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ABSTRACT

Glasses with compositions xPbO-RNa₂B₄O₇-(100-*R*-*x*)CAS (0.49CaO, 0.21Al₂O₃, 0.3SiO₂) with $0 \le x \le 50$ and $50 \le R \le 75$ mol% have been prepared using the normal melt quenching technique. The optical transmittance and reflectance spectrum of the glasses have been recorded in the wavelength range 300–1100 nm. The values of the optical band gap E_g^{opt} for indirect transition and refractive index have been determined for different compositions of the amorphous glass. The average electronic polarizability of the oxide ion α_0^{2-} and the optical basicity have been estimated from the values of calculated refractive indices. The compositional dependence of the different physical parameters such as the density, the optical band gap, the refractive index, the average electronic polarizability of the oxide ion and the optical basicity on PbO content have been analyzed and discussed.

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1. Introduction

Borosilicate glasses are widely used in a wide range of technological applications, from chemically resistant containers and piping to fiber composites, and from pharmaceutical, and sealing glasses to nuclear waste immobilization. The controlled heat treatment of borosilicate glasses especially those which have domain sizes in suboptical scale is interesting in controlling and designing the physical properties such as chemical durability, crystal nucleation rates, and high-temperature strength, and is of interest in some natural magmatic systems as well. Due to these interesting physical properties, borosilicate glasses can be used as laser host matrices after doping with rare earth oxides [1–8].

On the other hand, lead borosilicate glasses are chemically stable over a wide composition range, possess low melting points, a desirable electrical resistivity, and are considered as a basis for the manufacture of various coatings, semiconductor microelectronics, optical lenses, enamels, solder glasses, glass-ceramic cements, and as hard nuclear waste materials [9–12].

Therefore, the structure of silicate groups formed on the incorporation of SiO_2 in alkali borate glasses has been found to depend on the concentration and distribution of a variety of units containing BO_3 triangles, BO_4 tetrahedra that are bonded to a variety of cations of modifying oxides by non-bridging oxygens (NBOs) or by bridging oxygens (BO). Higher NBOs contents generally lower the viscosity of the precursor glass melt but

contribute to ease of corrosion; higher BO contents often lead to harder, more durable glasses that must be melted and worked at higher temperature [2,13].

Based on different studies, it has been established that the mutual influence between the coordination types of Si in lead borosilicate glasses were attributed to two reasons. Formation of Si–O–B linkages and increase in concentration of Q⁴ structural units of silicon (where Qⁿ represents silicon structural units having *n*-number of bridging oxygen atoms). At an intermediate range of PbO content in the lead borosilicate glasses, the mesounits (which are attributed to boroxol or metaborate rings) show a rapid decline in the amount of PbB₃O₆ units. This process was expected because of the continuous coupling of the silicate units with the borate ones, which removes the likelihood of formation of mesounits with three borate units. Hence, the decline of pure borate units is accompanied by the formation of B–Si mixed groupings [14–21].

The real UV transmission of borosilicate glasses is limited by extrinsic charge transfer and $s \rightarrow p$ absorption bands due to metal ions such as Pb^{2+} , etc., which are influenced by the glass matrix. It was shown that both intrinsic and extrinsic absorption are important for elucidating UV radiation-induced defects in the glasses (View Record in Scopus: cited in Scopus (57)) [22]. Also, the refractive index is closely related to the electronic polarizability of ions and the local field inside the material. Thus, it is a key parameter for optical device design [23].

On the basis of the aforementioned aspects, $xPbO-RNa_2B_4O_7-(100-R-x)CAS$ with $0 \le x \le 50$ and $50 \le R \le 75$ mol% glasses have been synthesized to explore the relationship between the structure and the macroscopic behavior of the glass for the design of materials suitable for specific applications. CAS # 0.49CaO,



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 $0.21 A l_2 O_3, \ 0.3 SiO_2. \ UV$ spectroscopy have been used for the present investigations.

2. Experimental procedures

Glass samples with the formula $xPbO-RNa_2B_4O_7-(100-R-x)$ CAS with $0 \le x \le 50$ and $50 \le R \le 75$ mol% were prepared by the melt-quenching technique. Required quantities of Analar grade $Na_2B_4O_7 \cdot 10H_2O$, PbO, CaCO₃, SiO₂, Al₂O₃ and were mixed together by grinding the mixture repeatedly to obtain a fine powder. The mixtures were melted in a platinum crucible in an electrically heated furnace under ordinary atmospheric conditions at a temperature of about 1423 K for 4 h to homogenize the melt. This temperature was sufficient to obtain clear, bubble-free melts for all glass compositions. The glass formed by quenching the melt on a pre-heated stainless-steel mold was immediately transferred to a muffle furnace where it was annealed at a temperature below the glass-transition temperature for 2 h. The annealing temperature was 623 K [9–12,22,23].



Fig. 1. Spectral behavior of transmittance and reflectance of the glass system $xPbO-RNa_2B_4O_7-(100-R-x)CAS$ with $0 \le x \le 50$ and $50 \le R \le 75$ mol%: (a) Y=75 mol% and (b) Y=50 mol%.

The composition refers to the nominal composition (the starting mixture), where the weight losses were found to be less than 0.5%. The obtained glasses were then grounded and optically polished to have suitable dimensions $(1 \times 1 \times 0.15 \text{ cm}^3)$ for the present optical measurements. Non-parallelism of the two opposite side faces was less than 0.01 mm.

X-ray diffraction patterns were recorded to check the noncrystallinity of the glass samples using a Philips X-ray diffractometer PW/1710 with Ni-filtered, Cu-K α radiation (λ =1.542 Å) powered at 40 kV and 30 mA.

The density ρ of the glass samples was determined using Archimedes technique by using toluene as an immersion fluid. At least, three samples of each glass were used to determine the density. A random error in the density values was found to be $\pm 1\%$.

The transmittance (*T*), and the reflectance (*R*) optical spectra of the glasses (Fig. 1), were recorded at room temperature in the wavelength range 300-1100 nm using a computerized double beam spectrophotometer, type SHIMADZU UV-2100.

3. Results and discussion

3.1. Density and molar volume

Density is a useful parameter to measure when investigating changes in the structure of glasses, as it is affected by structural softening/compactness, changes in geometrical configuration, coordination number, cross-link density and the dimensions of interstitial spaces in the structure.

It was found in the studied glass system that the density, as shown in Fig. 2, increases with increasing PbO content. The density of $75Na_2B_4O_7$ -25PbO and $50Na_2B_4O_7$ -50PbO glasses agree well with that reported elsewhere [24,25]. At the first sight, the increase in the density can be related to the replacement of SiO₂, CaO, and Al₂O₃ with oxide of greater molecular mass PbO, i.e., the increase in the density may be related to either the constitution of the glass or to the structural variation in the type of borate structural units. However, it must be indicated that is not the real reason in all cases. For example in Li₂O-B₂O₃ [26] or Li₂O-SiO₂ [27] glasses the density increases markedly with increasing Li₂O content at the expense of B₂O₃ or SiO₂, which



Fig. 2. Density ρ of *x*PbO–*R*Na₂B₄O₇–(100–*R*–*x*)CAS with $0 \le x \le 50$ and $50 \le R \le 75$ mol% in dependence of the PbO content. Lines are fitting plots of the data. A random error in the density values was found as $\pm 1\%$.

have greater molecular mass. The reason of density increase in those cases is related to the type of structural unites that form when Li₂O is incorporated into the glass structure. Depending on the composition of Li₂O-B₂O₃ glasses, Li₂O converts symmetric BO₃ triangles into BO₄ tetrahedra or converts the latter into asymmetric BO₃ triangles. Both the BO₄ tetrahedra and asymmetric BO₃ triangles are considerably denser than the symmetric BO₃ triangles [26]. In Li₂O-SiO₂ glasses, SiO₄ tetrahedra having one, two, three or four non-bridging oxygen ions are denser than those without non-bridging oxygen ions [27].

The increase of the density as shown in Fig. 2 can be explained by considering formation of BO_4 units due to introducing PbO in the borosilicate matrix. A compensation of the negative charge on the BO_4 tetrahedra would be verified from positively charged structural defects in the PbO₄ network or from one of the positive charges of Si⁴⁺, or Ca²⁺, or Al³⁺ cations for each BO₄ tetrahedron. The increase in density reveals that the former type of BO₄ units is denser than the latter.

3.2. Determination of optical band gap

The optical absorption coefficient, α , of a material can be evaluated from the optical transmittance and reflectance using the relation

$$\alpha = \frac{1}{d} \ln \frac{(1-R)}{T} \tag{1}$$

where *d* is the thickness of the sample, while the absorption coefficient $\alpha(v)$ as a function of photon energy (hv) for direct and indirect optical transitions, according to Pankove [28], is given by

$$\alpha h v = A_0 (h v - E_g^{opt})^p \tag{2}$$

For amorphous materials indirect transition is valid according to the Tauc relations [29], i.e., the power part p=2; hence, the values of indirect optical band gap energy (E_g^{opt}) can be obtained from Eq. (2) by extrapolating the absorption coefficient to zero absorption in the $(\alpha hv)^{1/2} - hv$ plot as shown in Fig. 3. The respective values of E_g^{opt} are obtained by extrapolating to $(\alpha hv)^{1/2}=0$ for the indirect transitions [29,30]. A plot of E_g^{opt} against PbO content shows that E_g^{opt} decreases linearly with an increase in the PbO content as shown in Fig. 4.

The decreasing values of E_g^{opt} upon increasing the PbO content can be understood in terms of the structural changes that are taking place in the studied glass systems. Addition of network modifiers (alkali/alkaline earth metal oxides) to borosilicate glasses results in the initial conversion of BO₃-BO₄ structural units [14,15]. With further increase in modifier concentration, BO_4 structural units in the glass are replaced by BO_3^- structural units (planar BO₃ structural units with one non-bridging oxygen atoms). Therefore, the structural units of lead borosilicate glasses may be formed from trigonally coordinated boron (BO₃), tetrahedrally coordinated boron (BO₄), silicon atoms with 3 and 4 bridging oxygen atoms, and Q^n units with Si-O-B/Si-O-Al linkages. Thus, the borate and silicate structures in the network may interact strongly, and mixed randomly with or without the presence of a modifier like PbO, with its two structural units PbO₄ and PbO₆ [16,17]. Therefore, as the PbO content increases in the studied borosilicate glasses, the BO₃ units will be destructed, and is converted into asymmetric BO₄ units associated with NBOs, while the interatomic distances may be increased. The Pb^{2+} ions may take up positions in the voids of the silicon-oxygen framework, and when all voids in the silicon-oxygen network are filled, the "excess" Pb^{+2} ions promote depolymerization of the network by creating isolated SiO₄ units [9,10,31]. The increase of PbO content will be accompanied by an increase in the



Fig. 3. Tauc's plots of the glass system $xPbO - RNa_2B_4O_7 - (100 - R - x)CAS$ with $0 \le x \le 50$ and $50 \le R \le 75$ mol%: (a) Y=75 mol% and (b) Y=50 mol%.

degree of ionic bonding in the glass which increases the anharmonism of atom vibrations. Thus, the lead became increasingly incorporated into borosilicate network via B–O–Pb, and Si–O–Pb bonding, and weaker Pb–O bond (low bond strength) replaces B–O bond (high bond strength) [32] in the glass matrix upon increasing the PbO content. These factors will decrease the values of E_g^{opt} .

3.3. Determination of the refractive index

According to the theory of reflectivity of light, the refractive index (n) as a function in the reflectance (R) and the extinction coefficient (k) can be determined by the quadratic equation

$$R = \frac{(1-n)^2 + k^2}{(1+n)^2 + k^2} \tag{3}$$

The extinction coefficient can be computed based on the wavelength (λ) and the calculated values of (α) according to the relation

$$\alpha = \frac{4\pi k}{\lambda} \tag{4}$$

It is clear that, the refractive index as shown in Fig. 5 decreases with increasing the wavelength of the incident photon, and on the contrary increases with the increase in the PbO content. Thus,



Fig. 4. Dependence of the indirect optical band gap of the glass system xPbO– $RNa_2B_4O_7-(100-R-x)CAS$ with $0 \le x \le 50$ and $50 \le R \le 75$ mol%: (a) Y=75 mol% and (b) Y=50 mol%.

the dependence of the refractive indices on the PbO content of the studied glasses can be explained as follows. According to the Lorentz–Lorenz [23,33] equation, the density of the material affects the refractive index in a direct proportion. Also, there are some factors influences the increase of the refractive index such as

- (i) The coordination number, Z, of the studied glasses. Introducing PbO causes as discussed previously, the increase in the coordination number of the glasses, as the BO₄ units will be increased and creating more NBOs. This in turn leads to an increase in the average coordination number of the studied glasses, which increases the refractive index [34].
- (ii) The creation of NBOs creates more ionic bonds which manifest themselves in a larger polarizability over the mostly covalent bonds of bridging oxygen providing a higher index value.

3.4. Polarizability of the glasses

The average electronic polarizability of ions is considered to be one of the most important properties of the materials, which is closely related to their applicability in the field of optics and electronics. It was found that, the optical non-linearity is caused by the electronic polarization of a material upon its exposure to intense light beams, thus, the non-linear response of the material is governed by the



Fig. 5. The refractive indices of the glass system $xPbO-RNa_2B_4O_7-(100-R-x)CAS$ with $0 \le x \le 50$ and $50 \le R \le 75$ mol%: (a) Y=75 mol% and (b) Y=50 mol% as a function of the wavelength.

electronic polarizability [35]. For this purpose, materials with higher optical non-linearity have to be found or designed on the basis of the correlation between the optical non-linearity and with some other easily understandable and accessible electronic properties.

For isotropic substance such as glasses, the average molar refraction (R_m) was given by the Lorentz–Lorenz equation

$$R_m = \left[\frac{n^2 - 1}{n^2 + 2}\right] \frac{M}{\rho} = \left[\frac{n^2 - 1}{n^2 + 2}\right] V_m \tag{5}$$

where the quantity $n^2 - 1/n^2 + 2$ is called the reflection loss [33]. The molar refraction is related to the structure of the glass and is proportional to the molar electronic polarizability of the material, (α_m) in cm³ × 10⁻²⁴ through the Clausius–Mosotti relation

$$\alpha_m = \left(\frac{3}{4\pi N}\right) R_m \tag{6}$$

where *N* is Avogadro's number. For various ternary oxide glasses with the general formula $y_1A_pO_q-y_2B_rO_s-y_3C_nO_m$, where *y*'s denote the molar fraction of each oxide, the electronic polarizability of oxide ion can be calculated on the basis of the values of the refractive indices using the following equations:

$$\alpha_0^{2-}(n_0) = \frac{\left[\frac{R_m}{2.52} - \sum \alpha_{cat}\right]}{(N_0^{2-})} \tag{7}$$



Fig. 6. The oxide ion polarizability $\alpha_o^{2-}(n)$ of the glass system xPbO-RNa₂B₄O₇-(100-R-x)CAS with $0 \le x \le 50$ and $50 \le R \le 75$ mol%: (a) Y=75 mol% and (b) Y=50 mol% as a function of wavelength in a wavelength range from 400 to 1100 nm.

where $\sum \alpha_{cat}$ denotes the molar cation polarizability given by $y_1p\alpha_A+y_2r\alpha_B+y_3n\alpha_c$ and N_0^{2-} denotes the number of oxide ions in the chemical formula given by $y_1q+y_2s+y_3m$. For the studied glasses, the values of $\sum \alpha_{cat}$ are given by $y_1q+y_2s+y_3m$ and the value of N_0^{2-} equals to 3.25. The values of $\alpha Ca=0.469$ Å³ for Ca²⁺ ions, of $\alpha Al=0.054$ Å³ for Al³⁺ ions, of $\alpha Pb=3.623$ Å³ for Pb²⁺ ions, of $\alpha Si=0.033$ Å³ for Si⁴⁺ ions and of $\alpha B=0.002$ Å³ for B³⁺ [36]. The wavelength dependence of electronic polarizability of the studied glasses is shown in Fig. 6. Therefore, the increase in PbO content causes an increase in the oxide ion polarizability along with an increase in the refractive index. Such behavior could be attributed to the increase in molar refraction which is giving rise to an increase in the measured refractive indices with increasing the electronic polarizability of ion oxide.

3.5. Optical basicity of the glasses

The optical basicity proposed by Duffy and Ingram [37,38] was used as a measurement of acid-base properties of the oxide glasses and is expressed in terms of the electron density carried by oxygen. Simultaneously, an intrinsic relationship exists between the electronic polarizability of the oxide ions and the



Fig. 7. Oxide ion polarizability $\alpha_0^{2-}(E_0)$ and molar cation polarizability $\sum \alpha_{cat}$ for the glass system *x*PbO–*R*Na₂B₄O₇–(100–*R*–*x*)CAS with $0 \le x \le 50$ and $50 \le R \le 75$ mol%: (a) *Y*=75 mol% and (b) *Y*=50 mol%.

optical basicity of the oxide medium. The increase in the oxide ion polarizability means stronger electron donor ability of the oxide ions and vice versa. Thus, in the present work, the average electronic polarizability of the oxide ions and the optical basicity of the oxide medium have been calculated on the basis of the property of light represented by the refractive indices of the studied glasses.

The theoretical optical basicity A_{th} can be calculated using basicities assigned to the individual oxides on the basis of the following equation proposed by Duffy and Ingram [37,38]:

$$A_{th} = X_1 A_1 + X_1 A_1 + \dots + X_n A_n \tag{8}$$

where $X_1, X_2, ..., X_n$ are equivalent fractions based on the amount of oxygen each oxide contributes to the overall glass stoichiometry and $A_1, A_2, ..., A_n$ are the basicities assigned to the individual oxides. In our present study, the values of $A_{CaO} = 1$, $A_{Al_2O_3} = 0.6$, $A_{PbO} = 0.95$, $A_{SiO_2} = 0.48$ and $A_{B_2O_3} = 0.42$ are used [36].

An alternative approach for the calculation of the optical basicity is the determination of the state of polarization of oxide ions in a glass matrix on the basis of refraction data. Duffy [39] has established the following correlation:

$$A = 1.67 \left(1 - \frac{1}{\alpha_o^{2^-}} \right)$$
(9)

The *A* values were estimated from the α_0^{2-} using Eq. (9) and are shown in Fig. 7. According to this relationship, an increase in the polarizability, will increase the optical basicity which in turn leads to an increase in the refractive index. Because PbO have a higher polarizability and higher optical basicity (due to its larger ionic radii) than that of the other oxides in the glass matrix. therefore, an increase in the high optical basicity means an increased ability of oxide ions to transfer electrons to the surrounding cations, i.e., a high electron donor ability of the oxide ions.

4. Conclusions

Increasing the PbO content at the expense of CAS in the studied glass system, reveals some remarkable features.

The density of the glass increases, which is due to the larger atomic weight of PbO compared with that of the other oxides in the glass matrix. As the PbO content increases, more NBOs were created and the bond-length of the borate structural units increase which in its turn increase the refractive indices, the polarizability, and the optical basicity of the studied glasses. Also, the observed decrease in the optical band gap was related to the weaker bond strength of the Pb-O compared with that of B-O and Si-O.

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