed vacuum silica bulbs ($10^{-3}$ Torr). Those are carried to 130°C during 24 hours, then to 900°C (at a speed of 3°C/mn approximately) and are maintained at this temperature during 24 hours. They, are finally soaked brutally in water-ice mixture. The x-rays diffractograms on powders made it possible to establish the vitreous state of the samples when they do not present any line but the very large waving characteristic to the vitreous state.

The thermal characteristic temperature such as the glass transition ($T_g$), the first crystallization temperature ($T_c$) and the first melting temperature ($T_f$) were measured in the Laboratoire de Chimie Physique, Minérale et Bioinorganique de la Faculté de Pharmacie de l’Université Paris XI (France) by using DSC 121 Setaram apparatus at a heating rate of 1°C mn$^{-1}$ in the (25 to 650°C) temperature range. For the studied glasses, glass-forming ability was estimated using the following numerical parameters: the reduced glass transition temperature $T_{rg} = T_g/T_f$ and $K_h = (T_c - T_g)/(T_f - T_c)$ parameters. The glass stability parameters were also estimated by $K_s = (T_c - T_g)/T_f$, $K_l = T_c/(T_g + T_f)$ and $\Delta T = T_c - T_g$. The degree of undercooling was evaluated by $\Delta T_r = (T_f - T_c)/T_f$ (Komatsu et al, 1997) [15]. In this study we calculated the values of glass-forming ability and glass stability parameters given by the above expressions.

3. Results and discussion

3.1 Glass-forming ability parameters

Using the data shown in Table 1, the reduced glass transition temperature ($T_{rg}$) values for glasses of $Sb_2Se_3-As_2Se_3$ system vary little from 0.5 (50 mol% $As_2Se_3$) to 0.47 (100 mol% $As_2Se_3$). On the ternary system $Sb_2Se_3-As_2Se_3-Sb_2Te_3$ with a constant concentration of 20 mol% $Sb_2Te_3$, the evolution of $T_{rg}$ is not linear. $T_{rg}$ has an optimal value equal to 0.53 (50 mol% $As_2Se_3$) and decreased values are observed when the $As_2Se_3$ concentration increases up 50 mol% (Table 1).

The parameter $K_s(T_c)$ calculated with the onset temperature $T_c$ (Table 1), increases from 0.17 to 1.79 on $Sb_2Se_3-As_2Se_3$ system (fig 1).

For $Sb_2Se_3-As_2Se_3-Sb_2Te_3$ glasses containing a constant $Sb_2Te_3$ concentration of 20 mol%, $K_s(T_c)$ parameter increases from 0.20 to 2.33 as shown in Fig. 2.
Table 1 Composition of glasses, thermal properties ($T_g$, $T_{c1}$ and $T_{f1}$) from DSC curves, values of glass-forming ability parameters ($T_{rg}$ and $K_h$), values of glass forming stability parameters ($K_w$, $K_l$ and $\Delta T$) and degree of undercooling $\Delta T_r$ for studied glass in the binary system Sb$_2$Se$_3$-As$_2$Se$_3$.

<table>
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<tr>
<th>Composition of glass to As$_2$Se$_3$</th>
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<th>$T_{c1}$</th>
<th>$T_{f1}$</th>
<th>$T_{rg}$</th>
<th>$K_h(T_{c1})$</th>
<th>$K_w(T_{c1})$</th>
<th>$K_l(T_{c1})$</th>
<th>$\Delta T(T_{c1})$</th>
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Fig. 1: Variation of $K_h(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$ binary system.

Fig. 2: Variation of $K_h(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 20 mol%
Variations in $T_{rg}$ and $K_b$ parameters are largely due to the variations in the thermal properties of glasses. These parameters seem to depend on compositions. Even if $T_{rg}$ does not give the width of the temperature interval it also determines how close to the liquidus temperature the decreasing mobility in the liquid starts to reduce the nucleation rate. $T_{rg}$ plays a crucial role in determining the glass-forming ability of an alloy because the higher is the ratio, the higher is the Glass-Forming Ability (GFA) according (Uhlmann, 1977) [16] and (Davies, 1975) [17]. It has been confirmed that $T_{rg} = 2/3$, the two thirds rule, holds well generally for wide variety of inorganic glass forming substances (Sakka and Mackenzie, 1971) [18]. Thus for the liquids having $T_{rg}$ equal to or more raised than $2/3$, the formation of glasses would be easy because it leads to low nucleation rates. As pointed out by (Tumbull, 1969) [19], these liquids are good glass formers. It is clearly shown that the two thirds rule holds well for the studied glasses of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ system.

**Fig. 3.** Variation of $K_w(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$ binary system.

**Fig. 4.** Variation of $K_l(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$ binary system.
The increase of $K_h(T_{c1})$ and $T_{rg}$ (except of $T_{rg}$ of 20% Sb$_2$Te$_3$) with increasing As$_2$Se$_3$ (covalent compound) incorporation supports the Glass-Forming Ability (GFA). In other words, GFA increases with increase in As$_2$Se$_3$ concentration. For these glasses $K_h(T_{c1})$ and $T_{rg}$ reflect the GFA effectively. GFA in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ system is conditioned by the presence of a glass forming compound As$_2$Se$_3$. This indicates that As$_2$Se$_3$ glass is the best glass forming system among the vitreous samples of the above system. There is in this case an obvious correlation between covalence and glass-forming ability. It is possible to suggest that the presence of covalent bondings gives flexibility (elasticity) to the structure that is a necessary factor for the topological disordering of the structure during glass formation. The enough large flexibility that permits to the elementary components (atoms, cations, coordination polyhedra) to occupy different positions one relative to another, which fact does not create long range order and does not lead to the simultaneous appearance of strains, that destroy the structure, gives the ability to oxide and chalcogenide system to form glasses. $T_{rg}$ evolution of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ glasses containing 20 mol% Sb$_2$Te$_3$ is not linear like $K_h(T_{c1})$ and $T_{rg}$ seen above. For these glasses, $T_{rg}$ cannot reflect the GFA. This behaviour was found in many bulk metallic glasses (Lu and Liu, 2002) [16] and phosphate glass systems (Ouchetto et al, 1991) [20].

**Fig. 5.** Variation of $K_h(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 20 mol%.

**Fig. 6.** Variation of $K_l(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 20 mol%.
3.2 Glass stability parameters

In this study, values of the supercooled liquid region $\Delta T(T_{c1})$ parameter and Lu-Liu parameter $K_l(T_{c1})$ used for stability assessment of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ glasses, Table 1 and 2. Generally, the difference $\Delta T(T_{c1}) = T_{c1} - T_g$, gives a measure of thermal stability of the glass (Kamboj and Thangaraj, 2003) [21]. For the present study, its values are found to be in the range 27-127°C on the binary system Sb$_2$Se$_3$-As$_2$Se$_3$ and 24-105°C on the ternary system Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ (containing 20 mol% Sb$_2$Te$_3$). Values of $K_w(T_{c1})$ parameter and Lu-Liu parameter $K_l(T_{c1})$ increase when As$_2$Se$_3$ concentration increase on the binary system Sb$_2$Se$_3$-As$_2$Se$_3$ (fig 3) and on the ternary system Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ with 20 mol% Sb$_2$Te$_3$ (fig 4). Even if $K_w$ and $K_l$ obtained at $T_{c1}$ don’t give the width of the temperature interval like $\Delta T(T_{c1})$, they can be suggested to represent the glass stability because they have similar trends when they are plotted versus As$_2$Se$_3$ concentration (fig 3-6). It is obvious that the thermally stable glasses in the above systems are obtained when the As$_2$Se$_3$ concentration increase. In other words, the thermal stability of these glasses against crystallization increases with increase in As$_2$Se$_3$ concentration. As can be shown in tables 1 and 2 the increases in $\Delta T(T_{c1})$ in the two systems mainly result from the increase of $T_{c1}$. This implies that the added As$_2$Se$_3$ acts effectively as an inhibitor of crystallization. The onset-crystallization $T_{c1}$ can serve as an important factor estimating the stability of glass (Hen et al, 1991) [22]. On the other hand, it is reasonable to assign that As$_2$Se$_3$ a covalent compound, acts as network breaking agents to decrease $T_g$ because it decrease with increasing As$_2$Se$_3$ concentration as shown in Table 1.

![Fig. 7. Evolution of the onset crystallization temperature $T_{c1}$, the first melting temperature $T_f$, and undercooled liquid region $T_f-T_{c1}$ versus the degree of undercooling $\Delta T(T_{c1})$ on the Sb$_2$Se$_3$-As$_2$Se$_3$ binary system](image-url)
Fig. 8. Evolution of the onset crystallization temperature $T_{c1}$, the first melting temperature $T_{f1}$ and undercooled liquid region $T_{f1}-T_{c1}$ versus the degree of undercooling $\Delta T_{r}(T_{c1})$ on the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 20 mol%.

Fig. 9. Variation of $\Delta T_r(T_{c1})$ with the As$_2$Se$_3$ concentration in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 0 and 20 mol%.
Increased values of $\Delta T(Tc_1)$ may indicate that the supercooled liquid can remain stable in a wide temperature range without crystallization. Thus, As$_2$Se$_3$ incorporation has a stabilizing effect because $\Delta T(Tc_1)$ becomes wide when its concentration increases. It is desirable to have $\Delta T$ as large as possible in order to achieve a large working range during operations such as perform preparation for fibre drawing according (Feng et al, 1999) [23]. The partial replacement of Sb$_2$Se$_3$ (less covalent) by As$_2$Se$_3$ (covalent), on Sb$_2$Se$_3$-As$_2$Se$_3$ and Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ (with 20 mol% Sb$_2$Te$_3$) systems, can exhibit a predominantly character which increases the resistance of the glass to devitrification. Thus, As$_2$Se$_3$ involves probably the formation of high stable network structure which may be due to the presence of covalent bondings. That means that As$_2$Se$_3$ acts to increase the homogeneity of glass and strengthening the glass network. In the same time it acts to increase the covalence character by forming stable units which ensure effectively the stability of the glasses. According to a previous study (El-Idrissi Raghni et al, 1995) [24], The Raman and IR bands of Sb$_2$Se$_3$-As$_2$Se$_3$ glasses are attributed to AsSe$_3$ and SbSe$_3$, the trigonal pyramidal units in which As and Sb obey the 8-N rule (N is the number of electrons needed to complete its valence shell). Results of $^{121}$Sb Mössbauer spectroscopy of 20 and 40 mol% Tl$_2$S sections of Sb$_2$S$_3$-As$_2$S$_3$-Tl$_2$S glasses (Durand et al, 1997) [25] and those of 10 mol% Sb$_2$Te$_3$ section of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ glasses (Leh Deli et al, 2005) [26], indicate that the isomer shifts are negative in all vitreous compounds studied by the above researchers. These isomer shifts are found to be between -5.06 and -4.56 mm sec$^{-1}$ for Sb$_2$S$_3$-As$_2$S$_3$-Tl$_2$S glasses and -5.16 and -4.79 mm sec$^{-1}$ for those of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$. Antimony (Sb) in these glasses exists only as Sb(III) species. So its coordination is pyramidal (SbSe$_3$). As there is no available information on the structure of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ glasses containing a constant concentration of 20 mol% Sb$_3$Te$_3$, we can suggest that these glasses also consist of mixed glass networks with the combination of SbSe$_3$ and AsSe$_3$ the trigonal pyramidal units as seen in the case of Sb$_2$Se$_3$-As$_2$Se$_3$ glasses. There is an obvious correlation between covalence and glass stability.
The calculated values of $\Delta T_r(T_{c1})$ at the onset crystallization temperature $T_{c1}$ for examined glasses are reported in table 1 and 2. It is indicated that $\Delta T_r(T_{c1})$ values depend on the thermal characteristics such as $T_{c1}$ and $T_f$. $T_{c1}$ increases but $T_f$ vary little when $As_2Se_3$ concentration increases on $Sb_2Se_3-As_2Se_3$ binary system and on 20 mol% $Sb_2Te_3$ section of $Sb_2Se_3-As_2Se_3-Sb_2Te_3$ ternary system. As indicated in fig 7 and 8, $T_{c1}$ decreasing and $T_f$ vary

3.3 Degree of undercooling parameter

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little imply the increasing of $\Delta T_r(T_c)$. So the higher value of $T_c$ (lower value of $T_f$) in each system corresponds the smaller value of $\Delta T_r(T_c)$ and a small undercooled region ($T_f- T_c$) is observed. At the smaller value of $T_c$ (higher value of $T_f$) corresponds to the higher value of $\Delta T_r(T_c)$ and a wide undercooled region is also observed. The degree of undercooling depends on the extended undercooled region which depends on $T_c$ and $T_f$. The variation of the degree of undercooling, $\Delta T_r(T_c)$, as a function of As$_2$Se$_3$ concentration is shown in Fig 9. $\Delta T_r(T_c)$ decreases exponentially on Sb$_2$Se$_3$-As$_2$Se$_3$ binary system and on 20 mol% Sb$_2$Te$_3$ section of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system whith increasing of As$_2$Se$_3$ concentration. In the two cases, it is seen that small values of $\Delta T_r(T_c)$ are obtained when the concentration of As$_2$Se$_3$ increases. The decreased values of $\Delta T_r(T_c)$ can be explained by progressive substitution of Sb$_2$Se$_3$ by As$_2$Se$_3$ as it is seen above in the cases of glass-forming ability and glass stability. Thus, added As$_2$Se$_3$ can diminish the degree of undercooling and the undercooled region. These results imply that the glasses with high concentration of As$_2$Se$_3$ have a possibility of a low frequency of homogeneous nucleation because $\Delta T_r(T_c)$ decreases in respect to the classical theory of homogeneous nucleation. A similar trend was reported in (30-x)K$_2$O-xNb$_2$O$_5$-70TeO$_2$) glasses (Komatsu et al, 1997) [15] when Nb$_2$O$_5$ concentration increases.

### Table 2 Composition of glasses, thermal properties ($T_g$, $T_c$, and $T_f$) from DSC curves, values of glass forming ability parameters ($T_{rg}$ and $K_h$), values of glass forming stability parameters ($K_w$, $K_l$ and $\Delta T$) and degree of undercooling $\Delta T_r$ for studied glass in the ternary system Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ at constant Sb$_2$Te$_3$ concentration of 20 mol%.

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<th>Composition of glass to As$_2$Se$_3$</th>
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<th>$T_f$</th>
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### 3.4 Correlation between the degree of undercooling, glass-forming ability and glass stability

Similar trends were observed when the glass-forming parameter, $K_h(T_c)$ and the glass-stability parameters, $K_w(T_c)$ and $K_l(T_c)$, are plotted versus As$_2$Se$_3$ concentration. So, we will use only values of $K_h(T_c)$ and $K_l(T_c)$ obtained with $T_c$ temperature during correlations in which these parameters are used. 

Correlation between $\Delta T_r(T_c)$ and $K_h(T_c)$, for studied glasses containing 0 and 20 mol% Sb$_2$Te$_3$ is shown in Fig 10 which indicates that $\Delta T_r(T_c)$ decreases when $K_h(T_c)$ increases. 

Correlation between $\Delta T_r(T_c)$ and $\Delta T(T_c)$ of glasses with 0 and 20 mol% Sb$_2$Te$_3$ shows that $\Delta T_r(T_c)$ decreases when $\Delta T(T_c)$ increases (Fig 11). Supercooled liquid having wide supercooled liquid region ($\Delta T(T_c)$ is wide) is characterized by a low degree of undercooling ($\Delta T_r(T_c)$ is low). That means that the supercooled liquid is stable in a wide temperature range without crystallization and with high resistance to the nucleation and growth of crystalline phase (KapaKlis et al, 2003) [27]. The same behaviour is observed when $\Delta T_r(T_c)$ is correlated with the other glass stability parameter such as $K_w(T_c)$ (Fig 12). At the higher values of $\Delta T(T_c)$, $K_w(T_c)$ corresponds with lower value of $\Delta T_r(T_c)$. Thus, when the glass stability parameter become higher, the frequency of homogeneous nucleation becomes lower ($\Delta T_r(T_c)$ decreases) and vice versa. 

The relationship between $\Delta T_r(T_c)$ and parameters ($\Delta T(T_c)$ and $K_w(T_c)$) shown in Fig 11 and 12 indicates a good correlation $\Delta T_r(T_c)$ is one of key parameter for the glass stability of As$_2$Se$_3$ based glasses.
Fig. 13. Correlation between $\Delta T(T_{c1})$ and $K_c(T_{c1})$ in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 0 and 20 mol%.

Fig. 14. Correlation between $\Delta T(T_{c2})$ and $K_a(T_{c2})$ in the Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ ternary system at constant Sb$_2$Te$_3$ concentration of 0 and 20 mol%.
Correlation between $\Delta T_{c(Tc_1)}$ and $K_{l(Tc_1)}$ of glasses with 0 and 20 mol% Sb$_2$Te$_3$ shows that $\Delta T_{c(Tc_1)}$ increases when $K_{l(Tc_1)}$ increases (Fig 13). Supercooled liquid having low glass stability parameter such as ($K_{l(Tc_1)}$ is low) is characterized by a wide degree of undercooling ($\Delta T_{c(Tc_1)}$ is wide).

Both $\Delta T_{c(Tc_1)}$, $K_{h(Tc_1)}$ of glasses with 0 and 20 mol% Sb$_2$Te$_3$ increase with the increasing in As$_2$Se$_3$ content (Fig 14). In another words, a wide supercooled liquid region shows high glass-forming ability. A large $\Delta T_{c(Tc_1)}$ value may indicate that the supercooled is stable in a wide temperature without crystallization, this leads to a larger GFA of the alloy (Inoue et al, 1993) [28]. There is a correlation between glass-forming ability and glass stability. As discussed above, the overall liquid phase stability is positively related to the quantity $\Delta T_{c(Tc_1)} = T_{c1} - T_{g}$ while the
crystallization resistance is proportional to $T_c$. The increase of $\Delta T(T_c)$ can lead to an increase of liquid phase stability at metastable state and hence an increase in the GFA. Therefore, the GFA is positively associated with the $\Delta T(T_c)$ for glasses of Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ system. So $K_h(T_c)$ can be used to represent $\Delta T(T_c)$ and vice versa for these glasses based on As$_2$Se$_3$. That means $\Delta T(T_c)$ is a good criterion for characterization of GFA for these glasses used in the experiment. This speculation has been well confirmed in several glass-forming alloy systems in which the supercooled liquid region correlates reasonably well with the GFA of alloys (Shen and Schawrz, 1999) [29]. The same trend is observed between $K_h(T_c)$ and the other parameters of glass stability ($K_w(T_c)$ and $K_i(T_c)$) of 0 and 20 mol% Sb$_2$Te$_3$ (Fig 15 and 16). According to (Hruby, 1972) [30], the higher is the value of $K_h$ for certain glass, the higher its stability against crystallization on heating and presumably, the higher the glass ability to vitrify on cooling. Glass which vitrifies easily (high $K_h(T_c)$) is a thermally stable glass ($\Delta T(T_c)$). Glass forming ability $K_h(T_c)$ governs the thermal stability of studied glasses. This behaviour is observed when each glass stability parameter is plotted as a function of $T_{rg}$ in Sb$_2$Se$_3$-As$_2$Se$_3$ binary system but not in the case of $T_{rg}$ of 20 mol% Sb$_2$Te$_3$ in Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ system. Thus $T_{rg}$ is not a good indicator of glass-forming ability for these glasses containing 20 mol% Sb$_2$Te$_3$.

4. Conclusions

Glass-forming ability parameters (except of $T_{rg}$ of 20 mol% Sb$_2$Te$_3$) and glass stability parameters increase but the degree of undercooling decreases when the content of As$_2$Se$_3$ increases in Sb$_2$Se$_3$-As$_2$Se$_3$-Sb$_2$Te$_3$ system. This implies that glasses can be obtained easily and they can become most stable against crystallization. A low frequency of homogeneous nucleation can be suggested. The correlations between the degree of undercooling, glass-forming ability and glass stability have shown that the degree of undercooling is an important parameter for the studied glasses.

References