

# Study of Physical Properties of Potassium-Borophosphate Glasses

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**ABSTRACT:** Glass samples of the Potassium-borophosphate  $42.5K_2O-(57.5-x)-B_2O_3-xP_2O_5$ ,  $x=0-57.5$  in the step of 5.75, were prepared by conventional melt-quench technique. The density and molar volume of glasses were determined in order to study their structure. The physical parameters like Oxygen packing density, Ionic concentration, Inter ionic distance, Polaron radius, Dielectric constant, Refractive index, Molar refractivity, Molar polarizability, Metallization, Electronic polarizability are studied. Average molecular weight, Molar volume, Inter-ionic distance, Polaron radius increases as mol % of  $P_2O_5$  increases where as, Oxygen packing density, ionic concentration, Molar polarizability decreases as mol % of  $P_2O_5$  increases. Dielectric constant, Refractive index and Molar refractivity, Molar refractivity, Electronic polarizability, Metallization varies monotonically. Variation in inter ionic distance & polaron radius, ionic concentration & inter ionic distance, molar refractivity & molar polarizability, electronic polarizability & metallization and refractive index & dielectric constants are linear. Inter ionic distance & polaron radius, molar polarizability & molar refractivity, dielectric constant & refractive index varies alike with increasing mol% of  $P_2O_5$ . Inter ionic distance & ionic concentration varies oppositely with increasing mol% of  $P_2O_5$ , crosses oppositely at 31.625 mol% of  $P_2O_5$ . Similarly electronic polarizability & metallization also varies reversely with increasing mol% of  $P_2O_5$ ; electronic polarizability is minimum equal to 0.258 & metallization is maximum equal to 0.348 at 51.75 mol% of  $P_2O_5$ .

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

## I. INTRODUCTION

Oxide glasses are classically described as a network composed by building entities such as  $SiO_2$ ,  $B_2O_3$ ,  $P_2O_5$ ,  $Sb_2O_3$ ,  $Bi_2O_3$ ,  $Ge_2O_3$  and modifiers such as alkaline oxides:  $Li_2O$ ,  $Na_2O$ ,  $K_2O$ ,  $Ag_2O$ , or alkaline earth oxides  $CaO$ ,  $MgO$ ,  $SrO$  [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.

The mixed glass former (MGF) and mixed glass former effect (MGFE) is defined as a nonlinear and non-additive change in the ionic conductivity with changing glass former composition at constant modifier composition. Potassium-borophosphate glasses are characterized by an interesting structure on account of the presence of two glass-forming components.

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 [3,4,5,6]. Several formulas have been derived to correlate the glass density to the glass composition [7,8,9,10,11]. 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 [12,13]. The density, molar volume and packing fraction [14, 15, 16] could be

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directly related to the short range structure of alkali oxide modified borate densities prove changes in both short glasses 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<sup>[17,18,19,20,21,22]</sup>, is a constitutive-additive property which represents the real volume of the molecules. That is calculated by the Lorenz-Lorentz formula. Electric polarizability<sup>[17-23]</sup>, 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<sup>[17,22,23]</sup>, inter ionic distance<sup>[17,22]</sup>. 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" polarons<sup>[22]</sup>. In present work, these parameters have studied to explain structural features.

## II. EXPERIMENTAL TECHNIQUES

### 2.1. PREPARATION OF GLASSES

The sodium-borophosphate glass samples having the general chemical formula  $42.5\text{K}_2\text{O}-(57.5-x)\text{B}_2\text{O}_3-x\text{P}_2\text{O}_5$ , ( $x=0-57.5$  in the step of 5.75) were prepared by conventional melt-quench technique from high-purity reagent grade  $\text{K}_2\text{CO}_3$ ,  $\text{H}_3\text{BO}_3$  and  $(\text{NH}_4)_2\text{HPO}_4$ . The raw materials were mixed up in the desired proportions and thoroughly ground in an agate mortar. The mixtures were fired in air in a porcelain crucible in the following manner: (a) reaching  $500^\circ\text{C}$  and keeping steady at this temperature for 30 min to allow the evaporation of water from the powders to be as complete as possible; (b) reaching  $800^\circ\text{C}$  and keeping steady at this temperature for 30 min to allow all the gases removal ( $\text{CO}_2$ ,  $\text{NH}_3$ ); (c) reaching  $1100^\circ\text{C}$  depending on composition., 4-5 hrs and keeping steady at this temperature for 30 min to allow a complete homogenization of the melt and attained desirable viscosity it was poured onto metal plate. The prepared samples was then annealed at  $300-400^\circ\text{C}$  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 smoothed 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 MEASUREMENTS

Density of all glass samples are measured at room temperature using toluene as the immersion liquid. Density is generally measured by the fluid displacement method depending on Archimedes principle. According the Archimedes principle, the buoyancy equals the weight of the displaced fluid. Archimedes Principle using toluene as the buoyant medium evaluated the density of the glass samples. The density was obtained by employing the relation:

$$\rho = W_a \rho_b / (W_a - W_b)$$

Where  $W_a$  is the weight of glass sample in air,  $W_b$  is the weight of glass sample in buoyant liquid,  $(W_a - W_b)$  is the buoyancy,  $\rho_b$  is density of buoyant. All the measurements were made using a digital balance.

### 2.3. MOLAR WEIGHT CALCULATIONS

Step I – Calculation of wt/mol: Weight/mole = molar weight of the constituents \* mol% / 100

Step II – Calculation of molecular weight of sample (M)

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

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Step III – Calculation of Molar volume ( $V_m$ ): Using molecular weight and density calculated as from above, the molar volume of the glass samples can be calculated from following expression:  $V_m = M/\rho$

Here,  $V_m$  is molar volume,  $\rho$  is the density of the sample and  $M$  is the molecular weight of the sample.

## 2.4. OXYGEN PACKING DENSITY (O)

The oxygen packing density of the glass samples were calculated using the following relation<sup>[24]</sup>

$$O = n(\rho/M)$$

Where  $\rho$ , is the density of desired glass samples,  $M$ , molecular weight of the sample and  $n$ , 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

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

## 2.6 INTER-IONIC DISTANCE (R)

Inter ionic distance (R) of the glass samples is given as,  $R = (1/N)^{1/3}$

Where  $N$  = ionic concentrations.

## 2.7 PHYSICAL PROPERTIES: RESULTS AND DISCUSSION

Average molecular weight ( $M$ ), Density ( $\rho$ ), Molar volume ( $V_m$ ), Oxygen packing density (O) Ionic concentrations (N), Inter-ionic distance (R) and polaron radius ( $r_p$ ) The mean atomic volume ( $V_M$ ) of each glass was obtained from the values of densities ( $\rho$ ) and mean atomic weights. The calculated values are given 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  $K_2O \cdot B_2O_3 \cdot P_2O_5$  glass system.

Glass system	Average molecular weight M (gm/mol)	Density $\rho$ (gm/cm <sup>3</sup> )	Molar Volume $V_m$ ( cm <sup>3</sup> /mol)	Oxygen packing density O (10 <sup>-6</sup> m <sup>3</sup> /mol)	Ionic Concentration N (10 <sup>21</sup> /cm <sup>3</sup> )	Interionic distance R (A <sup>0</sup> )	Polaron radius $r_p$ (A <sup>o</sup> )
KBP 1	80.0649	2.156	37.136	92.147	6.893	1.132	0.212
KBP 2	84.2236	2.242	37.566	90.377	6.814	1.136	0.213
KBP 3	88.3822	2.26	39.107	88.692	6.546	1.152	0.215
KBP 4	92.5409	2.29	40.411	87.622	6.334	1.164	0.218
KBP 5	96.6995	2.27	42.599	85.036	6.009	1.185	0.222
KBP 6	100.858	2.36	42.737	83.391	5.99	1.186	0.222
KBP 7	105.017	2.28	46.06	82.806	5.557	1.216	0.227
KBP 8	109.175	2.25	48.522	82.322	5.275	1.238	0.231
KBP 9	113.334	2.24	50.596	79.962	5.059	1.255	0.235
KBP 10	117.493	2.28	51.532	79.485	4.967	1.263	0.236
KBP 11	121.651	2.29	53.123	68.28	4.819	1.276	0.239

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Table 2.6.1 indicates the parameters values with increasing the mol% of P<sub>2</sub>O<sub>5</sub> at the cost of B<sub>2</sub>O<sub>3</sub> by keeping modifier K<sub>2</sub>O constant. Average molecular weight, Molar volume, Inter-ionic distance, Polaron radius increasing, Oxygen packing density, Ionic concentrations decreasing and Density, changes monotonically.

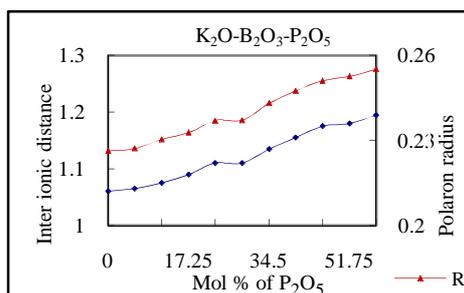


Figure: 2.6.1. Variation of Inter ionic distance polaron radius with mol% of P<sub>2</sub>O<sub>5</sub>

Inter ionic distance & polaron radius varies alike with increasing the mol% of P<sub>2</sub>O<sub>5</sub> (Figures 2.6.1), having linear relation with positive slope and R<sup>2</sup> is equal to 0.988; polaron radius having very small values than that of inter ionic distance. While ionic concentration & inter ionic distance varies reversely (Figures 2.6.2) with increasing the mol% of P<sub>2</sub>O<sub>5</sub> and crosses at 31.625 mol % of P<sub>2</sub>O<sub>5</sub>.

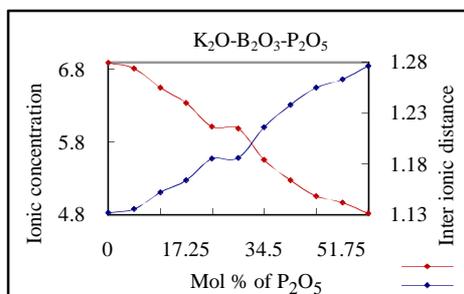


Figure: 2.6.2. Variation of Ionic concentration, Inter-ionic distance with mol% of P<sub>2</sub>O<sub>5</sub>.

Oxygen packing density, ionic concentration decreases, which suggests the increased free space within the glass structure,<sup>[25,26]</sup> it means that the glass structure becomes loosely packed<sup>[27]</sup>. The polaron comprises the electron plus its surrounding lattice deformation. Due to the increasing values of polaron radius the deformation extends over many lattice sites, and the lattice can be treated as a continuum. Average molecular weight, increases linearly as mol% of P<sub>2</sub>O<sub>5</sub> increases gives positive slope and R<sup>2</sup>=1. Transformation of BO<sub>3</sub> triangle units to BO<sub>4</sub> tetrahedral units can be expected to increase the network linkage of the glass which is reflected in the monotonically increasing density. Other physical parameters such as, Refractive index (n) of samples was calculated by using the following relation<sup>[28]</sup>. The dielectric constant was calculated from the refractive index of the glass using<sup>[29]</sup>.

$$\epsilon = n^2$$

The Lorentz-Lorenz equation<sup>[30,31,32,33]</sup> relates molar refraction R<sub>m</sub> to refractive index n and molar volume V<sub>m</sub> of the substance by,

$$R_m = \left[ \frac{(n^2 - 1)}{(n^2 + 2)} \right] V_m \quad \text{Where } V_m \text{ is equal to the molar volume.}$$

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This equation gives the average molar refraction of isotropic substances, i.e., for liquids, glasses and cubic crystals. The Lorentz-Lorenz equation presents the polarizability, i.e., the magnitude of the response of the electrons to an electromagnetic field.

On the other hand, Duffy<sup>[34]</sup> has obtained an empirical formula that relates energy gap  $E_g$  to molar refraction  $R_m$ .

$$R_m = V_m (1 - \sqrt{E_g / 20})$$

The ratio of  $R_m/V_m$  is called polarizability per unit volume. According to the Herzfeld theory of metallization<sup>[35]</sup>, If  $R_m/V_m > 1$  and  $R_m/V_m < 1$  samples predicting metallic or insulating. From Table 1.2 it is clear that present glass samples behave as non-metal. The difference  $M = 1 - R_m/V_m$  is so-called metallization criterion<sup>[36]</sup>. 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  $\alpha_m$  as

$$R_m = 4\pi\alpha_m A_v / 3$$

Where  $A_v$  is Avogadro's number introduced, with  $\alpha_m$  in ( $\text{\AA}^3$ ) this equation can be transformed to,

$$R_m = 2.52\alpha_m$$

Hence molar polarizability  $\alpha_m$  can be calculated.

The electronic polarizability ( $\alpha_e$ ) was calculated using the formula<sup>[37]</sup>.

$$\alpha_e = 3(n^2 - 1) / 4\pi A_v (n^2 + 2)$$

Where,  $A_v$  is the Avogadro number. The measured and calculated values of densities, molar volumes and polarizability of oxide ions of  $P_2O_5$  doped sodium-borate glasses are listed in the Table 2.7.1

Table: 2.7.1 Refractive index, Dielectric constant, Molar refractivity, Electronic polarizability, Metallization, Molar polarizability and Polarizability per unit volume.

Name of samples	Refractive index (n)	Dielectric constant( $\epsilon$ )	Molar Refractivity $R_m(\text{cm}^3)$	Electronic polarizability $\alpha_e * (10^{24} \text{ions/cm}^3)$	Metallization M	Molar polarizability $\alpha_m * (10^{24}/\text{cm}^3)$
KBP.1	2.851	8.128	26.012	0.279	0.300	10.322
KBP.2	2.849	8.117	26.448	0.279	0.297	10.495
KBP.3	2.869	8.231	27.641	0.280	0.293	10.969
KBP.4	2.890	8.352	28.699	0.281	0.290	11.389
KBP.5	2.869	8.231	30.108	0.280	0.293	11.948
KBP.6	2.880	8.294	30.278	0.281	0.292	12.015
KBP.7	2.795	7.812	31.973	0.275	0.306	12.688
KBP.8	2.869	8.231	34.294	0.280	0.293	13.609
KBP.9	2.610	6.812	33.367	0.261	0.341	13.241
KBP.10	2.574	6.625	33.650	0.258	0.348	13.353
KBP.11	2.728	7.440	36.240	0.270	0.318	14.381

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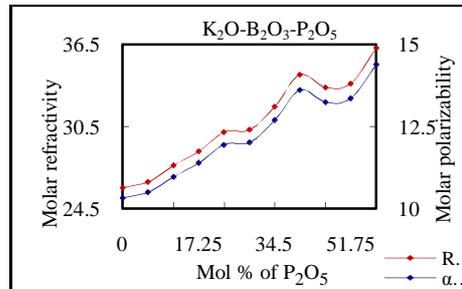


Figure: 2.7.1 Variation in Molar refractivity and Molar polarizability with mol % of P<sub>2</sub>O<sub>5</sub>

Molar polarizability and molar refractivity varies alike with increase of mol % of P<sub>2</sub>O<sub>5</sub>, relation between Molar polarizability and molar refractivity is linear with positive slope and R<sup>2</sup>=1, (Figure 2.7.1).

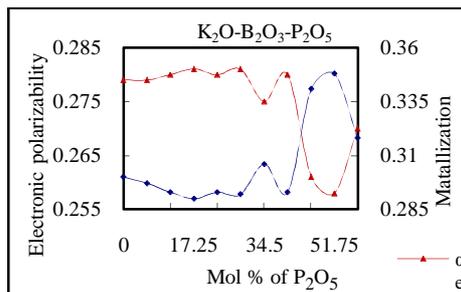


Figure: 2.7.2 Variation in electronic polarizability and metallization with mol % of P<sub>2</sub>O<sub>5</sub>

Electronic polarizability and Metallization varies inversely with increasing mol % of P<sub>2</sub>O<sub>5</sub>. Electronic polarizability is minimum equal to 0.258 and Metallization is maximum equal to 0.348 at 51.75 mol % of P<sub>2</sub>O<sub>5</sub>. There is linear variation between Electronic polarizability & Metallization, having R<sup>2</sup>=0.995.

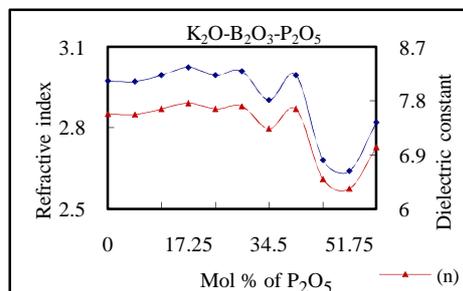


Figure: 2.7.3 Variation in dielectric constant and Refractive index with mol% of P<sub>2</sub>O<sub>5</sub>

Dielectric constant and refractive index varies alike with increasing mol % of P<sub>2</sub>O<sub>5</sub>. Dielectric constant and refractive index is minimum equal to 6.25 and 2.574 at 51.75 mol% of P<sub>2</sub>O<sub>5</sub> this is due to break down of borate & phosphate bonds to create non-bridging oxygen atoms. There is linear variation between Dielectric constant & refractive index having R<sup>2</sup> = 0.999.

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## III. CONCLUSIONS

Increasing the mol% of  $P_2O_5$  with respect to the  $B_2O_3$  by keeping modifier  $K_2O$  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. Deformation extends over many lattice sites and the lattice can be treated as a continuum. Dielectric constant and refractive index varies linearly. Molar refractivity behaviour is similar as that of molar polarizability, they also varies linearly. Electronic polarizability and metallization shows opposite behaviour, is also due to the break down of borate & phosphate bonds to create non-bridging oxygen atoms.

Polarizability per unit volume predicts present glass samples are non-metals. The formation of  $BO_4$  and  $PO_4$  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. Linear trends are found in Molar refractivity & the Molar polarizability, Metallization & Polarizability per unit volume, and in between Mol % of  $P_2O_5$  & Molar volume ( $V_m$ ). A nonlinear and non-additive change with changing glass former composition at constant modifier composition is due to the mixed glass former effect (MGFE).

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