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THERMAL AND DIELECTRIC STUDIES ON $Ge_{10}Se_{69}Tl_{21}$ CHALCOGENIDE GLASS

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Bulk $Ge_{10}Se_{69}Tl_{21}$ chalcogenide glass is prepared by melt quenching technique. Thermal analysis of bulk $Ge_{10}Se_{69}Tl_{21}$ glass has been undertaken using temperature modulated Alternating Differential Scanning Calorimetry (ADSC). The $Ge_{10}Se_{69}Tl_{21}$ glass is found to exhibit single glass transition temperature (T_g) and double stage crystallization reactions (T_{c1} & T_{c2}). The dependence of dielectric properties such as dielectric loss tangent ($\tan\delta$), dielectric constant (ε') and dielectric loss factor (ε'') on the frequency has been studied at the room temperature in the frequency range 10 kHz to 5 MHz. The dielectric parameters $\tan\delta$, ε' and ε'' are found to decrease with the increase in the frequency. Further, resistance of the $Ge_{10}Se_{69}Tl_{21}$ sample is also found to decrease with the increase in the frequency.

Keywords: *CHALCOGENIDE GLASS, THERMAL ANALYSIS, ALTERNATING DIFFERENTIAL SCANNING CALORIMETRY, DIELECTRIC PROPERTIES, DIELECTRIC CONSTANT, DIELECTRIC LOSS FACTOR.*

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1. INTRODUCTION

Chalcogenide glasses of metal containing Ge-Se system continue to attract a wide range of interest of the researchers as one of the main canonical systems for fundamental and applied research of vitreous semiconductors [1-3]. Potential applications of these materials cover a broad area of electronics, optoelectronics, photonics and telecommunication technologies. The feasibility of technological applications has stimulated the interest to investigate new chalcogenide glass compositions.

Differential Scanning Calorimetry (DSC) has proven to be a useful tool to characterize the thermal properties of different materials including glasses [4, 5]. In particular, the Temperature Modulated Differential Scanning Calorimetry (TMDSC) is being used substantially in the recent times to study the glass-transition behavior [6-9] and other physical and thermodynamic properties [10, 11] of glasses. This technique has allowed detailed probing of the nature of the glass transition and self-organization of molecular networks [12]. Alternating differential scanning calorimetry (ADSC) is a technique that operates analogous to TMDSC, working on the principle of superimposing a temperature modulation on a constant heating/cooling rate.

The binary Ge-Se is one of the most studied chalcogenide glassy systems and it has been established that the addition of thallium to binary Ge-Se is accompanied by a marked change in their structural and physical properties. Bulk Ge-Se-Tl glasses have attracted attention for their potential applications in acousto-optical devices [13, 14]. The study of dielectric behavior in chalcogenide glasses is likely to reveal structural information and is important to understand the nature and the origin of dielectric losses, which in turn, may be useful in the determination of structure and defects in these glasses.

In the present work, bulk $Ge_{10}Se_{69}Tl_{21}$ chalcogenide glass is prepared by melt quenching technique and its thermal properties have been studied using ADSC. Further studies on the, dielectric behavior of $Ge_{10}Se_{69}Tl_{21}$ glass, particularly in the frequency region 10 kHz to 5 MHz, has been undertaken.

2. EXPERIMENTAL DETAILS

Bulk $Ge_{10}Se_{69}Tl_{21}$ chalcogenide glass is prepared by melt quenching technique. Appropriate quantities of high purity constituent elements are weighed and transferred in to quartz ampoule, evacuated to 10^{-5} Torr and sealed. The ampoule is heated in a rotary furnace to a temperature of about 980 °C and maintained at this temperature for 24 hours. The ampoule containing the melt is rotated at 10 rpm to homogenize the melt. The homogenizing temperature is chosen well above the melting points of the constituent elements. After homogenizing, ampoule is subsequently quenched in a mixture of ice water and NaOH to obtain bulk glassy sample. The amorphous nature of the quenched sample is confirmed by X-ray diffraction studies.

ADSC experiment is carried out using a Mettler Toledo 822^c DSC instrument with the aid of STAR^e software. Temperature calibration is achieved using indium and zinc as standards, and the heat flow calibration is done using indium. A blank run is made prior to the actual measurements, with an empty aluminum pan on the reference side and an empty pan with a lid on the sample side, to calibrate the heat flow signal, to correct the amplitude and to eliminate cell asymmetry. The sample is polished into small piece, weighed (~ 15 mg) and sealed in aluminum pan. An empty aluminum pan is used as a reference. Experiment is undertaken in the temperature range of 30-300 °C, at the scanning rate of 3 °C/min and a modulation rate of 1/60 Sec. Argon is used as the purge gas at a flow rate of 80 ml/min.

Dielectric measurements were carried out at room temperature using impedance analyzer model PSM – 1735 (programmable) in the frequency range 10 kHz - 5 MHz.

3. RESULTS AND DISCUSSIONS

Figure 1 shows the X-ray diffraction pattern of $Ge_{10}Se_{69}Tl_{21}$ glass, which confirms the amorphous nature of the as quenched sample.

Figure 2 shows the total heat-flow curve obtained using ADSC for $Ge_{10}Se_{69}Tl_{21}$ glass. This result indicate the presence of a single glass transition temperature T_g at 86.29 °C and two distinct exothermic crystallization peaks

T_{c1} & T_{c2} at 163.83 °C and 192 °C respectively. The exothermic crystallization peak represents the glass-crystal transformation. The appearance of two distinct crystallization peaks indicates phase separation occurring in this glassy alloy.

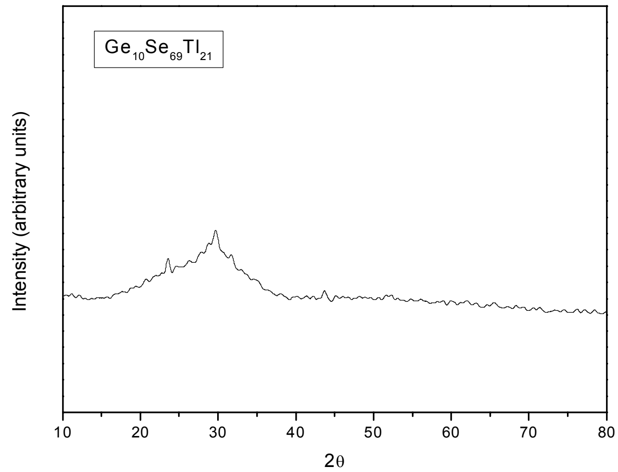

Fig. 1 *– X-ray diffraction pattern of as prepared* $Ge_{10}Se_{69}Tl_{21}$ *glass*

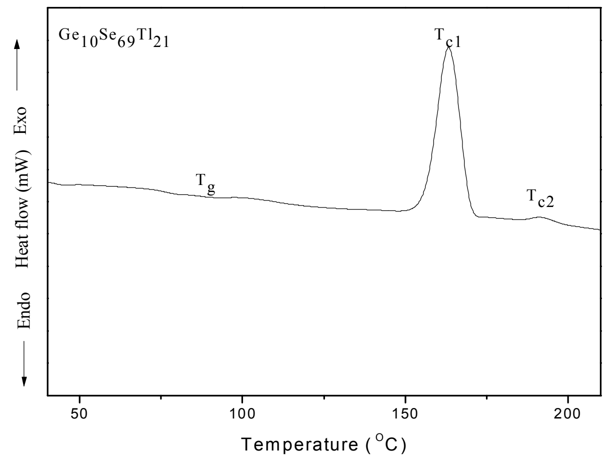

Fig. 2 *– Total Heat-flow curve of* $Ge_{10}Se_{69}Tl_{21}$ *glass*

Figure 3 displays the variation of dielectric loss tangent (tanδ), dielectric constant (ε'), dielectric loss factor (ε'') and resistance with the frequency at room temperature from 10 kHz to 5 MHz for $Ge_{10}Se_{69}Tl_{21}$ glass. The dielectric loss tangent (tanδ), dielectric constant (ε') and dielectric loss factor (ε'') are found to decrease with the increase in the frequency range 10 kHz - 5 MHz. The dielectric parameters (tanδ, ε' and ε'') are found to decrease rapidly at lower frequencies as compared to at higher frequencies. The resistance of the $Ge_{10}Se_{69}Tl_{21}$ glassy sample is found to decrease with the frequency in the frequency range 10 - 400 kHz. Further, resistance is found to be independent of frequency at higher frequencies.

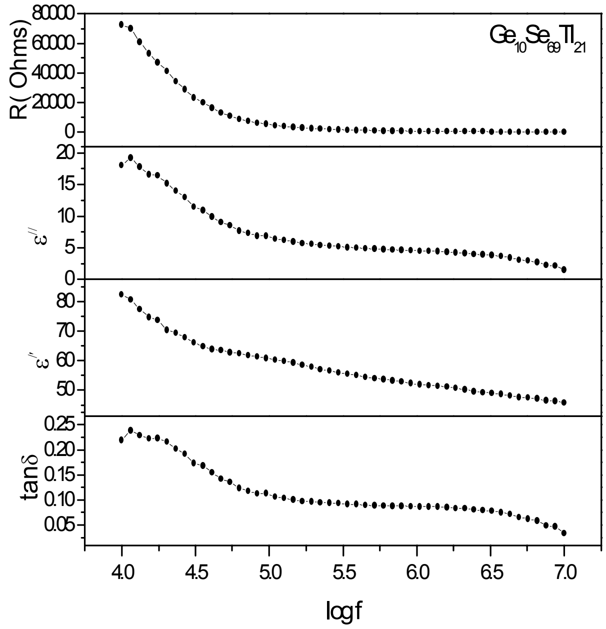

Fig. 3 *– Frequency dependence of tanδ, ε′, ε″ and resistance of $Ge_{10}Se_{69}Tl_{21}$ glass in the frequency range* 10 *kHz to* 5 *MHz*

The different behavior of dielectric parameters at low and high frequencies may be attributed to the fact that at lower frequencies the polarization follows the alternations of the field, whereas at higher frequencies the atomic nucleus ceases to follow the field and only the electronic polarization remains in the system. Further, at higher frequencies, dipolar and ionic contributions are small or almost negligible because of the inertia of the molecules or ions.

4. CONCLUSIONS

Thermal analysis of $Ge_{10}Se_{69}Tl_{21}$ chalcogenide glass has been undertaken using ADSC. The $Ge_{10}Se_{69}Tl_{21}$ glass is found to exhibit single glass transition temperature (T_g) and double stage crystallization reactions (T_{c1} & T_{c2}). The dielectric loss tangent (tanδ), dielectric constant (ε') and dielectric loss factor (ε'') are found to decrease with the increase in the frequency at the room temperature in the frequency range 10 kHz to 5 MHz. Observed dielectric behavior has been understood on the basis of frequency dependence of polarization in the chalcogenide glasses. Further, resistance of the $Ge_{10}Se_{69}Tl_{21}$ sample is also found to decrease with the increase in the frequency.

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