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Temperature Variation of Conductivity in Cuo Doped Borate Glasses

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Abstract: *A series of borate glasses doped with CuO have been synthesized by melt quenching method. The samples were annealed and their non-crystallinity has been confirmed by XRD studies. AC electrical conductivity has been measured in the temperature range from 315K to 610K and in the frequency range from 50Hz to 5MHz. The conductivity increased and activation energy decreased with increasing concentration of CuO. Both dc and ac conductivities increased with increasing temperature in all the samples which has been ascribed to semiconducting nature of the glasses. The temperature dependence of dc and ac conductivities at different frequencies was analyzed using Mott's SPH model and, the high temperature activation energies were determined. The frequency exponent, s behavior with temperature has been understood.*

Keywords: *borate glasses, ac conductivity, polaron hopping, activation energy*

1. INTRODUCTION

The study of Transition Metal Ions (TMI) doped glasses is important from fundamental and applications points of view. TMI doped borate glasses are mainly used for applications in electrochemical, electronic and electro-optical devices (1, 2 and 3). The conductivity in these glasses arises due to the presence of TMI in multivalent states (2, 11 and 12) it is particularly interesting to study ac conductivity of these glasses as it would offer a chance to understand hopping mechanisms of charge carriers i.e. polorons from low valance state to high valance state of TM ions (9, 10, 11, 12 and 14). Impedance spectroscopy has been traditionally applied to understand the frequency dependence of complex conductivity in various glassy materials (1, 11, 12 and 13). In the present paper, we report detailed study of conductivity in CuO doped borate glasses in the following compositions.

x(CuO) (1-x)(B_2O_3) (x=0.2,0.3,0.4, 0.5 and 0.6 labeled as ST1,ST2, ST3, ST4, and ST5).

Ac conductivity of CuO doped borate glasses has not reported so by any research group.

2. EXPERIMENTAL

The glasses were prepared by following melt quenching technique using the analytical grade Cuo and H_3BO_3. The relevant chemicals in the appropriate weight ratios were taken in a porcelain crucible and melted in a muffle furnace in the range of 1300K to 1400K. The melt was quickly quenched to room temperature. In order to relieve mechanical stresses the samples were annealed at 600K. The noncrystalline nature of the samples was confirmed by XRD studies. Silver electrodes were painted on to the two major surfaces of the samples and, the capacitance and dissipation factors have been measured using a computer controlled LCR HiTester (HIOKI, 3532-50) in the frequency range from 50Hz to 5MHz and temperature from 310K to 610K.

3. RESULTS AND DISCUSSION

The conductivity, σ_{Total} has been determined using the measured capacitance and dissipation factors as described in (2 and 5). The measured total conductivity is expressed as

$$\sigma_{Total} = \sigma_{dc} + \sigma_{ac} = \sigma_0 + A\omega^s$$

Where, σ_{dc} (=σ_0) is dc component, A is a temperature dependent constant, *s* is the frequency exponent and σ_{ac} (=$A\omega^s$) represents the pure ac component of the total conductivity, which depends both on temperature and composition of the sample. The above mentioned equation was fit to the data as shown in Fig.1 for ST2 glass. The best fits gave σ_0, A, and *s*. Similar regressional analysis has been performed on σ_{Total} of the remaining glasses in the present series**.** In Fig 1. It is observed that both σ_{dc} and σ_{ac} increases with increase in frequency. Similar kind of observation has been made in the remaining glasses.

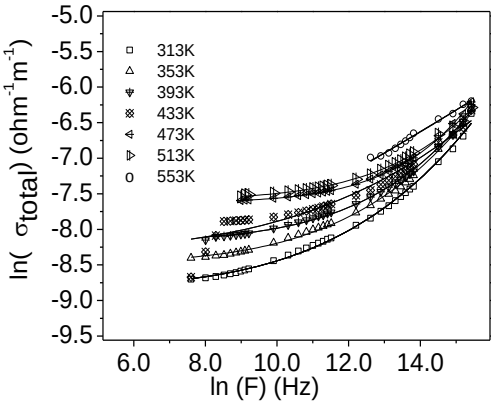

Figure1. *The plots of ln(σ_{Total}) vs ln(F) for ST2 glass. Solid lines are the best fits to $\sigma_{Total} = \sigma_o + A\omega^s$*

It is also noted that the glasses exhibited increase in conductivity with increase in the concentration of CuO. The variation of conductivity is observed to be a thermally activated process and is due to the hopping of polarons between the low valance state of Cu^+ to high valance state Cu^{2+} (2,11 and12). Fig.2 exhibits the temperature variation of σ_{ac} at 500kHz for all the glasses. The temperature variation of σ_{ac} at different frequencies for ST2 glass has been depicted in Fig.3. The σ_{ac} was observed to be increasing with increase in frequency. It can be noticed from Figs. (2 & 3) that the curves are linear at high temperature and non-linear at low temperature.

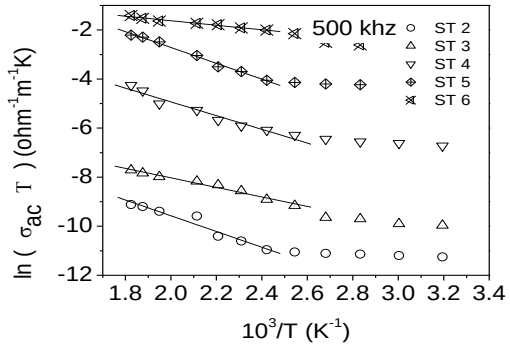

Figure2. *The plots of ln(σ_{ac}T) versus (1/T) at 500kHz.*

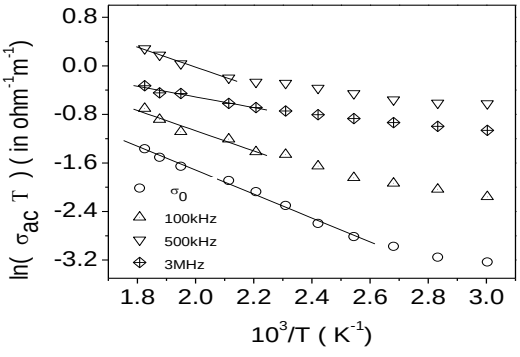

Figure3.*Variation of σ_{ac} with mole fractions of CuO.*

The activation energy for ac conductivity W_{ac} determined by fitting linear lines as per Mott's SPH model in the high temperature regions as shown in Figs.(2 & 3). W_{ac} is found to decrease with increase in concentration of CuO. And W_{ac} decreased with increase of frequency for ST2 glass as shown in Fig.4. Similar results are obtained for remaining glasses.

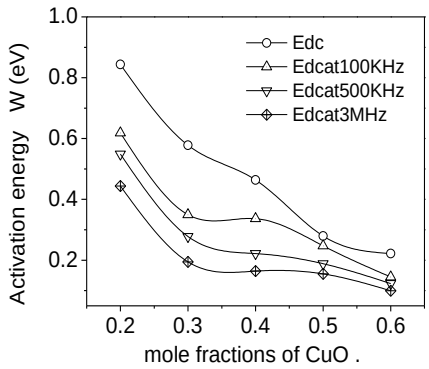

Figure4. *Activation energies at different frequencies for ST2 glasses.Activation energy for dc conductivity, W_{dc} is found to decrease with increase in CuO content.*

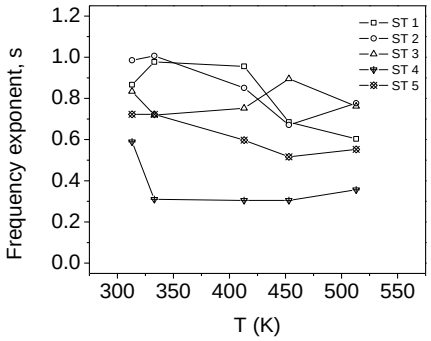

Figure5. *Temperature dependence of frequency exponent, s, for ST glasses.*

The temperature dependence of frequency exponent, *s*, for ST glasses *s*, values were determined to be lying between 0.35 and 1.01 in the studied range of temperature and not varied systematically as shown in Fig (5). The observed variation of s with T cannot be explained by either Quantum Mechanical Tunnelling or Correlated Barrier Hopping models.

The cross over frequency (F_0) from dc to dispersive like behavior has been found to be increasing with temperature. The frequency F_0 is defined through $\sigma'(F_0)=2\sigma_{dc}$ (10, 12). By shifting or scaling the conductivity isotherms along the straight line with slope one, the conductivity master curves were drawn and shown in Fig.6. for ST2 glass.

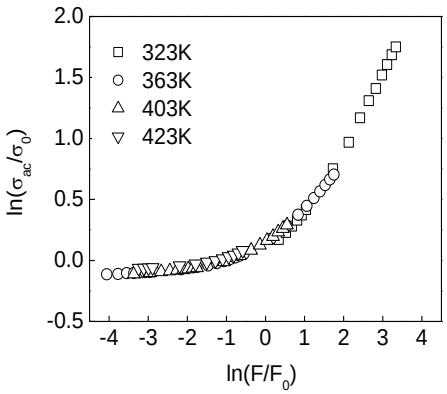

Figure6. *Master curves for ST2 glass.*

Master curves were obtained for all the remaining ST glasses. It can be observed from the master curves that the time-temperature superposition principle is fulfilled which in turn suggest the occurrence of a temperature independent relaxation mechanism is taking place (10, 12) in the present glasses.

4. CONCLUSIONS

A series of copper doped borate glasses were investigated for ac conductivity. The analyses of ac components of the conductivity, and frequency exponent, *s* lead to the following mentioned conclusions. (i)The ac conductivities at high temperatures were found to be varying as per Mott's SPH model. The activation energies were determined and discussed. (ii)Frequency exponent s exhibited non-systematic variation with temperature. Neither QMT nor CBH models can explain s variation. (iii) The time temperature superposition principle was found to be valid in these sets of glasses.

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