

Glass Parameters Affecting Radiative Lifetime for Electronic Transitions of Luminescent Dopants: Case of Magnesium Phosphate Glass

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Abstract Spectroscopic properties for $P_2O_5(0.55-x)-MgO(0.45)-xBi_2O_3$ glass system were studied, where the range of molar fraction was $0 \leq x \leq 0.07$ mol%. Glass series were prepared by melt quenching technique to be used as a host glass for upconversion applications. Both density and molar volume were determined. The structural changes have been studied by FT-IR spectroscopy. The adding up of Bi_2O_3 decreases the P—O—P covalent bond by forming P—O—Bi bonds which were associated with a conversion from Bi^{3+} into Bi^{6+} ions. The calculated optical dielectric constant was increased with increasing bismuth oxide contents due to the high polarizing power of Bi^{3+} and Bi^{6+} ions and their small field strength. The different factors that affect the radiative lifetime of electronic transitions for luminescent dopant ions impeded in the host glasses were studied as well.

Keywords Glass-ceramics, Spectroscopy, Refractive index

1. Introduction

Phosphate glasses have different applications in many modern technologies, e.g., photonics, fast ion conductors [1-3], and biomedical engineering [4]. The glasses possess interesting properties such as low glass transition temperature (T_g), low melting temperature, high thermal expansion coefficient and biocompatibility [5]. Depending on oxygen to phosphate ions ratio, the glass structure can be controlled.

Bi_2O_3 as a heavy metal oxide could change the physical properties of phosphate glasses [6]. Phosphate glass containing Bi_2O_3 would undergo structural modification when Bi^{3+} cation switched to stable linkage like P-O-Bi [7]. Bismuth based glasses have important applications in optoelectronic devices, sensors and superconductors [8-11]. Since the release of the first report on 1.3 μm optical amplification in the bismuth-doped silica glasses [12] and that on broadband infrared emission properties of bismuth-doped phosphate glasses, the bismuth-doped materials have attracted much attention. Magnesium bismuth phosphate glasses showed potential usefulness as light emitting devices [2].

The paper is prepared to determine the substitution

effects of replacing P_2O_5 by Bi_2O_3 on the spectroscopic properties and on the structure of magnesium phosphate host glasses. The results would support the use of the present glasses in advanced light emitting devices by controlling the radiative lifetime for luminescent dopant ions [1, 14].

2. Experimental Work

2.1. Glass Preparation

A series of Bismuth Magnesium Phosphate glasses of composition $P_2O_5(0.55-x)-MgO(0.45)-xBi_2O_3$ are prepared, where $0 \leq x \leq 0.07$ mol.%, where x is the oxide mol fraction. The used materials were of chemically pure grade, in the form of $NH_4H_2PO_4$, MgO , and Bi_2O_3 . The amount of the glass batch was 50 g melt⁻¹. The glass was prepared by melt quenching technique using porcelain crucibles in an electric furnace. The temperature of melting was 1100°C, whereas the duration of melting was one hour after the last traces of batches disappeared. To avoid the presence of bubbles in the glass melt, the melt was stirred continuously during the glass preparation.

The melt was poured onto stainless steel mould and annealed to around 350°C to remove the thermal strains. Optical slabs were prepared by grinding and polishing of the prepared samples with paraffin oil and minimum amount of water. The thickness of the glass slabs was about 3 mm. Polishing was completed with stannic oxide and paraffin oil

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to reach a minimum surface roughness that was tested by an interferometric method. The homogeneity of the glasses was examined using two crossed polarizers.

2.2. Density Measurements

The density of the prepared glasses, ρ , at room temperature was measured by the Archimedes principle using a sensitive microbalance with xylene as an immersion liquid. The error in measuring the glass density was ± 0.001 g/cm³.

2.3. Infrared Fourier-Transform Vibrational Spectroscopy

Using the Alkali halide disk technique, infrared absorption measurements were recorded for glass samples in the range of (4000–400 cm⁻¹) by using Jasco FT/IR-300E infrared spectrophotometer. Dried and ground glasses were mixed with dried infrared grade potassium bromide to obtain a homogeneous mixture of minimum particle size. At 70-MPa pressure the mixture was mechanically pressed to form a disk.

3. Results and Discussion

3.1. Density and Molar Volume

The density was calculated using the formula:

$$\rho = \frac{W_1}{W_1 - W_2} \times 0.86 \quad (1)$$

where W_1 was the weight of the sample in air, W_2 the weight of sample in xylene and 0.86 was the density of xylene. Fig. 1 shows the behavior of the glass density due to incorporation of molar fractions (x_b) of Bi₂O₃ on the expense of P₂O₅ content in the glass samples. The density of Bi₂O₃ is 8.90 g/cm³, while of P₂O₅ is 2.39 g/cm³. Therefore, increasing the Bi₂O₃ mole percentage led to an apparent increase of the glass density. The glass density was increased from 2.4095 g/cm³ (sample 1) into 3.1939 g/cm³ (sample 4). This provides a 32.5% increase of its value with only 0.07 Bi₂O₃ molar fraction (x_b).

Molar volume of glass V_m was determined via the following equation:

$$V_m = \sum_i M_i / \rho \quad (2)$$

where M_i denotes the molar mass of the glass which is equal to $C_i A_i$. Here C_i and A_i are the molar concentration and the molecular weight of the i th component, respectively. With $x_b = 0.07$ the glass molar volume decreased by 6.8 %, in other words, from 39.927 cm³/mol (sample 1) to 37.223 cm³/mol (sample 4). As shown in Fig. 1, the decrease in the molar volume of the glass with increasing the Bi₂O₃ content indicates that Bi₂O₃ has a contracting effect on the glass network. This can be attributed to a decrease in the bond length or inter-atomic spacing between the atoms due to an

increase in the bonds stretching force constant.

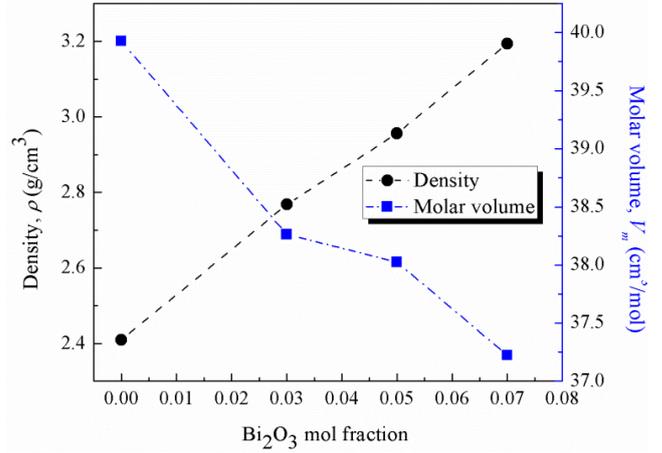


Figure 1. Variations of glass density and molar volume versus Bi₂O₃ molar fraction

3.2. Intermolecular Separation and Bond Density

The molar masses of Bi₂O₃ and P₂O₅ are 465.9 and 109.9 g/mol., respectively. Hence the replacement of P₂O₅ by Bi₂O₃ explains the increase in the glass density with increasing Bi₂O₃ content. Bi³⁺ state has ionic radius (0.65 Å) whereas Bi⁶⁺ has ionic radius 0.36 Å. Accordingly, the increase in the glass density reveals that a partial transformation was occurred for a part of (Bi³⁺) ion groups into the more dense (Bi⁶⁺) ion groups with its octahedral sites.

To be closer into the amendment of the glass network with the addition of Bi₂O₃, the average phosphate-phosphate separation $\langle d_{p-p} \rangle$ was calculated. The phosphate molar volume V_m^p was calculated by [15]:

$$V_m^p = \frac{V_m}{2(1 - x_p)} \quad (3)$$

where x_p is the molar fraction of P₂O₅ oxide. The average phosphate-phosphate separation $\langle d_{p-p} \rangle$ was calculated using the expression [16]:

$$\langle d_{p-p} \rangle = \left(\frac{V_m^p}{N_A} \right)^{1/3} \quad (4)$$

where N_A is Avogadro's number (6.0228×10^{23} g/mol). Fig. 2 illustrates that the increase in Bi₂O₃ content decreases the average phosphate-phosphate separation. The merging of Bi₂O₃ on the expense of P₂O₅ leads to a significant densification of the glass.

Moreover, the change in the number of bonds per unit volume, n_b , of the glass can explain the obtained decrease in the glass molar volume as Bi₂O₃ was incorporated into the glass network. The number of bonds per unit volume was calculated using the relationship [16]:

$$n_b = \frac{N_A}{V_m} \sum_i (n_{ci} x_i) \quad (5)$$

where n_{ci} and x_i were the coordination number and molar fraction of the cation, respectively. The used coordination number for P^{3+} , Bi^{3+} and Mg^{2+} were 3, 6 and 6, respectively. As shown in Fig. 3, the number of bonds per unit volume in the glass network increases with increasing the Bi_2O_3 content. The decrease in the glass molar volume is accompanied with an increase in number of bonds per unit volume, n_b , from $82 \times 10^{21} \text{ cm}^{-3}$ (sample 1) to $89 \times 10^{21} \text{ cm}^{-3}$ (sample 4). Accordingly, the insertion of Bi_2O_3 may lead to a decrease in the phosphate-phosphate intermolecular separation and an increase in the number of bonds per unit volume.

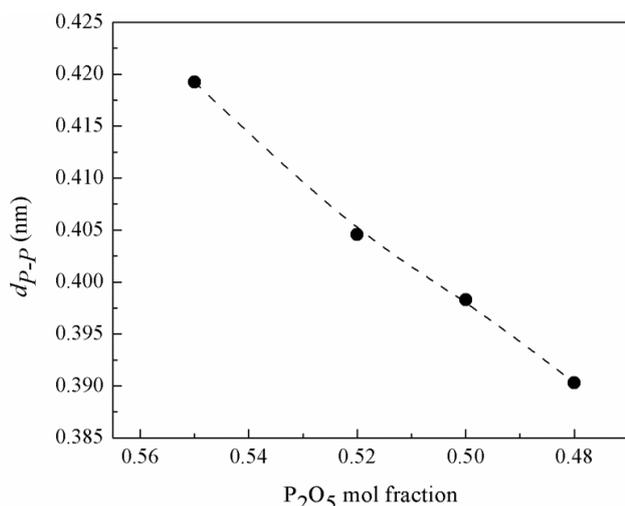


Figure 2. Variation of the average phosphate-phosphate separation (d_{p-p}) against the P_2O_5 molar ratio

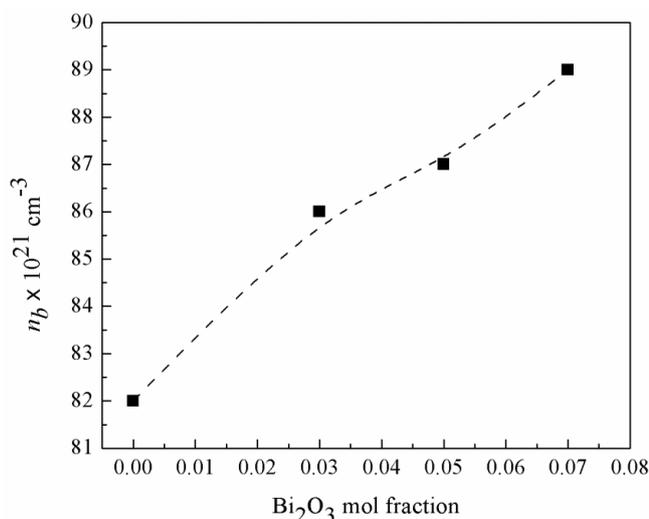


Figure 3. Variation of the number of bonds per unit volume, n_b , in the glass network versus bismuth oxide mol fraction

3.3. Optical Dielectric Constant and Oxide Polarizability

The dielectric constant, ϵ , is related to the permittivity. It expresses the ability of the glass to be polarized as a response to the applied electric field. The dielectric constant, ϵ , as a function of the glass density, ρ , and its molar electronic polarizability (α_m) is given by Lorentz-Lorenz equation where [17]:

$$\frac{(\epsilon - 1)}{(\epsilon + 2)} = \frac{4\pi}{3} N_A \alpha_m \frac{\rho}{M}, \quad (6)$$

Here M is the molar mass of glass composition and $(\epsilon - 1)/(\epsilon + 2)$ is defined as the molar reflectivity term. The glass molar electronic polarizability, α_m , is expressed as the summation of the electronic polarizabilities of constituent ions, α_i , and that of oxygen ion, $\alpha_{O_i}^{2-}$, by [18]:

$$\alpha_m = \sum_i x_i N_{O_i} \alpha_{O_i}^{2-} + \sum_i x_i N_i \alpha_i \quad (7)$$

where x_i oxide mole fractions, and N_{O_i} and N_i are the numbers of oxygen ions and cations, respectively, which can be obtained from chemical compositions. The cations electronic polarizabilities α_i were 0.021 \AA^3 , 0.094 \AA^3 and 1.508 \AA^3 for P^{3+} , Mg^{2+} and Bi^{3+} ions, respectively [19]. In the same sequence, the electronic polarizabilities of an oxygen ion, $\alpha_{O_i}^{2-}$, were 1.350 \AA^3 , 1.687 \AA^3 and 3.507 \AA^3 , respectively [19]. It can be seen that (Fig 4) increase in Bi_2O_3 mol. fraction increases the glass molar polarizability which increases the optical dielectric constant of the investigated glass. An increase in the optical dielectric constant in the order of 15.5% was obtained with the increase in the Bi_2O_3 mol fraction up to 7%. The calculated optical dielectric constant makes the glass appropriate for advanced photonic applications like microelectronic and luminescent devices [20].

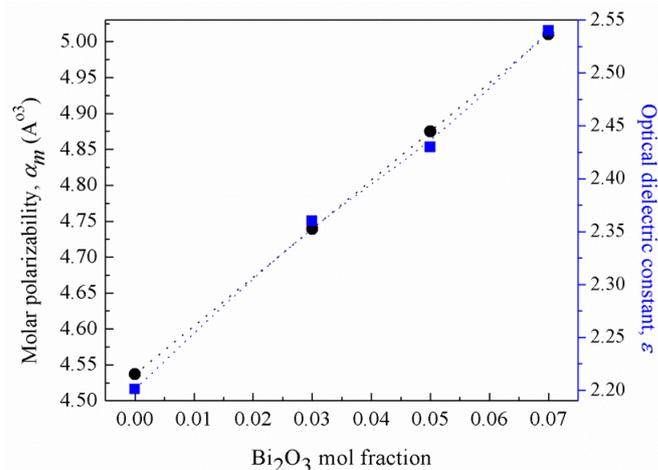


Figure 4. Variation of glass optical dielectric constant, ϵ , and the glass electronic molar polarizability, α_m , versus the increase in Bi_2O_3 mol. fraction

3.4. Infrared Fourier-Transform Vibrational Spectroscopy

As shown in Fig. 5, the assignment of IR spectra indicates the presence of;

1. Band at $1250\text{--}1300 \text{ cm}^{-1}$ assigned to asymmetric stretching modes, $\nu_{as}(\text{PO}_2)$, of the two non-bridging

oxygen atoms bonded to a phosphorus atom in a Q^2 phosphate tetrahedron and/or P=O groups, ν_{as} (P=O) [21, 22].

2. Asymmetric stretching vibrations of P – O⁻ in Q^2 units at 1084 cm^{-1} [23, 24].
3. Asymmetric stretching vibrations of P–O–P bonds at 929 cm^{-1} [24, 25].
4. Symmetric stretching vibration of P–O–P chains at 776 cm^{-1} [25].

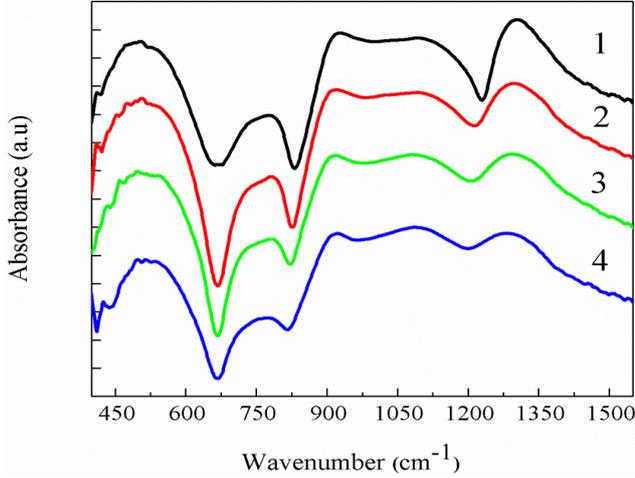


Figure 5. Infrared Fourier-transform vibrational spectra of the studied glasses

In glasses containing bismuth oxide, the presence of Bi^{6+} can be confirmed by the presence of highly distorted BiO_6 polyhedra band between 350 and 500 cm^{-1} [26-28]. The band which appears in the region of 450–550 cm^{-1} was assigned to the deformation vibrations of the phosphate groups [29]. With the addition of Bi_2O_3 , these bands can be assigned to the overlapping vibrations of the Bi–O bonds in the distorted BiO_6 octahedral with the deformation vibrations of the phosphate groups [26, 30]. The two bridged bands of P–O–P at 929 and 776 cm^{-1} may be considered as an indication of the existence of pyrophosphate units [31, 32].

With the continuous replacement of P_2O_5 by Bi_2O_3 a decrease in the amplitude of the band around 1305 cm^{-1} can be observed which was attributed to the increase in the intensity of vibrational band around 1091 cm^{-1} . This is assigned to P–O⁻ vibrations of PO_3^- groups [33-36] which is due to the cleavage of P=O bonds and formation of more ionic P–O–Bi and P–O⁻ bond group terminators that consist of non-bridging oxygens. A rapid depolymerization of the phosphate chains with increasing Bi_2O_3 contents is shown with the formation of P–O–Bi bonds and BiO_6 octahedral in the glass matrix. The stretching vibration of PO_3 groups was a sign of increasing covalent character of the P–O–Bi bonds. This covalent character supports the glass forming ability for the Bi^{6+} ions.

3.5. Estimation of Radiative Lifetime for Luminescence Dopants

Luminescence by near-ultraviolet radiation depends

basically on active centers, surrounding host composition and their interactions [37, 38]. Oxide glasses which has high quantum yield of luminescence can be act as hosts for rare-earth dopant ions. The effect of variation of optical constants of the host medium on the radiative lifetime (τ_R) of luminescence centers was investigated [39, 40]. The total lifetime, τ_t , of electronic excited state of an ion was given by [41]:

$$\frac{1}{\tau_t} = \frac{1}{\tau_R} + A_{nR} \quad (8)$$

where $A_{nR} = \frac{1}{\tau_{nR}}$ is the probability of non-radiative processes and τ_{nR} is the non-radiative lifetime. The radiative lifetime should be much shorter than the non-radiative lifetime to possess a competent luminescence. The expression giving the radiative lifetime for the electronic transitions of the embedded ion in the host glass was given by [42]:

$$\tau_R = 1.5 \times 10^4 \frac{\lambda_0^2}{f \varepsilon^{0.5} [1/3(\varepsilon + 2)]^2} \quad (9)$$

where λ_0 (in meter) is the average interband oscillator wavelength, f is the quantum mechanical oscillator strength for the electronic dipole transition of the dopant ion, and ε is the dielectric constant of the host glass. The dependence of radiative lifetime τ_R on the dielectric constant, ε , is due to the change in the electron density of states and due to modification in the electronic polarizability of the host glass as well. Fig. 6 and Fig. 7 demonstrate that the estimated radiative lifetime is decreased by 19% with increasing the dielectric constant by a 14.4%, and with increasing glass molar weight, M , by a 24%.

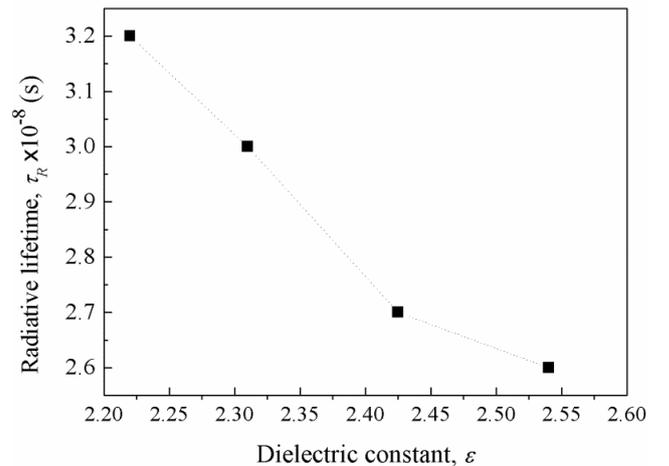


Figure 6. Radiative lifetime versus glass dielectric constant

Referring to Fig. 8 and Fig. 9, it can be seen that with increasing both of the oxide ion polarizability, $\alpha_{\text{O}^{2-}}$, (by 7.4%) and the glass electronic polarizability, α_m , (by 10.3 %) which were associated with increasing the Bi_2O_3 content, the radiative lifetime was decreased by 19%. The observed trend

confirms that the spectroscopic properties of doped ions will be strongly affected by the structure of host glass [43-45] and by the conversion of Bi^{3+} into Bi^{6+} ions (which have more polarizing power and smaller field strength).

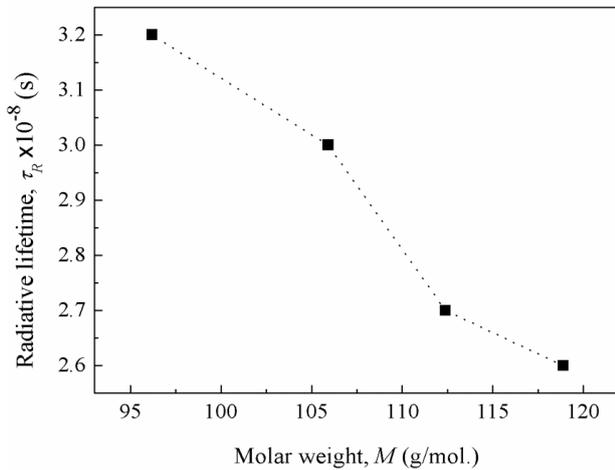


Figure 7. Radiative lifetime versus glass molar weight

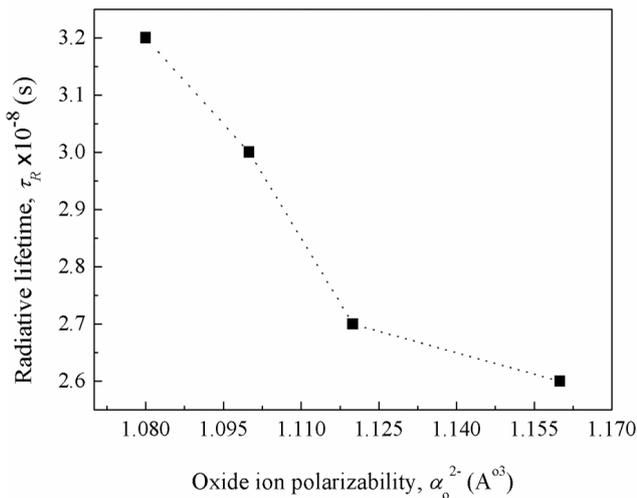


Figure 8. Radiative lifetime versus glass oxide ion polarizability

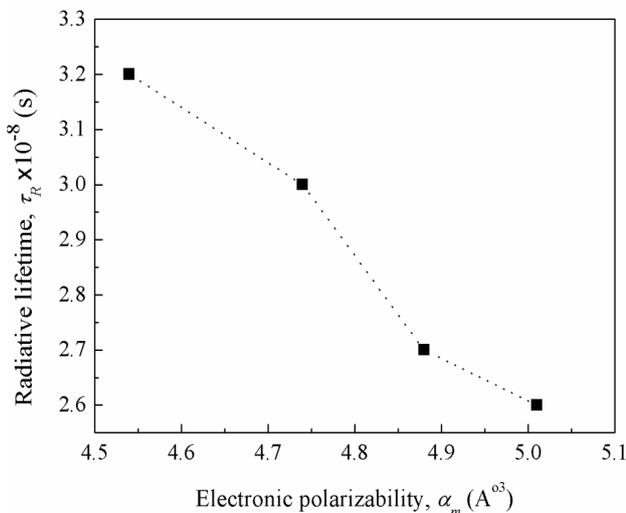


Figure 9. Radiative lifetime versus glass electronic polarizability

The glass optical basicity, A , is the average electron donor power of all the oxide atoms included in the glass system. It is an important spectroscopic property that affects the glass environment of the dopant ion sites. Optical basicity of glass gives a comparison of the acid-base nature of oxides and oxide glasses [46]. The increase of glass basicity leads to an increase in the negative charge on the oxygen atoms. Hence it increases the covalency of the oxygen-cation bonding in the glass network. Since the oxide ion polarizability provides the degree of negative charge on the oxygen atoms, the optical basicity offers indication of this charge.

The link between glass optical basicity, A , and the electronic polarizability of the oxide ion, α_o^{2-} , was given by [47]:

$$A = 1.67 \left(1 - \frac{1}{\alpha_o^{2-}} \right) \quad (10)$$

Fig. 10 illustrates the variation in radiative lifetime, τ_r , for luminescent dopants in parallel with the variation in A of the studied glass system. Optical basicity, A , depends on oxide ion polarizability that impinges on the included cations. It can be seen that with enlarging of the Bi_2O_3 content at the expense of P_2O_5 content, the radiative lifetime decreases by 19% with an increase in the optical basicity by 54%. The results offer a prospect variation in the values of the glass dielectric constant with the change in $\text{Bi}_2\text{O}_3/\text{P}_2\text{O}_5$ ratio.

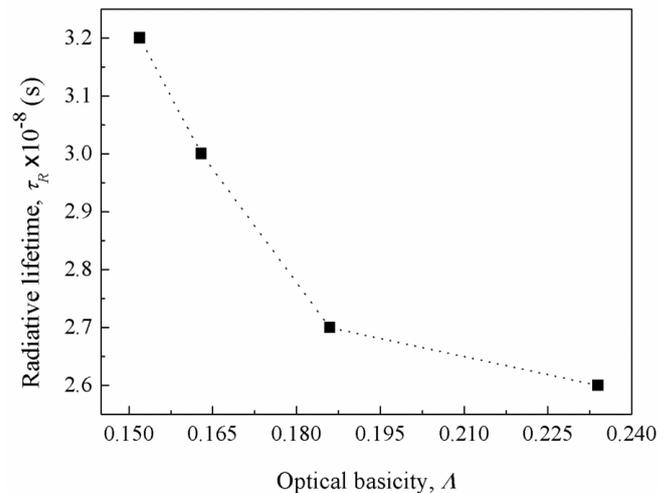


Figure 10. Radiative lifetime versus glass optical basicity

4. Conclusions

We studied the spectroscopic characteristics of the host glass (surrounding medium) which affect the radiative lifetime of dopant active centers. From the thorough analysis of the calculated results, it turns out that parameters such as the structure and the optical properties of the host glasses play important roles on estimation of radiative lifetime of the electronic transitions for luminescent dopant ions. For example, but not limited to, decreasing the radiative lifetime

for luminescent dopant ions (which is needed for the different luminescence devices) is accompanied with a clear increase in the following spectroscopic and structural host glass parameters;

- Glass density,
- Dielectric constant,
- Molar weight,
- Electronic polarizability,
- Oxide ion polarizability,
- Optical basicity.

The obtained results may be useful for the fabrication of doped glasses used in light emitting devices, continuous wave lasers and pulsed laser systems.

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