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RESEARCH ARTICLE

DIELECTRIC PROPERTIES AND CONDUCTIVITY STUDIES OF CUO DOPED BOROTELLURITE GLASSES

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ABSTRACT

CuO doped borotellurite glasses, $(B_2O_3)_{0.2}$-$(TeO_2)_{0.8-x}$-$(CuO)_x$, where x = 0.1, 0.2, 0.3, 0.4 and 0.5 were prepared by standard melt quenching method and their non-crystalline nature was confirmed by XRD studies. Dielectric properties as a function of frequency and temperature over the wide ranges have been measured. Observed decrease in dielectric constant and loss with increase in frequency and temperature has been ascribed to the decrease in ionic contribution. Dielectric parameters increased with increase in CuO content. Conductivity was estimated from dielectric data. The high temperature electrical conductivity has been analyzed using Mott's small polaron model and activation energy was determined. Activation energy decreased and conductivity increased with increase in CuO content. Conductivity also increased with increase in frequency. Frequency exponent of conductivity is found to be temperature dependent, and this contradicted Quantum mechanical tunneling (QMT) model's prediction. It is for the first time that CuO doped borotellurite glasses have been investigated for frequency dependent dielectric properties and ac conductivity and data analysed thoroughly.

INTRODUCTION

Tellurite glasses are considered to be the best optical materials as they have low melting temperatures and high dielectric constant (El-Damrawi, 2001). Oxide glasses containing transition metal ions such as, Fe, Co, V, Cu etc. exhibit semiconducting nature (Murawski *et al.,* 1979; Sayer and Mansingh, 1972). The glasses, xNa_2O – (1-x) B_2O_3 were investigated for frequency dependent conductivity in the temperature range 350K to 650K. At lower frequency conductivity was constant. Activation energy for conduction decreased with increasing Na content (Roling *et al.,* 1997). In (xFe_2O_3 – (70-x) V_2O_5 – 30 B_2O_3) glasses, conductivity decreased with increase in Fe_2O_3 content and increased with temperature (Virender Kundu *et al.,* 2008). V_2O_5 doped borate glasses were investigated for conductivity as a function of frequency and temperature. At low temperature, ac conductivity was found to be consistent with Quantum Mechanical Tunneling (QMT). Correlated Barrier Hopping (CBH) mechanism has been used to explained temperature dependence of frequency exponent at high temperature (Ashwini Ghosh, 1990-I; Ashwini Ghosh, 1990-II). The frequency dependent conductivity studies were reported for the glasses, x Li_2O – (1-x) V_2O_5 –2TeO_2, and $(V_2O_5)_{0.4}$ – $(Li_2O)_x$ – $(TeO_2)_{0.6-x}$ and $(V_2O_5)_{0.25}$ – $(Li_2O)_{0.25}$ – $(K_2O)_x$ – $(TeO_2)_{0.5-x}$, (Montani and Giusia, 2001; Prashant Kumar *et al.,* 2009).

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In both the sets of glasses, the conductivity increased with increase in frequency and decreased with increasing alkali content. Tellurite glasses doped with single and mixed TMI, $(V_2O_5)_x$ – $(TeO_2)_{1-x}$ and $(V_2O_5)_{0.4}$ – $(CoO)_x$ – $(TeO_2)_{0.6-x}$, have been studied for dielectric properties in the frequency range from 50 Hz to 5MHz and temperature range from 300 to 500K. The temperature dependence of ac and dc conductivities were analyzed using Mott's small polaron hopping model and the activation energies were determined. The activation energies were reported to be in the range 0.292 – 0.499 eV (Sankarappa *et al.,* 2008).

Electrical studies of single alkali and mixed alkali borotellurite glasses have been reported (Shaw and Ghosh, 2014; Harish Bhat *et al.,* 2004). Mixed alkali borotellurite glasses showed up MAE in terms of its ac conductivity. Silver ions doped borotellurite glasses have also been studied for conductivity studies (Sujatha *et al.,* 2010; Ramesh Kumar *et al.,* 2014) and noted that dielectric parameters decreased with increase in frequency and conductivity increased and activation energy decreased with increase in mole fractions of Ag_2O. It is noticed that there are no reports on dielectric and ac conductivity studies in CuO doped borotellurite glasses though it is reported that borotellurite glasses are chemically more durable and thermally stable (Mahraz *et al.,* 2014). CuO doped borotellurite glasses are ideal systems to investigate frequency and temperature dependent electrical properties as and probe conduction mechanism they are relatively clean containing only polarons to conduct.

MATERIALS AND METHODS

The copper doped boro-tellurite glasses, $(B_2O_3)_{0.2}$ $(TeO_2)_{0.8-x}$ $(CuO)_x$, where x = 0.1, 0.2, 0.3, 0.4 and 0.5, labeled as BTCU1, BTCU2, BTCU3, BTCU4 and BTCU5 were prepared by standard melt quenching technique using AR grade chemicals, H_3BO_3, TeO_2 and CuO. The raw materials were mixed up in the desired proportions and thoroughly ground. Mixer was taken in a silica crucible and heated in a SiC make furnace up to melting. The melt was maintained at 1500K for an hour and then quenched to room temperature by pouring onto SS plate and covering it up with another SS plate. The samples thus obtained were annealed at 623K for 12 hours to remove thermal strains in them. Non-crystallinity of the samples was confirmed by XRD studies.

The samples of thicknesses in the range from 2 to 3mm and cross sectional areas ranging from 20 to 80mm^2 were selected for dielectric measurement. Two large surfaces of the glasses were silver painted. Using Wayne Kerr (UK) make (Model - 6500B) Precision Impedance analyzer frequency dependent measurements of capacitance, C, and dissipation factor, tan δ, were measured for frequencies in the range 50 Hz to 5 MHz and temperature from 325 to 600K. The dielectric constant (ε′), dielectric loss factor (ε″) and ac conductivity (σ_{ac}) were determined as per the expressions given in (Sankarappa *et al.,* 2008; Shaw and Ghosh, 2014; Harish Bhat *et al.,* 2004; Sujatha *et al.,* 2010).

RESULTS AND DISCUSSION

Dielectric properties

The measured dielectric constant, ε′, varied in the range 6 to 7 and the dielectric loss, ε″, varied in the range 5 to 11. The typical plots of the variation of ε′ with frequency at different temperatures for BTCU2 glass is shown in Fig.1. Variation of ε″ is shown as inset to Fig.1 for the same glass. Similar nature of variation has been observed for the remaining glasses in the present series.

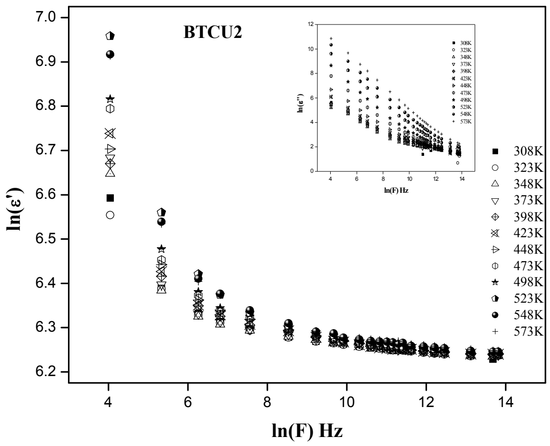


Fig.1. Plots of ln(ε′) versus ln(F) for the glass BTCU2 at different temperatures. Inset shows the frequency dependence of ε″ for the same glass

It found that both ε′ and ε″ decreases with increase in frequency and increases with increase in temperature for all the present glasses. These measured ε′ and ε″ values are in agreement with similar oxide glasses (Prashant Kumar *et al.,* 2009; Sankarappa *et al.,* 2008; Sujatha *et al.,* 2010). The increase in dielectric constant with increase in temperature can be accounted for decrease in bond energies (Bhagat and Abou-Zeid, 2001). Also, as the temperature increases the dipolar polarization weakens the intermolecular forces and thereby enhances orientational vibration (Prashant Kumar *et al.,* 2009; Sankarappa *et al.,* 2008). Decrease in ε′ and ε″ with increase in frequency can be due to decrease in ionic and orientational contributions to the polarizability with increase in frequency (Sankarappa *et al.,* 2008).

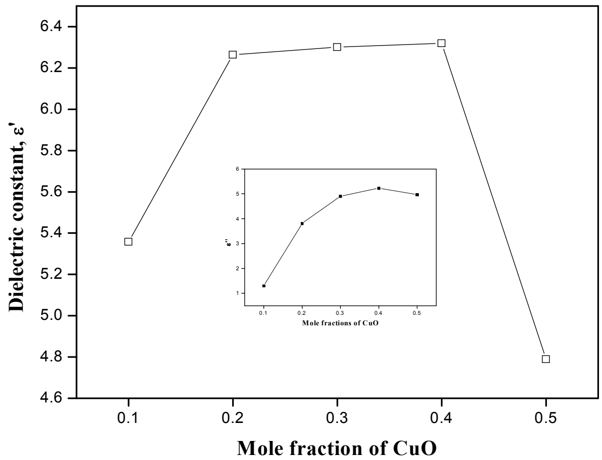


Fig.2. Variation of ε′ with CuO content at 500 kHz and temperature 573K. Inset shows variation of ε″ with CuO

Fig.2. shows change in dielectric constant, ε′ at a frequency of 500 kHz and temperature of 573K. Change in ε″ with CuO is shown as an inset of Fig.2. Both ε′ and ε″ are increasing with increase in CuO content. This may be due to the fact that as CuO content is increased, more number of ions and polarons which contribute to total polarization in the system increases. Therefore, dielectric parameter increases with CuO content.

Conductivity

Conductivity has been estimated using an expression, $\sigma = \varepsilon''\varepsilon_0\omega$, where ε_0 is the permittivity of free space (Prashant Kumar *et al.,* 2009; Sankarappa *et al.,* 2008; Sujatha *et al.,* 2010). Total conductivity calculated from dielectric data is the sum of dc and ac components. These two components were extracted by fitting to $\sigma_{Total} = \sigma_{dc} + A\omega^s$, where A is a constant and *s* is frequency exponent (Sankarappa *et al.,* 2008; Sujatha *et al.,* 2010). The nonlinear plots of ln(σ_{Total}) versus ln(ω) are shown for BTCU2 glass for different temperatures. Similar fits were achieved to the data on other glasses of the present series. Note that plots are shown on ln-ln scale in Fig.3 to show the quality of fits. It can be seen that the conductivity increases with increase in frequency and increase in temperature.

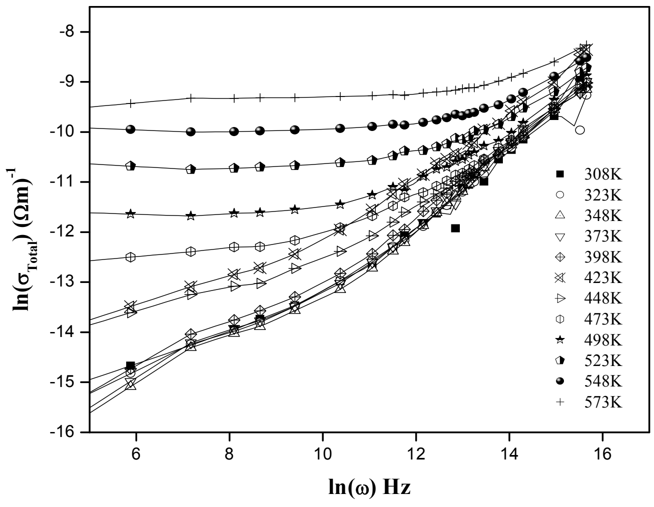


Fig.3. The plots of total conductivity, ln(σ_{Total}) versus ln(ω) for BTCU2 glass. Solid lines are the best fits to $\sigma_{Total} = \sigma_{dc} + A\omega^s$

The frequency exponent, *s,* obtained for the present glasses in the studied range of temperature, is in the range of 0.57 and 0.96. These *s* values indicate that the carrier transport is due to hopping of polarons (Sankarappa *et al.,* 2008). Both ac and dc conductivity increased with increase in temperature, which reflects on the semiconducting nature of the samples. Also, both the conductivity increased with increase in CuO concentration. Here only ac component is considered for analysis.

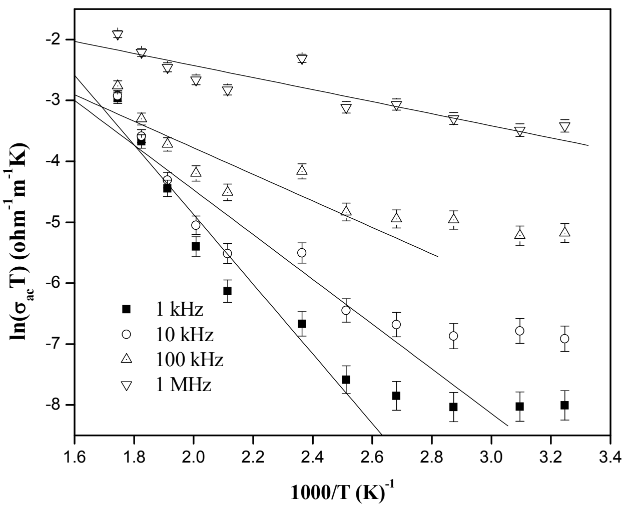


Fig.4. ln(σ_{ac}T) versus (1/T) for BTCU2 glass. Solid lines are the least square linear fits to the data in the high temperature region

According to Mott's SPH model (Mott, 1968) the conductivity in non-adiabatic regime is expressed as, $\sigma = (\sigma_0/T)\exp(-W/k_BT)$, where W is the activation energy and σ_0 is the pre-exponential factor. Mott plots of ac conductivity for BTCU2 glass for different frequencies are shown in Fig. 4. The least square linear lines were fit to the data at high temperatures. The activation energy, W_{ac} is estimated from the slopes (Table 1). The activation energy in present glasses is found to decrease with increase in mole fractions of CuO. As the CuO content is increased, more number of polarons are getting added to conduction band and thereby conductivity increases and activation energy decreases. Similar results have been reported for TMI doped tellurite glasses (Prashant Kumar *et al.,* 2009; Sankarappa *et al.,* 2008). Activation energy for ac conductivity for different frequencies obtains for BTCU glasses are tabulated in Table 1. It can be observed that activation decreases with increase in frequency and CuO content.

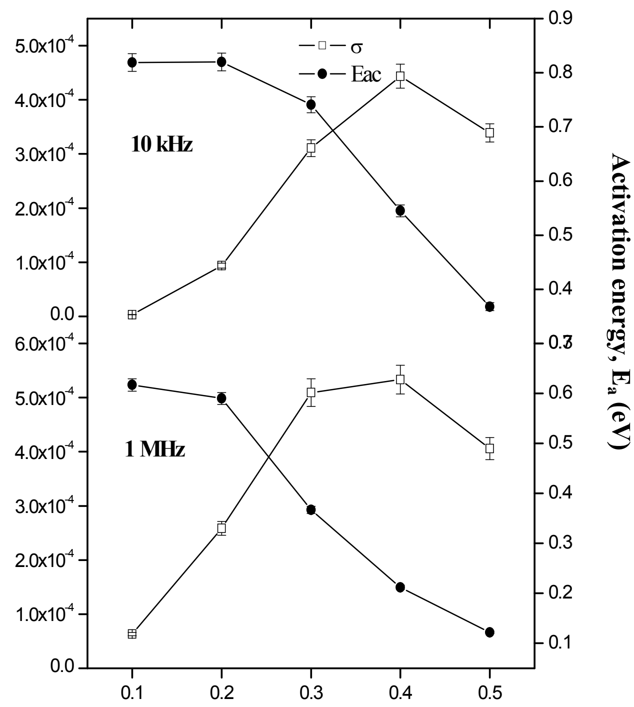


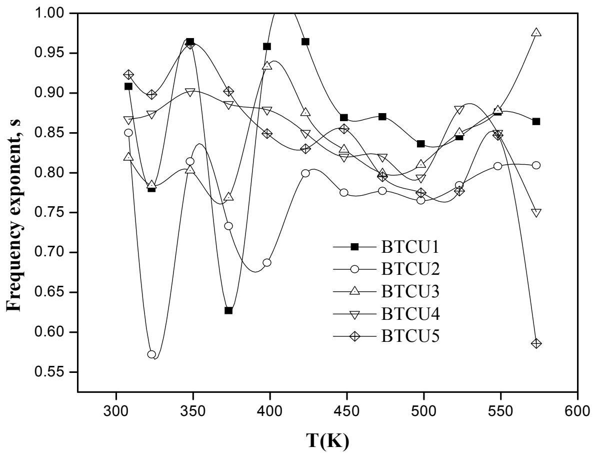
Fig. 5. Compositional dependence of activation energy, W_{ac}, and conductivity, σ_{ac}, at 473K for different frequencies

Table 1. Variation of ac activation energy, W_{ac} at different frequency, for BTCU glasses

Glass	W_{ac} (eV)			
	f = 1 kHz	f = 10 kHz	f = 100 kHz	f = 1 MHz
BTCU1	0.843	0.819	0.710	0.617
BTCU2	0.859	0.820	0.694	0.590
BTCU3	0.821	0.741	0.542	0.367
BTCU4	0.710	0.545	0.407	0.212
BTCU5	0.546	0.368	0.253	0.122

The temperature dependence of frequency exponent, s, for all the BTCU glasses is shown in Fig.6. It can be seen that s varies non linearly (non systematically) with temperature for all the glasses. This result negates the prediction of Quantum Mechanical Tunneling (QMT) model that s must be temperature independent.

The observed kind of variation of s with temperature may be due to different contributions from conducting and dielectric losses at different temperatures (Sankarappa *et al.,* 2008; Roumaih *et al.,* 2011).



Mole fractions of CuO

Fig.6. Temperature dependence of frequency exponent, s for BTCU glasses

Conclusion

Borophosphate glasses doped with CuO were synthesized and studied for dielectric properties and ac conductivity over a wide range of frequency (50Hz to 5MHz) and temperature (300K-625K). The dielectric constant and loss decreased with increase in frequency, increased with increase in temperature and CuO content. AC conductivity decreased with increasing temperature and increased with increase in CuO content. Variation of conductivity with temperature for different frequencies has been analysed using Mott's small polaron hopping model and activation energy was determined. Activation energy behaved in the opposite fashion to that of conductivity. Frequency exponent varied non systematically with temperature. For the first time the CuO doped borotellurite glasses have been investigated for dielectric properties and ac conductivity over a wide range of frequency and temperature and data analysed thoroughly.

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