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Physical Properties of Lithium-Sodiumborate Glasses

KEYWORDS: Density, Molar volume, Oxygen packing density, Dielectric constant, Refractive index, Molar refractivity, Molar polarizability; Metallization, Eelectronic polarizability, Polarizability per unit volume, glass composition, mixed glass former effect

M.D.Thombare	R.V.Joat	D.B. Thombre
Department of Physics, Research student, Vidya Bharati Mahavidyalaya, Amravati, India	Department of Physics, Associate, professor,Vidya Bharati Mahavidyalaya, Amravati, India	Department of physics, Associate professor, Jagdamba Mahavidyalaya, Achalpur City, India

ABSTRACT *Glass samples of the Lithium-sodiumborate (42.5-x) Li2O-xNa2O-57.5B2O3, x=0-42.5 in the step of 4.25, were prepared by conventional melt-quench technique.. Average molecular weight, Molar volume, Inter-ionic distance, Polaron radius increases; and Oxygen packing density, ionic concentration decreases on increasing the mol % of Na2O.Inter ionic distance & polaron radius increases. Ionic concentration & Oxygen packing density decreases on increasing the mol % of Na2O but linearly related with R2=1. Molar refractivity, Electronic polarizability, varies alike; minimum =0.629 cm3 and maximum =0.249(1024ions/cm3) at 25.5 mol % of Na2O; varies linearly with R2=0.999.Molar polarizability & Electronic polarizability varies inversely with respect to Metallization and minimum = 6.61(1024ions/cm3) & 0.249(1024ions/cm3), respectively for maximum metallization = 0.371 at same 25.5 mol % of Na2O. Refractive index & Dielectric constant, Molar refractivity & varies alike and minimum at 25.5 mol % of Na2O. The density and molar volume of glasses were determined in order to study their structure. Molar volume & density increases in reverse manner with increase in mol % of Na2O.The maximum variation of Metallization criterion is within the range of 0.274-0.574.*

1. INTRODUCTION

Oxide glasses are classically described as a network composed by building entities such as B_2O_3, SiO_2, P_2O_5,V_2O_5,Sb_2O_3, Bi_2O_3 and modifiers such as alkaline oxides: Li_2O, Na_2O, K_2O, Ag_2O, Rb_2O [1,2]. In such glasses, the oxygen from the metal oxide becomes part of the covalent glass network, creating new structural units. The cations of the modifier oxide are generally present in the neighborhood of the non-bridging oxygen (NBO) in the glass structure. The extent of the network modification obviously depends on the concentration of the modifier oxide present in the glass. A glass network affects various physical properties such as density, molar volume, glass transition temperature, polarization, etc.

A general theoretical approach for the mixed alkali effect in glasses is discussed based on the idea that the covalent host network creates different structural energy landscapes for different types of mobile ions. The model suggests that Coulomb forces have to be taken into account for explaining the differing behavior of activation energies in single modified and mixed ion glasses [3]. Whether in the single or mixed alkali glass, though there are structural changes. There exists a site mismatch energy, which can act as a barrier for diffusion of alkali ions. Alkali ions preferentially jump into sites that were previously occupied by the same type of alkali ions [4]. A weak electrolyte model for the mixed-alkali effect on electrical conductivity and ionic mobility in glass was developed for the dilute foreign alkali region. The basic assumptions are (a) that alkali transport in single-alkali glasses is due to a small concentration of mobile species [5]. A semi-empirical model has been applied to the densities in order to determine the volumes of the structural units present with volumes calculated from the ionic radii, were used in a general discussion of the filling of space by the structural units. The results indicate that the size increases of the structural groupings as one goes from small to large alkali are primarily due to the alkali being used [6]. Density can be used for finding out the structure of different types of glasses. The density of the glass is additive and can thus be calculated on the basis of the glass composition [7,8,9,10]. Several formulas have been derived to correlate the glass density to the glass composition [11,12,13,14,15]. The glass structure can be explained in terms of molar volume rather than density, as the former deals the spatial distribution of the ions forming that structure. The change in the molar volume with the molar composition of an oxide indicates the preceding structural changes through a formation or modification process in the glass network [16,17]. The density, molar volume and packing fraction [18,19,20] could be directly related to the short range structure of alkali oxide modified borate glasses. The densities prove changes in both short range order and co-ordination as the modification, while the molar volume is sensible in terms of size and packing. The packing of the borate based glasses with ions having volume smaller than the oxygen is considered to be covalent, controlled by oxygen covalent network, and heavily dependent on the glass former.

The molar refractivity [21,22,23,24,25,26], is a constitutive-additive property which represents the real volume of the molecules. That is calculated by the Lorenz-Lorentz formula. Electric polarizability [21-27], is the relative tendency of a charge distribution, like the electron cloud of an atom or molecule, to be distorted from its normal shape by an external electric field; similarly ionic concentration [21,26,27], inter ionic distance [21,26]. When an electron in the conduction band of a crystalline insulator or semiconductor polarizes or otherwise deforms the lattice in its vicinity. The polaron comprises the electron plus its surrounding lattice deformation. (Polarons can also be formed from holes in the valence band.) If the deformation extends over many lattice sites, the polaron is "large," and the lattice can be treated as a continuum. Charge carriers inducing strongly localized lattice distortions form "small"

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polarons [26]. In present work, these parameters have studied to explain structural features.

2. EXPERIMENTAL TECHNIQUES

2.1. PREPARATION OF GLASSES

The Lithium-sodiumborate glass samples having the general chemical formula (42.5-x) Li_2O-xNa_2O-57.5B_2O_3, x=0-42.5 in the step of 4.25, were prepared by conventional melt-quench technique from high-purity reagent grade B_2O_3 mol%.Li_2CO_3, and Na_2O reagents were purchased from Aldrich and appropriate amounts of the chemicals were well mixed and then dried in a vacuum oven at 150°C for 15 mins. Dried mixture samples were melted in Pt crucibles in an electric furnace at the temperature of 1000~1100°C. Melt was kept for 30 mins and rapidly quenched on stainless steel. The prepared samples was then annealed at 300-400 °C temperature for 2 hrs and then kept in vacuum desiccators to avoid possible moisture absorption before testing. The prepared glass samples are polished and the surfaces are made perfectly plane and smoothened by 120 No. emery paper. Thickness of the samples has been measured using digital vernier calipers with an accuracy of 0.0001mm.

2.2 DENSITY MEASUREMENT

The density was measured at room temperature by using Archimedes principle, with toluene as the buoyant medium.

$$\tilde{n} = W_a \tilde{n}_b / (W_a - W_b)$$

Where Wa is the weight of glass sample in air, Wb is the weight of glass sample in buoyant liquid, (Wa-Wb) is the buoyancy, ρb is density of buoyant.

2.3. MOLAR WEIGHT CALCULATIONS

Step I–Weight/mole = molar weight of the constituents * mol% of samples / 100

Step II–The molecular weight (M) of the sample is nothing but the summations of Wt/mole of its constituents.

Step III–The molar volume of the glass samples can be calculated from following expression:

$V_m = M/$ Here, ρ is the density of the sample and M is the molecular weight of the sample.

2.4. OXYGEN PACKING DENSITY (O)

Oxygen packing density of the glass samples were calculated using the following relation [27]

$O = n(\rho / M)$

where ρ, the density of desired glass samples, M, molecular weight of the sample and n is the number of oxygen atoms in the composition.

2.5 THE IONIC CONCENTRATIONS (N)

The ionic concentrations of the glass samples are determined using the following relation,

$$N = \left(\frac{6.023 \times 10^{23} \text{ mol}^{-1} * \text{mol\% of cation} * \text{valency of cation}}{\text{molar volume}} \right) \quad (4)$$

2.6 INTER-IONIC DISTANCE (R)

$R = (1/N)^{\frac{1}{3}}$

Inter ionic distance of the glass samples is given as, (5)

where N=ionic Concentration.

2.7 POLARON RADIUS (r_p)

$r_p = \frac{\delta}{2}\left(\frac{}{6N}\right)^{1/3}$ where N is the number of ions per unit volume.

(6)The values of the above parameter of Lithium-sodiumborate glasses are depicted in Table 2.6.1.

Table 2.6.1 Average molecular weight, Density, Molar volume, Oxygen packing density Ionic concentrations Inter-ionic distance and polaron radius for Li_2O-Na_2O·B_2O_3 glass system.

Name of Sample	Average molecular-weight M(gm/mol)	Density ρ(gm/cm³)	Molar volume V_m(cm³/mol)	Oxygen packing density O(cm³/mol)	Ionic concentrations N(10^{21}/cm³)	Inter-ionic distancer (A°)	Polaron radiusr$_p$ (A°)
LNB.1	52.731	2.26	23.332	92.147	21.942	0.77	0.144
LNB.2	54.095	2.92	23.526	92.002	27.935	0.765	0.141
LNB.3	55.46	2.31	24.008	89.552	21.324	0.777	0.145
LNB.4	56.824	2.27	25.032	85.889	20.452	0.788	0.147
LNB.5	58.188	2.3	25.299	84.983	20.236	0.791	0.148
LNB.6	59.552	2.34	25.45	84.481	20.116	0.792	0.148
LNB.7	60.916	2.34	26.033	82.589	19.666	0.798	0.149
LNB.8	62.28	2.38	26.168	82.161	19.564	0.8	0.15
LNB.9	63.644	2.42	26.299	81.751	19.466	0.801	0.15
LNB.10	65.009	2.41	26.974	79.705	18.979	0.808	0.151
LNB.11	66.373	2.35	28.244	76.123	18.126	0.82	0.153

Table 2.6.1 Indicates that the increasing mol% of Na_2O at the cost of Li_2O by keeping glass former B_2O_3 constant. The molar volume, Inter ionic distance, Polaron radius increasing; while Oxygen packing density, Ionic concentration decreases, which suggests the increased free space within the glass structure, [28-33], it means that the glass structure becomes loosely packed [30-34]. The polaron comprises the electron plus its surrounding lattice deformation. (Polarons can also be formed from holes in the valence band.) Due to the increasing values of Polaron radius the deformation extends over many lattice sites, and the lattice can be treated as a continuum.

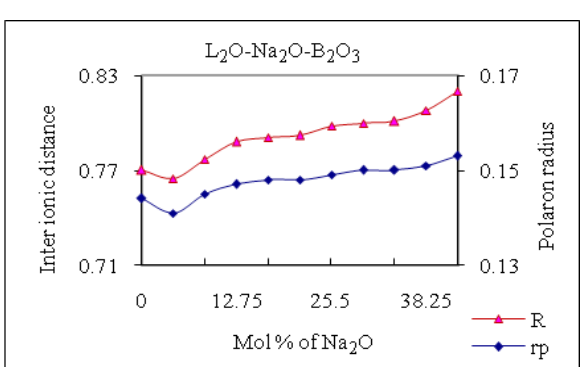

Figure: 2.6.1. Variation of Inter ionic distance & Polaron radius with mol% of Na_2O.

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Inter ionic distance & Polaron radius increases on increasing mol % of Na_2O, minimum =0.765 A° & 0.141 A° at 4.25 mol % of Na_2O (Figure 2.6.1).

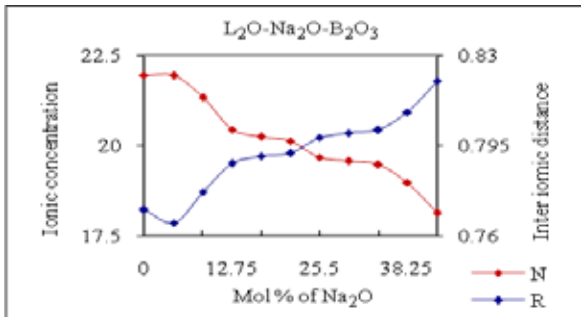

Figure: 2.6.2(a). Variation of Inter ionic distance Ionic concentration with mol% of Na2O.

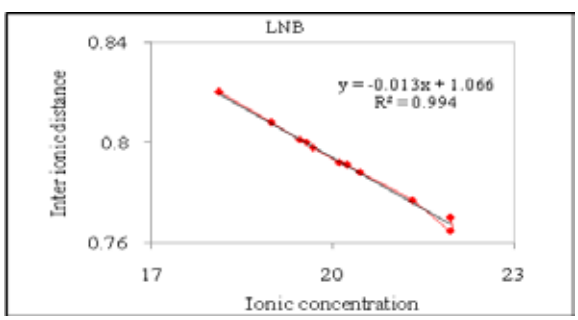

Figure: 2.6.2(b). Variation of Inter ionic Ionic distance & Ionic Concentration.

Ionic concentration goes on decreasing where as Inter ionic distance increasing with increasing mol % of Na_2O, i.e. they varies inversely and verifies from equation (4) and (5); and crosses at 21.25 Mol % of Na_2O, (Figure 2.6.2a) and varying linearly gives R^2 = 0.994.

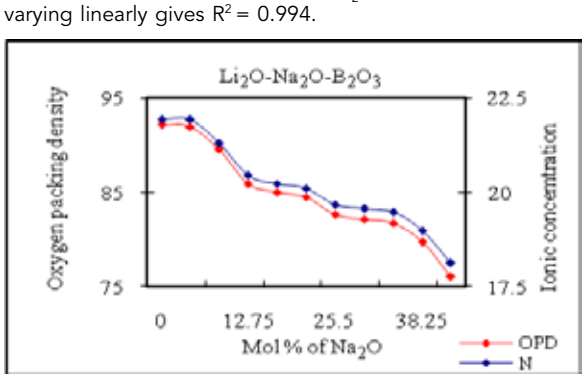

Figure: 2.6.3(a). Variation of Ionic concentration & oxygen packing density with mol% of Na2O

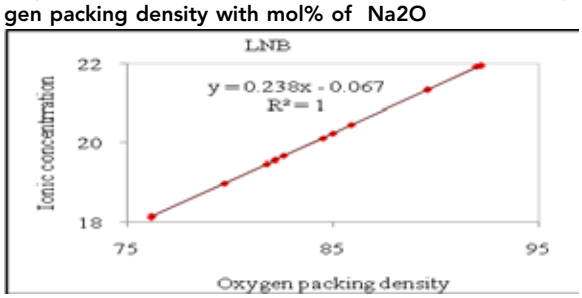

Figure: 2.6.3(b). Variation of Ionic concentration & oxygen packing density

Oxygen Packing Density & Ionic concentration both of them decreases on increasing the mol % of Na_2O (Figure 2.6.3a), which satisfies the equations (3) and (4); they also varies linearly with R^2=1(Figure 2.6.3b).

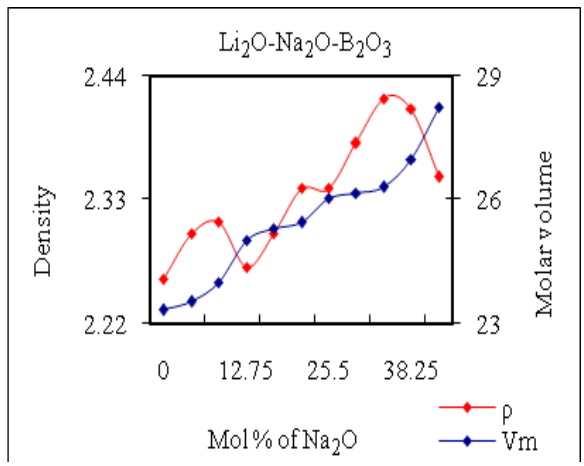

Figure: 2.6.4. Variation of Density & Molar volume with mol% of Na_2O.

Density and Molar volume increases monotonically with increasing mol % of Na_2O depicted in Figure 2.6.4.

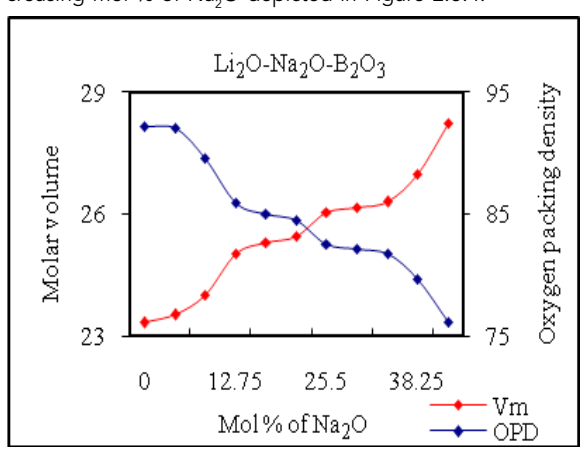

Figure: 2.6.5(a). Variation of Oxygen Packing Density & Molar volume with mol% of Na2O.

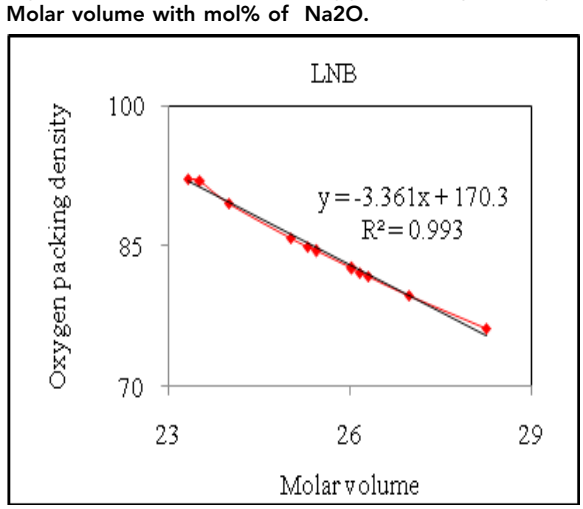

Figure: 2.6.5(b). Variation of Molar volume and Oxygen Packing Density

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Molar volume increases where as Oxygen packing density decreases which satisfies the equations (2) and (3); and crosses at 21.25 mol % of Na_2O, on increasing mol % of Na_2O (Figure 2.6.5a). The monotonically variations is due to the transformation of BO_3 triangle units to BO_4 tetrahedral units can be expected to increase the network linkage of the glass.

2.7 PHYSICAL PROPERTIES: RESULTS AND DISCUSSION

Other physical parameters such as, Refractive index (n) of samples was calculated by using the following relation [35].

$$\left(\frac{n^2-1}{n^2+2}\right)=\left(1-\sqrt{\frac{Eg}{20}}\right)$$

(1) Where, E_g is the energy gap.

The dielectric constant (Є) was calculated from the refractive index of the glass using [36].

Є = n² where n = Refractive index

On the other hand, Duffy [37] has obtained an empirical formula that relates energy gap E_g to molar refraction R_m.

$$\left(1-\sqrt{\frac{E_g}{20}}\right)=\left(\frac{R_m}{V_m}\right)$$

The ratio of Rm/V_m is called polarizability per unit volume. According to the Herzfeld theory of metallization [38], If R_m/V_m >1 and R_m/V_m <1 samples predicting metallic or insulating. From Table 2.7.1 it is clear that present glass samples behave as non-metal. The difference M = 1 – R_m/V_m is so-called metallization criterion [39].

Materials with large M close to 1 are typical insulators. The small value of M close to zero means that the width of both valence and conduction bands become large, resulting in a narrow band gap and increased the metallicity of the solid. The molar refraction R_m, can be expressed as a function of molar polarizability α_m as

$R_m = 4\pi\alpha_m A_v/3$ Where A_v is Avogadro's number introduced, with α_m in (Å³) this equation can be transformed to, $R_m = 2.52\alpha_m$ Hence molar polarizability α_m can be calculated.

.The electronic polarizability (α_e) was calculated using the formula [40].

$$\alpha_e = \frac{3\left(n^2-1\right)}{4\pi A_v\left(n^2+2\right)}$$

by using equation (1) and (2) equation (3) can be written as

$$\alpha_e = \left(\frac{3}{4\pi A_v}\frac{R_m}{V_m}\right)$$

Where, A_v is the Avogadro number. The measured and calculated values of densities, molar volumes and polarizability of oxide ions of Na_2O doped Lithium-borate glasses are listed in the Table 2.7.1

Table: 2.7.1 Refractive index, Dielectric constant, Molar refractivity, Metallization, Electronic polarizability (α_e*10^{-24} cm3), Molar polarizability (α_m*(10^{24}/cm³) and Polarizability per unit volume.

Refractive index (n)	Dielectric constant (Є)	Molar Refractivity R_m(cm³)	Electronic polarizability α_e	Metallization M	Molar polarizability α_m	Polarizability per unit volume R_m/V_m
2.993	8.958	0.726	0.288	0.274	6.714	0.726
2.934	8.608	0.717	0.284	0.283	6.745	0.717
2.912	8.48	0.714	0.283	0.286	6.825	0.714
2.695	7.263	0.676	0.268	0.324	6.572	0.676
2.624	6.885	0.662	0.262	0.338	6.861	0.662
2.515	6.323	0.64	0.253	0.361	6.877	0.64
2.466	6.079	0.629	0.249	0.371	6.61	0.629
2.502	6.259	0.637	0.252	0.363	6.771	0.637
2.791	7.79	0.694	0.275	0.307	7.293	0.694
2.879	8.289	0.709	0.281	0.292	7.928	0.709
2.969	8.815	0.723	0.286	0.277	8.344	0.723

The plots of Dielectric constant & Refractive index; (Figure 2.7.1a) varies alike; minimum value of dielectric constant = 6.079 and minimum value of refractive index = 2.466; at 25.5 mol% of Na_2O; they also varies linearly with R^2 = 0.999, with positive slope (Figure 2.7.1b).

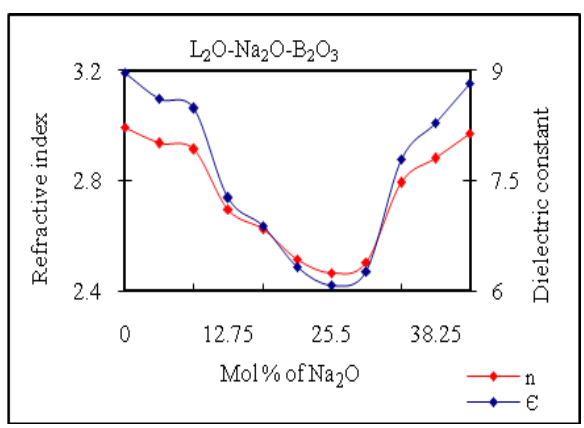

Figure 2.7.1(a) Variation of Dielectric constant & Refractive index with mol% of Na2O

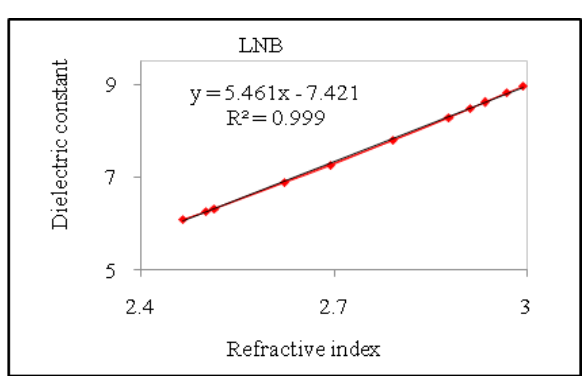

Figure2.7.1 (b) Variation of Dielectric constant & Refractive index.

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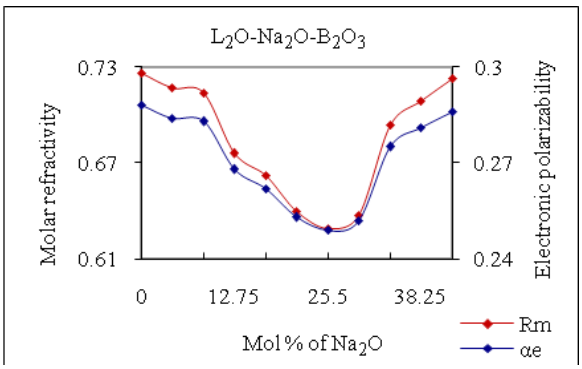

Figure2.7.2(a).Variation of Electronic polarizability & Molar refractivity with mol% of Na2O.

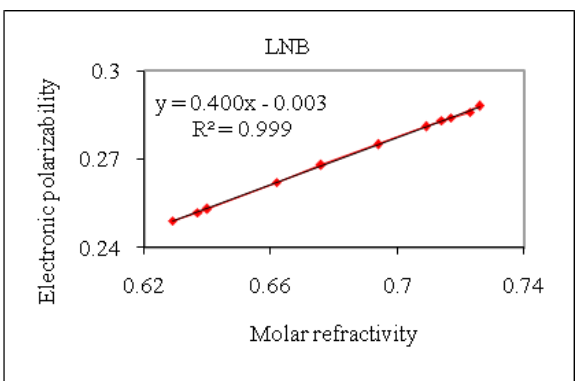

Figure2.7.2: Variation of Molar refractivity & Electronic polarizability

Molar refractivity and Electronic Polarizability (Figure 2.7.2a) varies alike; minimum value of molar refractivity and electronic Polarizability = 0.629 cm^3 & 0.249 (10^{24}ions/cm^3); at 25.5 mol% of Na_2O; with increasing mol% of Na_2O both of them also varies linearly having R^2=0.999 with positive slope (Figure 2.7.2b).

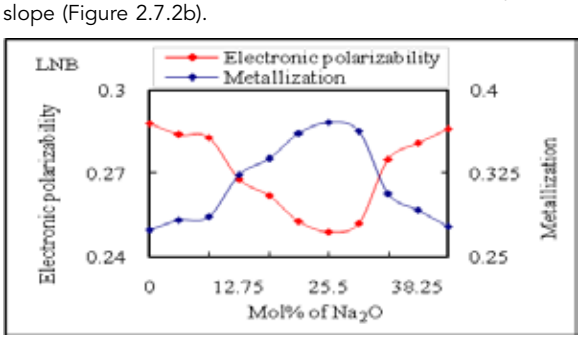

Figure 2.7.3(a). Variation of Electronic polarizability & Metallization with mol% of Na2O

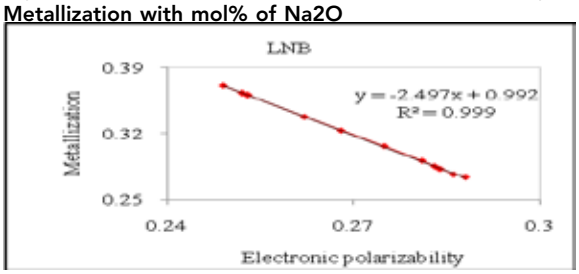

Figure 2.7.3(b). Variation of Metallization

Electronic polarizability & Metallization (Figure 2.7.3a), varies inversely minimum value electronic polarizability = 0.249 (10^{24}ions/cm^3) and maximum value of metallization 0.371; at 25.5 mol % of Na_2O, on increasing mol % of Na_2O. They also varies linearly having R^2=0.999 with negative slope.

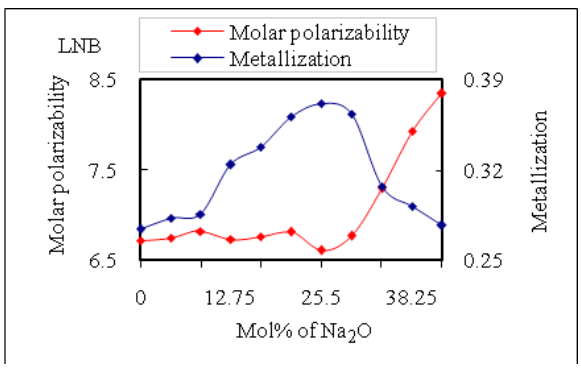

Figure 2.7.4: Variation of Molar polarizability and Metallization with mol% of Na_2O.

Molar polarizability and Metallization varies inversly minimum value of Molar polarizability is = 6.61 (10^{24}ions/cm^3); where as maximum value of Metallization is = 0.371; at 25.5 mol % of Na_2O, on increasing mol % of Na_2O.

CONCLUSIONS

Increasing the mol% of Na_2O with respective to Li_2O by keeping B_2O_3 glass former constant, the molar volume, inter ionic distance, polaron radius increasing, suggests the increased free space within the glass structure, it means that the glass structure becomes loosely packed. Oxygen packing density, ionic concentration monotonically decreases, because the polaron comprises the electron plus its surrounding lattice deformation. Hence, the deformation extends over many lattice sites and the lattice can be treated as a continuum. Transformation of BO_3 triangle units to BO_4 tetrahedral units can be expected to increase the network linkage of the glass which is reflected in the monotonically increasing density. Dielectric constant & refractive index, Molar refractivity behavior is alike as that of Molar polarizability, they also varies linearly. Electronic polarizability and metallization shows reverse behavior, is also due to the break down of borate & sodium bonds to create non- bridging oxygen atoms. Polarizability per unit volume predicts present glass samples are non-metals. The formation of BO_4 and BO_3 which will modify the glass structure by creating NBOs in the network gives parallel variation in Dielectric constant, Refractive index, Molar refractivity and Molar polarizability. Liner trends are found in Molar refractivity & the Molar polarizability, Metallization & Polarizability per unit volume, and mol % of Na_2O & Molar volume. Finally it is conclud that as glass former is kept constant the mixed alkali does not effect in different ways that is it behave as alike which is due to smallar Coulomb forces. In the single or mixed alkali glass, though there are structural changes. There exists a site mismatch energy, which can act as a barrier for diffusion of alkali ions. Alkali ions preferentially jump into sites that were previously occupied by the same type of alkali ions.

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