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Correlation between structural and thermodynamic properties of some selenium based phase-change materials

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ABSTRACT

In this study, we prepared novel selenium rich multi-component glasses by incorporating In, Cd and Sb as foreign elements in an Sn containing Se—Te system in order to study their metal-induced effects on the thermal properties of the parent ternary glass. In particular, we determined the thermodynamic parameters of $Se_{80}Te_{18}Sn_2$ and $Se_{80}Te_8Sn_2M_{10}$ (M = Cd, In, Sb) glassy semiconductors in a non-isothermal environment using the differential scanning calorimetry. Calorimetric measurements were obtained in the glass transition regions for $Se_{80}Te_{18}Sn_2$ and $Se_{80}Te_8Sn_2M_{10}$ (M = Cd, In, Sb) glasses to determine their thermodynamic parameters such as the specific heat, enthalpy, and entropy during glass transition. We analyzed the variation in the specific heat before and after the heat capacity jump in these alloys. The metal-induced effects of foreign elements on the thermodynamic properties of the parent glass were also investigated in terms of the influence of the elemental specific heat of the added elemental metal as well as the thermal stability and glass-forming ability of the glasses.

1. Introduction

In recent decades, there has been great progress in the development of new compositions of chalcogenide glasses. These compositions are potential candidates for applications in the design of optical systems, including optical memory applications, fibers for telecommunications, and lasers [1–7]. In these applications, the thermal and thermodynamic properties are important for determining the figure of merit of the optical glasses [8–11]. The determination of these properties for a glass-forming material requires measurements of a range of physical parameters using a combination of techniques. The specific heat and related thermodynamic properties can provide useful information about the glass structure, phonon energy, and heat diffusion coefficients [12–14]. Previous studies of the specific heat indicates that it is a significant parameter for providing information regarding the solubility, and thus it is directly related to the thermal transport properties of glass-forming melts after doping with ions [15]. Some studies have shown that this property play a key role in the structural modification of glasses in the glass transition range [14–16]. However, the variations in the specific heat values before and after thermal relaxation are highly diverse.

Thus, specific heat measurements can be used to understand the glass transition behavior of chalcogenide glasses [8–16], but further experimental studies are still required. Specific heat studies have been conducted for various two-component (i.e., binary) and three-component (i.e., ternary) glassy systems by investigating the effects of specific additives on the thermodynamic properties of binary and ternary glasses after varying their compositions. In the present study, we obtained calorimetric measurements for $Se_{80}Te_{18}Sn_2$ and $Se_{80}Te_8Sn_2M_{10}$ (M = Cd, In, Sb) glasses after changing the additive element rather than varying the composition.

2. Experimental

Ternary $Se_{80}Te_{18}Sn_2$ glass and quaternary $Se_{80}Te_8Sn_2M_{10}$ (M = Cd, In, Sb) glass were prepared using the well-known quenching method, as described previously [17]. Details of the compositions are given in Table 1.

The compositions of the as-prepared samples were analyzed using energy-dispersive X-ray spectroscopy (EDX). Fig. 1 shows the EDX patterns obtained for the glassy $Se_{80}Te_8Sn_2In_{10}$ and $Se_{80}Te_8Sn_2Sb_{10}$ alloys, which indicates that the uncertainties of the compositions of the as-prepared samples were not significant i.e., the final compositions were almost the same as the initial concentrations introduced into the batch.

Using X-ray diffraction (XRD), we confirmed that the final structures

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Table 1
Contents of glassy $Se_{80}Te_{18}Sn_2$ and $Se_{80}Te_8Sn_2M_{10}$ (M = Cd, In, Sb) alloys.

System.	Element	Atomic mass	Atomic %	Atomic mass %	Actual Weight (gm)
$Se_{80}Te_{18}Sn_2$	Se	78.96	80	6316.8	3.5684
	Te	127.6	18	2296.8	1.2974
	Sn	118.71	2	237.42	0.1341
$Se_{80}Te_8Sn_2Cd_{10}$	Se	78.96	80	6316.8	3.6307
	Te	127.6	8	1020.8	0.5867
	Sn	118.71	2	237.42	0.1365
	Cd	112.41	10	1124	0.6461
$Se_{80}Te_8Sn_2In_{10}$	Se	78.96	80	6316.8	3.6206
	Te	127.6	8	1020.8	0.5851
	Sn	118.71	2	237.42	0.1361
	In	114.82	10	1148.2	0.6581
$Se_{80}Te_8Sn_2Sb_{10}$	Se	78.96	80	6316.8	3.5921
	Te	127.6	8	1020.8	0.5805
	Sn	118.71	2	237.42	0.1350
	Sb	121.75	10	1217.6	0.6924

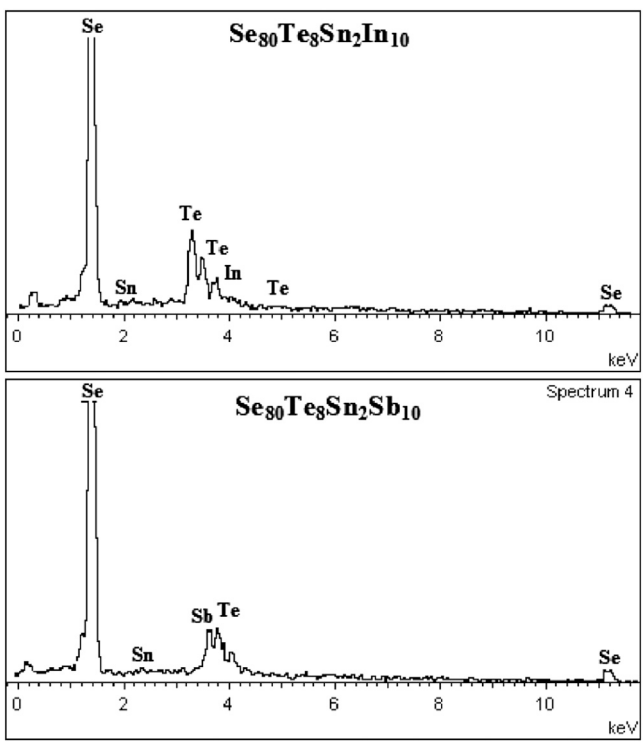

Fig. 1. EDX patterns obtained for the glassy $Se_{80}Te_8Sn_2In_{10}$ and $Se_{80}Te_8Sn_2Sb_{10}$ alloys.

of the as-prepared materials were glassy in nature (Fig. 2). The glasses were then ground to obtain fine powdered samples for differential scanning calorimetry (DSC) analyses. The advantages of DSC compared with other thermo-analytical techniques are as follows: (1) DSC is a more straightforward approach; (2) it only requires a finely ground powder; (3) it provides relatively better insights; and (4) unlike other methods, it does not depend on the size and shape of the sample. The thermal activities were determined using a DSC instrument (TA instrument USA; Model Q20), which had a temperature accuracy of ~ ± 0.1 K and a mean standard error of ~ ± 1 K in the measured value ranges (glass transition and crystallization temperatures). We heated 5–10 mg of sample at a rate of 20 K/min and the changes in the heat flow were recorded with respect to a blank pan. We obtained the measurements under almost the same conditions. We used the program provided with the DSC instrument to determine the dependence of the specific heat (C_p) on the temperature. Integrating the C_p curve using the DSC software provided the enthalpy value.

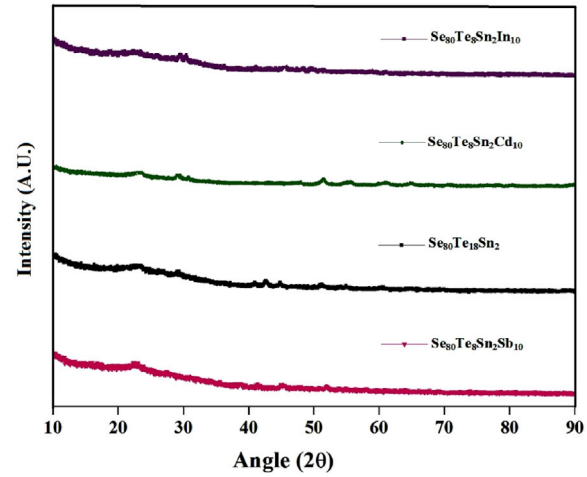

Fig. 2. XRD patterns obtained for the glassy $Se_{80}Te_{18}Sn_2$ and $Se_{80}Te_8Sn_2M_{10}$ (M = Cd, In, Sb) alloys.

The Q20 DSC instrument was calibrated using the calibration wizard over a temperature range of 273 K–673 K under standard conditions (a heating rate of 20 K/min for baseline and sapphire calibration runs, and a 10 mg indium sample (99.999% pure) was also crimped in aluminum pans and heated at 10 K/min).

The Calibration Wizard is a calibration setup process in a step-by-step fashion, which allowed us to choose a Calibration method and then perform the calibration. The calibration was performed before obtaining the specific heat measurements. After T-zero and cell constant calibrations, the indium sample was reloaded and run. We carefully prepared and weighed 6–10 mg samples in standard aluminum pans. After crimping the pans containing the reference material, the pans were further flattened to improve the thermal contact, especially for the higher temperature melting standards (where a good thermal contact is particularly important). The melting heats and temperatures were calculated, where the experimental enthalpies were within 0.9% of the calibrated values using indium. The experimental heat capacities were within 1% of the calibration values using sapphire.

3. Theoretical basis

DSC was used to monitor the thermally-induced phase transitions. DSC records the heat flow difference between the sample in the pan and a reference pan (which is usually empty) with identical temperature increases. Furthermore, we plotted this difference as a function of temperature. The temperature of both the sample and reference increased at a constant rate. The experiment was performed at constant pressure and thus the heat flow was comparable to the variations in enthalpy, as follows.

$$\frac{dH}{dt} = \left(\frac{dq}{dt}\right)_p \tag{1}$$

The difference in the heat flow in both pans of the DSC cell is given by the following expression.

$$\Delta\left(\frac{dH}{dt}\right) = \left(\frac{dH}{dt}\right)_{sample} - \left(\frac{dH}{dt}\right)_{reference} \tag{2}$$

The absorption of heat occurs during endothermic reactions such as glass transitions and melting. Thus, the heat flow through the reference pan is smaller than that to the sample pan, i.e. $\Delta\ (dH/dt) > 0$ for an endothermic reaction. By contrast, the liberation of heat occurs during the exothermic reactions such as crystallization. Thus, the heat flow to the sample pan is lower than that through the reference pan, i.e. $\Delta(dH/$

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