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Dielectric parameters in Se₇₀Te₃₀ and Se₇₀Te₂₈Zn₂ chalcogenide glasses

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Abstract

Temperature and frequency dependence of dielectric constant (ϵ') and dielectric loss (ϵ'') are studied in glassy Se₇₀Te₃₀ and Se₇₀Te₂₈Zn₂. The measurements have been made in the frequency range (8–500 kHz) and in the temperature range 300 to 350 K. An analysis of the dielectric loss data shows that the Guintini's theory of dielectric dispersion based on two-electron hopping over a potential barrier is applicable in the present case.

No dielectric loss peak is observed in glassy Se₇₀Te₃₀. However, such loss peaks exist in the glassy Se₇₀Te₂₈Zn₂ in the above frequency and temperature range. The Cole–Cole diagrams have been used to determine some parameters such as the distribution parameter (α), the macroscopic relaxation time (τ_0), the molecular relaxation time (τ) and the Gibb's free energy for relaxation (ΔF). © 2008 Elsevier B.V. All rights reserved.

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1. Introduction

The study of dielectric behaviour of chalcogenide glasses is expected to reveal structural information, which in effect can be useful for the understanding of the conduction mechanism as well. In addition, a study of temperature dependence of dielectric permittivity particularly in the range of frequencies where dielectric dispersion occurs can be of great importance for the understanding of the nature and origin of losses occurring in these materials.

Most of the dielectric measurements for chalcogenide glasses have been reported at audio and/or radio frequencies. Literature survey of the dielectric measurements shows that relatively very few measurements have been reported on these glasses as compared to electrical properties. It is also interesting to note the diversity in findings reported by different investigators for the same material [1–15].

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The dielectric relaxations are important to understand the nature and origin of the dielectric losses, which in turn may be useful to determine the nature of defects in solids. Measurements of the dielectric properties of different substances have been the subject of many researches and various models have been proposed to interpret the experimental results [1-15].

In view of the above, we have made dielectric measurements in glassy $Se_{70}Te_{30}$ and $Se_{70}Te_{28}Zn_2$ alloys to study the effect of Zn incorporation in binary $Se_{70}Te_{30}$ alloy on dielectric behaviour.

The reason for the selection of Zn as chemical modifier in Se–Te system is based on its attractive and important applications in chalcogenide glasses [16–21]. Like silver, Zn can also be used for photo-doping of chalcogenide glassy semiconductors [16–19]. There are reports of successful doping of $ZnSe_xTe_{1-x}$ in the literature [20] that results in the development of light-emitting diodes and lasers. Development of $ZnSe_xTe_{1-x}$ p-type contacts for highefficiency tandem structures has been recently reported [21] for the tandem solar cell devices, which have been under

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development in various configurations because of their promise of high efficiency.

2. Experimental details

2.1. Preparation of glassy alloys

Glassy alloys of $Se_{70}Te_{30}$ and $Se_{70}Te_{28}Zn_2$ were prepared by the quenching technique. High-purity Se, Te and Zn materials (5 N) were weighed according to their atomic percentages, and were sealed in quartz ampoules (length ~5 cm and internal diameter ~8 mm), with a vacuum ~10⁻⁵ Torr. The sealed ampoules were kept inside a furnace where the temperature was raised to 1000 °C, at a rate of 3–4 °C/min. The ampoules were frequently rocked for 10 h at the maximum temperature to make the melt homogeneous. Quenching was done in ice water and the glassy nature was verified by X-ray diffraction (XRD). The XRD pattern of $Se_{70}Te_{28}Zn_2$ alloys is shown in Fig. 1. Absence of any sharp peak confirms the glassy nature of the sample. Similar XRD pattern was obtained for the binary alloy.

The glassy alloys thus prepared were ground to a very fine powder and pellets (diameter $\sim 10 \text{ mm}$ and thickness $\sim 1 \text{ mm}$), which were obtained by compressing the powder in a die at a load of 5 ton. The pellets were coated with vacuum-evaporated indium film to ensure good electrical contact between the sample and electrodes.

2.2. Dielectric measurements

The pellets were mounted in between two steel electrodes of a metallic sample holder for dielectric measurements. The temperature measurement was facilitated by a copper–constantan thermocouple mounted very near to the sample. A vacuum of $\sim 10^{-2}$ Torr was maintained over the entire temperature range (300–350 K). The temperature dependence of ε' was studied in a heating run at a heating rate of 1 K/min. Dielectric measurements were made using a digital LCR meter. The parallel capacitance and dissipation factor were measured and then ε' and ε'' were



Fig. 1. XRD pattern of the glassy Se₇₀Te₂₈Zn₂ alloy.

calculated. Three terminal measurements were performed to avoid stray capacitances. We preferred to experiment on the pellet rather than the bulk as macroscopic effects (gas bubbles, etc.) may appear in the bulk during preparation. It has been shown by Goyal et al. [2], both theoretically and experimentally, that bulk ingots and compressed pellets exhibit similar dielectric behaviour in chalcogenide glasses for the suspected inhomogeneities in case of compressed pellets in these materials.

3. Results

Guintini et al. [22] have proposed a model for dielectric dispersion in chalcogenide glasses. This model is based on the Elliott's idea [23] of hopping of charge carriers over a potential barrier between charged defect states (D^+ and D^-). Each pair of site (D^+ and D^-) is assumed to form a dipole which has a relaxation time depending on its energy [24,25]; the latter can be attributed to the existence of a potential barrier over which the carriers hop [26].

According to the above model [22], ε'' at a particular frequency, in the temperature range where the dielectric dispersion occurs, is given by

$$\varepsilon''(\omega) = (\varepsilon_0 - \varepsilon_\infty) 2\pi^2 N (ne^2/\varepsilon_0)^3 k T \tau_0^m W_m^{-4} \omega^m \tag{1}$$

Here, m is a power of angular frequency and is negative in this case and is given by

$$m = -4kT/W_{\rm m} \tag{2}$$

n is the number of electrons that hop, *N* is the concentration of localized sites, ε_0 and ε_{∞} are the static and optical dielectric constants, respectively, $W_{\rm m}$ is the energy required to move the electron from a site to infinity.

It has been reported [1-15] that, in chalcogenide glasses, the temperature dependence of ε' and ε'' is appreciable only in a certain temperature range. At lower temperatures, ε' is almost constant and ε'' is negligibly small. After a certain temperature, ε' and ε'' increase appreciably with temperature. Therefore, the present measurements have been made only in the high-temperature region where dielectric dispersion is quite appreciable. The temperature dependence of ε' and ε'' is studied at various frequencies (8–500 kHz) for all the glassy alloys studied here. The glass transition temperature of the alloys used is around 350 K; hence the temperature range of measurements has not been more than 350 K.

3.1. Temperature dependence of dielectric constant (ε') and dielectric loss (ε'')

Figs. 2 and 3 show the results of the dielectric constant (ε') and the dielectric loss (ε'') measurements at different frequencies for glassy Se₇₀Te₃₀ and Se₇₀Te₂₈Zn₂ alloys below the glass transition temperature. From Figs. 2 and 3, it is clear that the temperature dependence of ε' and ε'' is appreciable in the glass transition region. In these glassy samples, dielectric constant increases with the increase in

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