Stability Analysis of IV-V-VI Chalcogenide Glasses Using Glass Transition and Crystallization Temperature

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Abstract. Selenium based chalcogenide glasses are attractive candidates for IR devices due to their low transmission loss. Thermal studies for $Ge_{19}Se_{81-x}Sb_x$ (x = 0, 4, 8, 12, 16, 17.2, 20) have been carried out using differential thermal analysis. Glass transition temperature has been calculated using Tanaka's relation. Marseglia's and Ozawa's methods have been used for the calculation of activation energy for crystallization. Effect of *Sb* addition on *GeSe* base system shows that resistance to devitrification increases up to x = 17.2.

Keywords: Differential thermal analysis, Glass transitions
PACS: 81.70Pg; 64.70.P-

INTRODUCTION

Chalcogenide glasses have received great attention due to their high photosensitivity, high crystallization and glass transition temperature [1]. Fast switching between amorphous and crystalline states make these glasses promising candidates for rewritable data storage applications [2]. All optical switching devices are considered as next-generation devices due to high speed optical communication [3]. Chalcogenide glasses show transparency in infrared region, so that they are able to be drawn in fibers [4]. These fibers can be used for high speed telecommunication systems such as in optical switching, amplification, optical regenerators, etc. due to their high nonlinear refractive index [5]. *Ge-Se-Sb* shows large glass forming area and having applications in optical region as they have low transmission loss [6].

In the present paper, the effect of *Sb* on *GeSe* has been studied in terms of glass transition temperature and crystallization temperature. T_g has been calculated using Tanaka's relation. Marseglia's and Ozawa's methods have been used to calculate activation energy for crystallization.

EXPERIMENTAL DETAILS

Melt quench technique has been used for synthesis of $Ge_{10}Se_{81-x}Sb_x$ (where x = 0, 4, 8, 12, 16, 17.2, 20) samples. Detailed sample preparation technique is given elsewhere [7]. Amorphous nature of both bulk and thin films was examined by X ray diffraction spectra using X-ray diffractometer (X'Pert Pro). The prepared samples were ground to fine powder and were taken in alumina pan for DTA studies (EXSTAR/TG/DTA 6300) at different heating rates (α = 5, 10, 15, 20 K/min). All measurements were made under non-isothermal conditions.

RESULTS AND DISCUSSION

Glass transition temperature (T_g) is one of the most important parameters for characterization of glassy state. T_g represents the strength and rigidity of network and has been calculated using theoretical approach. T_g is associated with mean coordination number which has been expressed by Tanaka [8].

$$\ln T_g \cong 1.6Z + 2.3. \qquad (1)$$

where Z expresses the average coordination number. The values of T_g have been found to increase with increasing content of *Sb* (Table 1).

The activation energy for crystallization (E_c) deals with the nucleation and growth process that dominates the devitrification of most glassy solids. Based on the experimental results, the theoretical approach is set up on two models to analyze E_c.

First model: The activation energy of amorphous to crystalline transformation has been calculated on the basis of Marseglia's theory [9].

$$\ln (T_c/\alpha) = E_c^M + const. \qquad (2)$$

where T_c is crystallization temperature, α is heating rate. The plot of T_c/α *vs.* $1000/T_c$ for all compositions

is shown in Figure 1. Activation energy can be deduced by fitting the straight line.
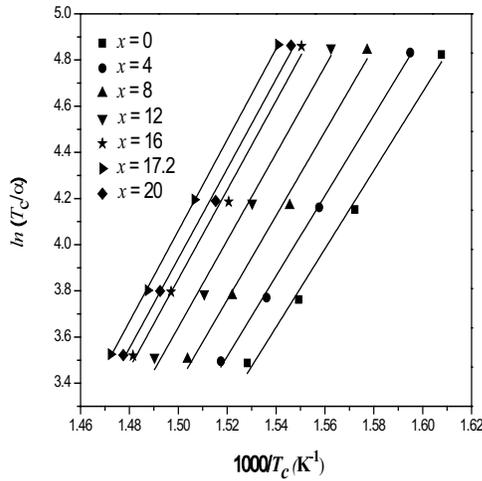

FIGURE 1. Plot of $ln(T_c/\alpha)$ vs. $1000/T_c$ for $Ge_{19}Se_{81-x}Sb_x$ ($x = 0, 4, 8, 12, 16, 20$)

Second model: Ozawa's method has been used to calculate activation energy of crystallization [10].

$$\ln \alpha = const. - 1.052\, E_c^{\,O}/RT_c. \quad (3)$$

TABLE 1. Values of T_g, E_c^M, E_c^O for $Ge_{19}Se_{81-x}Sb_x$ (where $x = 0, 4, 8, 12, 16, 17.2, 20$).

At. %	T_g (K)	E_c^M (kJmol^{-1})	E_c^O (kJmol^{-1})
x=0	449.44	141.27	146.52
x=4	479.14	144.93	150.34
x=8	510.81	151.67	157.08
x=12	544.57	156.67	162.16
x=16	581.56	160.66	166.15
x=17.2	591.82	163.82	169.31
x=20	618.93	161.57	167.07

The slope of straight line in Figure 2 gives the value of E_c^O. The values of E_c calculated from both the
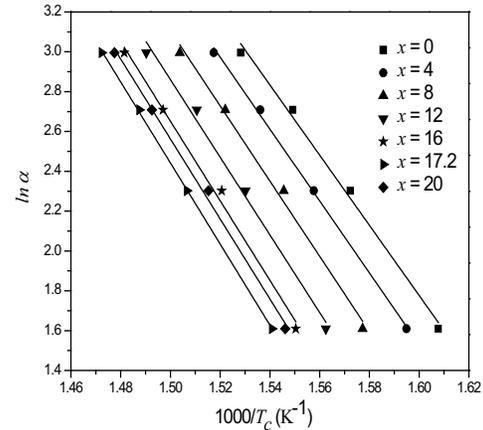

FIGURE 2. Plot of $ln\ \alpha$ vs. $1000/T_c$ for $Ge_{19}Se_{81-x}Sb_x$ (where $x = 0, 4, 8, 12, 16, 17.2, 20$)

methods have been listed in Table 1. Increase in glass transition temperature shows that the rigidity of system increases. There is a delay in activation energy of crystallization indicating maximum for $x = 17.2$ and hence, thermally most stable composition.

CONCLUSION

Glass transition temperature has been found to increase with increasing content of *Sb*. Activation energy of crystallization has been found to increase up to $x = 17.2$ and then decreases. Results indicate that $Ge_{19}Se_{63.8}Sb_{17.2}$ composition is thermally stable and may be explored for memory devices.

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