Structural, and Radiation Shielding Simulation of B2O3-SiO2-LiF- ZnO-TiO2 Glasses

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Structural, and radiation shielding simulation of B₂O₃-SiO₂-LiF- ZnO-TiO₂ glasses

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Abstract:

Quaternary glasses with a 59B₂O₃-29SiO₂-2LiF-(10 – x) ZnO-xTiO₂ composition using the melt-quench techniques were prepared. XRD examined the nature of prepared glasses. The FT-IR spectra was studied for the changes in the structure of these glasses. While the density is increased, the molar volume of the glass system is reduced. The velocities and elastic modulus of these glasses were experimentally and theoretically based on the Makishima-Mackenzie model evaluated. Besides, for the studied glasses, the radiation shielding efficiency was investigated by Phy-X/PSD and XCOM software. These glasses were found to have an abnormal attenuation, structural, and density relationship. The mass attenuation coefficient (μ/ρ), linear attenuation coefficient (LAC), half-value layer (HVL), tenth value layer (TVL), and effective atomic number (Z_{eff}), of glasses, have been designed to simulate for gamma photon energies between 0.015 and 15 MeV. MAC values calculated using Phy-X/PSD and XCOM were compared and was observed in good agreement with the other.

Keywords: Phy-X/PSD, Elastic modulus, FT-IR.

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1-Introduction

The specifications of materials that can serve a double function over the last century can be regarded as among the disguised targets of several researchers. Transparent glasses can be used in radiation shielding materials as a dual function. For the development of optically transparent radiation shielding materials, significant numbers of glass research labs are rising day-by-day. These materials are used in optical communication, modern optical devices, and radiation shielding materials where protection is needed from radiation [1-8].

Due to its remarkable features, such as good thermal stability, hardness, chemical stability, and so on, borosilicate glasses have become the best substitute for concrete shielding, and borosilicate glasses have improved physical characteristics like transparency and refractive index. One of the most important additives in the glass systems can be considered: TiO_2 , including borate, silicate, borosilicate, and phosphate glasses. The addition of TiO_2 has been reported to increase the glass system's network stability and mechanical characteristics. Glasses having a higher amount of titanate confirmed the structural unit TiO_4 , TiO_5 , and TiO_6 and their physical features depend on their number of coordinates [1-8].

To modify the characteristics of the glass, glass modifiers are incorporated into the glass network. Alkali halides [9-11] such as LiF , and some transition metals like ZnO , and TiO_2 are included in modifiers. The melting temperature of glass and its viscosity can be affected by glass modifiers while retaining its chemical structure, as well as affecting its optical, mechanical, thermal, and shielding ability. Halides such as NaF , LiF , are introduced into the glass matrix to generate mobile ion species, Li^+ , Na^+ , etc. So that, halide glasses are excellent reagents for metal ions. For a long time, glasses combining halide ions were studied due to their unique physical characteristics.

The emergence of TiO_2 into the glass network improved the glass's optical, mechanical, thermal, and shielding characteristics [12-13]. Because of good conductivity of these glasses in ionic terms, it is extremely probable to use them in UV optics, solid-state batteries, and radiation protection. Depending on their concentration in the glass matrix, intermediate oxides such as TiO_2 can act as either a glass modifier or former. TiO_2 improves the mechanical strength and radiation protection of the host glass matrices. These glasses possess lower photon energy and a greater refractive index than other glasses. The significant development of lithium fluoride zinc titanate borosilicate glasses is very important scientifically and technologically.

It's the best study for the preparation of lithium fluoride zinc titanate borosilicate glasses and their structural, mechanical, shielding radiation, and optical properties. Thus, it is possible to find the lithium fluoride zinc titanate borosilicate glasses are suitable for use in environments exposed to radiation. The purposes of this research are to identify the attenuation proficiency of lithium fluoride zinc titanate borosilicate glasses using Phy-X/PSD [15] and XCOM software and to identify the mechanical and structure of these glasses to determine their suitability as gamma-ray shielding materials.

2. Experimental processes and techniques.

Glass samples in Table 1 were formulated using the melt-quench method in the chemical formula $29\text{SiO}_2 - 2\text{LiF} - 59\text{B}_2\text{O}_3 - (10-x) \text{ZnO} - x \text{TiO}_2$, where $x: (0 \leq x \leq 10)$ mol %. SiO_2 , LiF , H_3BO_4 , ZnO , and TiO_2 are the initial materials for obtaining these glasses. All the initial materials have been acquired from Sigma-Aldrich Company. Through grinding the blend repetitively to obtain a fine powder, the starting materials were blended. First, to eliminate H_2O and other impurities, the base materials were heated to 650°C for 1 h. The heat was increased for 45 minutes to 1200°C . To reduce the internal stresses, samples were annealed at 450°C for 2 h and left to cool slowly to ambient temperature.

The glass sample's amorphous state was checked using X-ray diffraction (A Philips X-ray diffractometer PW/1710). To obtain FTIR spectrums for glass samples, a Fourier transform infrared spectrometer (JASCO, FT/IR-430, Japan) was used. For this purpose, each glass powder was blended with KBr at a rate of 1:100 (by weight) and pressed into a pellet via a hand press. Through 4 cm⁻¹ resolution in the 4000-400 cm⁻¹ wavenumber range. The consequent spectra has curve-fitted to obtain quantitative values using the computer program fitting peak for the band areas of extensively overlapped bands.

The density of glass samples was estimated by Archimedes' code by using toluene as the immersion liquid. $\rho = \rho_0 \left(\frac{M}{M - M_1} \right)$ where M and M_1 are the weightings of glasses in the air and liquid, the glass density is ρ and the density of toluene is ρ_0 (0.865 g. cm⁻³) with error ± 0.001 g cm⁻³. The molar volume, V_m can be evaluated as $= \frac{M}{\rho}$ where M the molar weight of the glass.

Using the pulse-echo technique, ultrasonic velocities, longitudinal (v_L) and shear (v_T), at ambient temperature were evaluated. In this technique, x-cut and y-cut transducers were used together with a digital ultrasonic flaw detector (KARL DEUTSCH Echograph model 1085) functioning at 4 MHz with error ± 10 m s⁻¹. Besides the density, the velocities were used to evaluate elastic moduli. longitudinal waves $L = \rho v_L^2$, transverse waves $G = \rho v_T^2$, Young's modulus $Y = (1 + \sigma)2G$, bulk modulus $K = L - \left(\frac{4}{3}\right)G$ [15 – 32].

Elastic moduli of samples were evaluated using the exemplary based on packing density $V_i = \left(\frac{3\pi}{4}\right) N_A (mR_A^3 + n R_O^3) m^3.mol^{-1}$, and dissociation energy $G_i = \left(\frac{1}{V_m}\right) \sum_i G_i X_i$, the metallic and oxygen Pauling ionic radii are R_m and R_O . Longitudinal waves $L = K + \left(\frac{4}{3}\right)G$, transverse waves $G = 30 * \left(\frac{V_i^2 G_i}{V_i}\right)$ Young's modulus $Y = 8.36 V_i G_i$, bulk modulus $K = 10 V_i^2 G_i$. Poisson's ratio $\sigma = \frac{1}{2} - \left(\frac{1}{7.2 * V_i}\right)$. Acoustic Impedance; $Z = v_L \rho$.

Micro Hardness; $H = \frac{(1-2\sigma)Y}{6(1+\sigma)}$. Debye Temperature: $\theta_D = \frac{h}{k} \left(\frac{9N_A}{4\pi V_m} \right)^{\frac{1}{3}} M_s$, Where h and k are the constants of Planck and Boltzmann and N_A is the number of Avogadro [33-34].

Average velocities, $M_s = \frac{1}{3} \left(\frac{\frac{2}{v_T^3}}{\frac{1}{v_L^3}} \right)^{\frac{1}{3}}$. Thermal coefficient of expansion,

$\alpha_P = 23.2 (v_L - 0.57457) \cdot$ Volume of the oxygen molar, $V_o = \left(\frac{M}{\rho} \right) \left(\frac{1}{\sum x_i n_i} \right)$, Density of Packing,

$OPD = \left(\frac{1000 C}{V_m} \right) \left(\frac{Mol}{L} \right)$.

In this article, radiation parameters have been computed using Phy-X/PSD software and these parameters are calculated using the following equations: the coefficient of mass

attenuation samples $(\mu/\rho) = \sum_i x_i (\mu/\rho)_i$. Effective atomic number $Z_{eff} = \frac{\sum_i f_i A_i (\mu/\rho)_i}{\sum_j f_j \frac{A_j}{Z_j} (\mu/\rho)_j}$. Half

and tenth value layer (HVL), and (TVL): $HVL = \frac{0.693}{LAC}$, $TVL = \frac{2.3}{LAC}$

3. Results and Discussions

3.1 XRD studies

No discrete lines, no sharp peaks, were confirmed by XRD patterns as exemplified in Fig.1 and indicate that the samples have a high amorphous state. The width of the halo varies from one sample to another, but no indication of crystalline phase has been shown in all the samples.

3.2 Investigations of FT-IR

FT-IR spectra are exemplified in Fig. 2 for titanium borosilicate glasses. To obtain accurate band positions in the FT-IR spectrum, Deconvoluted process is used. Residue results were plotted to get the quality in the FT-IR deconvolution fitting, and the variation is less than 0.02% in the experimental and simulated graphs. Gaussian fit of FT-IR spectrum of these glasses is exemplified in Fig. 3 and Table 2, respectively. The structural units of the network in

these glasses were identified and summarized as: The band at $\sim 444 - 490 \text{ cm}^{-1}$ is related to the network structure's deformation modes. Bands in the $562\text{-}570 \text{ cm}^{-1}$ region had been ascribed to (LiO_6) , (ZnO_6) , and (TiO_4) vibrations and they overlap with the O-Si-O unit bending vibrations. The band at $\sim 680 - 691 \text{ cm}^{-1}$ is assigned to the bending vibrations of the bridging oxygen atom. The band at $\sim 890 \text{ cm}^{-1}$ is assigned to the tri-, tetra-, and penta-borate groups' stretching vibrations. The band at $\sim 1120 - 1044 \text{ cm}^{-1}$ is assigned to B-O stretching BO_4 tetrahedra vibrations. The band at $\sim 1120 - 890 \text{ cm}^{-1}$ is assigned to B-O, bridge in the BO_4 , and a major component of asymmetric stretching of BO_4 tetrahedron. The band at $\sim 1240 - 1260 \text{ cm}^{-1}$ is assigned to trigonal BO_3 units of B-O bond stretching. The observed band at $\sim 1360 - 1350 \text{ cm}^{-1}$ is assigned to B-O stretching vibrations that mainly involve divers' groups of connected oxygen. B-O symmetric stretching vibrations of different borate groups are due to the band at $\sim 1650\text{-}1500 \text{ cm}^{-1}$. FT-IR spectral causes a shift to higher wavenumbers with an increase in titanium concentration. Also, the structure becomes more compact by incorporating TiO_2 , as TiO_4 species are produced. Boron transforms from BO_3 into BO_4 tetrahedra after alkali metal halide is incorporated [25-32]. This increases the coherence of the glass network and the structure stiffening.

3.3 Investigations of Mechanical

Glass density raised as TiO_2 content increased and molar volume reduced as Fig. 4. The density increase is ascribed to the transformation of the BO_3 triangles into the tetrahedral BO_4 of the glass network with the progressive replacement of ZnO with TiO_2 . Rising density results affect the stiffness of the glass network and the increase in rigidity. Enhanced density values also reflect an increase in the density of cross-links. The density increase may also be due to the rise in oxygen in the glass network. The mixed oxide effect may be contributed to a variation in density and molar volume. These results are like the FT-IR results.

The velocity of prepared glasses with different amounts of TiO_2 was illustrated in Fig.

4. Both velocities (v_L and v_T) were increased, as shown in Table 3, by an increase in TiO_2 , and (v_L) values higher than (v_T). This increase in the estimated ultrasonic velocity is possible to explain by considering variables:

- (i) Increasing TiO_2 will enhance the amorphous network by increasing the concentration of TiO_4 and TiO_6 structural units with higher coordination in comparison with ZnO structural unit.
- (ii) Consequently, there was an increased polymerization of the glass coordination number, cross-link density, and connectivity within the glass network.
- (iii) Because of the increase in internal energy, the velocities were increased.

Experimentally and theoretically, elastic modules were evaluated for prepared glasses and exemplified in Figs 6 & 7. The elastic moduli exactly as noticed of velocities as exemplified in Fig. 6 & 7 i.e., it depends on the nature of bonds in the glass and the cross-link density. With the increase of TiO_2 , the elastic moduli value shows an increasing trend. The rise in elastic modules as the number of coordinates raised, and bond strength of $\text{Ti}-\text{O}$ (73KCal/mol) is higher than $\text{Zn}-\text{O}$ (36 KCal/mol). As the modification role of TiO_2 in the glass system, all mechanical parameters as revealed in Table 4 are increased with the increase in TiO_2 content.

3.4 Photon Shielding Features

Concerning mass attenuation coefficient values (MAC) obtained by Phy-X/PSD software with a photon energy range of 0.015-15 MeV. Because MAC is characterized by the material absorbed to attenuate radiation, a higher value indicates a more impenetrable shield. Phy-X/PSD and X-com performance, MAC values concerning the energy and TiO_2 mol % were exemplified in Figs. 8&9. More clearly, it is from Figs. 8&9 that the impact of the glass structure plays an important role in improving the MAC values of these glasses. The

highest values of MAC are found at low energy, and with the shift towards greater energy, MAC decreased rapidly. The MAC of the glasses reduces as energy increases as more photons can absorb through the sample, reducing its absorbency, and reducing MAC. This sequence also shows that samples are the most efficient at lower energies. Their shielding capacity becomes less efficient when energy increases. Besides, Ti has a lower atomic number (22) than Zn (33). The atomic number correlates positively with MAC, causing the highest radiation shielding capability in the glass sample G 1. Even so, the G1 glass can be the most superior attenuation abilities to the other glasses [35-42].

As shown in Table 5, MAC values calculated using Phy-X/PSD and XCOM are compared. The other is in excellent agreement with these values. The Phy-X/PSD and XCOM deviations were calculated as $Deviation \% = \left[\frac{(\mu/\rho)_{Phy-X} - (\mu/\rho)_{XCOM}}{(\mu/\rho)_{Phy-X}} \right] \times 100$, The dev. % acquired is small, which provides the Phy-X results [43].

LAC is a useful method for calculating the glasses' attenuation shielding efficiency, which also demonstrates the same trend as MAC. Figs.10 exemplified LAC with the photon energy. The atomic number plays a critical role in improving LAC values. In general, with the decrease in atomic number LAC and MAC are decreased. The identified LAC values follow the $G1 > G2 > G3 > G4 > G5 > G6$ trend for all energies. These data indicated that the LAC values are associated with the atomic number of titanates, causing G1 to be the highest LAC. It is possible to describe the reduction using the same reasoning as for MAC.

Figure 11 exemplified *HVL* as a photon energy function. It was observed that *HVL* values an increasing with energy. So, G1 the most attractive shield than the other samples examined. This result suggests that, at a lower energy level, glasses are much more effective, while reducing efficiency as excess energy. Figure 12 exemplified *TVL* as a

photon energy function. The *TVL* values increased with the increase in photon energy in all the examined glasses. *TVL* was analogous with *HVL* values.

Figure 13 exemplified Z_{eff} as a photon energy function. Z_{eff} symbolizes the mean atomic number, with a higher value suggesting a good shield. G1 has been reported the highest value. This behavior can be explained as Ti has a lower atomic number (22) than Zn (33). This behavior indicates that it is further needed to increase the titanate in the glasses. The Z_{eff} values decreased with the increase in photon energy in all the examined glasses. The photoelectric effect is strongly dependent on the atomic number, therefore the glasses containing Zn have a quickly reducing Z_{eff} since Zn has an atomic number of 30. The G6 glass sample shows the smallest potential for shielding than other samples.

4. Conclusions

In the current study the glass containing different amounts of titanate with the formula $59\text{SiO}_2 - 29\text{SiO}_2 - 2\text{LiF} - (10-x) \text{ZnO} - x\text{TiO}_2$ where $x = (0 \leq x \leq 10)$ conventional melt-quenching methods have been manufactured. The structure, mechanical, and shielding variables have been examined for these glasses. The findings showed the following objects:

- 1- XRD measurements established the amorphous nature of glasses.
- 2- As TiO_2 content rises, the density of the samples raised while the molar volume reduced.
- 3- Ultrasonic velocities of these glasses were increased with increasing TiO_2 concentration.
- 4- Gamma shielding characteristics of these glasses were predictable by the Phy-X / PSD and XCOM program between 0.015-15 MeV. The effect of the addition of TiO_2 on the shielding ability of the glasses was discussed and we found that: (i) The mass attenuation coefficient decreased with the increase of the concentration of TiO_2 from 0 mol. % to 10 mol. %, (ii) The sample coded as G 1 possesses the lowest

HVL while highest Z_{eff} . (iii) Lithium fluoride zinc titanate borosilicate glasses were found to have an abnormal attenuation, structural, and density relationship.

The results obtained have shown that the increase in TiO_2 concentration in the glass system can lead to a significant improvement in the attenuation, structural, and mechanical properties. Furthermore, it is possible to use this glass in aircraft bodies, a shield from radiation in the x-ray centers, facades of houses.

Author contributions: Kh. S. Shaaban: performing XRD, UV measurements and analysis, Writing-review, writing the manuscript, Methodology, Software, and writing – discussion.

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Figures

Figure 1

XRD of the studied glasses.

Figure 2

FT-IR spectra of prepared glasses.

Figure 3

The Gaussian fit of the FT-IR spectrum of prepared glasses.

Figure 4

Glass density and molar volume as a function of TiO_2 content.

Figure 5

Ultrasonic velocities of prepared glasses with varying quantities of TiO_2 .

Figure 6

Experimentally elastic modules of prepared glasses with varying quantities of TiO₂.

Figure 7

Theoretically elastic modules of prepared glasses with varying quantities of TiO₂.

Figure 8

Mass attenuation coefficient prepared glasses a function of photon energy according to Phy-X/PSD.

Figure 9

Mass attenuation coefficient prepared glasses a function of photon energy according to X-COM.

Figure 10

Linear attenuation coefficient prepared glasses a function of photon energy according to Phy-X/PSD.

Figure 11

Have value layer prepared glasses a function of photon energy according to Phy-X/PSD.

Figure 12

Tenth value layer of prepared glasses a function of photon energy according to Phy-X/PSD.

Figure 13

Effective atomic number of prepared glasses a function of photon energy according to Phy-X/PSD.