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Original Article

Gamma Ray Shielding Study of Barium–Bismuth–Borosilicate Glasses as Transparent Shielding Materials using MCNP-4C Code, XCOM Program, and Available Experimental Data

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ABSTRACT

In this work, linear and mass attenuation coefficients, effective atomic number and electron density, mean free paths, and half value layer and 10^{th} value layer values of barium–bismuth–borosilicate glasses were obtained for 662 keV, 1,173 keV, and 1,332 keV gamma ray energies using MCNP-4C code and XCOM program. Then obtained data were compared with available experimental data. The MCNP-4C code and XCOM program results were in good agreement with the experimental data. Barium–bismuth–borosilicate glasses have good gamma ray shielding properties from the shielding point of view.

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1. Introduction

Today's application of radiation sources and radioactive materials in various fields, such as nuclear power plants, nuclear medicine, as well as industry and agriculture, has made it essential to study different parameters related to shielding against harmful and dangerous radiations [1–5].

Concretes are the most common radiation shielding materials, because they are inexpensive and easily adapted to any types of construction, so they are commonly used against ionizing radiations [6–8]. However, concrete has many disadvantages and can be damaged by many processes, such as the expansion of aggregates, freezing of trapped water, fire or radiant heat, bacterial corrosion, leaching, physical and chemical damage, and considerable variability in its composition and water content [9]. In addition, concrete is opaque to visible light, and with the increasing use of gamma rays in the industry of medicine and agriculture, it is important to develop transparent radiation shielding materials. Glass materials are a good option for this purpose because they are 100% recyclable, can be transparent to visible light, and their properties can be modified and changed by adding other compounds [10,11].

Various types of glasses have been introduced to different nuclear applications. In the present work, barium–bismuth–borosilicate glass has been considered. Borosilicate glass is a type of glass with silica and boron oxide constituents [12]. These glasses are well known for their very low thermal expansion coefficients, resistance to thermal shock, and ability for transmission to visible light. Bismuth contributes to the stabilization of glass structure and improves chemical durability [11]. Moreover, bismuth and barium, due to their high atomic numbers, promote gamma ray shielding properties of the glass. The linear and mass attenuation coefficients, effective atomic number and electron density, means free paths, and half value layer (HVL) and 10^{th} value layer (TVL) values of barium–bismuth–borosilicate glasses were calculated for ^{60}Co (1,173 keV and 1,332 keV) and ^{137}Cs (662 keV) gamma rays on the basis of the elemental composition of glass samples using MCNP-4C code (Los Alamos National Laboratory, New Mexico, United States) and XCOM program (National Institute of Standards and Technology (NIST), Gaithersburg, Maryland, United States). The MCNP code is a general-purpose Monte Carlo radiation transport code for modeling the interaction of radiation with matter [13].

In addition, the theoretical values for mass attenuation coefficients of the different elements, compounds and mixtures have been provided by Hubbell and Seltzer [14] and given in the form of XCOM program at energies 1 keV to 100 GeV by Gerward et al. [15]. Therefore, XCOM program was used for the determination of shielding characteristics and for comparison with MCNP results too. Also, in order to verify and validate simulated and calculated values, the obtained results were compared with available experimental data [16].

2. Materials and methods

2.1. *Geometry of glass samples*

Cylindrical geometries were employed for the modeling of glass samples. Eight sections of subcylinders, 15 cm in diameter and 2 cm in thickness, were considered for every type of sample and set on the z axis in tandem.

2.2. *Source specification*

Attenuation coefficients of the glass samples were measured in a narrow beam transmission geometry using planar sources with collimated and monoenergetic beam and uniform distribution of radioactive material upon them, which emit gamma rays perpendicular to the front face of the shields (in the direction of z axis). A disc source with 2 cm diameter, which was parallel to the *x/y* plane and the origin of which was on the z axis, was defined in an MCNP data card with ERG, PAR, POS, and DIR commands for energy, type of particle, position, and direction, respectively.

2.3. *Material specification of glass samples*

The elemental composition of glass samples depends mainly on the mix proportions and chemical composition of the materials used. According to the experimental condition [16], the barium–bismuth–borosilicate glass samples were considered as 50BaO–xBi$_2$O$_3$–(50–x) borosilicate glass, where x is expressed in terms of mol% (x is 0, 5, 10, 15, and 20). The chemical composition and densities of glass samples and borosilicate glass are shown in Tables 1 and 2, respectively. Also, the percentages by weight of each element in the glass samples used in the material card of MCNP are presented in Table 3.

2.4. *Detector geometry and tally definition*

A small cylinder, 2 cm in diameter and 2 cm in length, was considered as the detector volume and set inside a detector collimator 33 cm away from the source. The collimator is

Table 1 – Chemical composition and densities of glass samples.

Glass samples	Density (g/cm^3)	Composition (mol%)		
		BaO	Bi_2O_3	Borosilicate glass
S1	3.45	50	0	50
S2	3.67	50	5	45
S3	3.81	50	10	40
S4	3.97	50	15	35
S5	4.21	50	20	30

Table 2 – Chemical composition (by weight) of borosilicate glass.

Compound	%
B_2O_3	20.20
Na_2O	8.21
Al_2O_3	17.35
SiO_2	48.51
K_2O	5.73

Table 3 – Percentage of atomic composition of five barium–bismuth–borosilicate glass samples.

Element	Atomic number	Glass samples				
		S1	S2	S3	S4	S5
Boron	5	1.94	1.48	1.14	0.88	0.68
Oxygen	8	22.96	19.97	17.77	16.07	14.74
Sodium	11	1.88	1.43	1.11	0.86	0.66
Aluminum	13	2.83	2.16	1.67	1.29	1.00
Silicon	14	7.00	5.34	4.12	3.19	2.45
Potassium	19	1.47	1.12	0.86	0.67	0.51
Barium	56	61.92	52.51	45.58	40.27	36.06
Bismuth	83	0	15.99	27.75	36.77	43.90

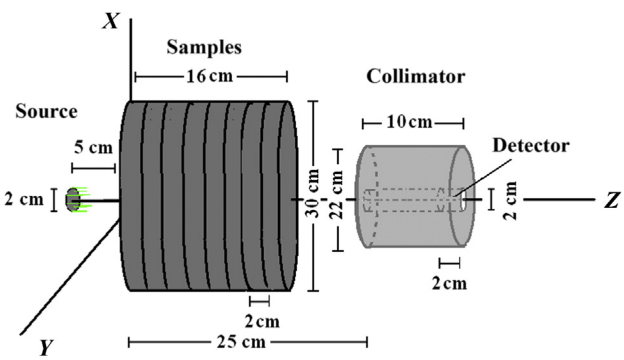

Fig. 1 – Geometry of modeled configuration (sizes are not on scale).

made of a 10-cm-long lead cylinder, with 22 cm and 2 cm outer and inner diameters, respectively. Fig. 1 shows the geometry of the system used for simulation.

Tally F4 was used to obtain MCNP-4C simulation data. This tally calculates average flux in a cell (detector volume) for only one gamma photon that enters the cell.

Simulations were performed with 100,000 to 1 million histories depending on the type and thickness of glass samples. All results simulated by MCNP-4C code were reported with less than 0.1% error.

3. Results and discussion

3.1. *Transmission factor*

The transmission factor of any type of glass sample, $T(E, d)$, for gamma ray of energy E through thickness d (cm) of the shielding glass sample, was obtained by dividing the average flux value in a detector, $\Phi(E, d)$, attained by Tally F4, by the average flux value in the same detector volume, $\Phi(E, 0)$, in the absence of any shielding material, as shown in Eq. (1):

$$T(E, d) = \frac{\Phi(E, d)}{\Phi(E, 0)} \quad (1)$$

Transmission factors for 662 keV, 1,173 keV, and 1,332 keV gamma rays as a function of thickness of glass samples, for all glass types, are shown in Figs. 2–4, respectively.

It is obvious from these figures that the S5 glass sample (20 mol% of Bi_2O_3) with the highest density and S1 glass sample (0 mol% of Bi_2O_3) with the lowest density have, respectively, the most and the least attenuation (least and most transmission) compared with other types of glass samples. It is also found that the transmission factors of glass samples based on the gamma ray energy span from 1 to $<10^{-2}$.

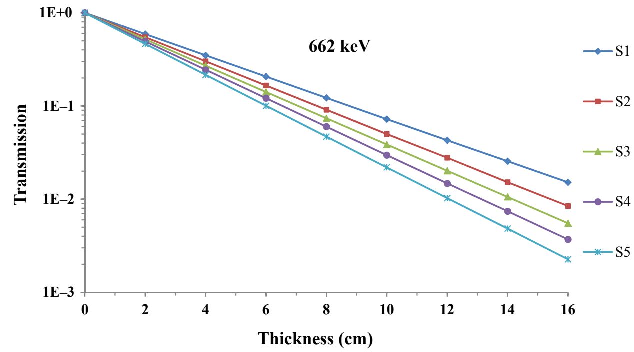

Fig. 2 – Transmission factor of glass samples for 662 keV gamma ray of ^{137}Cs.

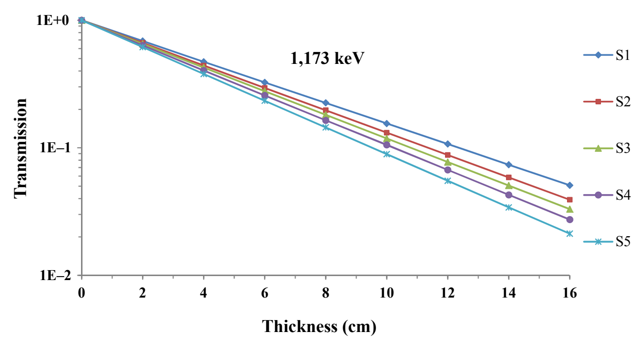

Fig. 3 – Transmission factor of glass samples for 1,173 keV gamma ray of ^{60}Co.

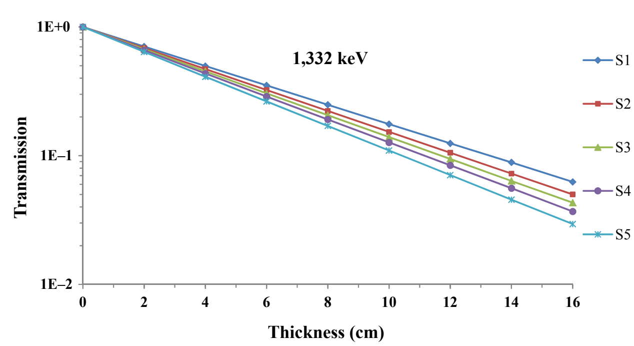

Fig. 4 – Transmission factor of glass samples for 1,332 keV gamma ray of ^{60}Co.

In order to compare the transmission rates of the studied photon energies through identical glass samples, transmission factors of these three gamma rays through an S5 glass sample are shown in Fig. 5.

It can clearly be seen from Fig. 5 that glass of higher thickness is needed for gamma rays with higher energy. The

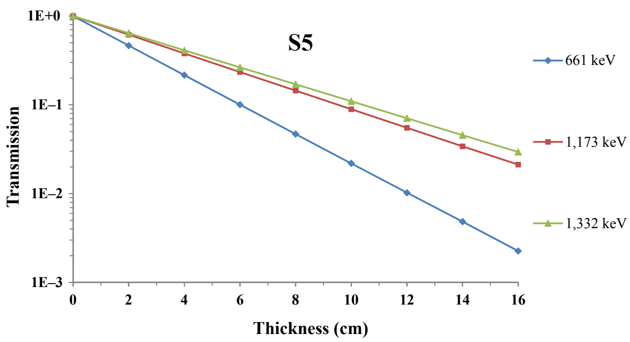

Fig. 5 – Transmission factor of studied photon energies through an S5 glass sample.

differences between transmission factors of studied gamma rays become larger for glass samples of greater thickness.

3.2. Linear and mass attenuation coefficients of glass samples

Linear and mass attenuation coefficients of glass samples (μ and μ_m) for the studied gamma rays were derived from transmission factor curves through fitting Lambert law ($I = I_0e^{-\mu t}$) using MATLAB software (version 7.10.0.499; The MathWorks, Inc. Natick, Massachusetts, United States) with excellent correlation coefficient R^2. Lambert law is described by the following equation:

$$I = I_0e^{-\mu t} \quad (2)$$

In the equation, I_0 and I denote incoming and outgoing intensities of photons through attenuator, t is sample thickness, and μ denotes the linear attenuation coefficient. The mass attenuation coefficients were calculated by dividing the linear attenuation coefficient of each sample of barium–bismuth–borosilicate glass by its density.

Alternately, mass attenuation coefficients of glass samples were calculated using XCOM program data by Eq. (3), in which w_i and $\mu_{m,i}$ (obtained directly from XCOM program) are the percentage by weight and mass attenuation coefficient of the i^{th} element in the concrete, respectively [1]:

$$\mu_m = \sum_{i=1}^{n} w_i \times \mu_{m,i} \quad (3)$$

Mass attenuation coefficients of glass samples obtained by MCNP-4C code and XCOM program for photon energies of interest in this research are presented in Table 4. In addition, obtained data are compared with available experimental data in this table. The simulation and calculation values of linear attenuation coefficients along with experimental values are shown in Fig. 6.

In Fig. 7, relative deviation (RD) values, the differences between simulation and theoretical results with experimental data of mass attenuation coefficients at 662 keV, 1,173 keV, and 1,332keV photon energies, are plotted for MCNP-4C code and XCOM program using Eq. (4):

$$RD = \{\text{theoretical} - \text{experimental}\} \times 100/\text{experimental} \quad (4)$$

The RD values range from 0.64% to 7.99% and from 0.38% to 5.19% for MCNP and XCOM results, respectively, for all the glass samples. The RD values were found to be < 8% for all barium–bismuth–borosilicate glass samples. Average differences in the MCNP and XCOM data with the experimental results of μ_m were 3.75% and 2.57%, respectively. In addition, it was found that the RD values in Fig. 7 were roughly independent of glass samples.

A good agreement was observed between experimental and theoretical values; the discrepancies are considered not to be very large because the differences are in the range of the reported experimental errors, which is <3.5% [16].

As shown in Fig. 6, the results of linear attenuation coefficients increase with increase in the Bi_2O_3 content (or density) of glass system, which may be due to an increase in the weight fraction of a higher-atomic-number constituent (Bi) as compared with other elements (weight fraction of barium element, as that of other higher-atomic-number constituents is constant for all glass samples). This increase is more intense with lower energies, so that the μ_{S5}/μ_{S1}ratios for 662 keV, 1,173 keV, and 1,332 keV gamma rays for experimental and theoretical results are approximately 1.4, 1.3, and 1.2, respectively. It could be due to the photoelectric effect that is favored by low-energy photons and high-atomic-number absorbers.

It should be noted that the discrepancies between MCNP and XCOM values can also be attributed to differences in employed techniques and databases for each method.

3.3. Mean free paths, and HVL and TVL values of glass samples

The mean free path (MFP), which is defined as the average distance between two successive interactions of photons in an

Table 4 – Mass attenuation coefficients (cm^2/g) of barium–bismuth–borosilicate glass samples.

Glass Samples	662 keV			1,173 keV			1,332 keV		
	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.
S1	0.0763	0.0752	0.0721	0.0543	0.0540	0.0525	0.0505	0.0505	0.0502
S2	0.0816	0.0800	0.0789	0.0555	0.0550	0.0533	0.0512	0.0512	0.0503
S3	0.0857	0.0836	0.0807	0.0560	0.0557	0.0549	0.0519	0.0517	0.0504
S4	0.0888	0.0864	0.0824	0.0570	0.0563	0.0555	0.0523	0.0521	0.0506
S5	0.0909	0.0886	0.0842	0.0575	0.0567	0.0565	0.0527	0.0525	0.0515

Exp., experiment.

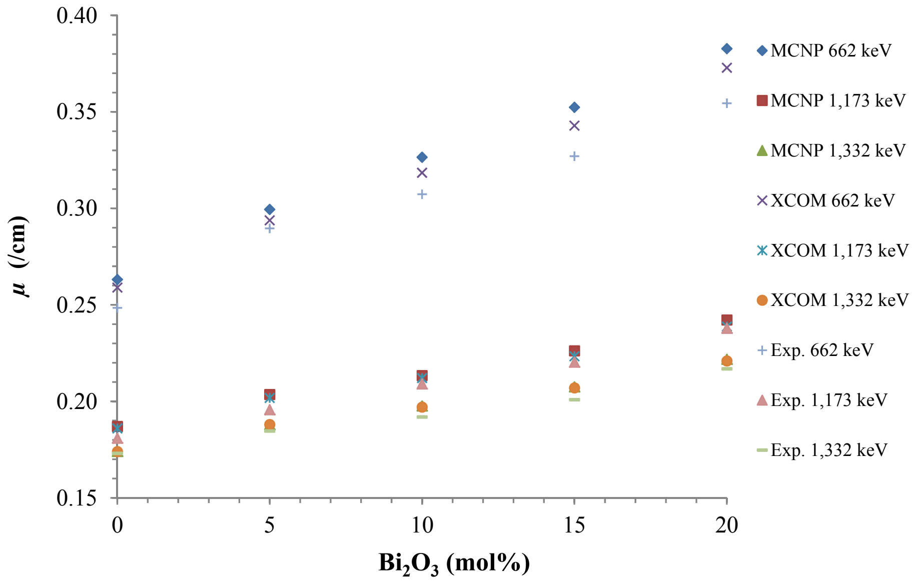

Fig. 6 – Dependency of linear attenuation coefficient of barium–bismuth–borosilicate glass samples on mol% of Bi_2O_3. (Comparison is made between simulation, theoretical and experimental values.)

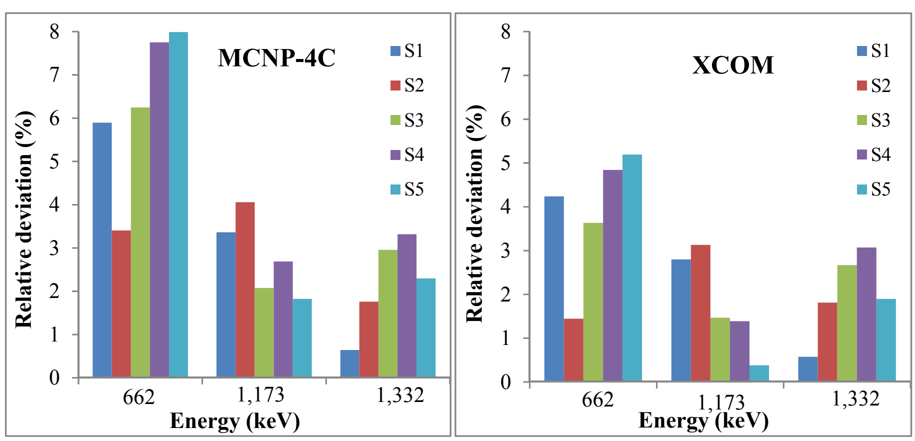

Fig. 7 – Difference (%) between experimental data and MCNP and XCOM results.

absorber [17], was calculated for glass samples using the following equation:

$$MFP = \frac{1}{\mu} \quad (5)$$

These values are shown in Table 5 together with MFP values of ordinary and barite concretes for comparison. Table 5 shows that the minimum value of the MFP is observed in S5 glass sample. It is observed that having 10 mol% of Bi_2O_3 content or higher at glass sample, the value of MFP is even lower than barite concrete as an appropriate high density shielding concrete.

The HVL and TVL of glass samples are shown in Figs. 8 and 9, respectively. HVL and TVL quantities are defined as the thickness of the attenuator that reduces photon density to, respectively, half and 10^{th} of its initial intensity. These figures show that HVL and TVL values of the glass samples decrease with an increase in the mol% of Bi_2O_3 content (or density of glass sample) and increase with incident photon energy. Finally the MFP, HVL, and TVL experimental values are in good accordance with simulation and theoretical results.

3.4. *Effective atomic number and electron density of glass samples*

The total atomic cross sections, σ_a, for glass samples are calculated from the simulation and experimental values of μ_m using the following relation [20].

$$\sigma_a = \frac{\mu_m N}{N_A} \quad (6)$$

where N is the atomic mass of glass samples and N_A is the Avogadro's number. The total atomic cross sections, σ_a, and

Table 5 – Mean free path of glass samples.

Glass samples	Density (g/cm^3)	662 keV			1,173 keV			1,332 keV		
		MCNP	XCOM	Exp.	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.
S1	3.45	3.80	3.86	4.02	5.35	5.38	5.53	5.74	5.75	5.78
S2	3.67	3.34	3.40	3.45	4.91	4.96	5.11	5.32	5.32	5.42
S3	3.81	3.06	3.14	3.25	4.69	4.71	4.78	5.06	5.07	5.21
S4	3.97	2.84	2.92	3.06	4.42	4.48	4.54	4.82	4.83	4.98
S5	4.21	2.61	2.68	2.82	4.13	4.19	4.20	4.51	4.53	4.61
Ordinary concrete	2.46	5.52	5.52	5.35 [18]	7.30	7.25	—	7.81	7.75	6.06 [19]
Barite concrete	3.463	3.88	3.85	3.37 [19]	5.38	5.35	—	5.78	5.71	5.52 [6]

Exp., experiment.

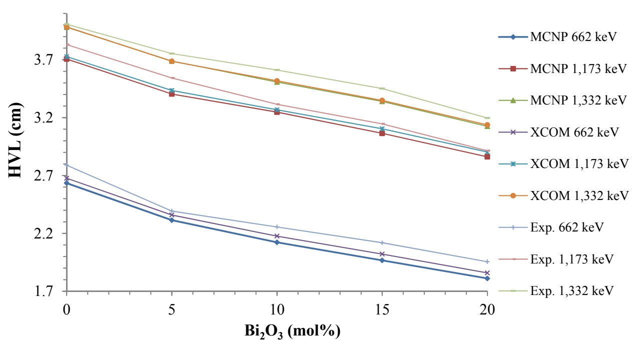

Fig. 8 – HVL values of studied glass samples for selected photon energies. HVL, half value layer.

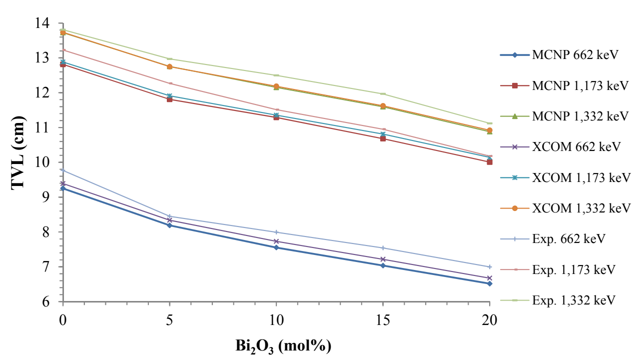

Fig. 9 – TVL values of studied glass samples for selected photon energies. TVL, 10th value layer.

total electronic cross sections, σ_e, for XCOM program are calculated from the following mixture equations [21]:

$$\sigma_a = \frac{1}{N_A}\sum f_i N_i \mu_{m.i} \quad (7)$$

$$\sigma_e = \frac{1}{N_A}\sum \frac{f_i N_i \mu_{m.i}}{Z_i} \quad (8)$$

In these equations, f_i denotes the fractional abundance of the ith element with respect to the number of atoms such as $f_1 + f_2 + f_3 + \ldots + f_i = 1$, and Z_i and N_i are the atomic numbers and atomic mass of the ith element, respectively. Finally, the effective atomic number (Z_{eff}) and effective electron density (N_{eff}) of glass samples are calculated from Eqs. (9) and (10), and are given in Tables 6 and 7, respectively [16]:

$$Z_{eff} = \frac{\sigma_a}{\sigma_e} \quad (9)$$

$$N_{eff} = \frac{\mu_m}{\sigma_e} \quad (10)$$

From the obtained values of Z_{eff} given in Table 6 for all three groups of data, it was found that the values of Z_{eff} increase as the amount of bismuth oxide increases and decreases as photon energy increases. This is due to the ratio of high atomic number elements in the composite material and indicates that the composite materials having high Z_{eff} values will effectively absorb incoming photons. The average differences between the MCNP and XCOM data, and experimental results of Z_{eff} were 3.76% and 11.30%, respectively. MCNP-4C results showed better agreement with experimental data in comparison with XCOM program.

It is evident from Table 7 that the effective electron density (N_e) varies in the range of (2.6–3.2) × 10^{23} electrons/g. It is almost independent of the glass sample composition and decreases slowly as photon energy increases.

As shown in Table 4 and Fig. 6, the linear and mass attenuation coefficients of the barium–bismuth–borosilicate glass samples calculated by MCNP-4C code and XCOM programs were always higher than the experimental results. This rule also applies to other studied parameters, which are calculated using linear and mass attenuation coefficients [for MFP, HVL, and TVL values inversely (Table 5, and Figs. 8 and 9) and for Z_{eff} and N_{eff} values directly (Tables 6 and 7)].

The MCNP code and XCOM program calculate parameters of interest based on theoretical calculations and equations. Although it was tried to design the experiment setup precisely, there were some inaccuracies that were always present and these affected factors were eliminated in simulation and theoretical calculations. Therefore, the calculated values (MCNP and XCOM data) are mostly higher than the experimental results.

It should be noted that some observed differences in results could be due to the MCNP code and the model itself, such as physical and mathematical models, uncertainties in the nuclear/atomic data, improper modeling of source energy and actual geometry and errors in the material compositions, etc., and also from experimental situations such as nuclear electronic setups and related errors, physical condition of environment (pressure, humidity, and temperature), and errors in measurement of

Table 6 – MCNP-4C, XCOM, and experimental values of effective atomic numbers (Z_{eff}) of glass samples.

Glass samples	662 keV			1,173 keV			1,332 keV		
	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.
S1	17.58	18.56	16.60	16.74	17.85	16.19	16.63	17.83	16.53
S2	20.98	22.24	20.29	19.36	20.73	18.60	19.09	20.64	18.76
S3	24.15	25.59	22.73	21.69	23.43	21.25	21.49	23.28	20.87
S4	27.00	28.66	25.05	24.09	25.95	23.46	23.66	25.76	22.90
S5	29.54	31.48	27.34	26.24	28.33	25.77	25.76	28.09	25.18

Exp., experiment.

Table 7 – MCNP-4C, XCOM, and experimental values of electron density ($N_{eff} \times 10^{23}$ electrons/g) of glass samples.

Glass samples	662 keV			1,173 keV			1,332 keV		
	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.	MCNP	XCOM	Exp.
S1	2.88	2.84	2.72	2.74	2.73	2.65	2.73	2.73	2.71
S2	3.01	2.95	2.91	2.78	2.75	2.67	2.74	2.74	2.69
S3	3.10	3.02	2.91	2.78	2.76	2.72	2.76	2.75	2.68
S4	3.14	3.06	2.92	2.80	2.77	2.73	2.75	2.75	2.67
S5	3.16	3.08	2.92	2.81	2.77	2.76	2.76	2.74	2.69

Exp., experiment.

physical quantities such as dimensions, densities, elemental composition of materials, intensity of sources, etc.

These considerations are the limitations of modeling; however, the suggested model estimates the shielding characteristics of barium–bismuth–borosilicate glasses to a high extent. Efforts were made to modify the MCNP code details and geometry setup, and to derive shielding parameters (HVL, TVL, MFP, Z_{eff}, and N_{eff} values) using linear attenuation coefficients.

4. Conclusion

In the present work, gamma ray shielding properties of barium–bismuth–borosilicate glasses were studied with 662 keV, 1,173 keV, and 1,332 keV gamma rays using MCNP-4C code, XCOM program, and available experimental data.

It was found that the results by XCOM, MCNP, and the experiment are in good agreement with each other. In addition, the differences in the used geometry of simulation relative to experimental geometries lead to little discrepancy in calculation and measured values. The simulation results demonstrated that the S5 glass sample of high density (4.21 g/cm^3) and with constituents of relatively high-atomic-number elements relative to other mentioned glass samples is a more effective shield.

Conflicts of interest

The authors declare that no conflict of interest exists.

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