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INVESTIGATION OF THE RADIOLOGICAL SHIELDING PROPERTIES OF PHOSPHATE GLASSES WITH ADDED BISMUTH OXIDE

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ABSTRACT

The addition of specific additives to glass compositions can enhance their shielding properties. This study aimed to investigate shielding parameters such as the linear attenuation coefficient, half-value layer (HVL), tenth-value layer (TVL), and saturation thickness in phosphate glass doped with bismuth oxide. The research was conducted using Monte Carlo computational simulations, which enable accurate modeling of real experiments and analysis of various parameters. The bismuth oxide-doped glasses studied in this work exhibited superior shielding parameters compared to commonly used shielding materials, such as barite concrete.

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INTRODUCTION

Exposure to ionizing radiation poses significant health risks, including cellular damage and increased cancer risk, making the development of effective shielding materials essential for medical, industrial, and research applications. The selection of shielding materials is crucial in the development of radiation protection devices, requiring a careful evaluation of their physical, chemical, and radiological properties, as well as considerations of availability and cost. Traditionally, materials such as lead, barite concrete, and metal-based composites have been widely used for radiation shielding due to their high density and attenuation capabilities. However, these materials present several drawbacks, including toxicity, environmental concerns, excessive weight, and mechanical limitations (Bijanu *et al.*, 2021; Daungwilailuk *et al.*, 2022; Ihsani *et al.*, 2024). Recent studies indicate that combining multiple layers of distinct materials can significantly enhance shielding effectiveness, providing more robust protection (Nascimento *et al.*, 2018). Additionally, the geometry of shielding structures plays a key role in radiation dose distribution, necessitating meticulous design to optimize protection. Among the emerging materials for radiation shielding, glass has gained attention due to its unique combination of transparency, mechanical strength, and compositional versatility. Unlike traditional shielding materials, glass can be engineered to exhibit specific mechanical, optical, and thermal properties by incorporating different additives (Kavaz *et al.*, 2019; Karpuz, 2023; Kurtulus, 2024). In particular, doping glass with high atomic number elements enhances its radiation attenuation capabilities while maintaining advantages such as lightweight properties and ease of manufacturing. Bismuth oxide (Bi_2O_3) has been widely studied as an alternative to lead due to its high density, non-toxic nature, and excellent photon attenuation properties. Glass doped with bismuth oxide is produced through melting and rapid cooling processes, resulting in a material with a high refractive index and increased density (Singh &Karmakar, 2012). These characteristics contribute to enhanced radiation attenuation, making bismuth oxide-doped glass a strong candidate for medical and industrial applications, such as protective barriers in radiology rooms, observation windows in nuclear facilities, and shielding elements in portable radiation protection devices (Cheewasukhanont *et al.*, 2020; Kamislioglu, 2021; Alzahrani *et al.*, 2023; Thabit *et al*, 2023). Computational modeling Monte Carlo simulations, have facilitated the precise evaluation of shielding parameters, allowing for optimized material design before experimental validation. These techniques enable the assessment of crucial properties such as the linear attenuation coefficient, half-value layer (HVL), and tenth-value layer (TVL), contributing to the development of more efficient and lightweight shielding materials. The combination of advanced materials, strategic design, and innovative technologies highlights the importance of ongoing research in the field of ionizing radiation shielding. In this context, bismuth oxide-doped glasses present a promising alternative. In this research we further explore these properties, evaluating the effectiveness of bismuth oxide-doped phosphate glasses in radiation protection applications.

MATERIALS AND METHODS

Monte Carlo Simulation: FLUKA (FLUktuierende KAskade) version 4-3.0, installed on Ubuntu 20.04.6 LTS, was used to carry out the simulations in this article.It is a fullycomputational simulation tool that uses the Monte Carlo method to calculate how different particles interact with matter (Battistoni *et al.*, 2015). It is widely used to simulate particle propagation in materials, allowing precise analysis of how they behave when passing through different substances. This is essential for various applications, such as radiotherapy and particle physics, to understand radiation effects and optimize the design of radiological protection systems. Additionally, FLUKA includes an advanced user interface called FLAIR (Vlachoudis, 2009), which simplifies the editing of input files, the construction of simulation geometries, and the execution of codes. It also integrates the Gnuplot graphics utility, enabling the generation and visualization of output data through graphical representations.

Radioactive Sources: For the attenuation experiments, four radioactive sources emitting gamma rays with different energies were used, representing a wide spectrum of radiations found in real-world applications (Table 1). Moreover, these gamma sources are among the most commonly used commercially and have long half-lives, ensuring excellent operational durability.

Table 1. Isotopes Energy

Isotopes	Energy (keV)
Americium (Am 241)	59.5
Barium (Ba 133)	356
Cesium (Cs 137)	662
Cobalt (Co 60)	1337

Glass samples doped with bismuth oxide were simulated, varying the sample thickness according to the isotope. The variation in thickness is shown in Table 2:

Table 2. Variation of Simulated Thicknesses

Isotopes	Thickness (cm)
Americium (Am 241)	0.1 cm to 1.2 cm, with an increment of 0.1 cm
Barium (Ba 133)	0.1 cm to 4 cm, with an increment of 1 cm
Cesium (Cs 137)	2 cm to 28 cm, with an increment of 2 cm
Cobalt (Co 60)	2 cm to 40 cm, with an increment of 2 cm

The target in FLUKA is composed of a mixture of $0.7P_2O_5$ and $0.3Bi_2O_3$. This composition consists of phosphorus (P), oxygen (O), and bismuth (Bi), with their respective mass percentages being 18.13% phosphorus, 29.44% oxygen, and 52.43% bismuth.

Simulation Setup: In the experiment, a gamma-ray beam is directed at the target material, and the photons that manage to pass through the material without interacting are collected by a detector positioned on the opposite side (Figure 1 and 2). These transmitted photons are then counted and used to calculate attenuation parameters. The detector is encapsulated in a lead collimator.

Figure 1. FLUKA Monte Carlo simulation setup used for mass attenuation coefficients calculations of bismuth oxide-doped glass.

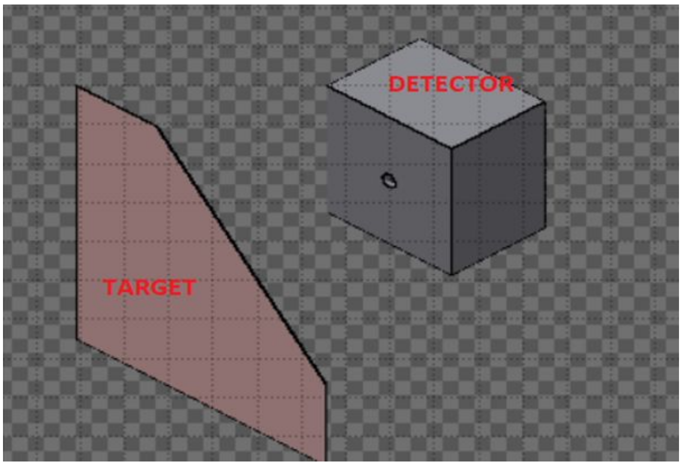

Figure 2. **3D of the simulation geometry used**.

Exponential Data Fit: The attenuation coefficients were obtained for each energy by fitting the exponential graph of counts versus thickness, allowing for the calculation of shielding parameters (HVL, TVL, MFP). The experimentally obtained linear attenuation coefficients were compared with theoretical values generated by the Phy-X software (Şakar *et al.*, 2020). Lambert (1870) observed that light transmission is related to the thickness of the absorbing medium layer. When a beam of monochromatic light passes through a homogeneous transparent medium, each layer of the medium absorbs a constant fraction of the light that passes through it, regardless of the intensity of the incident light. This led to the formulation of Lambert's Law: "The intensity of emitted light decreases exponentially as the thickness of the absorbing medium increases arithmetically (Guimarães *et al.*, 2008)." Thus, when a beam of monoenergetic photons strikes a material of thickness x, parts of the incident photons are removed from the primary beam. Letting I_0 represent the intensity of the incident beam, it is related to the intensity I of the transmitted beam based on Lambert-Beer's Law, also known as the exponential attenuation law, represented by:

$$\boldsymbol{I(x) = I_0 e^{-\mu x}} \quad (1)$$

where $I(x)$ is the transmitted intensity after passing through thickness x, I_0 is the initial intensity, and μ is the linear attenuation coefficient.The photon count data was plotted in graphs for each energy. An exponential fit was performed using the Lambert-Beer Law equation.

Calculation of Shielding Parameters: Greater material thickness leads to increased radiation absorption, reducing the intensity of the beam that passes through it. Absorption follows an exponential law, causing the intensity to gradually decrease without ever being eliminated. The absorption efficiency varies depending on the material, explained by the absorption coefficient "μ", which indicates the probability of a photon being removed from the beam per unit length, either by absorption or scattering (Tauhata *et al.*, 2003). Using the μ values obtained for each energy and material, the following parameters were calculated:

Half-Value Layer (HVL): The thickness needed to reduce the radiation intensity by half, calculated as:

$$HVL = \frac{\ln(2)}{\mu} \quad (2)$$

where μ is the linear attenuation coefficient.

Tenth-Value Layer (TVL): The thickness needed to reduce the intensity to one-tenth of the initial value, calculated as:

$$TVL = \frac{\ln(10)}{\mu} \quad (3)$$

where μ is the linear attenuation coefficient.

Mean Free Path (MFP): The average distance a photon travels in the material before interacting, calculated as:

$$MFP = \frac{1}{\mu} \quad (4)$$

where μ is the linear attenuation coefficient.

Comparison with Theoretical Values: The experimentally obtained linear attenuation coefficients were compared with theoretical values generated by the Phy-X software. Phy-X is an online tool that calculates various essential parameters for radiation shielding, such as the attenuation coefficient, half-value layers, tenth-value layers, and mean free path over a wide range of energies from 1 keV to 100 GeV (Şakar *et al*., 2020). These theoretical parameters will be compared with simulated results, enabling a more accurate analysis of the tested materials' efficiency and validating the methods used.

RESULTS AND DISCUSSION

Fluence measures the number of particles passing through a unit area. Using fluence graphs generated by FLUKA, it is possible to analyze the scattering pattern of the beam when it hits the doped glass. The fluence graph (Figure 3) shows the beam scattering upon hitting the doped glass. The incident beam is focused on the central direction of the external detector, while scattering occurs in various directions after colliding with the glass.

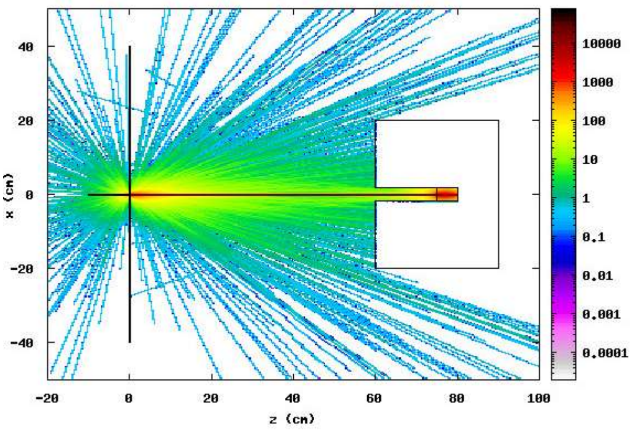

Figure 3. Photon Fluence

The count versus sample thickness graphs are shown in Figure 4,5,6 and 7. The red line represents the exponential fit performed with the points obtained from the simulation. The equation obtained from the fit, which directly provides the linear attenuation coefficient, is also indicated in each graph.

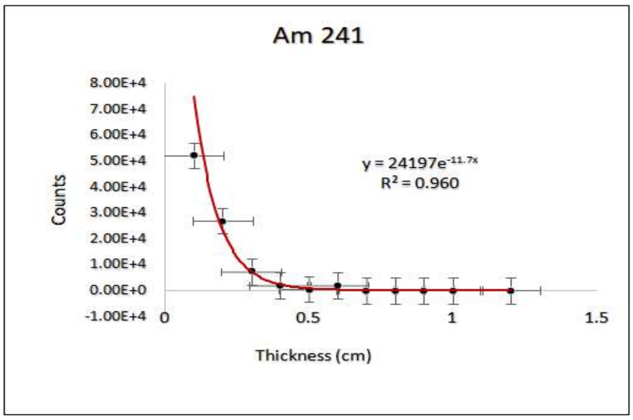

Figure 4. Photon count for Am 241 isotope by target material thickness

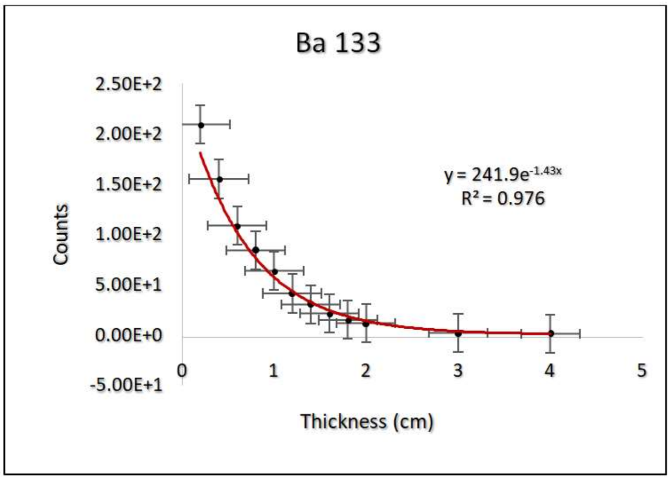

Figure 5. Photon count for Ba 133 isotope by target material thickness

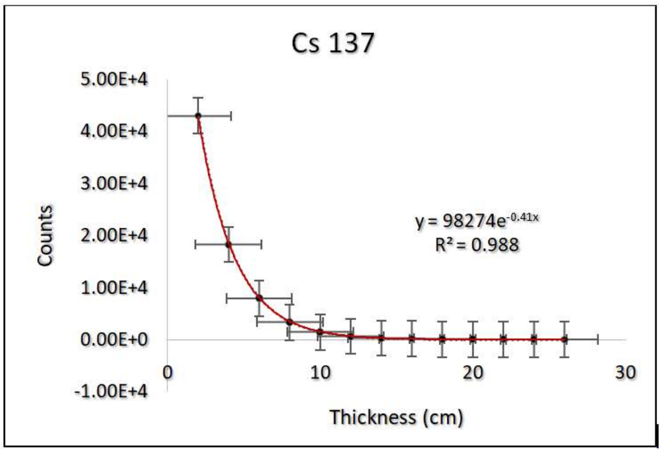

Figure 6. Photon count graph for Cs 137 isotope by target material thickness

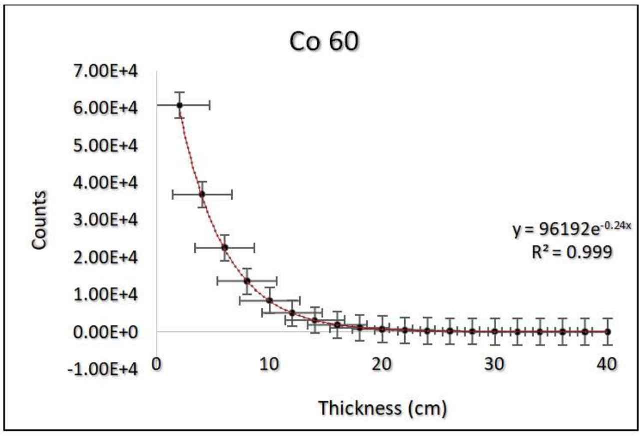

Figure 7. Photon count graphs for Co 60 isotope by target material thickness

In Table 3,4,5 and 6, the shielding parameters calculated based on the linear attenuation coefficient obtained through the exponential curve fit are compared with the theoretical values provided by the Phy-X software. Comparing the experimentally obtained values with the theoretical values from Phy-X, the following observations can be made. The experimental linear attenuation coefficient for Am-241, calculated from the exponential fit, was approximately half of the theoretical reference value, suggesting an unsatisfactory fit. This discrepancy is attributed to the high attenuation of the low-energy Am-241 beam in the target material. For thicknesses above 0.5 cm, the beam is completely attenuated, as shown in Figure 4.

Table 3. Comparison of experimental and theoretical linear attenuation coefficient values

Isotope	Coefficient (μ) (cm^{-1}) (exp)	Coefficient (μ) (cm^{-1}) (Phy-X)
Am 241	11.74	22.155
Ba 133	1.437	1.262
Cs 137	0.417	0.495
Co 60	0.246	0.256

Table 4. Comparison of experimental and theoretical HVL values

Isotope	HVL (cm) (exp)	HVL (cm) (Phy-X)
Am 241	0.059	0.0312
Ba 133	0.482	0.549
Cs 137	1.661	1.401
Co 60	2.817	2.708

Table 5. Comparison of experimental and theoretical TVL values

Isotope	TVL (cm) (exp)	TVL (cm) (Phy-X)
Am 241	0.196	0.104
Ba 133	1.602	1.824
Cs 137	5.521	4.65
Co 60	9.360	8.996

Table 6. Comparison of experimental and theoretical MFP values

Isotope	MFP (cm) (exp)	MFP (cm) (Phy-X)
Am 241	0.085	0.0451
Ba 133	0.695	0.792
Cs 137	2.395	2.021
Co 60	4.065	3.907

For the other isotope energies (Ba-133, Cs-137, and Co-60), the experimental values closely matched the theoretical ones, indicating good agreement. The calculated values for the Half-Value Layer (HVL), Tenth-Value Layer (TVL), and Mean Free Path (MFP) suggest that the phosphate glass with bismuth oxide evaluated in this study exhibits high efficiency in gamma radiation shielding.When compared to materials commonly used for this purpose, such as barite concrete, its shielding performance proves to be even superior, as supported by the literature (Gausmann& Boldo, 2023). Therefore, glasses doped with heavy oxides emerge as highly promising materials for radiation protection in both medical and industrial environments.

CONCLUSION

Glasses doped with bismuth oxide have proven to be highly effective in shielding low-energy radiation, such as that emitted by Americium-241, although their efficiency decreases as radiation energy increases, as expected. Despite some discrepancies between experimental and theoretical values for Am-241, the results for higher-energy isotopes (Ba-133, Cs-137, and Co-60) showed strong agreement with Phy-X calculations, reinforcing the reliability of the experimental methodology. The calculated shielding parameters, including the Half-Value Layer (HVL), Tenth-Value Layer (TVL), and Mean Free Path (MFP), confirm that phosphate glass with bismuth oxide exhibits superior radiation attenuation compared to conventional materials such as barite concrete. These findings highlight the potential of doped glasses as advanced shielding materials, particularly for medical and industrial applications where transparency, reduced weight, and customizable composition offer significant advantages.

Furthermore, the combination of experimental measurements and Monte Carlo simulations effectively validates the use of computational modeling in evaluating shielding efficiency. This underscores the importance of integrating theoretical and experimental approaches to optimize the development and practical application of bismuth oxide-doped glasses in real-world radioprotection scenarios.

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