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Structural and transport properties of amorphous Se–Sb–Ag chalcogenide alloys and thin films

K.S. Bindra [a], Nikhil Suri [a], M.S. Kamboj [b], R. Thangaraj [a,*]

[a] Semiconductors Laboratory, Department of Applied Physics, Guru Nanak Dev University, Amritsar-143005, Punjab, India
[b] Department of Electrical Computer Engineering, Ryerson University, 350 Victoria Street Toronto, Ontario, Canada M5B 2K3

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Abstract

Bulk $Se_{80-x}Sb_{20}Ag_x$ ($8 \le x \le 14$) chalcogenide glasses have been prepared by conventional melt quenching technique. Thin films of the above composition have been prepared by thermal evaporation of the bulk material. X-ray diffraction studies have been performed to investigate the structure of the thin films. The presence of only broad features and the absence of any sharp peaks in the X-ray diffractogram confirm that the films are amorphous in nature. The addition of Ag to the Se–Sb host has been found to segregate as Ag_2Se nanophase. Scanning electron microscopy images show that both the Ag-rich and Sb-rich nanophases co-exist in this system. The glass transition temperature and the crystallization temperature of the bulk samples have been determined by Differential Scanning Calorimetric analysis performed under non-isothermal conditions. The dark electrical conductivity of the $Se_{80-x}Sb_{20}Ag_x$ ($8 \le x \le 14$) thin films has been measured as a function of temperature and has been found to increase with increase in temperature as well as with increase in Ag content. It has been found to be activated over the entire temperature range (273–333 K). The activation energy for electrical conduction and the optical bandgap are found to decrease with increase in Ag content and follow the same trend. It has been observed that the electrical activation energy is nearly half of the optical bandgap indicating that there are trap states in the gap that pins the Fermi level.
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Keywords: Chalcogenide; Differential Scanning Calorimetry; Glass transition; Activation energy

1. Introduction

Chalcogenide glassy semiconductors have several useful properties that can be employed in various solid-state devices. They show a continuous change in physical properties with change in composition [1]. Many researchers have studied the structure, electrical properties, photoconductivity, glass formation and crystallization kinetics of the glassy system [2–4]. Impurity effects in chalcogenide glasses have importance in fabrication of glassy semiconductors. Several workers have reported the impurity effect in Se-based chalcogenide glasses.

The aim of the present work is to investigate the structural and transport properties of $Se_{80-x}Sb_{20}Ag_x$ ($8 \le x \le 14$) chalcogenide glasses. We have chosen Sb as the first additive material as it has been reported that addition of small percentage of Sb is sufficient to cause fast crystallization of Se [5] which is a useful property for phase change optical recording material. Ag has been used as a second additive due to the ionic nature of conduction [6,7]. At present, considerable research has been focused on mixed electronic–ionic conductors. This is associated with the specific feature of mutual effect of the electron-hole and ionic mechanisms of charge transfer [8].

2. Experimental details

Bulk samples of $Se_{80-x}Sb_{20}Ag_x$ ($8 \le x \le 14$) were prepared by melt quenching technique. High-purity (99.99%) elements were weighed according to their atomic percentage and sealed in a quartz ampoule (length ~10 cm and internal diameter ~0.6 cm), in a vacuum of $\sim 10^{-5}$ mbar. The ampoule was kept in a vertical furnace for 48 h. The temperature was raised up to 1273 K, at a rate of 4–5 K/min. The ampoule was inverted at regular intervals (~1 h) to ensure homogenous mixing of the constituents, before quenching in an ice bath. The material was separated from the quartz ampoule by dissolving the ampoule

* Corresponding author.
E-mail address: rthangaraj@rediffmail.com (R. Thangaraj).

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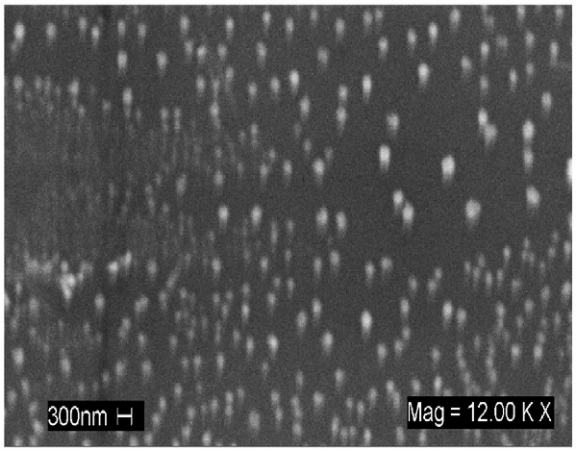

Fig. 1. SEM image of $Se_{80}Sb_{20}$ thin film showing Sb_2Se_3 nanophase.

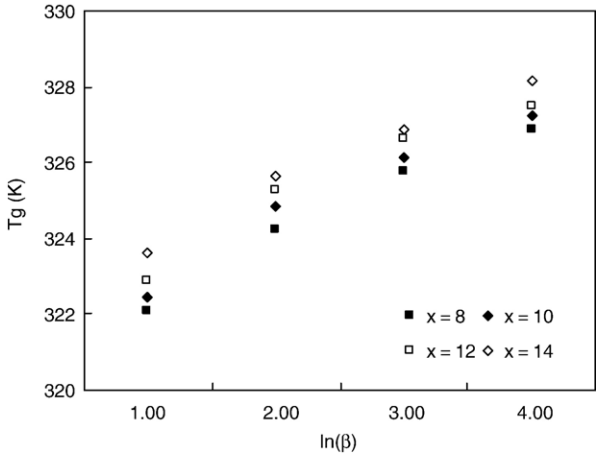

Fig. 3. Variation of T_g with ln β for $Se_{80-x}Sb_{20}Ag_x$ (x=8, 10, 12, 14).

into a solution of $HF+H_2O_2$ for approximately 48 h. Using this as source material, thin films were deposited onto well-degassed glass substrates by thermal evaporation technique in a vacuum better than 10^{-5} mbar using a Hind High Vacuum coating unit (model 12A4D). The thickness of the films was measured by Tolansky interference method [9]. Thickness was found to be ~2300 Å. The amorphous nature of the thin films was confirmed by taking XRD scans using X-ray diffraction technique (Model: Philips PW1610, Goniometer: Philips1710, detector CuK_α). The surface microstructure of the films was examined using a scanning electron microscope (SEM). DSC runs were carried out using Mettler Toledo Star^e instrument. DSC thermograms for all the compositions were taken at different heating rates (β) viz. 5,10,15,20 K/min. In all the scans we get three peaks first endothermic peak is glass transition temperature (T_g), exothermic peak is of crystallization temperature (T_c) and the third one is melting temperature peak (T_m). The glass transition temperature is defined as the temperature corresponding to the intersection of two linear portions adjoining the transition elbow of DSC trace of first endothermic peak as shown in Fig. 2. The T_c is taken as the temperature corresponding to the onset of crystallization. The conductivity measurements were carried out in the temperature range 273–333 K in a running vacuum of 10^{-3} mbar. Electrical contacts with an electrode gap of ~2 mm in a coplanar geometry were made using silver paint. The current was measured using a digital Pico ammeter (DPM-111 Scientific Equipments, Roorkee). The ohmic nature of the contact was verified by straight line passing through the origin of voltage versus current plot. The optical transmission spectrum was recorded at room temperature for all samples using UV–visible spectrophotometer (UV-160A Shimadzu, Japan) in the wavelength range 300–1100 nm. The absorption coefficient, α was calculated using the relation $\alpha=(1/t)*\ln(100/T)$, where t is the thickness of the films and T is the percentage transmission.

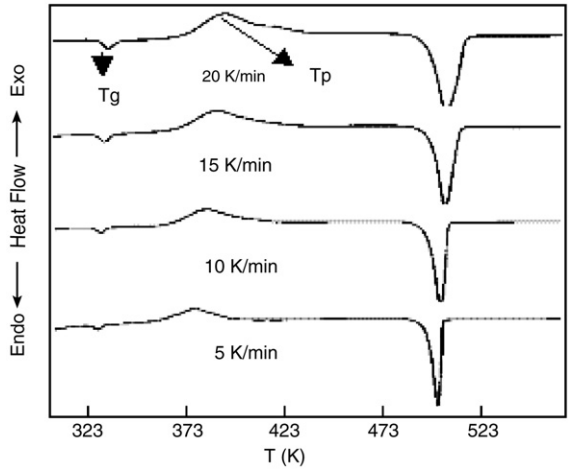

Fig. 2. DSC thermogram for $Se_{72}Sb_{20}Ag_8$ at different heating rates.

3. Results and discussions

The presence of only broad features and the absence of any sharp peaks in the X-ray diffractograms indicate that the samples are amorphous in nature. The surface morphology of the samples was examined using SEM. The scanning electron micrograph of $Se_{80}Sb_{20}$ film is shown in Fig. 1. The SEM picture shows the presence of nanoparticles embedded in an amorphous background. The presence of this nanophase in films with x=0

Table 1
The values of A and B along with E_t and E_c at a heating rate of 15 K/min for $Se_{80-x}Sb_{20}Ag_x$ (x=8, 10, 12, 14) glassy system

Alloy	A (K)	B (K)	E_t (kJ mol^{-1}) Kissinger	E_c (kJ mol^{-1}) Kissinger / Matusita
$Se_{72}Sb_{20}Ag_8$	315±1	3.66±0.4	238±0.5	114±4.5 116±1.0
$Se_{70}Sb_{20}Ag_{10}$	316±1	3.50±0.4	240±0.8	111±3.4 113±1.0
$Se_{68}Sb_{20}Ag_{12}$	317±1	3.34±0.3	342±0.7	109±2.0 111±0.8
$Se_{66}Sb_{20}Ag_{14}$	319±1	3.22±0.4	245±0.4	98±1.5 110±1.1

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