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INFLUENCE OF TMO ADDITION ON THE PROPERTIES OF LITHIUM BOROSILICATE GLASS

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ABSTRACT

The influence of Fe_2O_3 addition on the variation in electrical conductivity, density, T_g and CTE has been studied in Li_2O:B_2O_3:SiO_2 glass system. The results have been explained in the light of the NBOs present in the glass, the electronic contribution to the total conductivity, the size and mass of the Fe^{3+} ion which is substituted for B_2O_3 and SiO_2 in the glass.

KEYWORDS:

electronic conductivity, NBOs,
density, CTE

INTRODUCTION:

Lithium conducting glasses have been studied widely since last few decades due to their potential application in electrochemical devices [1,2]. However, there is a problem of their stability and achieving high ionic conductivity in them. Lithium borosilicate glass system shows better glass formability and chemical stability which are essential for any potential solid electrolyte material in solid state batteries. The electrical, thermal, and optical properties of a few transition metal oxide doped glass systems have been studied earlier [3-5]. It has been reported that the addition of transition metal oxides in the glasses increases the electronic conductivity. However, systematic investigations in this regard are still lacking. Hence in the present work, the influence of Fe_2O_3 addition on the properties of lithium borosilicate glass has been studied.

EXPERIMENTAL METHODS:

The Glasses studied in the present work can be represented by the general formula$30Li_2O$: XFe_2O_3: (70 – X)(1/7 SiO_2:6/7 B_2O_3) where X = 0, 1.0, 2.5, 5, 7.5, 10.0. The finely mixed powders of Li_2CO_3, Fe_2O_3, B_2O_3, SiO_2 of high purity (>99.5%) were melted in platinum crucible at 1050^0C for one and a half hour. The melts were stirred from time to time to attain homogeneity and quenched in an aluminum mould at room temperature in air. The glasses so prepared were annealed for two hours. XRD studies confirm the amorphous nature of the samples prepared.

The Electrical conductivity of the glass samples was studied in the temperature range from 473K to 613K using high resolution dielectric analyzer. The glass transition temperature T_g was determined by DTA and the coefficient of thermal expansion of these glasses was measured by Dilatometer. The densities of these samples were measured by

Archimedes' Principle with toluene as immersion liquid. The Transport number measurements were done by Keithley electrometer, using dc polarization technique.

RESULTS AND DISCUSSION:

The XRD patterns of all the samples with Fe_2O_3 addition show no sharp peaks which confirm that the quenched samples are in amorphous network which are shown in fig (1).

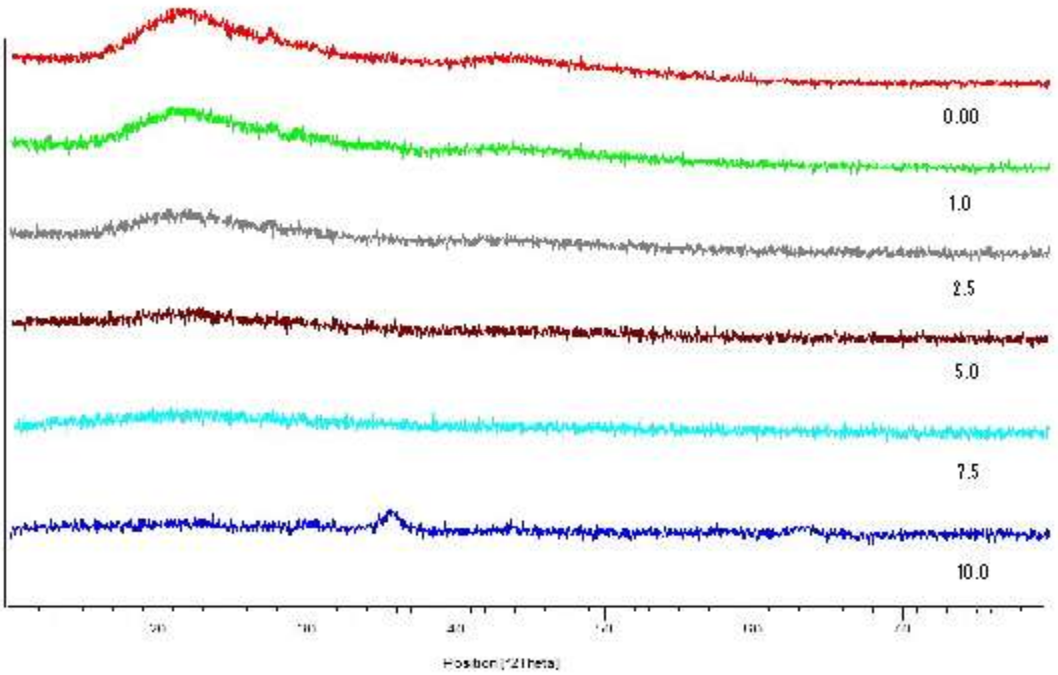

Fig (1) XRD patterns of samples with Fe_2O_3 addition at levels X=10.0, 7.5,5.0, 2.5, 1.0, 0.00

The variation of electrical conductivity and activation energy as a function of Fe_2O_3 content is shown in Fig (2). It is observed that the electrical conductivity, as a function of Fe_2O_3 content, decreases up to 7.5 mol% and then increases. The variation of activation energy shows a behavior opposite to that of conductivity. The lithium content in all the glass samples is kept fixed. Hence, the variation in the electrical conductivity can be understood on the basis of changes in the glass structure in terms of the number of non-bridging oxygens (NBOs) which govern the mobility of the lithium ions and also the fraction of electronic contribution to the total conductivity [6]. The decrease in ionic conductivity up to 7.5 mol% Fe_2O_3 may be attributed to the decrease in the mobility of the Li ions in the altered glass network.

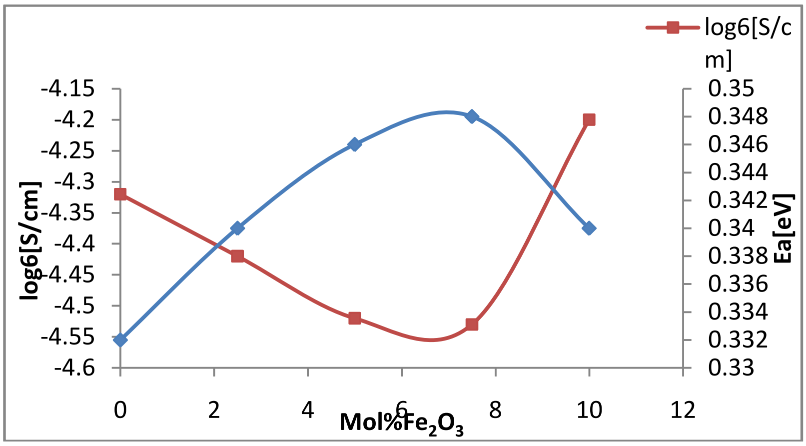

Fig (2) variation of logσ and Ea with mol% of Fe_2O_3.

The addition of Fe_2O_3 at the cost of glass formers initially may alter the glass structure in such a way so as to reduce the number of non-bridging oxygens (NBOs). This in turn may reduce the potential sites which lithium ions can move, thereby reducing the mobility.

The increase in conductivity beyond 7.5 mol% Fe_2O_3 may be due to electronic contribution to the total conductivity. This has been confirmed by the transport number measurements which reveal that the electronic transport number is more for glass samples with Fe_2O_3 > 7.5 mol%. It has been reported that like alkali contents, Fe^{3+} oxide involves in sharing of oxygen between the glass forming networks. Wu et al and Zhong et al have proposed that the borate groups consume oxygen to form tetrahedral boron units. It may lead to an increase in NBOs in the matrix.

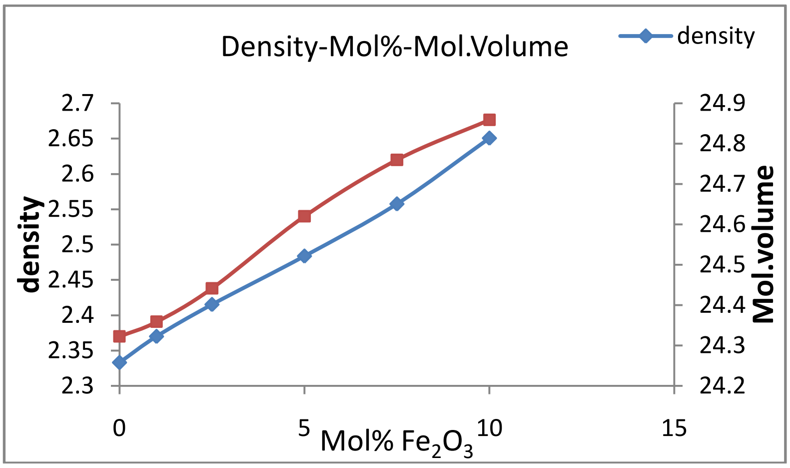

Fig (3) Variation of density and molar volume with mol % of Fe_2O_3.

Fig (3) depicts the variation of density and molar volume as a function of Fe_2O_3 for the entire glass series. The observed increase in the values of density and molar volume may be attributed to the heavier mass and bigger size of Fe ions as compared to those of boron and silicon.

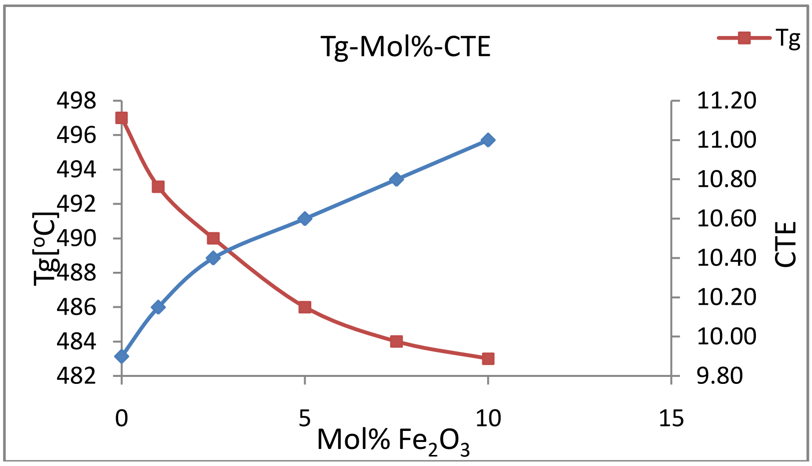

Fig (4) Variation of T_g and CTE with mol % of Fe_2O_3.

Fig (4) illustrates the variation of T_g and CTE as a function of Fe_2O_3 content for the glass samples. The glass transition temperature T_g determined from DTA shows continuous decrease with increase of Fe_2O_3 addition indicating the weakening of glass structure. This is supported by the observed increase in CTE values with addition of Fe_2O_3. This is also supported by the increase in molar volume observed. The increase in density of glass in spite of increase in the molar volume has to be essentially due to heavier Fe ion. The specific heat (ΔCp) values at T_g are obtained from DTA studies. It is observed that it decreases up to 7.5 mol% Fe_2O_3 and then started to increase. It may be attributed to the presence of ions in Fe^{3+} state in the glass network.

CONCLUSION:

It can be concluded from the present work that the addition of Fe_2O_3 increases the electronic conductivity of the glass in general. The total conductivity depends on the number of NBOs present in the glass sample and the fractional contribution of electronic conductivity. The variation in molar volume, density, T_g and CTE of the glass samples are governed by the size and the mass of the Fe^{3+} ion which is added in the glass, apart from the NBOs present therein.

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