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Calculation of LAC and HVL values of newly developed barium-borotellurite glass containing different heavy metal oxides using Phy-X/PSD

Farklı ağır metal oksitler içeren yeni geliştirilen baryum-borotellürit camının Phy-X/PSD programı kullanılarak LAC ve HVL değerlerinin hesaplanması

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Abstract

This paper examined the radiation shielding characteristics as linear attenuation (LAC) and half-value layer (HVL) of barium-borotellurite glass (BBT), 20BaO-20B_2O_3-60TeO_2, reinforced with 2.5 mol% of different heavy metal oxides (HMOs), X_2O_3 (X: Bi, Gd, La, Sm). For this purpose, five different glass systems (BBT: reference, BBTB: Bi_2O_3, BBTG: Gd_2O_3, BBTL: La_2O_3, and BBTS: Sm_2O_3) were explored by performing the newly developed Phy-X/PSD program for theoretical computations. The LAC and the HVL were found out in the photon energies of 0.015 to 15 MeV. Eventually, the findings were compared with some heavyweight concretes and commercial radiation shielding glasses to make a deeper sense. One can report that all HMOs addition contributed to increasing LAC while decreasing HVL thicknesses in our newly developed BBT system. In particular, the BBTB glass provided the best effectiveness in radiation shielding. Further, the BBTB glass system can compete with commercially available glasses, particularly, it could accomplish to overtake lead-oxide containing ones. This study revealed that BBT glasses with differing HMOs can effectively be used in radiation shielding applications.

Keywords: Borotellürit cam, Gama-ışını sönümleme, Kurşunsuz cam, Phy-X/PSD, Radyasyon zırhlama.

Öz

Bu çalışma, %2.5 mol farklı ağır metal oksitler (HMO'lar), X_2O_3 (X: Bi, Gd, La, Sm) ile güçlendirilmiş baryum-borotellurit (BBT), 20BaO-20B_2O_3-60TeO_2 camının doğrusal zayıflatma katsayısı (LAC) ve yarı-değer katmanı (HVL) olarak radyasyon zırhlama özelliklerini incelemiştir. Bu amaçla, teorik hesaplamalar için yeni geliştirilen Phy-X/PSD programı uygulanarak beş farklı cam sistemi (BBT: referans, BBTB: Bi_2O_3, BBTG: Gd_2O_3, BBTL: La_2O_3 ve BBTS: Sm_2O_3) araştırılmıştır. Doğrusal zayıflatma katsayısı (LAC) ve yarı değer katmanı (HVL) 0.015 ila 15 MeV foton enerjilerinde bulundu. Sonunda, bulguları anlamlandırmak için bazı ağır betonlar ve ticari radyasyon koruyucu camlarla karşılaştırıldı. Yeni geliştirdiğimiz BBT sistemimizde HVL kalınlıklarını azaltırken, tüm HMO'ların ilavesinin LAC'nin artmasına katkıda bulunduğu söylenebilir. Özellikle BBTB camı, radyasyondan korunmada en iyi etkinliği sağladı. Ayrıca BBTB cam sistemi, ticari olarak bulunan camlarla rekabet edebilir, ve hatta kurşun oksit içeren camları geçmeyi başarabilir. Bu çalışma, farklı HMO'lara sahip BBT camlarının radyasyondan korunma uygulamalarında etkili bir şekilde kullanılabileceğini ortaya koymuştur.

Anahtar kelimeler: Borotellurite glass, Gamma-rays attenuation, Lead-free glass, Phy-X/PSD, Radiation shielding.

1 Introduction

Due to the growing interest in research activities via numerous devices capable of high energies or the increasing need for applications such as medical diagnostics or nuclear energy production, the usage of shielding material against high photon energies (e.g. X-rays or gamma-rays) irradiated from these applications has emphasized the utmost importance of protecting onsite workers [1]-[5]. The reason is that high ionizing energy rays can damage living tissues, mutate DNA codes, and burn skin [6],[7]. With this in mind, protective actions such as standing an optimum distance away from the ray source, minimizing exposure to ray dose, or decreasing duration exposed to rays have completely been put into action [8]. Considering the best protection concerns towards radiation energies, the usage of shielding materials has become vital. For this, the lead metal as protecting material has intensively been used owing to its high density (11.34 g/cm^3) and easy shaping ability [9]. However, the toxicity of lead proved by many studies [10],[11] has restricted its common use. Heavy-weight concrete having high-density and dimension flexibility has principally been implemented in surrounding walls of irradiating devices [12],[13]. Yet, the utilization rate has eventuated in limited to technical deficiencies of concrete materials like cracking tendency as well as production difficulties [14],[15]. Nonetheless, the essential lack for both mentioned materials is that neither metallic lead nor heavy-weight concrete can provide transparency in visible light. However, there is a critical design parameter that almost all radiation applications require an observation window in accordance with radiation protection standards [16],[17]. At this point, glass materials come forward due to their transparency in visible light as well as effectiveness in radiation shielding.

Recently, glass materials have extensively been used in almost every area from daily lives to advanced technology applications [18]. Radiation shielding applications is one of these fields since attenuating high energy photons (e.g. X-rays or gamma rays) can effectively be carried out thanks to the glass materials. With compositional flexibility in glasses, various types such as silicate, borate, or phosphate have traditionally come to the forth till now [19]. These types of glasses containing several high-density oxides have extensively been studied by many

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researchers in the past decade for radiation shielding [20], [21], [22]. Though satisfactory consequences could be obtained in comparison to some standard or heavy-weight concrete materials, the effectiveness of these glass systems is highly narrowed due to limited transmission, optical losses, and nominal rare-earth solubility [23]. That is why studies have been put on heavy metal oxide glasses like gadolinium, bismuth, tellurite ones thanks to their superior properties including wide transmission range and good optical features. Most particularly, tellurite glasses stand out among others because of the low melting point, high refractive index, good thermal characteristics, and compositional flexibility [24],[25]. Therefore, researches conducting radiation shielding characteristics of tellurite glasses have dramatically increased so as to obtain a high-performance shielding material.

Tellurium dioxide (TeO_2) is not recognized as a glass network former but can form glass with some oxides additions [38]. Starting from this condition, numerous constituents of alkalis (e.g. Na_2O) or heavy metal oxide (e.g. Bi_2O_3, La_2O_3) contributions have been made, and thus it was considered as an alternative shielding glass [39]-[42]. For instance, in tellurite glasses, BaO addition is a good choice for improving radiation shielding effectiveness [26], or B_2O_3 can predominately provide glass-forming ability due to being a glass former [27]. Additionally, heavy metal oxides can contribute to the enhancement in radiation shielding properties since their high atomic weight and high densities increase the overall density of the tellurite glasses [28].

On the other hand, there are many studies focusing on different oxide additions in tellurite glasses [29], [30]. For glass property determination, Grelowska et al. [26] studied TeO_2-BaO-Na_2O and TeO_2-BaO-WO_3 glasses to observe structural and optical aspects. As BaO content increased in the tellurite glass system, the glass transition temperature was not directly affected whereas the refractive index and the transparency window (200 to 2500 nm) increased. Another study conducted by Ersundu et al. [31] evaluated the physical, structural, and shielding parameters on the K_2O-WO_3-TeO_2 glass system experimentally and theoretically. They showed the effectiveness of varying amounts of K_2O and WO_3 in tellurite glasses and concluded that the K30W60T10 sample (30, 60 and 10 in mol% for K_2O, WO_3 and TeO_2, respectively) provided the best radiation shielding in terms of mass attenuation coefficient. Lakshminarayana et al. [32] prepared a sodium zinc barium borotellurite glass system doped with Er_2O_3 and Pr_6O_{11} to reveal vibrational, thermal, and radiation shielding characteristics against gamma-rays. They utilized theoretical (WinXCom) and simulational (MCNP5 code) calculations followed by experimental measurements. According to their findings, TeO_4, TeO_{3+1}, TeO_3, BO_3, and BO_4 structural clusters occurred in the glass network. Besides, the produced glass was found to be transparent in visible light with ensuring high density (up to 5.402 g/cm^3), a very low HVL value, and possessing a good agreement in WinXCom and MCNP5 code calculations. Similarly, Tekin et al. [33] investigated B_2O_3-Bi_2O_3-SiO_2-TeO_2 glasses via the MCNPX Monte Carlo code. The authors pointed out that the increasing amounts of Bi_2O_3 paved the way for increasing the mass attenuation coefficient (LAC) while diminishing HVL thicknesses. Sayyed et al. [23] reported different heavy metal oxide such as Bi_2O_3, MoO_3, Sb_2O_3, WO_3, and ZnO contributions in the TeO_2-$PbCl_2$ glass system against high photon energies. They found out that whole contents improved the radiation shielding characteristics of the glass system, but Bi_2O_3 and WO_3 additions provided more. In addition to heavy metal oxide contributions in tellurite glass systems, many investigations were conducted on Gd_2O_3, La_2O_3, or Sm_2O_3 additions in different glass types [34],[35]. They revealed that all oxide ensured to obtain better shielding characteristics.

In the present study, 20BaO-20B_2O_3-60TeO_2, a barium-borotellurite (BBT) glass, was selected. Bi_2O_3, Gd_2O_3, La_2O_3, and Sm_2O_3 contents were separately added in the amount of 2.5 mol%, and radiation shielding characteristics against high ionizing energies were calculated theoretically via the newly developed and user friendly Phy-X/PSD program in the energy range of 0.015 to 15 MeV. Besides, the sofware findings were compared with some standard heavyweight concrete as well as commercially available radiation shielding glasses.

2 Materials & Methods

2.1 Glass composition design

In this work, five different glass systems were investigated via the Phy-X/PSD program within the energy range of 0.015 to 15 MeV. The glass compositions and density values are summarized in Table 1. According to Table 1, the given amounts of HMOs were introduced into the reference glass system (BBT). Afterward, the density values were calculated as differing from 4.7562 to 4.9260 g/cm^3 by applying Inaba and Fujino [36] relation given in Eq. 1.

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

Additionally, the chemical composition of glass systems can fairly be seen from Table 2. As HMOs content increases at the expense of TeO_2 amount, the elemental composition of each glass system differs, accordingly.

Just after the compositional design, the authors explored the radiation shielding characteristics for the glass systems. The calculations of shielding parameters were accomplished via the newly developed Phy-X/PSD program [37]. Basically, it is possible to obtain photon cross-section data for a single element, compound, or mixture (a combination of elements and compounds) thanks to this software. In the below sections, each parameter was explained in detail.

2.2 Radiation shielding parameters

The effectiveness of radiation shielding is independent of the type of materials used. In other words, any material can be used for shielding purposes if the necessary thickness values are assured. There is no doubt that the material's density takes a significant role in effective shielding, as well. To bring out shielding effectiveness, the linear attenuation coefficient (LAC) is calculated by the following expression, called the Beer-Lambert equation:

$$I = I_0 exp^{-\mu t} \tag{2}$$

The terms, I_0 and I, define the incident intensity on material and transmitted intensity from the material, respectively, whereas μ is the linear attenuation coefficient, and t is the thickness of the material.

Table 1. Chemical glass compositions (mol%) and densities (g/cm³) of the investigated barium-borotellurite glass systems.

Code	TeO_2	B_2O_3	BaO	Bi_2O_3	Gd_2O_3	La_2O_3	Sm_2O_3	Density (g/cm³)
BBT	60.0	20	20	0	0	0	0	4.7562
BBTB	57.5	20	20	2.5	0	0	0	4.9260
BBTG	57.5	20	20	0	2.5	0	0	4.8490
BBTL	57.5	20	20	0	0	2.5	0	4.8041
BBTS	57.5	20	20	0	0	0	2.5	4.8352

Table 2. Weight fraction of elements in each glass composition.

Code	Te	B	Ba	O	Bi	Gd	La	Sm
BBT	0.5455	0.0308	0.1957	0.2280	-	-	-	-
BBTB	0.4957	0.0292	0.1856	0.2189	0.0706	-	-	-
BBTG	0.5045	0.0297	0.1889	0.2228	-	0.0541	-	-
BBTL	0.5077	0.0299	0.1901	0.2242	-	-	0.0481	-
BBTS	0.5057	0.0298	0.1893	0.2233	-	-	-	0.0518

Afterward the half-value layer (HVL), another essential paramete, is calculated. The HVL is very useful to present the required material thickness value since the term is defined as the thickness value of a substance which is necessary to decrease the intensity of rays down to 50% of the initial situation. The following relation is used to calculate the HVL value.

$$HVL = \frac{ln2}{\mu} \tag{3}$$

3 Results & Discussions

3.1 Linear attenuation coefficient (LAC)

The barium-borotellurite (BBT) glass system doped with four different HMOs was explored via the Phy-X/PSD. Figure 1 represents the LAC of five different glass systems at different photon energies ranging from 0.015 to 15 MeV. According to Figure 1, all glass samples showed a similar trend towards increasing photon energy. That is, the LAC values rapidly reduced as photon energy increased up to 0.1 MeV. This phenomenon may be attributed to the photoelectric interaction with matter. In the higher photon energies, i.e. from 0.1 to 1 MeV, Campton scattering situation took place irrespective of atomic numbers of substances. This led to obtaining approximate LAC values for each of the glass systems. On the other hand, as the photon energy raised greater than 1 MeV, the pair production situation came forward, and thus the LAC values started to increase with the photon energy increment.

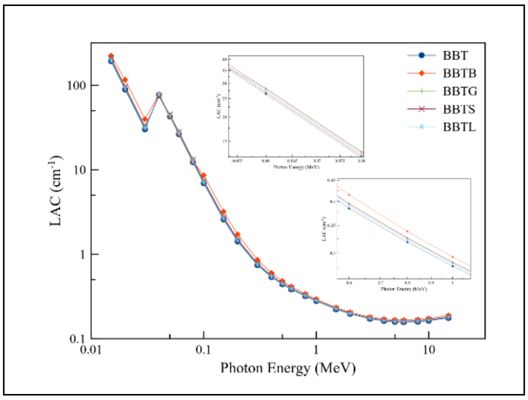

Figure 1. Linear attenuation coefficient (LAC) for the glass systems in the energy range 0.015 to 15 MeV.

Furthermore, the LAC values changed with differing HMOs additions. That is, the BBT glass had the lowest LAC value whereas the doped glasses provided higher values. To illustrate, in the intermediate energy region, e.g. at 0.5 MeV, the LAC value of BBT was calculated as 0.441 cm^{-1} whereas 0.482 cm^{-1} for BBTB, 0.454 cm^{-1} for BBTG, 0.452 cm^{-1} for BBTS, and 0.447 cm^{-1} for BBTL were found. From these findings, one can say that HMO contributions showed a reasonable effect on LAC increment at intermediate photon energy. Moreover, BBTB glass system had the highest LAC value while BBTG, BBTS, and BBTL glasses followed in the decreasing order. This might be attributed to the atomic numbers of constituents in the order of La<Sm<Gd<Bi. Although studies in literature do not cover these oxides altogether in one work, numerous seperate findings revealed that Bi_2O_3 [38], Gd_2O_3 [39], Sm_2O_3 [40], and La_2O_3 [41] can improve radiation shielding characteristics of glass systems in comparison to their non-doped situations. Our findings seems to have similar results to those ones. As a result of LAC investigations, BBTB and BBTG glass systems were found to be more effective in radiation shielding applications in comparison to BBTL and BBTS as well as BBT glasses.

3.2 Half-value layer (HVL)

In radiation shielding applications, the HVL is an essential indicator of radiation shielding effectiveness. In this study, the HVL values were calculated with respect to the given formula, and the results were graphically demonstrated in Figure 2 within 0.015 to 15 MeV photon energies.

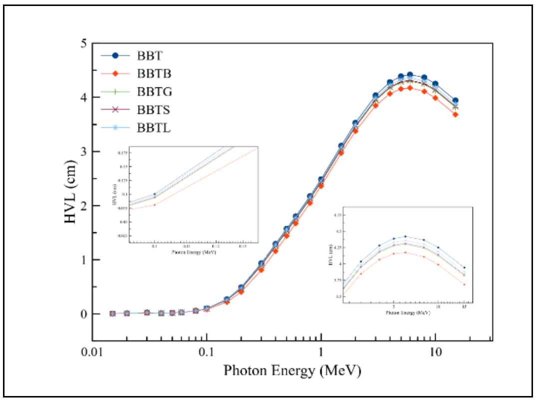

Figure 2. HVL values for the glass systems in the energy range 0.015 to 15 MeV.

It is considered that the lower the HVL value is, the higher the radiation shielding ability will be. With this in mind, it was evidently seen that the HVL values for all-glass systems were found very small thicknesses, namely around 3.30×10^{-3} cm at 0.015 MeV. As the photon energy was increased to higher values, for instance to 5 MeV, the HVL values become higher which is greater than 4.4 cm. In detail, the lowest HVL thickness was achieved by BBTB glass sample with 4.16 cm of HVL. Therefore one can deduce that bismuth oxide addition can improve radiation shielding properties of glass systems.

When it comes to evaluating the effects of different heavy metal oxide additions, all constituents reduced the HVL thicknesses compared to BBT glass. To illustrate this, at 0.1 MeV photon energy, BBT glass had the thickness value of 0.100 cm while BBTB, BBTG, BBTS, and BBTL possessed 0.081, 0.093, 0.095, and 0.097 cm, respectively. This means that Bi2O3 addition ensured to have the lowest HVL values among others. In conclusion, one can say that the HVL thickness is highly dependent on heavy metal oxide additions which also means that the atomic numbers, as well as the density values, are very effective on HVL thicknesses.

3.3 Comparison of HVL values

We performed the LAC and the HVL calculations to our glass systems. The findings were discussed with each other, however, it is more essential to make sense of the values with other shielding materials, for example, concrete materials and commercial ones. At this point, Table 3 clearly lists some shielding materials and their HVL thicknesses at 0.662 and 1.250 MeV photon energies. According to Table 3, concrete materials as standard and hematite-serpentine [42] provide 3.88 and 3.62 cm thicknesses, respectively at 0.662 MeV. At the same photon energy, our glass systems have by far lower thickness values, namely almost half of the concrete materials. Therefore, even though no other heavy metal oxide addition is employed in the glass system, the BBT has very lower HVL values.

In other respects, a comparison between our findings and commercially available ones have become more of an issue. By implying commercial products, we have meant RS series radiation shielding glasses produced by Schott company [43]. In that series, RS253 and RS253G18 glasses do not contain any lead-oxide (PbO) content whereas the remaining have it in the amounts of 33, 45, and 71 wt%, respectively. The lead-oxide-free glass systems of our study can be called as environmentally-friendly and non-toxic systems. At 0.662 MeV photon energy, all-glass systems of this study showed by far the lower HVL values when compared to RS253, RS253G18, RS323G18, and RS360 glasses. This may be attributed to the higher density values of our glass systems. Nothing but RS520 has a lower thickness than our glass systems, however, we have considered that BBTB glass can compete with RS520 in terms of HVL values. Although relatively higher thickness value was observed in our glasses compared to RS520 at 0.662 MeV, the difference in thickness values obviously decreased as the photon energy became higher, for instance, 1.250 MeV. That is to say, the difference between BBTB and RS520 at 0.662 MeV was calculated as 0.28 cm whereas 0.66 cm seemed obtainable at 1.250 MeV. For this reason, it can be deduced that BBTB can compete stronger at higher photon energies with commercial radiation shielding materials.

4 Conclusions

To sum up, a barium-borotellurite glass, $20BaO$-$20B_2O_3$-$60TeO_2$, was investigated. Different HMOs of Bi_2O_3, Gd_2O_3, La_2O_3, and Sm_2O_3 as 2.5 mol% were separately introduced in the glass system, and radiation shielding characteristics were calculated via the Phy-X/PSD program in the energy range of 0.015 to 15 MeV. It was found out that the LAC values changed with differing heavy metal oxide additions. The BBTB glass system had the highest LAC value while BBTG, BBTS, and BBTL glasses followed in the decreasing order. The atomic weights of HMOs imposed the LAC values. Further, the authors figured out that the HVL value increased as the photon energy became higher. When it comes to evaluating the effects of different heavy metal oxide additions, all constituents reduced the HVL thicknesses compared to BBT glass. In the lower photon energies, BBTB glass ensured to have the lowest HVL value whereas BBTL provided the least value for HVL in the higher photon energies.

Table 3. A comparison for the HVL of some concretes, commercial glasses, and the investigated glasses.

Density (g/cm³)	Shielding Material	*Energy (MeV)*	
		0.662	*1.250*
2.400	Standard concrete	3.88	-
2.500	Hematite serpentine concrete	3.62	-
2.495	Commercial window glass	4.73	-
2.500	RS253	3.65	4.950
2.520	RS253G18	3.65	4.950
3.260	RS323G18	2.48	3.850
3.600	RS360	2.17	3.300
5.180	RS520	1.39	2.310
4.756	BBT	1.80	3.105
4.926	BBTB	1.67	2.969
4.894	BBTG	1.75	3.040
4.804	BBTL	1.78	3.072
4.835	BBTS	1.76	3.048

As a result of comparison with heavyweight concrete materials and commercially available radiation shielding materials, we determined that all glass systems of our study showed a very lower HVL thickness in comparison to concrete materials. Further, none but RS520 had a lower thickness than our glass systems. In conclusion, contributions of different HMOs in our newly developed barium-borotellurite glass system have promising results in radiation shielding applications.

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6 Author contribution statements

Recep KURTULUŞ, Creating the idea, designing, literature review, writing and critical review. Taner KAVAS, Supervision, collecting data, writing and critical review.

7 Ethics committee approval and conflict of interest statement

There is no need to obtain permission from the ethics committee for the article prepared. Additionally, there is no conflict of interest with any person / institution in the article prepared.

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