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A comparative study on the glass-forming ability of some alloys in the Sb–As–Se system by differential scanning calorimetry

D. García-G. Barreda, J. Vázquez *, P.L. López-Alemany, P. Villares, R. Jiménez-Garay

Departamento de Física de la Materia Condensada, Facultad de Ciencias, Universidad de Cádiz, Apartado 40, 11510, Puerto Real (Cádiz), Spain

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Abstract

The glass-forming ability and devitrification of alloys in the Sb–As–Se system have been studied by differential scanning calorimetry (DSC). A comparison of various simple quantitative methods to assess the level of stability of glassy materials in the above-mentioned system is presented. All these methods are based on the characteristic temperatures, obtained by heating of the samples in non-isothermal regime, such as the glass transition temperature, T_g, the temperature at which crystallization begins, T_{in}, the temperature corresponding to the maximum crystallization rate, T_p, or the melting temperature, T_m. In this work, a kinetic parameter $K_r(T)$ is added to the stability criteria. The thermal stability of some ternary compounds of $Sb_xAs_{0.60-(2x+y)}Se_{0.40+x+y}$-type has been evaluated experimentally and correlated with the activation energies of crystallization by this kinetic criterion and compared with those evaluated by other criteria.

Keywords: A. Amorphous materials; A. Glasses; C. Differential scanning calorimetry (DSC); D. Phase transitions; D. Semiconductivity

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1. Introduction

Knowledge of amorphous materials is one of the most active fields of research in the physics of condensed matter today [1]. The great interest in these materials is largely due to their ever-increasing applications in modern technology. Their possibilities in the immediate future are huge based on the characteristic properties such as electronic-excitation phenomena, chemical reactivity and inertia, and superconductivity. Therefore, the advances that have been made in the physics and chemistry of the quoted materials have been very appreciated within the research community. Glassy alloys of chalcogen elements were the initial objects of study because of their interesting semiconducting properties [2] and more recent importance in optical recording [3]. The As_xSe_{1-x} system is probably one of the most studied among the binary systems of semiconducting chalcogenide glasses [4–6] and there is a great number of works on the effect of addition of different elements on the physical properties of the stoichiometric $As_{0.4}Se_{0.6}$ glass [7–9]. The ternary Sb–As–Se glasses are formed by substituting As atoms by Sb atoms in the As–Se system. The corresponding substitution does not alter drastically the basic structure of glass, since both As and Sb are isovalent elements. Addition of appropriate amounts of Sb to the As–Se system could form good ternary glasses from the point of view of the thermal stability. The glass transition temperature, optical energy gap, thermal diffusivity, elastic behaviour and electrical conductivity of the Sb–As–Se system have been measured and analysed by different workers in the last decades [10–16]. The maximum Sb content that forms glass in this ternary system is 20 at.%, according to the literature [17]. Bearing in mind that the recording materials must be stable in the amorphous state at low temperature, to use alloys of the above-mentioned system as recording materials, it is very important to know the glass-forming ability and chemical durability of the solids of quoted system.

In order to evaluate the level of stability of the glassy alloys, different simple quantitative methods have been suggested. Most of these methods [18–22] are based on the characteristic temperatures such as the glass transition temperature, T_g, the crystallization temperature, T_p, or the melting temperature, T_m. Some of them [23, 24] are based on the reaction rate constant, K. Some of the others [25–27] are based on the crystallization activation energy. These thermal parameters [28] are easily and accurately obtained by differential scanning calorimetry (DSC) during the heating processes of glass samples. The first thorough study on the glass thermal stability of various compounds was done by Sakka and Mackenzie [29], using

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* Corresponding author. Tel.: +34 956 016 323; fax: +34 956 016 288.
E-mail address: jose.vazquez@uca.es (J. Vázquez).

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the ratio T_g/T_m. Dietzel [18] introduced the glass criterion, $\Delta T = T_{in} - T_g$ (T_{in} is the onset temperature of crystallization), which is often an important parameter to evaluate the glass-forming ability of the materials. By the use of characteristic temperatures, Hruby [21] developed the Hr criterion, $\mathrm{Hr} = \Delta T/(T_m - T_p)$, and compositional dependencies of the Hruby coefficient were survived by Sestak [30]. On the basis of the Hr criterion, Saad and Poulain [22] obtained two other criteria, weighted thermal stability H' and S criterion, $H' = \Delta T/T_g$, $S = (T_p - T_{in})\Delta T/T_g$, respectively.

In the present work, the above-mentioned criteria have been applied to the alloys $\mathrm{Sb}_x\mathrm{As}_{0.60-(2x+y)}\mathrm{Se}_{0.40+x+y}$, where $x=0.08$, $y=0$ (S1), $x=0.12$, $y=0$ (S2) and $x=0.16$, $y=0.06$ (S3), and it is found that the parameters ΔT, Hr, H' and S increase with decreasing antimony content. Bearing in mind that the values of these parameters increase with increasing stability, it is possible to suggest that the lower the antimony content of the alloy, the greater is its glass thermal stability [17]. In addition, a kinetic parameter, $K_r(T)$, with an Arrhenian temperature dependence, is introduced to the stability criteria. Decreasing values of the above-mentioned parameter have been found for the alloys with decreasing antimony content. This fact confirms that S1 alloy is the most stable one.

2. Theoretical background

The theoretical basis for interpreting DSC results is provided by the formal theory of transformation kinetics. This theory describes the evolution with time, t, of the volume fraction crystallized, x, in terms of the crystal growth rate, u

$$x = 1 - \exp\left[-gN\left(\int_0^t u(t')\,\mathrm{d}t'\right)^n\right] = 1 - \exp(-gNI_1^n). \quad (1)$$

Here g is a geometric factor, N is the number of pre-existing nuclei per unit volume, and n is an exponent, which depends on the mechanism of transformation. In Eq. (1) it is assumed that the nucleation process takes place early in the transformation and the nucleation frequency is zero thereafter. This case has been referred in the literature [31–33] as 'site saturation'.

Although, in general, the temperature dependence of the crystal growth rate is not Arrhenian when a broad range of temperature is considered [34]; however, over a sufficiently limited range of temperature (such as the range of crystallization peaks in DSC experiments), u may be described in a zeroth-order approximation by

$$u \approx u_0 \exp(-E/RT) \quad (2)$$

where E is the effective activation energy for crystal growth, u_0 is a pre-exponential factor and R is the ideal gas constant.

Taking the derivative of Eq. (1) with respect to time and substituting Eq. (2) in the resulting expression, the crystallization rate is obtained as

$$\mathrm{d}x/\mathrm{d}t = n(gN)(1-x)I_1^{n-1}u_0 \exp(-E/RT) \quad (3)$$

The maximum crystallization rate in a non-isothermal process with a heating rate $\beta = \mathrm{d}T/\mathrm{d}t$ is found by making $\mathrm{d}^2x/\mathrm{d}t^2 = 0$, thus obtaining the relationship

$$nC_p(I_1^n)|_p = \beta E(I_1)|_p/RT_p^2 + (n-1)u_p \quad (4)$$

where the quantity values, which correspond to the maximum crystallization rate, are denoted by the subscript p, and where the function $C_p = gNu_p$ has a dimension equation which can be expressed as $[C_p] = [\mathrm{L}]^{1-n}[\mathrm{T}]^{-1}$.

By means of the substitution $y' = E/RT'$, the integral I_1 can be expressed, according to the literature [35], by the sum of the alternating series

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

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 $y' = E/RT' \gg 1$ (usually $E/RT' \geq 25$), it is possible to use only the first two terms of this series and the error introduced is not greater than 1%. In addition, if it is assumed that $T_0 \ll T$ (T_0 is the starting temperature), so that y_0 can be taken as infinity [34], the integral I_1 becomes

$$\begin{aligned} I_1 &= u_0E(\beta R)^{-1}\mathrm{e}^{-y}y^{-2}(1-2y^{-1}) \\ &= RT^2u(\beta E)^{-1}(1-2RTE^{-1}). \end{aligned} \quad (5)$$

Substituting the last expression of I_1 in Eq. (4), one obtains

$$(I_1)|_p = (gN)^{-1/n}(1-2RT_p/nE)^{1/n}$$

relationship that when it is equated to Eq. (5), for the maximum crystallization rate, gives

$$\begin{aligned} &RT_p^2(\beta E)^{-1}K_0\exp(-E/RT_p) \\ &\quad = (1-2RT_p/nE)^{1/n}(1-2RT_p/E)^{-1} \end{aligned} \quad (6)$$

where $K_0 = (gN)^{1/n}u_0$ is the frequency factor, related to the probability of effective molecular collisions for the formation of the activated complex and whose dimension equation is $[K_0] = [\mathrm{T}]^{-1}$. The logarithmic form of the last expression can be written as

$$\ln(T_p^2/\beta) + \ln(K_0R/E) - E/RT_p = (2RT_p/E)(1-n^{-2}) \quad (7)$$

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.

It should be noted that for most crystallization reactions, the right-hand side (RHS) of Eq. (7) is generally negligible in comparison to the individual terms on the left-hand side for common heating rates ($\leq 100\ \mathrm{K\,min^{-1}}$). Therefore, the approximation in Eq. (7) (RHS=0) might introduce a 3% error in the value of E/R in the worst cases, and Eq. (7) becomes

$$\ln(T_p^2/\beta) = E/RT_p + \ln(E/RK_0). \quad (8)$$

This equation represents a straight line with slope, E/R, and intercept, $\ln(E/RK_0)$. Then, one can obtain E, K_0 and $K(T)$.

In order to evaluate the thermal stability of glassy materials, Surinach et al. [23] introduced a $K(T_g)$ criterion, and Hu and Jiang [24] developed the $K(T_p)$ criterion, $K(T_g) = K_0 \exp(-E/RT_g)$ and $K(T_p) = K_0 \exp(-E/RT_p)$, respectively. Thus, the values of these two parameters indicate the tendency of glass to devitrify on heating. The larger their values, the greater is the tendency to devitrify. The formation of a glass is a kinetic process, therefore, it is reasonable to assess the glass stability by a kinetic parameter, $K(T)$. The Hr parameter itself is a stability factor based on the characteristic temperatures. Here a stability criterion is defined as $K_r(T)$:

$$K_r(T) = K_0 \exp(-\mathrm{Hr}E/RT) \tag{9}$$

where T is any temperature between T_g and T_p. The theoretical background for the definition of the new parameter $K_r(T)$ would be based on the analysis of the relation between the parameters $K(T)$ and $K_r(T)$. Differentiating the expressions of both parameters results in

$$\mathrm{d}K_r = \mathrm{Hr}EK_r(RT^2)^{-1}\mathrm{d}T, \qquad \mathrm{d}K = EK(RT^2)^{-1}\mathrm{d}T$$

and the relative variation in each parameter per Kelvin is

$$\frac{1}{K_r}\frac{\Delta K_r}{\Delta T} = \frac{\mathrm{Hr}E}{RT^2}, \qquad \frac{1}{K}\frac{\Delta K}{\Delta T} = \frac{E}{RT^2}.$$

It should be noted that the above-mentioned variation of the parameter $K_r(T)$ is Hr times the variation in parameter $K(T)$, which could justify the accuracy of this new parameter.

Just like the $K(T)$ criteria, the smaller the values of $K_r(T)$, the greater is the thermal stability of the glass. The obvious advantage of this method is that it can evaluate the glass stability over a broad temperature range other than at only one temperature such as T_g or T_p.

3. Experimental procedure

The alloys were prepared in bulk form by the standard melt quenching method. High-purity (99.999%) antimony, arsenic and selenium in appropriate atomic percent proportions were weighed (total 7 g per batch) into quartz glass ampoules. The contents were sealed under a vacuum of 10^{-2} Pa, heated to 1225 K for about 24 h and continuously rotated in a furnace to homogenize the material. The ampoules were quenched in water, which supplied the necessary cooling rate for obtaining the glass. The amorphous state of the material was checked through a diffractometric X-ray scan in a Siemens D500 diffractometer. The calorimetric measurements were carried out in a Perkin–Elmer DSC7 calorimeter with an accuracy of ± 0.1 K. Temperature and energy calibrations of the instrument were performed using the well-known melting temperatures and melting enthalpies of high-purity indium and zinc supplied with the instrument. For non-isothermal experiments, glass samples weighing about 20 mg were sealed in aluminium pans and scanned at room temperature through their T_g at different heating rates of 2, 4, 8, 16, 32 and 64 K min^{-1}. An empty aluminium pan was used as a reference, and in all cases a constant 60 ml min^{-1} flow of nitrogen was maintained in order to provide a constant thermal blanket within the DSC cell, thus eliminating thermal gradients, and ensuring the validity of the applied calibration standard from sample to sample. Moreover, the nitrogen purge allows to expel the gases emitted by the reaction, which are highly corrosive to the sensory equipment installed in the DSC furnace. The glass transition temperature was considered as a temperature corresponding to the inflection point of the lambda-like trace on the DSC scan.

4. Results and discussion

The characteristic temperatures from DSC scans are given in Table 1. The glass-forming ability of the three alloys studied

Table 1
Characteristic parameters of the alloys S1, S2 and S3

Alloy	β (K min^{-1})	T_g (K)	T_{in} (K)	T_p (K)	T_m (K)	ΔT (K)	Hr	H'	S (K)
S1	2	424.3	516.4	537.1	642.0	92.1	0.8780	0.2171	4.4932
	4	429.9	523.3	544.9	650.8	93.4	0.8820	0.2173	4.6928
	8	435.5	530.2	552.9	659.6	94.7	0.8875	0.2175	4.9361
	16	441.1	537.1	561.2	668.3	96.0	0.8964	0.2176	5.2451
	32	445.5	542.8	569.7	677.2	97.3	0.9051	0.2184	5.8751
	64	452.1	550.9	578.5	686.1	98.8	0.9182	0.2185	6.0316
S2	2	427.5	512.7	533.2	647.7	85.2	0.7441	0.1993	4.0856
	4	433.1	519.5	542.6	657.6	86.4	0.7513	0.1995	4.6083
	8	441.4	529.5	553.4	668.5	88.1	0.7654	0.1996	4.7703
	16	447.6	537.0	562.6	678.8	89.4	0.7694	0.1997	5.1131
	32	454.7	545.6	574.9	693.0	90.9	0.7697	0.1999	5.8574
	64	469.8	564.0	597.8	716.3	94.2	0.7949	0.2005	6.7773
S3	2	393.1	471.2	484.9	592.9	78.1	0.7231	0.1987	2.7219
	4	399.6	479.0	494.4	603.3	79.4	0.7291	0.1987	3.0600
	8	406.7	487.6	504.2	615.0	80.9	0.7301	0.1989	3.3020
	16	412.4	494.5	514.9	627.2	82.1	0.7311	0.1991	4.0612
	32	418.8	502.3	526.9	640.9	83.5	0.7325	0.1994	4.9047
	64	424.8	509.6	540.8	656.4	84.8	0.7336	0.1996	6.2282

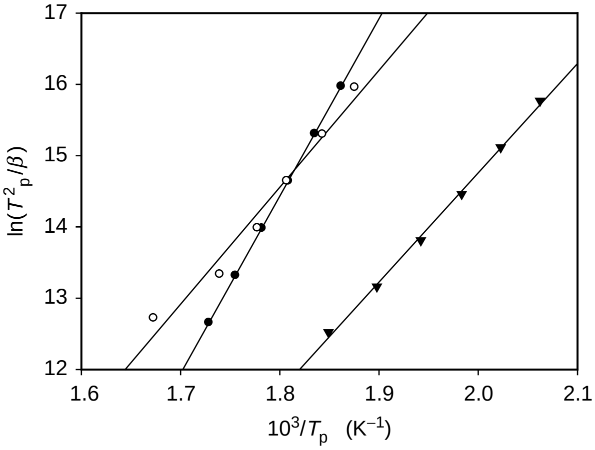

Fig. 1. Plots of $\ln(T_p^2/\beta)$ versus $10^3/T_p$ and straight regression lines for three glassy alloys S1 (●), S2 (○), S3 (▼).

can be assessed using these characteristic temperatures, T_g, T_{in}, T_p, and T_m. The existing stability criterion parameters based on these characteristic temperatures are also listed in Table 1.

To obtain the kinetic parameters of the crystallization, Eq. (8) is applied. Fig. 1 represents the evolution of $\ln(T_p^2/\beta)$ versus $1/T_p$ for the three alloys. The plots were found to be straight lines in accordance with Eq. (8). The activation energy, E, and frequency factor, K_0, are then evaluated by least-squares fitting method. Table 2 summarizes the values determined by these calculations. After knowing the values of E and K_0, the kinetic parameters $K(T)$ and $K_r(T)$ of studied alloys were calculated by using the relationship $K(T)=K_0\exp(-E/RT)$ and Eq. (9), respectively. These calculations were carried out in order to compare the stability sequence of the studied materials from the quoted parameters with the corresponding sequence deduced from stability criteria based on the characteristic temperatures. The values of $K(T)$ and $K_r(T)$ for the temperatures T_g and T_p are listed in Table 3. Fig. 2 represents the plots of $K_r(T)$ versus T. It is found that $K_r(T)$ of S1 varies slowly with increasing T and the values are on the T-axis, indicating a relatively high stability, while $K_r(T)$ of the other two samples varies more rapidly with increasing T, which signifies a minor stability. These considerations verify the thermal stability order of the above-mentioned glassy alloys.

It is known that these existing criteria of glass stability allow predicting the glass-forming ability of a material. It is possible to suggest that the larger their values, the greater should be the glass thermal stability. According to these suggestions, the parameters, ΔT, Hr, H' and S, in Table 1 show that the S1 glass sample is more stable than the other two samples. Also, it is possible to obtain a consistent stable order for these glasses by the reaction rate constant. According to literature [23,24] ($K(T_g)$ and $K(T_p)$ criteria), smaller the values of these parameters, better should be the glass-forming ability of the material. So the data for both $K(T_g)$ and $K(T_p)$ in Table 3 indicate that S1 glass sample is the most stable, and the stability orders at different heating rates are S1 > S2 > S3. In addition, by using Eq. (9), the data of $K_r(T_g)$ and $K_r(T_p)$ are calculated

Table 2
Straight regression lines (SRL) fitted to the values of $\ln(T_p^2/\beta)$ and kinetics parameters of the analysed alloys

Alloy	SRL	E (kcal mol^{-1})	K_0 (s^{-1})	r
S1	$24.8682\times10^3/T_p-30.3305$	49.74	3.70×10^{17}	0.9889
S2	$16.4141\times10^3/T_p-14.9888$	32.83	2.12×10^{11}	0.9876
S3	$15.6128\times10^3/T_p-16.4678$	31.23	8.86×10^{11}	0.9996

r is the correlation coefficient.

Table 3
Kinetic parameters $K(T)$ and $K_r(T)$ for three alloys

Alloy	β (K min^{-1})	$K(T_g)$ (s^{-1})	$K(T_p)$ (s^{-1})	$K_r(T_g)$ (s^{-1})	$K_r(T_p)$ (s^{-1})
S1	2	1.30×10^{-8}	2.87×10^{-3}	1.65×10^{-5}	8.16×10^{-1}
	4	2.78×10^{-8}	5.58×10^{-3}	2.56×10^{-5}	12.17×10^{-1}
	8	5.85×10^{-8}	1.08×10^{-2}	3.61×10^{-5}	17.02×10^{-1}
	16	1.21×10^{-7}	2.10×10^{-2}	4.16×10^{-5}	20.70×10^{-1}
	32	2.11×10^{-7}	4.07×10^{-2}	4.21×10^{-5}	25.61×10^{-1}
	64	4.76×10^{-7}	7.90×10^{-2}	4.28×10^{-5}	26.60×10^{-1}
S2	2	4.47×10^{-6}	9.04×10^{-3}	8.28×10^{-2}	23.8581
	4	7.35×10^{-6}	1.54×10^{-2}	9.11×10^{-2}	28.5358
	8	1.50×10^{-5}	2.78×10^{-2}	9.22×10^{-2}	29.2678
	16	2.51×10^{-5}	4.52×10^{-2}	1.18×10^{-1}	37.7521
	32	4.45×10^{-5}	8.43×10^{-2}	1.81×10^{-1}	60.5076
	64	1.42×10^{-4}	2.52×10^{-1}	1.84×10^{-1}	70.2935
S3	2	4.97×10^{-6}	9.16×10^{-3}	2.97×10^{-1}	0.68×10^{2}
	4	9.48×10^{-6}	1.70×10^{-2}	3.75×10^{-1}	0.88×10^{2}
	8	1.87×10^{-5}	3.14×10^{-2}	5.93×10^{-1}	1.34×10^{2}
	16	3.19×10^{-5}	5.98×10^{-2}	8.42×10^{-1}	2.08×10^{2}
	32	5.68×10^{-5}	1.19×10^{-1}	12.20×10^{-1}	3.31×10^{2}
	64	9.63×10^{-5}	2.56×10^{-1}	17.23×10^{-1}	5.60×10^{2}

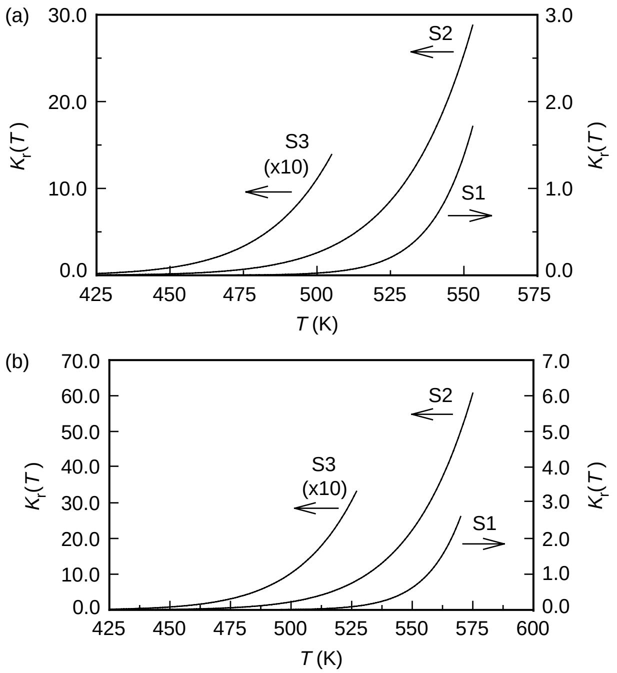

Fig. 2. Plots of $K_r(T)$ versus T for three glassy alloys to verify the stable order: (a) $\beta = 8$ K min^{-1}, and (b) $\beta = 32$ K min^{-1}.

and given in Table 3, showing that S1 alloy is also the most stable, and the orders of stability are also $S1 > S2 > S3$ at various heating rates. This stability result agrees with that of the $K(T_g)$ and $K(T_p)$ criteria.

The above-mentioned stability orders agree satisfactorily with literature [17], where it is noted that the crystallizing ability of glassy arsenic selenides increases substantially when antimony is introduced.

5. Conclusion

The glass-forming ability of some alloys in the Sb–As–Se system has been evaluated using various thermal stability criteria, based on the characteristic temperatures. Moreover, in the present work, the $K_r(T)$ criterion has been considered for the evaluation of glass stability from DSC data. It includes both the kinetic parameters and critical temperatures. Therefore, it is reasonable to think that the obtained data from the quoted criterion agree satisfactorily with the values, which result from the existing criteria based on the characteristic temperatures and $K(T)$ criteria. A high value of $K_r(T)$ means poor stability of the glass. In the present work, the non-isothermal devitrification of three glassy alloys in the above-mentioned system has been studied at different heating rates and various temperatures. By means of the quoted study, it has been verified that the $K_r(T)$ criterion is slightly affected both by the heating rate and by the temperature, while the other criteria show a bigger variation with the heating rate. Among the three glassy alloys, $K_r(T)$ of S1 glass sample is smallest, so this glass composition is the most stable. Finally, the stability order of these three glass samples is $S1 > S2 > S3$.

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