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Refractive Index, Molar Refraction and Polarizability Studies of ZnO-Doped Sodium Phosphate Glasses

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Abstract: Ternary glasses of composition 42.5Na_2O:(57.5-x)P_2O_5:xZnO (x = 0, 0.5, 1, 2, 3, 4 and 5 mol%) were prepared by the conventional melt-quenching technique. X-ray diffraction (XRD) confirmed the amorphous nature of all samples, evidenced by broad diffuse humps and the absence of sharp crystalline peaks over the 20–80° 2q range. The refractive index (n) of each glass was measured using a point laser source (l = 6500 Å) by the ratio of the sine of the angle of incidence to the sine of the angle of refraction. The refractive index increased systematically from 1.340 (x = 0) to a maximum of 1.680 (x = 4 mol%), before showing a slight decrease at x = 5 mol%. Using the experimentally obtained refractive index and the measured molar volume, the molar refraction (RM), specific refractivity of the isotropic dielectric (e), molecular dipole polarizability (a), and the number of molecular chains (N) were calculated using the Lorentz–Lorenz and related formalisms. Molar refraction and polarizability were found to increase with increasing ZnO content (9.109 to 17.188 (mol.cm)-1 and 3.612 to 6.816 ×10-23 cm3, respectively), while the number of molecular chains decreased slightly. These trends are interpreted in terms of the higher polarizability of the Zn2+ modifier ion relative to Na+, and and the corresponding network expansion and increase in non-bridging oxygen content with ZnO substitution.

Keywords: *Sodium Phosphate Glass; ZnO; Refractive Index; Molar Refraction; Polarizability; Lorentz–Lorenz Equation.*

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I. INTRODUCTION

The optical properties of oxide glasses, particularly the refractive index and the related quantities of molar refraction and electronic/ionic polarizability, are of both fundamental and applied interest, since they are directly governed by the polarizability of the constituent ions and the compactness of the glass network. In phosphate glass systems, the substitution of a modifying oxide for the glass-forming oxide (P_2O_5) alters the non-bridging oxygen (NBO) population and hence the polarizability of the surrounding oxide ions, which is directly reflected in the refractive index and molar refraction of the glass (Tomihara et al., 2020; Algradee et al., 2022). Zinc oxide is a widely used intermediate oxide in phosphate glass formulations because the Zn^{2+} ion, owing to its relatively high field strength and polarizability compared to alkali ions such as Na^+, can significantly alter the optical basicity, refractive index, and non-linear optical response of the host glass (Mondal et al., 2020; El-Mallawany et al., 2020; Rammah et al., 2020). The molar refraction and polarizability, calculated from the refractive index and molar volume via the Lorentz–Lorenz relation, provide a quantitative measure of this effect and have been used extensively to interpret structural changes in ZnO- and other modifier-doped phosphate glass systems (Ahmed et al., 2020; Meena et al., 2023). In the present work, the refractive index, molar refraction, specific refractivity of the isotropic dielectric, dipole polarizability, and number of molecular chains of 42.5Na_2O:(57.5-x)P_2O_5:xZnO glasses (x = 0–5 mol%) are evaluated and discussed in relation to the compositional substitution of ZnO for P_2O_5.

II. EXPERIMENTAL

➢ *Materials and Method of Preparation*

High-purity (99.9%) Na_2CO_3, P_2O_5, and ZnO were used as starting materials. A series of ternary sodium phosphate glasses with the nominal composition 42.5Na_2O:(57.5-x)P_2O_5:xZnO (x = 0, 0.5, 1, 2, 3, 4, 5 mol%) were prepared by the conventional melt-quenching technique. Batch compositions of 10–15 g were prepared by accurately weighing the requisite amounts of the starting materials according to their predetermined molar proportions using a monoplane balance. The weighed powders were thoroughly homogenized and subsequently transferred to a high-quality porcelain crucible for melting in an electrically heated furnace. The samples were heated at a controlled rate, and the furnace temperature was maintained approximately 20 K above the corresponding melting temperature, within the range of 800–

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900 °C, for 40 min to ensure complete melting and adequate homogenization of the glass melt. Following complete homogenization, the molten glass was rapidly quenched between two polished aluminium blocks to obtain transparent, bubble-free glass discs. This conventional melt-quenching technique is a highly reliable and widely adopted route for synthesizing homogeneous, non-crystalline phosphate-based glassy matrices [2,3]. The resulting samples were approximately 2 cm in diameter and 0.1–0.2 cm in thickness. No visible contamination or reaction products originating from the aluminium blocks were observed on the prepared glass specimens. Porcelain crucibles were used in preference to platinum to avoid extended cleaning times, with precautions taken to prevent crucible-derived impurities from entering the melt. For refractive index measurement, samples were polished on both faces.

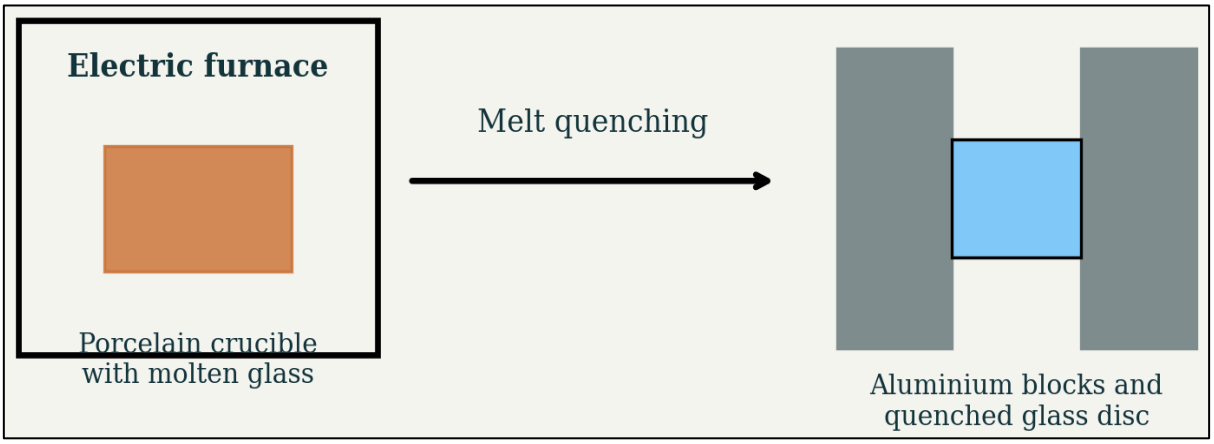

Fig 1 Schematic Representation of the Melt-Quenching Procedure Used to Prepare the Glass Discs.

➢ *X-ray Diffraction (XRD)*

The XRD patterns of the prepared glass samples was examined using a Seifert XRD 3003 T/T diffractometer with Cu Kα radiation (λ = 1.541 Å) and a nickel filter, revealed a characteristic amorphous profile for all investigated compositions. Instead of exhibiting well-defined Bragg reflections, the diffraction patterns consisted of broad, diffuse halos in the (2θ) range of approximately 25–30°, which originate from the short-range atomic ordering of the phosphate-tetrahedral backbone in the absence of long-range periodic lattice planes [2,3]. The absence of sharp diffraction peaks indicates that the samples lack long-range structural periodicity, confirming their amorphous nature [2,3]. The persistence of this diffuse scattering feature across all ZnO concentrations further suggests that the incorporation of ZnO up to 5 mol% does not induce detectable crystalline phase formation within the phosphate glass matrix. Thus, the XRD results confirm the successful formation of a homogeneous amorphous glass network for all compositions studied.

➢ *Refractive Index Measurement*

The refractive index of each polished glass sample was determined using a point laser source (λ = 6500 Å, red light). The angle of incidence in air (θ_{air}) and the angle of refraction within the glass (θ_{glass}) were measured geometrically, and the refractive index was calculated as:

$$n = \frac{sin\theta air}{sin\theta glass} \quad (1)$$

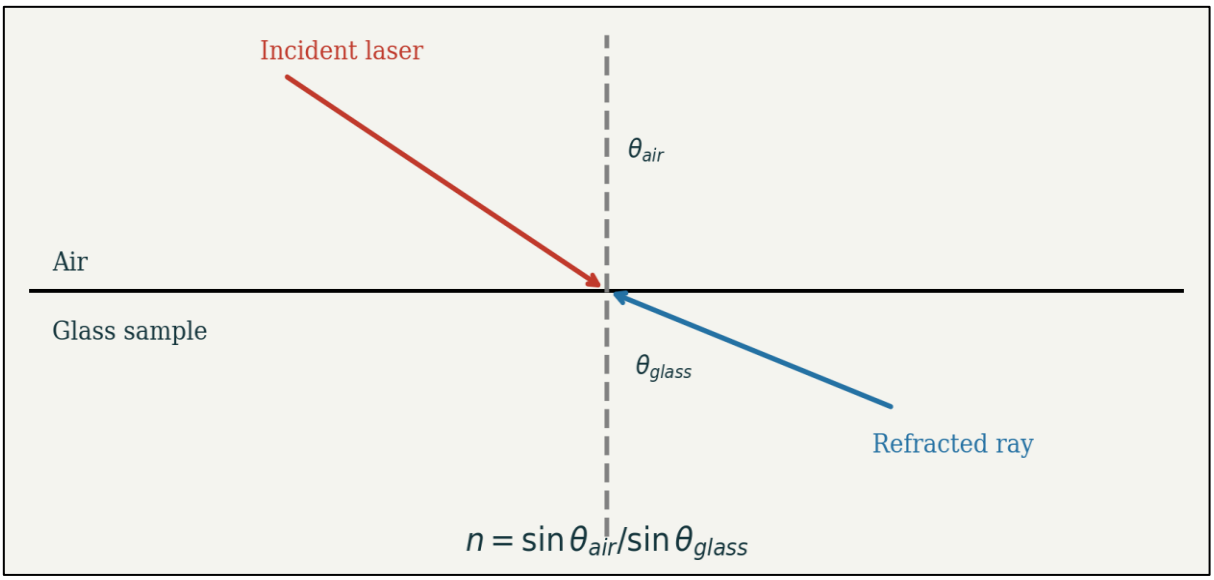

Fig 2 Principle of Refractive-Index Determination Using the Measured Incidence and Refraction Angles.

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➢ *Calculation of Molar Refraction, Polarizability and Related Parameters*

Using the experimental refractive indices (n) and the measured molar volumes (V), the molar refraction (R_M) was calculated from the Lorentz–Lorenz relation [1,4]:

$$R_M = \frac{(n2-1)}{(n2+2)} \times V \quad (2)$$

The specific refractivity of the isotropic dielectric (ε) was obtained from the de Vries relation, (n2 -1)/(n2 + 2) = εd, where d is the glass density. The mean molecular dipole polarizability (α), representing the dipole moment induced by the electric field, was calculated using the Lorentz–Lorenz formula (n2 - 1)/(n2 + 2) = 4πNα/3, and the number of molecular chains (N) was obtained from $N = N_A d/M$, where N_A is Avogadro's number, d is density and M is molecular weight.

III. RESULTS AND DISCUSSION

➢ *Structural (XRD) Confirmation*

XRD patterns recorded for representative compositions (x = 0.5 and x = 3 mol% ZnO) showed only broad diffuse humps cantered near 2θ » 25–30°, with no sharp Bragg reflections across the full 20–80° 2θ range, confirming the amorphous nature of the glass network for all prepared compositions. This structural confirmation underpins the optical property measurements discussed below, since the refractive index and Lorentz–Lorenz-derived quantities reported here are valid only for a homogeneous, isotropic (i.e., non-crystalline) glass network.

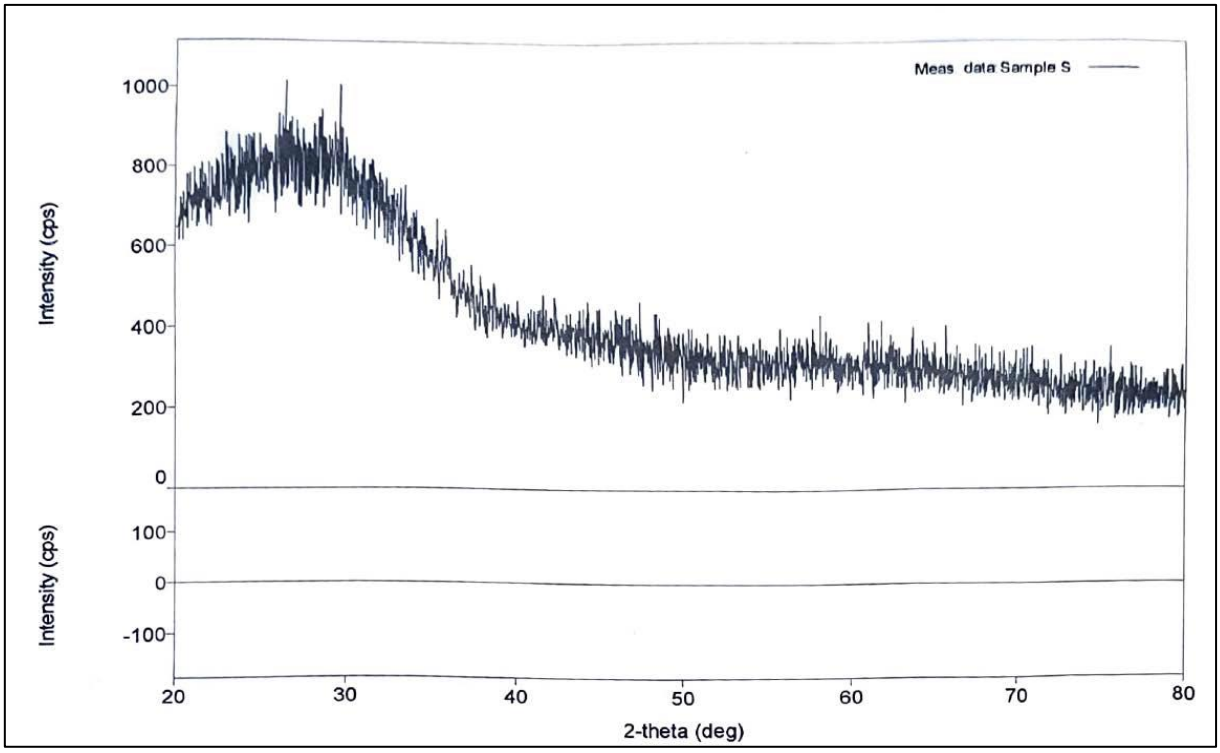

Fig 3 XRD Pattern of Sample Na 42.5%:P 54.5%: Zn 3%.

➢ *Density, Molar Volume, Refractive Index and Related Optical Parameters*

Table 1 summarizes the density, molar volume, refractive index, polarizability, molar refraction, isotropic dielectric constant, and number of molecular chains for all seven glass compositions.

Table 1 Density, Molar Volume, Refractive Index, Polarizability, Molar Refraction, Isotropic Dielectric Constant and Number of Molecular Chains of $42.5Na_2O:(57.5-x)P_2O_5:xZnO$ Glasses.

Composition (mol% ZnO)	Density (g/cm3)	Molar Vol. (cm^3/mol)	R.I. (n)	Polariz. ($x10^{-24}$ cc)	Molar Refr. (R_m)	Isotropic Dielectric	N $x10^{22}$
0	2.4842	43.458	1.340	3.612	9.109	0.0844	1.386
0.5	2.4264	44.368	1.462	4.836	12.195	0.1132	1.358
1	2.4037	44.668	1.462	4.869	12.278	0.1144	1.348
2	2.3805	44.842	1.536	5.543	13.976	0.1309	1.343
3	2.3500	45.167	1.618	6.276	15.824	0.1490	1.334
4	2.3203	45.484	1.680	6.816	17.188	0.1629	1.324
5	2.3075	45.547	1.667	6.724	16.954	0.1616	1.322

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Unlike binary sodium phosphate glasses where the addition of modifier oxides typically increases density and contracts molar volume [9], the current ternary system exhibits a continuous density decrease and molar volume increase as ZnO substitution increases. This behaviour reflects a localized structural network expansion, where Zn^{2+} incorporation deforms and expands the interstitial cavities of the sodium ultra phosphate matrix [3,4].

➢ *Refractive Index*

The refractive index increases from 1.340 at x = 0 mol% ZnO to a maximum of 1.680 at x = 4 mol%, followed by a slight decrease to 1.667 at x = 5 mol%. This overall increasing trend is consistent with the progressive replacement of P_2O_5 by the more polarizable ZnO. This refractive index enhancement is directly governed by the ratio of the total electronic polarizability of the oxide ions to the molar volume of the glass network [3,4] and mirrors the refractive-index behaviour reported for other ZnO-modified phosphate glass systems, where increasing modifier-oxide content raises the electronic polarizability of the network and, correspondingly, the refractive index [5,3]. Furthermore, oxide glasses synthesized with high refractive indices are highly sought after for optoelectronic applications, such as radiation shielding, optical fiber amplifiers and photonic devices [2,6].

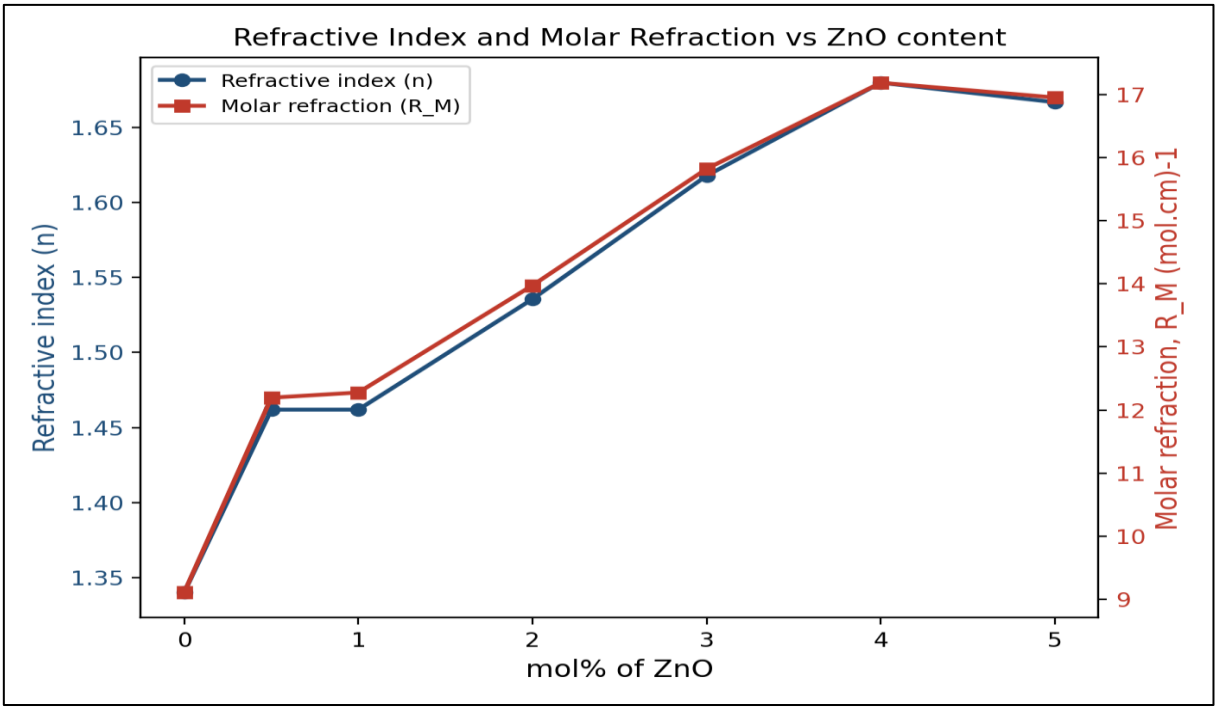

Fig 4 Refractive Index and Molar Refraction of $42.5Na_2O:(57.5\text{-}x)P_2O_5:xZnO$ Glasses as a Function of ZnO Content.

➢ *Molar Refraction and Polarizability*

Both molar refraction (RM) and and dipole polarizability (a) increase monotonically with ZnO content, from 3.612 to 6.816 $\times 10^{-24}$ cm^3, respectively, over the 0–4 mol% range.This behavior is attributed to the creation of highly polarizable non-bridging oxygen (NBO) atoms that depolymerize the metaphosphate chains, modifying the single bond strength and transferring the bond nature from a covalent to a more ionic form [1,4]. This is attributed to the higher electronic polarizability of the Zn2+ modifier ion relative to Na+ [1,4], and to the increasing molar volume (network expansion) observed with ZnO substitution, both of which increase molar refraction through the Lorentz–Lorenz relation. A similar correlation between polarizability, refractive index and molar volume has been reported for zinc-containing phosphate and phospho-tellurite glass systems [1,2,7].

➢ *Isotropic Dielectric Constant and Number of Molecular Chains*

The specific refractivity of the isotropic dielectric (ε) increases from 0.0103 to 0.1629 over the composition range, consistent with the increase in both refractive index and polarizability [2,4]. The number of molecular chains (N), calculated from the density and molecular weight, decreases slightly from 1.386 $\times 10^{22}$ to 1.322 $\times 10^{22}$, reflecting the increase in average molecular weight and molar volume per structural unit as ZnO progressively replaces P_2O_5. It should be noted that the slight decrease in refractive index, molar refraction, and isotropic dielectric constant observed at x = 5 mol% (relative to x = 4 mol%) breaks the otherwise monotonic trend; this should be verified against raw angle-of-refraction measurements before being treated as a genuine compositional effect rather than an experimental deviation.

IV. CONCLUSION

The refractive index, molar refraction, polarizability, isotropic dielectric constant, and number of molecular chains of $42.5Na_2O:(57.5\text{-}x)P_2O_5:xZnO$ glasses (x = 0–5 mol%) were evaluated. Refractive index, molar refraction and polarizability all increase with increasing ZnO substitution for P_2O_5 up to 4 mol%, consistent with the higher polarizability of Zn2+ relative to Na+ and the accompanying network expansion. These results are in agreement with optical trends

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reported for related ZnO-modified phosphate glass systems and add a new composition data set within the sodium ultra phosphate region.

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