



## The role of $B_2O_3$ in lithium-zinc-calcium-silicate glass for improving the radiation shielding competencies: A theoretical evaluation via Phy-X/PSD

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### ABSTRACT

In this study, a preliminary theoretical investigation on lithium-zinc-calcium-silicate (LZCS) glass with a composition of  $(15-x)Li_2O-10ZnO-10CaO-65SiO_2-xB_2O_3$  where  $x$ : 0, 3, 6, 9, 12, and 15 mol% was performed to understand the effect of  $B_2O_3$  on physical, optical, and radiation shielding properties. For this purpose, the L15B0 to L0B15 glass series was designed for evaluating glass density ( $\rho_{glass}$ ), refractive index ( $n$ ), mass attenuation coefficient ( $\mu_m$ ), and half-value layer ( $\Delta_{0.5}$ ) parameters. The theoretical calculations showed that the increasing amount of  $B_2O_3$  increased the overall  $\rho_{glass}$  from 2.9195 to 2.9865 g/cm<sup>3</sup>. Further, the addition of  $B_2O_3$  in substitution for  $Li_2O$  enhanced the  $n$  parameter from 1.6882 to 1.7626. Additionally, BatchMaker software aided to investigate viscosity behavior with the increasing temperature. We found out that the melting point of LZCS glass series ascends with the addition of  $B_2O_3$ , namely from 1309 to 1624 °C. On the other hand, the newly developed Phy-X/PSD software computations paved the way for ascertaining  $\mu_m$  and  $\Delta_{0.5}$ . According to the  $\mu_m$  computations, one can clearly state that an increasing trend is observable against the increasing photon energy, but the L0B15 possessed an enhanced shielding ability than that of the remaining at all photon energies. Moreover, we found out that the  $\Delta_{0.5}$  increased with respect to the ascending photon energies, however, the  $\Delta_{0.5}$  was effectively improved with the addition of  $B_2O_3$  in the order of L0B15<L3B12<L6B9<L9B6<L12B3<L15B0. Lastly, a comparison for  $\Delta_{0.5}$  variations between L0B15 and commercially available RS253 G18 evidently demonstrated that L0B15 achieves 4.11 cm while RS253 G18 fulfills 4.95 which in turn confirms that the proposed glass system can be utilized in radiation shielding applications. All in all,  $B_2O_3$  has promising effects on radiation shielding features in LZCS glass series.

### 1. Introduction

In today's world, the radiation term has become a major concern among the scientific community since artificial radiation sources have been exponentially increasing with the developing technological areas [1,2]. From medical diagnosis to industrial non-destructive tests, or from food sterilizations to communication systems the humanity has been facing with variety of irradiation including alpha, beta, gamma, or the like [3-5]. In fact, these rays have a strong probability of damage to human health with respect to time, exposure, and duration parameters. DNA mutations, skin burns, or even cancer problems may raise as a result of exposure to these irradiations [6,7]. From this point of view, the usage of a radiation shielding material against incoming rays has become a major action in order to be protected from the above-mentioned health risks.

Radiation shielding material can, therefore, minimize the incoming photon energy thanks to the attenuation characteristics they have, and thus workers and/or patients can be safeguarded [8].

Conceptually, attenuation of incoming photon energy is directly associated with the atomic mass, atomic radius, and density value of the substance used. This is because higher molecular mass or higher atomic radius can contribute enhancing the attenuation competencies of the radiation shielding material, which in turn facilitating the interaction between incoming rays and materials. Within the scope of practically preferred radiation shielding materials, metallic lead finds a broad application area due to its appreciable density value (11.32 g/cm<sup>3</sup>) [9,10]. The rooms where these devices with the capability of irradiations are placed have been covered with the use of metallic lead in accordance

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with the regulations. Despite the great benefits of metallic lead in terms of advanced attenuating characteristics in particular to the higher photon energies (i.e. gamma-rays), the toxicity issue on humans and the environment has recently begun to be restricted its wide preference [11]. An alternative material, heavy-weight concrete containing aggregates, has become of interest because the building blocks can be fabricated by considering the possible effects of photon energies [12,13]. In fact, heavy-weight aggregates such as barite can be inserted in concrete blends for improving radiation shielding features [14]. Nevertheless, the cracking problem during operation is the primary deficiency of heavy-weight concrete materials. Even though the mentioned problems are valid for these shielding materials the employment has still in progress due to the lack of alternative materials. That's why scientists have extensively been continuing to search for more options.

Glass materials offer spectacular features in many different applications including from daily needs to advanced optical fibers. One of the most essential properties of glass materials is the transparent appearance which metallic lead and heavy-weight concrete could not have. Additionally, compositional flexibility, being environmentally-friendly, well-known melting and shaping techniques, as well as relatively lower production costs make glass materials preferable in radiation shielding applications [15]. Commercially, there are diverse glass types involving silicates, borates, phosphates, and so on. Among these, lithium-zinc-silicate (LZS) glass is regarded as an essential system for various applications such as sealing, dental, and dielectrics. This is because, wide thermal expansion range ( $5$  to  $20 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ ), ability for glass-ceramic formations, relatively good glass viscosity behavior, high electrical insulation, and considerable density values can be obtained with the use of LZS glasses which in turn possessing great potential for radiation shielding applications [16,17]. Furthermore, calcium oxide (CaO) which has a relatively high density value ( $3.34 \text{ g/cm}^3$ ) can aid to improve radiation shielding characteristics in the LZS glass system since its positive effect on different glass system has been confirmed by numerous literature studies [18,19]. On the other hand, boron oxide ( $\text{B}_2\text{O}_3$ ) can ensure a lot of benefits including low thermal expansion coefficient, facilitating melting conditions, increasing chemical durability, and good glass formation ability [20]. From the perspectives of radiation shielding features,  $\text{B}_2\text{O}_3$  presents superior shielding abilities, particularly against neutron scatterings. Additionally, Turkey is the leading country in terms of boron reserves, and hence value-added applications involving boron compounds can bring countless benefits both economically and strategically [21, 22]. In conclusion for the selection of LZS glass system and  $\text{B}_2\text{O}_3$  content, it is worth to investigate the role of  $\text{B}_2\text{O}_3$  in the LZS glass systems for enhancing radiation

shielding competencies.

Literature is generally focused on the understanding the characteristics of LZS glass system with respect to physical, chemical, mechanical, and thermal properties. However, it is evident that the radiation shielding competencies is lack of investigation, in this regard. From the perspective of lithium-zinc-added glass systems, we come forward numerous studies on radiation shielding properties [23-25]. Moreover, silicate-based glass systems are of interest by many researchers for doping and adding various oxide contents [26-28]. As can be appreciated from the literature works, no further investigations have been made on LZS or LZCS glass systems. Indeed, the role of  $\text{B}_2\text{O}_3$  in the LZCS glass system has not been experiences both theoretically and experimentally. Therefore, the authors strongly believe that any findings will make difference in the literature for further explorations.

In the present study, a lithium-zinc-calcium-silicate (LZCS) glass system reinforced with  $\text{B}_2\text{O}_3$  was examined with the use of Phy-X/PSD software. For this, the dopant was designed in the glass system of  $(15-x) \text{Li}_2\text{O}-10\text{ZnO}-10\text{CaO}-65\text{SiO}_2-x\text{B}_2\text{O}_3$  where  $x$ : 0, 3, 6, 9, 12, and 15 mol% to obtain the L15B0 to L0B15 glass series. Glass density ( $\rho_{\text{glass}}$ ) and refractive index ( $n$ ) calculations were conducted to determine the possible alterations. Furthermore, viscosity versus temperature profiles was drawn with the use of BatchMaker software for understanding glass melting characteristics. Lastly, a comparison between the LZCS series and commercial radiation shielding materials were done to make a deeper sense for our study. As a result, this study reported the new results on LZCS containing  $\text{B}_2\text{O}_3$  glass system which has not previously been investigated in the literature studies.

## 2. Materials and Methods

### 2.1. Compositional Design

To understand the role of  $\text{B}_2\text{O}_3$  in LZCS glass system, we designed a distinct composition series as listed in Table 1. L15B0 represents the oxide content with molar percentage for lithium oxide and boron oxide as 15 and 0 mol%, respectively. The remainings are also signified with respect to these codes.

**Table 1.** The compositional design for LZCS glass system in mol%.

Code	Li <sub>2</sub> O	ZnO	CaO	SiO <sub>2</sub>	B <sub>2</sub> O <sub>3</sub>
L15B0	15,00	10,00	10,00	65,00	0,00
L12B3	12,00	10,00	10,00	65,00	3,00
L9B6	9,00	10,00	10,00	65,00	6,00
L6B9	6,00	10,00	10,00	65,00	9,00
L3B12	3,00	10,00	10,00	65,00	12,00
L0B15	0,00	10,00	10,00	65,00	15,00

## 2.2. Calculations for Physical and Optical Properties

Glass density ( $\rho_{glass}$ ) parameter plays an essential characteristics on the investigated glass systems. To find out the changes in  $\rho_{glass}$  with the insertion of  $B_2O_3$  in LZCS glass system, it is beneficial to apply empirical relation stated by Inaba and Fujino [29] as expressed in Equation 1. Here,  $M_i$ ,  $V_i$  and  $x_i$  indicate molecular weight (g/mol), packing density factor ( $cm^3/mol$ ), and molar fraction of  $i^{th}$  component, respectively.

$$\rho = (0,53) \cdot \frac{(\sum M_i \cdot x_i)}{(\sum V_i \cdot x_i)} \quad (1)$$

Another essential parameter, refractive index ( $n$ ), is the key factor determining optical properties of glass materials. To calculate this parameter, energy band gap ( $E_g$ ) values of each oxide is employed throughout the Equation 2 [30].

$$\frac{n^2-1}{n^2+2} = 1 - \sqrt{\frac{E_g}{20}} \quad (2)$$

## 2.3. Glass Viscosity Behavior

Estimating the glass viscosity behavior with respect to temperature changes takes great importance before experimental studies. In literature, there are few works focusing on viscosity alterations for glass systems in the perspective of computational aspects. For this reason, BatchMaker software which utilizes numerous property calculations based on glass oxide contents emerges as an efficient tool for determining viscosity versus temperature profiles [31]. The software takes into consideration the findings of Lakatos et al. [32] and Fluegel [33] in this respect. We employed a calculation set up for our LZCS glass series, and drew viscosity versus temperature profiles to make a deeper understanding on  $B_2O_3$  insertion in substitution for  $Li_2O$ .

## 2.4. Radiation Shielding Computations

Owing to the newly developed user-friendly software, Phy-X/PSD, the radiation shielding parameters can be computed in an efficient way. Within the scope of Phy-X/PSD, one can simply input glass composition and the related glass density to the software. After this, the software begins for computing the variables for figuring out the different radiation shielding parameters [19]. Here, we evaluated some essential parameters including mass attenuation coefficient ( $\mu_m$ ) and half-value layer ( $\Delta_{0.5}$ ). As is known, the  $\mu_m$  defines the interaction between incoming ray and the matter of unit mass per unit region whereas  $\Delta_{0.5}$  indicates the thickness where the energy of incoming photon is attenuated to its half value. Equations 3 and 4 were used

aid to calculate the parameters.

$$\mu_m = \frac{\mu}{\rho_{glass}} \quad (3)$$

where  $\mu$  is the linear attenuation coefficient, and  $\rho_{glass}$  is the overall glass density for each samples.

$$\Delta_{0.5} = \frac{\ln 2}{\mu} \quad (4)$$

## 3. Results and Discussion

### 3.1. Physical and Optical Properties

In literature, almost all researchers have identified the one of the most important parameter, glass density ( $\rho_{glass}$ ). This is because radiation shielding competencies are straight-forwardly associated with the  $\rho_{glass}$  value. As a general concept, it is expected to have higher  $\rho_{glass}$  value as possible for enhancing the radiation shielding features [34,35]. From this point of view, the authors calculated the theoretical  $\rho_{glass}$  values for the examined glass series. On the other hand, refractive index ( $n$ ) is a measure of the interaction between incident ray and the matter. The incident ray may be encountered with refraction, absorption, or transmission with respect to the interaction phenomenon. We know that the higher the  $n$  value for the glass system the more efficient the radiation shielding will [36]. For understanding the possible alteration in  $n$  value for our glass systems, we evaluated the theoretical  $n$  calculations for each glass samples. Furthermore, since both  $\rho_{glass}$  and  $n$  parameters have a parallel behavior Figure 1 depicts the changes in these properties with regard to the  $B_2O_3$  addition. One can firmly deduce that both parameters are in increasing trend with the increasing insertion ratio of  $B_2O_3$ . For instance, L15B0 sample has a  $\rho_{glass}$  value of 2.9195  $g/cm^3$  whereas L0B15 sample gains 2.9865  $g/cm^3$ . This situation may be attributed to the higher molecular mass of  $B_2O_3$  (69.62

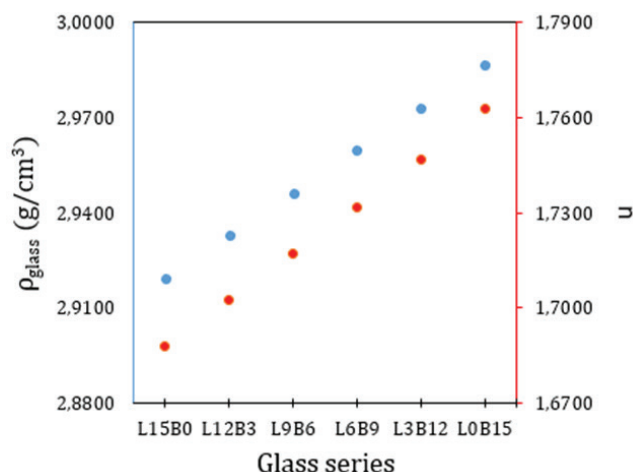


Figure 1. Alteration of  $\rho_{glass}$  (blue dots) and  $n$  (red dots) with respect to the  $B_2O_3$  addition for the investigated LZCS glass series.

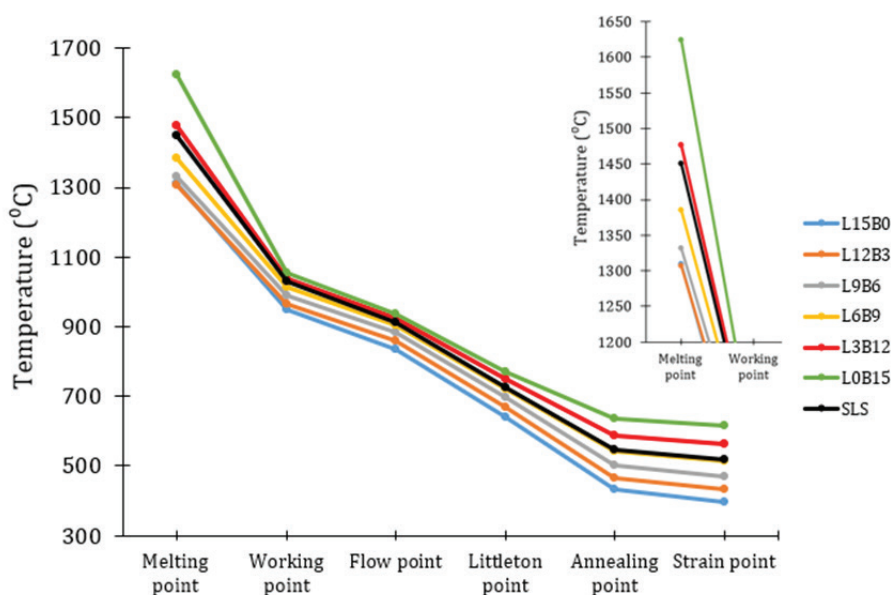


Figure 2. Viscosity versus temperature profiles for the investigated LZCS glass series.

g/mol) in comparison to  $\text{Li}_2\text{O}$  (29.88 g/mol). Similar findings were also stated with some literature studies conducted by [37,38]. On the other hand, the contribution of  $\text{B}_2\text{O}_3$  in place of  $\text{Li}_2\text{O}$  enhance the  $n$  parameter in the order of  $\text{L0B15} > \text{L3B12} > \text{L6B9} > \text{L9B6} > \text{L12B3} > \text{L15BO}$ . This is because the  $E_g$  value for  $\text{B}_2\text{O}_3$ -added series are becoming lower, and thus the incident rays could not transmitted throughout the glass substance. In other words, much more incoming photon energy becomes interacted with the glass substance, particularly, herein, boron atoms. Therefore, the insertion of  $\text{B}_2\text{O}_3$  possesses successful conclusions on both parameters.

### 3.2. Viscosity versus Temperature Profiles

The investigated LZCS glass series are evaluated in terms of viscosity behaviour with respect to the varying temperature via using BatchMaker software in Figure 2. For making a sensible comments on the behavior of the LZCS series, a well-known glass type as soda-lime-silica (SLS) is also depicted. From the profiles, it is obvious that the increasing amount of  $\text{B}_2\text{O}_3$  has an adverse effect on melting temperature. To exemplify, L15B0 specimen has a value of 1309°C for melting point whereas L0B15 acquires 1624°C. At this point, SLS glass possesses nearly 1450°C as a melting point. The reason behind this phenomenon may be referred to the glass-forming characteristics of  $\text{B}_2\text{O}_3$  rather than fluxing agent behavior of  $\text{Li}_2\text{O}$ . The decrease in the amount of fluxing agent in the LZCS glass system ascends the liquidus point through lack of sufficient liquid phase. Therefore, we can report that the insertion of  $\text{B}_2\text{O}_3$  in substitution for  $\text{Li}_2\text{O}$  in the LZCS glass system elevates the glass melting point.

### 3.3. Radiation Shielding Features

For highlighting the radiation shielding features of the

examined LZCS glass series, we evaluated the most essential parameters as  $\mu_m$  and  $\Delta_{0.5}$ . Figure 3 displays the mass attenuation coefficients ( $\mu_m$ ) with respect to incoming photon energies. It can be observed that the  $\mu_m$  is found to be in increasing trend as a function of the increasing photon energy. In the lower photon energy levels, *i.e.* <0.015 MeV, the  $\mu_m$  values range around 15  $\text{cm}^2/\text{g}$ . However, a sudden decrease can be observable towards the increasing photon energy, *i.e.* 0.1 MeV. In this case, the  $\mu_m$  descends through 0.20  $\text{cm}^2/\text{g}$ . For the higher photon energies, *i.e.* >1 MeV, we gain the  $\mu_m$  around 0.02  $\text{cm}^2/\text{g}$ . Such a behavior may be attributed to the well-known three mechanisms as photoelectric absorption at low energy, Compton scattering at intermediate energy, and pair production process at high energy. On the other hand, when it comes to evaluate the effect of  $\text{B}_2\text{O}_3$  in replacement for  $\text{Li}_2\text{O}$ , it is apparent that the  $\text{B}_2\text{O}_3$  is directly enhancing the  $\mu_m$  values at all photon energy levels. The  $\mu_m$  is increased from 0.628 to 0.631  $\text{cm}^2/\text{g}$  with the insertion ratio of  $\text{B}_2\text{O}_3$  from 0 to 15 mol% at 1.30 MeV. This

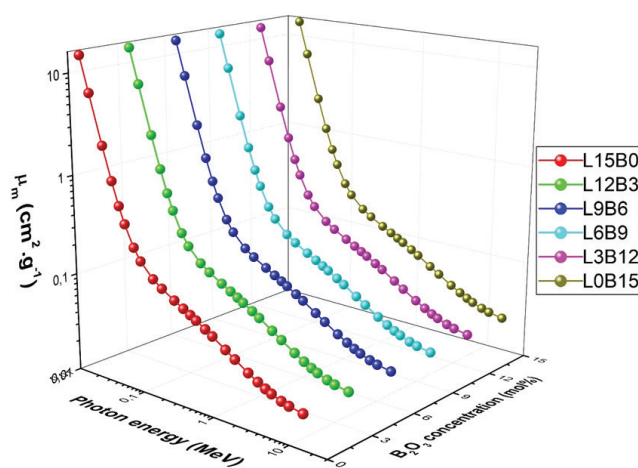
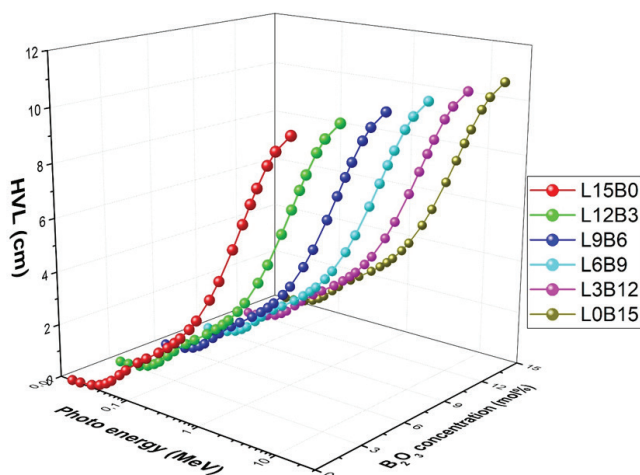


Figure 3. Mass attenuation coefficient of LZCS glass series with respect to photon energies.

increase is associated with the higher density value of  $B_2O_3$  ( $2.46 \text{ g/cm}^3$ ) than that of  $Li_2O$  ( $2.01 \text{ g/cm}^3$ ). The higher the glass density the better the radiation shielding competencies were confirmed by various studies in the literature [39,40]. Since an increasing trend is required for improving radiation shielding competencies we can report that  $B_2O_3$  can provide this enhancement in an efficient way.

Yet another significant parameter, half-value layer ( $\Delta_{0.5}$ ), signifies the required thickness value to attenuate the half of incoming energy. From this perspective, Figure 4 reveals the variations in  $\Delta_{0.5}$  for different photon energies. It is clear that the required thickness value for all LZCS glass series ascends with the increasing photon energy. However, L0B15 dominates over the remaining in all photon energies. For instance, L15B0 requires the  $\Delta_{0.5}$  value of 3.39 cm while L0B15 yields 3.30 cm. That means the lesser thickness can attenuate the half of the incoming energy owing to the contribution of  $B_2O_3$ . In fact, the higher molecular mass of  $B_2O_3$  ( $69.62 \text{ g/mol}$ ) compared to  $Li_2O$  ( $29.88 \text{ g/mol}$ ) is the reason facilitating this advancement. Moreover, a comparison between L0B15 and commercial radiation shielding glasses (*i.e.* Schott RS series) can make a better understanding of the figured out  $\Delta_{0.5}$  values. Schott RS series have distinct type of glasses for the required photon energies [41]. Here, it may be more plausible to highlight the higher energy levels such as gamma-rays occurred at 1.25 MeV ( $^{60}\text{Co}$ ). Besides, it may be more meaningful to select the lead-free RS series such as RS 253 G18 since our LZCS glass series do not contain any lead oxide substance. When it comes to compare  $\Delta_{0.5}$  values, L0B15 specimen provides 4.11 cm while RS 253 G18 ensures 4.95 cm. From these values, we can firmly report that L0B15 sample achieves better radiation shielding features than that of commercially available RS 253 G15 glass. Therefore, our glass system can make a difference in a commercial manner.



**Figure 4.** Half-value layer alterations with respect to photon energies for the LZCS series.

#### 4. Conclusions

The present study focused on the theoretical calculations for evaluating the effects of  $B_2O_3$  addition on the physical, optical, and radiation shielding properties of the lithium-zinc-calcium-silicate (LZCS) glass system. For this,  $(15-x)Li_2O-10ZnO-10CaO-65SiO_2-xB_2O_3$  where  $x$ : 0, 3, 6, 9, 12, and 15 mol% glass system was designed, and L15B0 to L0B15 glass series were examined. The glass density ( $\rho_{glass}$ ) calculations presented that  $B_2O_3$  insertion is an efficient way to increase the overall  $\rho_{glass}$ . Additionally, the refractive index ( $n$ ) value was enhanced owing to the  $B_2O_3$  contribution. For understanding the viscosity behavior of the investigated glass series, BatchMaker software was utilized. It showed that the addition of  $B_2O_3$  increased the melting point of the glass series due to the glass-forming role. In the perspective of radiation shielding features, the newly developed Phy-X/PSD software provided to compute mass attenuation coefficient ( $\mu_m$ ) and half-value layer ( $\Delta_{0.5}$ ). We found out that the increasing content of  $B_2O_3$  increased the  $\mu_m$  and decreased the  $\Delta_{0.5}$ . These alterations ensured to improve radiation shielding characteristics for  $B_2O_3$ -reinforced LZCS series. More essentially, the L0B15 sample can compete with the commercial radiation shielding glass, particularly the lead-free series of RS 263 G18. In conclusion, the authors reported that the insertion of  $B_2O_3$  is a good candidate for the LZCS glass system for the obtainment of advanced physical, optical, and radiation shielding features in comparison to the undoped situation.

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