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DC conductivity of heavy metal oxide (Bi_2O_3) boro-tellurite glasses: Effect of Eu_2O_3

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Abstract

A novel investigation on structural and DC conductivity of Eu_2O_3 activated heavy metal oxide boro-tellurite (BBTE) glasses were analyzed. The boro-bismuth-tellurite glass samples doped with europium trioxide were fabricated by the conventional melt quenching method. The microstructural and structural studies of the glasses have been done by scanning electron microscope (SEM) and X-ray diffraction (XRD), respectively. The DC conductivity of the BBTE samples has been studied at the frequency range 40Hz-6MHz and in the temperature range 303-453K. The XRD and SEM, confirm the non-crystalline and homogeneous properties of prepared glasses. The DC conductivity of glasses obeys Arrhenius behavior and DC conductivity decreases with increasing Eu_2O_3 concentration. A very less amount of DC conductivity was noticed in glasses with various temperatures and it is due to less availability of oxide ions.

1. Introduction

Due to their high conductivity and high thermal stability, a significant number of investigations were committed to develop many types of transition metal ions (TMIs) based on fast ion conducting (FIC) materials. Therefore FIC compounds exhibit potential applications in solid-state ionic batteries, fuel cells, sensors, capacitors as electrolyte materials [1,2]. Glass modifiers such as Ag_2O, Cu_2O, and Ag_2S give strong exothermic chemical reactions when mixed with glasses [1-5]. Glasses having heavy metal oxides (HMO^s)like PbO, Bi_2O_3 retain outstanding and appreciated physical and chemical properties. Hence these glasses find applications in optoelectronics, optical devices, photonics [6-9]. For example; Bi_2O_3 and PbO can form stable glasses upto 85% with B_2O_3, P_2O_5, and SiO_2 because these two HMOs behave like either network modifiers (BiO_6, PbO_6) or network formers (BiO_3, PbO_4) [6-9]. In addition, glasses containing bismuth oxide are strongly suggested for significant application in thermal sensors, mechanical sensors, high energy physics, scintillation detectors for reflective windows [10,11], and more recently bone applications as bioactive glasses. [12]. Borate glasses possessing HMOs and transition metal oxides (TMO^s) reveal technological applications in mechanical sensors, optical-electronic devices, reflecting windows, thermal sensors [13-16]. Commonly, glasses containing TMO^s exhibited greater semiconducting properties [15-21]. Therefore, such glasses show important applications in modern optical and electronic devices [15,21] and electrode materials in batteries [22,23]. The TeO_2, in its pure form, cannot give glasses directly due to its conditional glass nature. But tellurite based glasses show good chemical durability, low melting point, better optical properties, and good mechanical strengths. Glass forming ability of TeO_2 can be enhanced by adding other glass formers, and among all available glass formers, boron oxide is well suited to TeO_2 due to its low melting, high thermal stability, and high glass-forming ability even with regular quenching rates [24-28]. Additionally, TeO_2-based glasses [12,13], unlike the transition metal ions (TMI), have been measured to have higher electrical conductivity than silicate, phosphate, and borate samples. Several electrical studies on glass materials have revealed that glasses do not necessarily have to be insulators but in some cases super-ionic conductors [29-31]. TeO_2 glass systems have been explored for diverse conduction studies [32,33]. The presence of a weak hopping method in glass materials containing HMOs was shown by Mogus-Milankovic *et al* [34]. Moustafa reported the conduction process and electrical properties of semiconducting iron bismuth glasses [35]. Recently, the Effect of temperature and frequency on mixed transitions metals doped semiconducting bismuth-phosphate glasses was reported by Nanao Ningthemcha *et al* [36]. They reported that the DC conductivity was is described with Mott and Greaves VRH models shows reduction of DC conductivity with the increases in MoO_3 concentration. Sunil Dhankhar *et al* reported electronic transport and relaxation studies in bismuth-modified zinc boro-tellurite glasses [37]. The effect of silver ion on transport properties were explored in boro-tellurite samples [38]. Dielectric properties were practically investigated on boro- tellurite glasses, $(B_2O_3)0.2$-$(TeO_2)0.3$-$(CoO)x$-$(Li_2O)0.5$-x (x=0.05-0.5) [39]. The effect of mixed glass has been studied in a set of lithium oxide doped boro-tellurite samples [40]. The polaronic conductivity and ionic conduction outcomes were studied for alkali boro-tellurite glasses [41] and Bi_2O_3-B_2O_3-TeO_2 [42] respectively. The eminent level application in optical fiber amplifiers, diffractometer display monitors, and planner waveguides [43,44] was reported on oxide glasses embedded with rare earth. Among available rare earth, the europium is a renowned element due to its significant optical device applications. There is not much research available on the dielectric and conductivity properties of rare-earth Eu_2O_3 doped oxide samples.

The present study aims to novel investigate of structural and DC conductivity property of the oxide samples containing two glass formers (B_2O_3 and TeO_2) by replacing heavy metal oxide Bi_2O_3 and rare earth Eu_2O_3. The DC conductivity of prepared glasses was studied by keeping Bi_2O_3 and TeO_2 constant.

2. Materials and methods

2.1 Synthesis of Glass

Concerning previous work on the function of Eu^{3+} ions on luminescence [45], thermal and structural [46] properties of boro-tellurite glasses, a new set of glass with batch compositions (65-x)B_2O_3-25TeO_2-10Bi_2O_3-xEu_2O_3, where x= 0.1, 0.2,0.3,0.4, and 0.5 mol%, and indicated as BBTE1, BBTE2, BBTE3, BBTE4, and BBTE5 respectively, were synthesis using melt quenching method. The high purity raw materials procured from Sigma Aldrich, B_2O_3, TeO_2, Bi_2O_3, andEu_2O_3 were used for preparation after mixing dried powders in the preferred weight fractions. The 50 g batches of dried and thoroughly mixed chemicals were taken in the porcelain crucibles for heating. Then crucibles were kept in an electrical high-temperature muffle furnace for heating. The chemical is heated at 1000°C for 1 h and the molten liquid is mixed at regular intervals to obtain a homogeneous mixture. The melt was delivered in a preheated brass mold and pressed from another brass mold for rapid cooling. The glasses were annealed at 390°C for 3 h to remove the thermal strains caused by the heating process. The obtained glasses were engraved and cut to a suitable shape and thickness for further analysis.

2.2 Characterizations

Glass samples BBTE1-5 were made powder with pestle and agate mortar to analyze the amorphous character by X-ray diffraction technique. For this analysis, a Bruker D8 focus XRD enclosing Cu-Kα radiation of wavelength 1.54 Å was employed for measurement. These measurements were employed at room temperature with 2θ range of 10° to 70° and operating voltage of 40 kV and 30 mA.

The microstructure surface analysis of the BBTE glasses was conducted at 5 kV by scanning electron microscope (SEM). The gold is coated on glasses by Bal-Tec SCD-005 sputter coater.

The electrical conductivity analysis of obtained BBTE samples was made by taking glasses in the form of a circular disc with thickness and diameter, 0.11 cm and 1cmrespectively. The surfaces of the glasses were polished with emery paper and then silver paste was coated and dried for about 7 h at 320 K. Further, samples were sandwiched between electrical leads made of silver. The electrical conductivity of glasses have been analyzed using a Precision impedance analyzer (Agilent-4294A) in which a home-built cell assembly, a Pt-Rh thermocouple, to measure temperature, placed much closed to the samples were used. At the frequency range,40Hz-6MHz, the corresponding capacitance (CP) and conductance (G) were measured at the temperature range 303 K to 453 K.

3. Results and discussion

3.1 XRD and SEM analysis

The non-crystalline nature of the prepared BBTE glasses has been studied by XRD results. X-ray diffraction patterns are shown in Figure 1. The occurrence of a broad bulge around 31° and the absence of sharp peaks can be observed. This is the characteristic property of non-crystalline materials. XRD analysis reveals the non-crystalline nature of BBTE glasses. The above non-crystalline nature is further confirmed by scanning electron microscopy (SEM). Figure 2 depicts the scanning electron micrographs (SEM) of BBTE glasses. In the SEM image, there is a plain surface with non-appearance of any microstructure. The absence of micrograph and microstructure confirms the amorphous and homogeneous nature of the prepared samples, respectively.

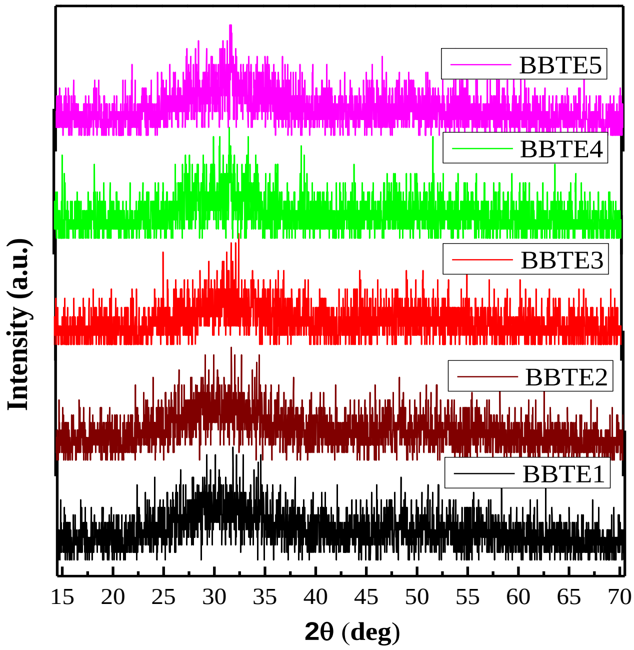

Figure 1. XRDgraph of B_2O_3-TeO_2-Bi_2O_3-Eu_2O_3 glasses.

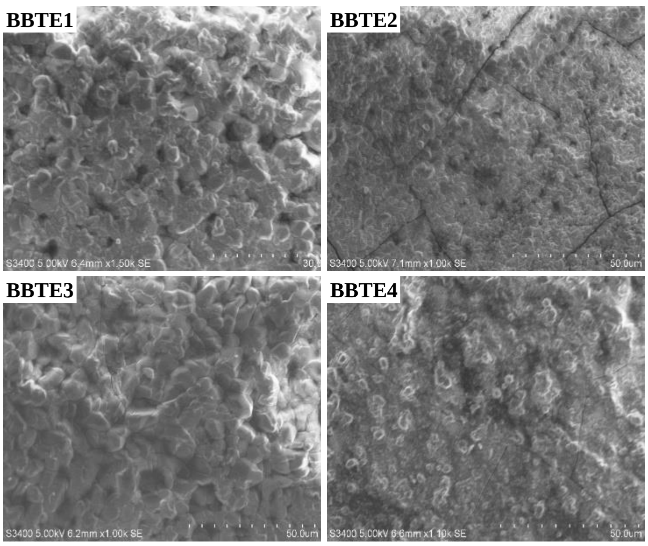

Figure 2. SEM micrographs of B_2O_3-TeO_2-Bi_2O_3-Eu_2O_3 glasses.

3.2 DC conductivity

The measurement of DC conductivity of glass samples cannot be made directly, not only due to the effects of polarization on the sample-electrode interface and also due to practical difficulties in finding suitable electrodes. These types of problems can be determined by the AC impedance spectroscopy method [47]. The capacitance (C_p) and conductance (G) were measured for all the prepared glass samples in the temperature and frequency range 303 K to 453K and 40 Hz to 6 MHz. Using the mathematical relationships (1), (2), and (3) and obtaining the conductivity and retention data, the real(Z′) and imaginary (Z″) parts of the complex impedance are estimated [13].

$$Z^* = Z' + JZ'' = \frac{1}{G + J\omega C_P} \quad (1)$$

$$Z' = \frac{G}{G^2 + \omega^2 C_P^2} \quad (2)$$

$$Z'' = \frac{\omega C_P}{G^2 + \omega^2 C_P^2} \quad (3)$$

Where ω denotes the angular frequency.

The DC conductance is calculated by semicircular complex impedance (Z" versus Z') plots. The intersection values taken at the low-frequency end on the Z′ axis give the residual impedance and the DC conductivity was estimated using the mathematical equation (4).

$$\sigma_{DC} = \frac{1}{R}\frac{d}{A} \quad (4)$$

Here, R indicates the intersection taken along the Z′ axis in the Cole-Cole plot, and reciprocal R is taken as residual resistance. In equation (4), A and d denotes the area and thickness of the glass respectively [13]. The Cole-Cole plot of BBTE4 glass is illustrated in Figure 3. It has been observed that the semicircle over the entire range of temperatures studied shows a typical characteristic nature of the conduction mechanism [48]. Furthermore, with increasing temperatures, the semicircle intersection points shift towards lower Z′ values for all BBTE glass samples and therefore DC conductivity is a thermally activated process. The semicircles in the impedance plot are well defined and indicates non-Debye type relaxation mechanism, because centers of the semicircle lies below Z' axis [33-35,39,46]. From the measured DC conductivity values, it is noted that the conductivity values decrease with the increase of Eu_2O_3 concentration because the glasses lose their conductivity. In Table 1, the calculated conductivity values and DC activation energy values (E_{DC}) of all prepared glass samples are recorded. The temperature-dependent DC conductivity glasses were shown in the form of log (σ_{DC}) against 1000/T plot as shown in Figure 4. A single linear variation of log (σ_{DC}) versus 1000/T was observed in all the samples. From Figure 4 it is assumed that the DC conductivity is increasing with the increase of temperature which confirms the Arrhenius behavior [48,49].

$$\sigma_{DC} = A\exp^{\left(\frac{-E_{DC}}{kT}\right)} \quad (5)$$

Here, k is Boltzmann's constant; E_{DC} is the activation energy for the DC conductivity, and A is the pre-exponential factor. The activation energy for electrical conduction is estimated by using data of each sample by fitting it to least-square fit to a straight line. For BBTE glasses, the DC activation energies, E_{DC} were determined using the slopes of the log (σ_{dc}) against 1000/T plot. The obtained E_{DC} values concerning the different concentrations of Eu_2O_3 are depicted in Figure 5. It is seen from Figure 4 and 5 that, the DC conductivity decreases gradually with a considerable increase in activation energy values hence it confirms the dependency of conductivity with temperature. The obtained values of activation energy and conductivity indicate a decrease in conductivity due to the low presence of oxide ions in the glass [50-52]. Structural modification in the glass network with the addition of Eu_2O_3 and the reduction of the molar concentration of Bi_2O_3 may result in lower oxide ions [52].

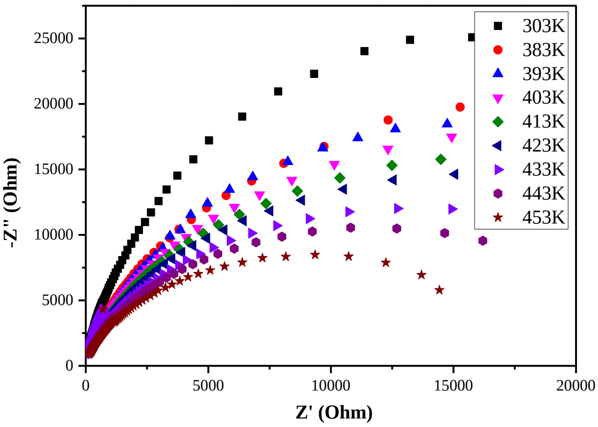

Figure 3. Semicircular complex impedance graph of BBTE4 sample at various temperatures.

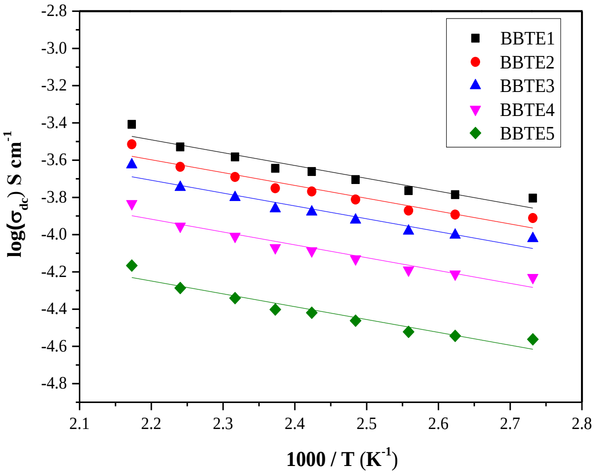

Figure 4. The log (σDC) against 1000/T plot of B_2O_3-25TeO_2-10Bi_2O_3-Eu_2O_3 glasses.

Table 1. DC conductivity (σ_{DC}) DC activation energy (E_{DC}) of B_2O_3-TeO_2-Bi_2O_3-Eu_2O_3samples.

Sample Name	(σ_{DC}) on 453K ($S \cdot cm^{-1}$)	E_{DC} (eV)
BBTE1	1.29×10^{-4}	0.907
BBTE2	9.31×10^{-5}	0.923
BBTE3	7.28×10^{-5}	0.937
BBTE4	5.37×10^{-5}	0.949
BBTE5	2.62×10^{-5}	0.975

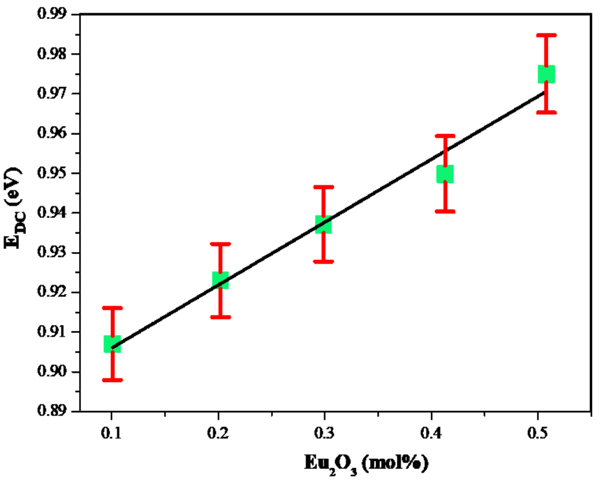

Figure 5. A plot of DC activation energy and Eu_2O_3 (mol%) of B_2O_3-TeO_2-Bi_2O_3-Eu_2O_3 glasses.

Moreover, the decrease in DC conductivity is may be owing to the sitting of Eu^{3+} ions on the lattice sites of TeO_2/Bi_2O_3. This is due to relatively heavy mass of Eu^{3+} ions than mass of TeO_2/Bi_2O_3. Hence conductivity of glasses decreases and corresponding activation energy increases with increasing Eu_2O_3 concentration. Though the considerable and less amount of conductivity was found in individual glasses concerning the addition of Eu_2O_3 it could be owing to the presence of oxide ions TeO_2/Bi_2O_3. From Figure 5, the existence of the linear reliance among the reciprocal of the absolute temperature and log of DC electrical conductivity was noticed, generally, this is the characteristic nature of ionic conductivity, in which the activation energy does not vary with temperature. In addition, if the conductivity present in glasses owing to polarons then activation energy differs with temperature, and the conduction due to the polarons is a distinct dependence on the activation energy.

4. Conclusions

Structural and DC conductivity of Boro-bismuth-tellurite samples prepared by melt quenching process were studied: impact of Eu_2O_3. The non-crystalline structure of glasses was confirmed with X-ray diffraction. The homogeneity and amorphous nature of the prepared glasses were shown by scanning electron microscopy. Analysis of DC conductivity and Arrhenius behavior for prepared glasses has been performed in the temperature range 303 K to 453 K and frequency range 40 Hz to 6 MHz.It is depicted from the obtained values that the DC conductivity of the glasses decreases with the reduced mobility of the oxide ions due to the large mass of Eu^{3+} ions. Furthermore, the values of DC conductivity in glass indicate the temperature-dependent ionic conductivity behavior. These glasses can therefore be used for the development of solid-state device applications.

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