ISSN: 2322-0015

RESEARCH ARTICLE

Electrical conduction in CuO-MnO₂-B₂O₃ glasses

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Manuscript Details

Received: 28 may, 2014 Revised: 14 June, 2014 Revised received: 27 June, 2014

Accepted: 15 July, 2014 Published: 27 July, 2014

ISSN: 2322-0015

Editor: Dr. Arvind Chavhan

Cite this article as:

Gawande WJ, Yawale SS and Yawale SP. Electrical conduction in $CuO-MnO_2-B_2O_3$ glasses, *Int. Res. J. of Sci. & Engg.*, 2014; 2 (4): 145-149.

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ABSTRACT

Due to the technological importance of CuO-MnO₂-B₂O₃ glasses, dcconductivity measurement with increasing concentration of CuO (in the range of 5-30 mol%) have been reported in the temperature range of 313-573 K in the present study. A plot of $-\log \sigma$ versus 1/T shows two different regions of conduction suggesting two types of conduction mechanisms switching from one type to another occurring at knee temperature. The DC conductivity increases with increase in temperature of the sample and also with increase of mol% of CuO. Activation energy calculated from both regions (LTR and HTR) is below 1 eV. Thus electrical conduction is electronic. Activation energy in LTR and HTR are temperature independent but composition dependent. The values of dielectric constant at different temperature (313-573K) at a constant frequency of 1 KHz are reported. It is observed that the dielectric constant is independent of temperature upto certain temperature range, but after that the dielectric constant increases with temperature rapidly. The dielectric constant of all the samples studied is found to be composition dependent. In the glasses studied dipole relaxation phenomenon is observed.

Keywords: CuO-MnO₂-B₂O₃ glasses, DC-conductivity, dielectric constant.

1. INTRODUCTION

Now a day's glasses have a prominent role in the field of electronics and have wide applications in industry, space research, computer memories etc. Since 1954 when the electronically conducting oxide glasses were discovered, glass formation and properties in transition metal oxide systems have been extensively studied due to their important semiconducting behavior (Austin and Mott, 1969, Chaudhury, 1995, Denton, et al, 1954, Mott, 1968, Sayer and Mansingh, 1972) have discussed in brief the general procedure for making glass ceramic superconductors and some of their physical properties. All the glasses which become superconductors after properly annealing at higher temperatures are in

general transition metal oxides (TMO) with copper ions. Ghosh and Chaudhury, 1984 discussed the results of dc-conductivity of semiconducting vanadium bismuth oxide, containing 80-95 mol% vanadium pentaoxide in the 300-500 K temperature range on the basis of polaronic hopping model similarly they observed adiabatic hopping conduction. The electrical properties of V₂O₅-B₂O₃ glasses are discussed on the basis of small polaron hopping model by Culea and Nicula, 1986. The charge transfer mechanism plays a dominant role in semiconducting glasses. Dcconducting and hopping mechanism in Bi₂O₃-B₂O₃ glasses has been studied by Yawale and Pakade,1993. The physical and transport properties such as density, hopping distance, polaron radius, dc-conductivity and activation energy are reported by them. The small polaron hopping model is applied to the glass system. Dc-conductivity, density and infrared investigation have been carried out on Zno-PbO-B2O3 glasses by Doweldar et al, 1994, Mandal et al, 1987 have reported the dielectric behaviour of glass system BaO-PbO-TiO₂- B_2O_3 -SiO₂. The electric relaxation study of V_2O_5 - B_2O_3 glasses has been done by Singh and Tarsikka, 1988.

2. MATERIALS AND METHODS

2.1 Preparation of glass samples:

The glass samples were prepared in a fireclay crucible. The muffle furnace used was of Heatreat Co. Ltd. (India) operating on 230 volts A.C. reaching upto a maximum temperature of $1500 + 10^{\circ}$ C. Glasses were prepared from A R grade chemicals. Homogenous mixture of an appropriate amounts of CuO, MnO₂ and B₂O₃ (mole%) in powder form was prepared. Then, it was transferred to fire-clay crucible which was subjected to melting temperature (1300° C). The duration of melting was generally two hours. The homogenized molten glass was cast in steel disc of diameter 2 cm and thickness 0.7 cm. Samples were quenched at 200° C and obtained in glassy state by sudden quenching method. All the samples were annealed at 350° C for two hours.

2.2 Electrical Measurement:

The dc resistance of the glass samples was measured by using D.C. microvoltmeter, Systronics 412 India; having an accuracy of $\pm 1~\mu V$ and input impedence 10 M Ω , by voltage drop method given by Kher and Adgaonkar, 1972. Before electrical measurements all the samples were polished to smooth surfaces using fine quality emery paper. After application of conducting silver paint at either sides, the samples

were used for electrical measurements. The silver paint acts like electrodes for all the samples.

2.3 Dielectric Constant:

The dielectric constant of the glass samples was measured by measuring the capacitance of the samples at constant frequency 1 KHz in the temperature range 313 to 573 K. Digital LCR meter 925, systronics made (India), was used for the measurement of capacitance. The accuracy in the capacitance measurement was $\pm 0.1 \, \mathrm{pF}$

3. RESULTS AND DISCUSSION

3.1: Dc-electrical Conductivity:

D.C. electrical conductivity of the glass samples is measured in the temperature range 313 to 573 K. The value of d.c. conductivity is found to be of the order of 10^{-10} to 10^{-11} ohm⁻¹ cm⁻¹ at 313 K. Fig 1 shows the plot of -log σ versus 1/T. It is observed that, the conductivity of all the glass samples studied increases with increasing temperature.

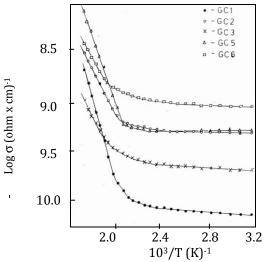
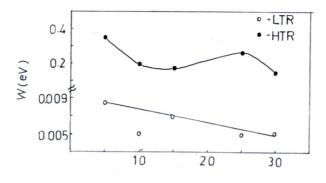


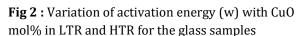
Fig. 1:Temperature Dependence of dc-electrical conductivity for the glasses of different compositions of CuO-MnO₂-B₂O₃

This plot is found to consist of two distinct straight linear regions called as low temperature regions (LTR) (313 to 413 K) and high temperature region (HTR) (523 to 573 K). In LTR conductivity increases linearly with increasing temperature at very slow rate where as in HTR conductivity increases linearly with increasing temperature at a faster rate. Obviously two activation energies and two conduction mechanisms are associated with electronic conduction in all the glasses studied. The same type of dc conductivity behaviour is reported in literature (Giridhar and Mahadevan, 1989,

Glass No.	CuO-MnO ₂ -B ₂ O ₃	Activation energy W (eV)		Kink	Activation	Pre-exponential
		LTR (W _L)	HTR (W _h)	Tempera-ture $\theta_c(K)$	energy at θ_c W (eV)	factor σ_0 (ohm x cm) ⁻¹ 10 ⁻⁹
G C1	5-20-75	0.0086	0.345	485	0.1245	12.5
G C2	10-20-70	0.0051	0.198	478	0.0862	8.31
G C3	15-20-65	0.0069	0.181	476	0.0739	5.24
G C5	25-20-55	0.0051	0.276	469	0.0862	45.7
GC6	30-20-50	0.0051	0.163	471	0.0984	10.9

Table 1 : Activation energies, Kink temperature and Pre- exponential factor σ_0 of CuO-MnO₂-B₂O₃ glasses.





Marshall and Owen, 1975, Yawale and Pakade, 1993). The activation energies are obtained from slope of the plot of -log σ versus 1/T in both the regions and reported in table 1.

It is observed that the activation energy is temperature independent but depends on composition. The activation energies obtained are found to be of order of borate vanadate and other semiconducting glasses reported in literature (Ghosh and Chaudhury, 1986, Kulkarni et al,1984, Nassar, 1982, Sayer and Mansingh, 1972, Singh and Tarsikka, 1988,). Activation energy calculated for both regions (LTR and HTR) is found to be less than 1 eV, thus the electrical conduction is electronic (Karimi and Gupta, 1987). The kink temperature θ_{c} is the temperature at which the Arrhenius plot is divided in to two linear regions of different slopes. The kink temperature (θ_c) is determined from the plot of $-Log \sigma$ versus 1/T and is reported in table 1. The kink temperature θ_c for the series of glasses studied decreases with increasing mol% of CuO. The activation energy is also calculated at kink temperature and the values are reported in table 1. The inetecept on $-\log \sigma$ axis of $-\log \sigma$ versus 1/T plot gives the values of pre-exponential factor ($-\log \sigma_0$).

Table 1 reports the values of activation energy, kink temperature pre-exponential factor of CuO- MnO_2 - B_2O_3 glasses. The values of different

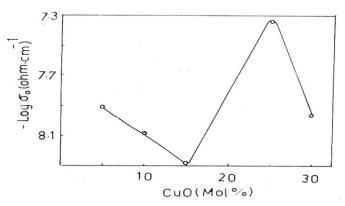


Fig 3: Variation of Pre-exponential factor $(-\log \sigma_0)$ versus Composition for the glass samples.

parameters reported in the table agreed with the values reported for semiconducting glasses in the literature (Ghosh and Chaudhury, 1986, Kulkarni et al,1984, Mori et al, 1993, Nassar, 1982, Sayer and Mansingh, 1972, Singh and Tarsikka, 1988, Yawale and Pakade,1993) Fig 2 shows the variation of activation energy (w) with CuO mol% in LTR and HTR for the glass samples. Fig 3 shows variation of pre-exponential factor (-log σ_0) versus composition for the glasses studied.

3.2 Dielectric Constant:

The variation of dielectric constant (\in ') at different temperature (313-573K) at a constant frequency of 1 KHz for the glass samples is shown in Fig. 4. It is observed that the dielectric contant (\in ')is independent of temperature upto certain temperature range, but after that the dielectric constant increases with temperature rapidly. A similar trend has been reported for different transition metal oxide glasses by Mansingh et al, 1983, Sayer and Mansingh, 1972. This increase in dielectric constant is partly due to a change in electronic structure and partly due to thermal expansion. In glasses rise of temperature may increase the free carrier density to introduce

conduction losses. The change in dielectric constant at high temperature is a characteristics of Debye type relaxation process where symmetrical distribution of relaxation time takes place. The rapid rise is likely to arise from the other sources of polarization possibly from enhanced electrode polarization as temperature rises. More sharp rise of (\in ') at high temperature was also observed in other oxide glasses by Singh and Tarsikka, 1988 and Sunder and Rao, 1982.

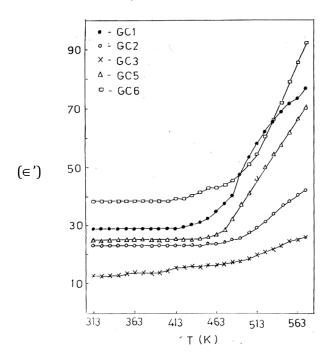


Fig 4: Variation of dielectric constant with temperature at constant frequency of 1KHz for the glass samples.

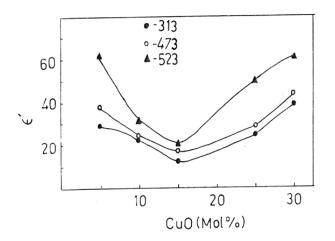


Fig 5: Variation of dielectric constant with composition at a constant temperature for the glass samples.

The dielectric constant of all the sample studied is found to be composition dependent. Variation of dielectric constant with composition at a constant temperature is shown in figure 5. A dip is observed at 15 mol% of CuO. In these glasses dipole relaxation phenomenon is observed

4. CONCLUSION

D.C. conductivity of $CuO-MnO_2-B_2O_3$ glass system is studied in the temperature range 313-573K. The activation energy are found to be in the range of semiconducting glasses. The electrical conduction is electronic. The dielectric constant of the glass samples is found to be temperature and composition dependent. In the glasses dipole relaxation phenomenon is observed.

ACKNOWLEDGEMENT

The authors express their sincere thanks to the Director and Head, Department of Physics, Govt. Vidarbha Institute of Science and Humanities, Amravati for providing laboratory facilities.

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148 www.irjse.in

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