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Comparative Study of Gamma Radiation Shielding Parameters for Different Oxide Glasses

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Abstract:

The goal of this work was investigated materials that can be used as good gamma shielding materials. In this paper a comparative study of the barium bismuth and barium lead borosilicate glass samples as radiation shielding parameters and optical properties was presented. Bismuth oxide glasses are very useful for developing as lead-free, high density and radio shielding glasses. The eleven cylindrical geometries barium bismuth and barium lead borosilicate glass samples have been prepared in different percentages and investigate for radiation shielding properties of gamma rays from, ^{137}Cs and ^{60}Co radioactive point sources. All glass samples have been prepared by melt-quenching method. The mass attenuation coefficients (μ_m) for all glass samples are measured experimentally and also calculated theoretically by WinXCom program and Monte Carlo Model MCNP-5. The mean free path (mfp), effective atomic numbers (Z_{eff}) and the effective electron densities (N_{eff}) have been determined for all glass samples at different photon energies. Theoretical optical basicity (Λ_{th}) of the glasses and the % of heaviness with lead normalized to 100% for all glasses samples shielding materials have been calculated. The obtained results show that the experimental values of the glass samples are found to be in a good agreement with the theoretical values. From the results it has been observed that the values of μ_m, mfp, Z_{eff}, N_{eff}, Λ_{th} and % of heaviness for all PbO and Bi_2O_3 studied glasses samples at

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the same concentration are approximately equal. So, Bi_2O_3 and BaO glass will open new possibility for a lead-free radiation protecting glass with non-toxicity to our environment.

Key words: Oxide glasses, gamma radiation shielding, attenuation coefficients, XCOM Program, MCNP-5 code**.**

1-INTRODUCTION

Radiological protection is the science and practice of protecting people and the environment from the harmful effects of ionizing radiation. Ionizing radiation is widely used in industry and medicine, and can present a significant health hazard. The effectiveness of shielding is dependent on the stopping power of radiation particles, which varies with the type and energy of radiation and the shielding material used. Oxide glasses are the good systems which are used as shielding materials [1].

Today's application of radiation sources and radioactive materials in various fields has made it essential to study different parameters related to shielding against harmful and dangerous radiations [2-6]. Shielding, or the attenuation of gamma radiation, occurs through the interaction of the gamma radiation with matte. The degree to which gamma radiation is attenuated is dependent upon the energy of the incident gamma radiation, the atomic number and density of the elements in the shielding material, and the thickness of the shielding. Composite materials may offer additional benefits in chemical resistance, physical durability and portability [1]. There are several mathematical descriptions of different interaction processes that are dependent on the atomic number, Z [7].

Glass materials are 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 [8]. They are possible alternatives for radiation shielding materials with two advantages brought by their transparency to visible light, and their properties can be modified by using composition and preparation techniques [9].

Lead oxide (PbO) is a promising gamma ray shielding materials due to its strong absorption of gamma rays [10]. Glasses containing heavy metal oxide such as PbO have many advantages due to their high density and high refractive index. These properties are making them important materials for development of advanced optical telecommunication and gamma-ray shielding materials [11-13]. Due to toxicity of lead on human beings as well as on the environment, commercial lead –free glasses based on other heavy elements such as barium and bismuth offer comparable gamma radiation shielding. The samples colors turned from colorless to yellow with increasing $Bi_2 O_3$ contents [14].

Duffy and Ingram have made a great contribution for development of the Lewis acid-base principles by introducing the "optical basicity" concept. The concept was developed for quantitative determination of the acid-basic properties of a large number of oxides, glasses, alloys, slags, molten salts, etc. In the contents of this concept the oxygen behaves as base in the Lewis sense and the metal ions behave as Lewis acids. The ability of oxygen to transfer electron density to surrounding cations depends from the degree of its polarization. Increase in optical basicity results in increasing ability of oxide ions to donate electrons to surrounding cation. This suggests that the present glasses are more basic [15].

Glasses formed with heavy metal oxides (HMO) (atomic weight >100) have received significant attention because of their interesting physical and optical properties. In this perspective, bismuth (atomic weight = 209) oxide containing glasses is one of the most important members of this family. Bismuth contributes to the stabilization of glass structure and improves chemical durability [16].

2. THEORY BACKGROUND

2.1 Gamma Radiation Shielding Parameters

The measured intensity (I) of the transmitted gamma ray through layer of material thickness *(x)* and the initial intensity of gamma ray (I_o) are related to the linear attenuation coefficients (μ) and can be given by the following expression:

$$I = I_0 \, e^{-\mu x} \tag{1}$$

Mass attenuation coefficient (μ_m) of the material is obtained by dividing μ by the material density (ρ). The mass attenuation coefficient, for a compound or mixture is given by [17]:

$$\mu_{m=} \sum_i w_i \, (\mu_m)_i \tag{2}$$

Where w_i and $(\mu_m)_i$ are the weight fraction and mass attenuation coefficient of the i_{th} is constituent element, respectively.

Attenuation of X-rays and gamma-rays in matter is related to density and atomic number. The effective atomic number (Z_{eff}) of compounds and composite materials plays a crucial role in representing the attenuation of X-rays and gamma-rays, particularly for dose calculations in radiation therapy. This parameter has gained composite materials. Z_{eff} can be calculated based on knowledge of the total atomic cross-section (σa) for materials that can be obtained from the measured values of μ_m using the following relation [18, 19]:

$$\sigma_a = \frac{\mu_m}{N_A \sum_i \frac{w_i}{A_i}} \tag{3}$$

Where N_A is Avogadro's number and A_i is the atomic weight of i_{th} element. Similarly, the total electronic cross section is given by [18, 19]:

$$\sigma_{el} = \frac{1}{N_A} \sum f_i \, \frac{A_i}{Z_i} (\mu_m \ )_i \tag{4}$$

Where fi denotes the fractional abundance of the element i with respect to the number of atoms such that $f1 + f2 + f3 + . . . + fi = 1$, Z_i is the atomic number of i_{th} element.
The effective atomic number (Z_{eff}) can be defined as [18, 19]:

$$Z_{eff} = \frac{\sigma_a}{\sigma_{el}} \quad (5)$$

The effective electron number or electron density (N_{eff}) (number of electrons per unit mass) can be given by the formula [19]:

$$N_{eff} = \frac{\mu_m}{\sigma_{el}} \quad (6)$$

The mean free path (MFP), which is defined as the average distance between two successive interactions of photons in an absorber [20], was calculated for glass samples using the following equation:

$$\mathrm{MFP} = \frac{1}{\mu} \quad \mathrm{cm} \quad (7)$$

Where μ is linear attenuation coefficient of the material at a given photon energy.

2.2 Theoretical Optical basicity

The optical basicity addresses the ability of oxide glass in contributing the negative charges in the glass matrix. In other words it defines the electron donating power of the oxygen in the oxides glass. The theoretical optical basicity can be calculated by the equation proposed by Duffy and Ingram [21].

$$\Lambda_{th} = X_1 \Lambda_1 + X_2 \Lambda_2 + \ldots + X_n \Lambda_n \quad (8)$$

where X_1 , X_2 , X_3 , X_4 , , X_n are equivalent fraction based on the amount of oxygen each oxide contributes to the overall glass stoichometry and Λ_1 , Λ_2 , Λ_3 , Λ_4 , , Λ_n are basicities assigned to the individual oxides. The values of optical basicity of each oxide are: Λ (B_2O_3) = 0.425, Λ (Bi_2O_3) = 1.19, Λ (PbO) =1.18, Λ (Na_2O) = 1.55, Λ (BaO) = 1.33, Λ (Al_2O_3) =0.61, Λ (SiO_2) = 0.926, Λ (K_2O) = 1.4.

2.3 Heaviness

To verify the heaviness of the glass samples, lead was assumed as standard and normalized to 100%. With reference to lead, the % of heaviness of the other predictable shielding materials was evaluated using the following relation [22]:

$$\% \text{ of heaviness} = \frac{\text{Density of given material}}{\text{Density of lead}} X100\ \% \quad (9)$$

3. MATERIALS AND METHODS

A- Samples preparation

The eleven barium bismuth and barium lead borosilicate glass samples were prepared in different percentages with 5 mm thickness. The glass samples were prepared by melt quenching technique. Melts of the aforesaid systems with different compositions were obtained in electrically heated furnace (at around 1050-1100^0 C). Dry Oxygen was bubbled through melt using quartz tube to ensure homogeneity. The melt was annealed in another furnace at 270^{0}C in preheated copper mould. Samples were grounded and polished by using different grades of silicon carbide and aluminum paper respectively. Density of these samples was measured by Archimedes' principle using benzene as the immersion liquid. The chemical composition and densities of glass samples are shown in Tables 1 and 2, respectively.

Table 1. The fractional by weight and densities of the barium bismuth and barium lead borosilicate glass samples

Sample code	Density (g/cm^3)	Bi_2O_3	PbO	BaO	Borosilicate glass
Pb-Bi 0%	1.83	0	0	40	60
Pb5%	2.31	0	5	40	55
Pb10%	2.78	0	10	40	50
Pb15%	3.25	0	15	40	45
Pb20%	3.72	0	20	40	40
Bi 5%	2.27	5	0	40	55
Bi10%	2.60	10	0	40	50
Bi15%	2.96	15	0	40	45
Bi20%	3.51	20	0	40	40
Pb10Bi10%	3.68	10	10	40	40
Bi 15 Pb 5%	3.44	15	5	40	40

Table 2. Chemical composition by weight of borosilicate glass

Component	%
B_2O_3	19.2
Na_2O	9.3
Al_2O_3	16.5
SiO_2	47.4
K_2O	7.6

B- Experimental setup

The linear attenuation coefficients of the investigated samples were measured for gamma rays of energies 661.6, 1173 and 1333 keV photon energies which have been obtained from ^{137}Cs

point source and ^{60}Co point source. The experiment has been performed using gamma ray spectrometer which consists of 3'' × 3'' NaI (Tl) Scintillation detector (Oxford model), amplifier and 16 k multi-channel analyzer at Egyptian Nuclear and Radiological Regulatory Authority. For each sample, the gamma ray spectrum was recorded as a function of the thickness of the material. And the area under the photo peak of the spectrum is used to evaluate the intensity (I) of the transmitted beam by using the initial intensity (I_o) which is the area under the photo peak obtained without any sample between detector and source. The experimental setup for mass attenuation coefficient determination is shown in Fig. (1).

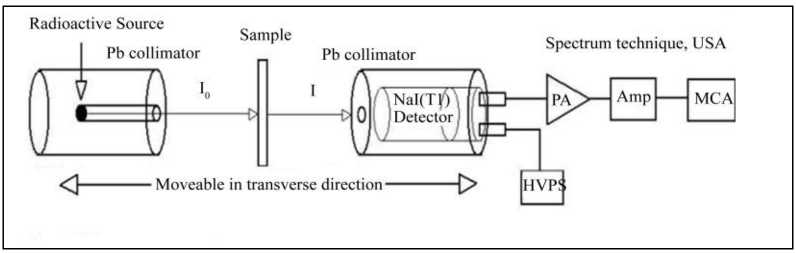

Fig. (1): Experimental setup for mass attenuation coefficient determination

3.2 XCOM program

The μ/ρ values of the samples were calculated by mixture rule

$$(\mu / \rho)_{composite} = \sum w_i (\mu / \rho)_i$$

Where w_i is the proportion by weight and $(\mu / \rho)_I$ is mass attenuation coefficient of the i_{th} element by using XCOM [23].

The uncertainties in μ/ρ values is about 1% for low-Z (1<Z<8) in Compton region (30 keV to 100 MeV). Below 30 keV energy, the uncertainties are as much as 5-10% because of correction to experiments for high-Z impurities and departure of Compton cross section from Klein-Nishina theory. Also above 100 MeV photon energy, uncertainties in μ/ρ values may be 5-10%. Uncertainties in photon energy absorption coefficient may be slightly greater values. The gamma sources of photon energies above 5 keV are being used in medical, biological, industrial, radioactive source transportation and other shielding applications. Hence uncertainty in the result may not have any impact for practical applications [24].

3.3 MCNP-5 Theoretical Model

In this study, Monte Carlo code has been used for investigations on mass attenuation coefficients of the barium bismuth and barium lead borosilicate glass samples. In present simulation, each simulation parameters have been defined in input file by considering the material properties and experimental setup. In this study, the geometrical forms and physical parameters for simulation have been defined in cell card and surface card of MCNP-5 input file.

Cylindrical geometries were employed for the modeling of eleven glass samples with 5 mm thickness. MCNP-5 simulation geometry for calculations mass attenuation coefficients is shown in Fig. (2). A point source with collimator was putting on z axis and defined in an MCNP-5 data card with ERG commands for energy, type of particles respectively. The point detector tally F2 for the model was used to calculate the photon intensity. This tally calculates average gamma ray flux over the surface. The percentages by weight of each element in the glass samples used in the material card of MCNP-5. Simulations were performed with 1 million histories and all results simulated by MCNP-5 code were reported with less than 0.1% error.

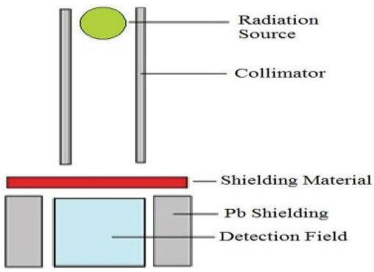

Fig. (2): MCNP-5 simulation geometry

4. RESULTS AND DISCUSSIONS

The mass attenuation coefficients have been calculated and measured at the photon energies 662, 1173 and 1333 KeV. Both calculated and measured results are recorded in Table (3) and displayed in Fig. (3).

Table 3. The experimental mass attenuation coefficient and theoretical by MCNP and XCOM for different concentration of all studies borosilicate glass samples at different energies.

Samples code	Mass attenuation coefficient at different energy MCNP and XCOM μ_m (cm^2 /g) $\times 10^{-2}$								
	662 (KeV)			1173 (KeV)			1333 (KeV)		
	EXP.	MCNP	XCOM	EXP.	MCNP	XCOM	EXP.	MCNP	XCOM
Pb-Bi 0%	7.621	7.650	7.682	5.611	5.621	5.601	5.239	5.251	5.245
Pb5%	7.840	7.852	7.831	5.629	5.634	5.621	5.261	5.259	5.253
Pb10%	8.001	7.902	7.950	5.646	5.655	5.661	5.271	5.269	5.268
Pb15%	8.136	8.151	8.141	5.671	5.665	5.653	5.278	5.281	5.276
Pb20%	8.312	8.321	8.423	5.690	5.685	5.675	5.291	5.289	5.287
Bi 5%	7.851	7.852	7.861	5.652	5.647	5.559	5.228	5.262	5.258
Bi10%	8.011	8.012	8.021	5.649	5.661	5.654	5.281	5.279	5.278
Bi15%	8.162	8.164	8.161	5.681	5.675	5.665	5.290	5.285	5.291
Bi20%	8.330	8.328	8.311	5.701	5.694	5.686	5.291	5.301	5.311
Pb10Bi10%	8.312	8.315	8.332	5.692	5.689	5.677	5.261	5.269	5.272
Bi 15 Pb 5%	8.321	8.320	8.311	5.693	5.689	5.682	5.286	5.291	5.301

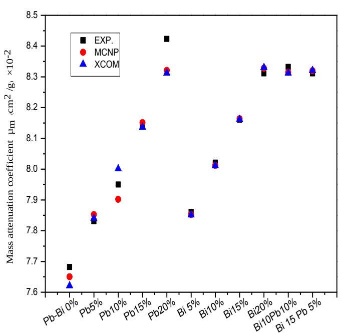

Fig. (3a) : Mass attenuation calculated by MCNP and XCOM with respect to experimet at 662 keV in glass samples

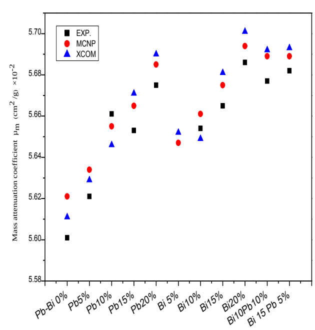

Fig. (3b) : Mass attenuation calculated by MCNP and XCOM with respect to experimet at 1173 keV in glass samples

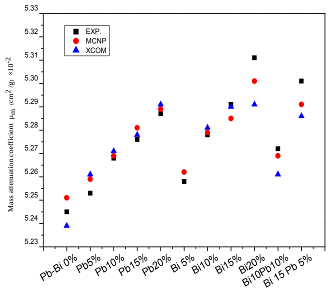

Fig.(3c): Mass attenuation calculated by MCNP and XCOM with respect to experimet at 1333 keV in glass samples

In fig. 3 (a-c), the mass attenuation coefficients decrease with the increase in photon energies. This is because the interaction mechanism of photons with the matter is different for different photon energy [25]. In this work, the total mass attenuation coefficients of the glasses increase with increasing Bi_2O_3 and PbO concentration because of increasing photoelectric absorption interaction of all glass samples. The total mass attenuation coefficients of the Bi_2O_3 glasses and the PbO glasses are comparable.

This indicates that Bi_2O_3 can be used for radiation shielding glasses. The Bi_2O_3 and BaO glass will open new possibility for a lead-free radiation protecting glass with non-toxicity to our environment. The theoretical values of gamma ray attenuation coefficients were generally in good agreement with experimental values.

The obtained results of barium bismuth and barium lead borosilicate glasses have been compared in terms of mean free path (mfp) values with standard radiation shielding ordinary concretes used by Bashter [26] and given in table (4) and plotted in fig. (4). It is notice that the mfp was decreasing with increasing Bi_2O_3 and PbO concentration. This table shows that the minimum value of the mfp is observed in Pb20% and Pb10Bi10% glass samples**.** It is observed that for the glass samples, having 10 mol% of bismuth oxide content or higher, the value of mean free path (mfp) is even lower than ordinary concrete. Therefore, we report here that the lower mean free path (mfp) values of the BaO–Bi_2O_3 glasses are the better radiation shielding materials in comparison to standard shielding concretes.

Table 4. The Mean Free Path for different concentration of all studies Borosilicate glass samples at energies 662, 1173 and 1333 KeV

Sample code	Mean free path (cm)		
	662 KeV	1173 KeV	1333 KeV
Pb-Bi 0%	7.17	9.74	10.43
Pb5%	5.52	7.69	8.23
Pb10%	4.53	6.42	6.87
Pb15%	3.82	5.48	5.88
Pb20%	3.25	4.75	5.11
Bi 5%	5.74	7.97	8.62
Bi10%	4.80	6.81	7.28
Bi15%	4.14	5.95	6.39
Bi20%	3.57	5.22	5.63
Pb10Bi10%	3.39	4.95	5.35
Bi 15 Pb 5%	3.49	5.11	5.50
Ordinary [26]	5.58	7.33	7.82

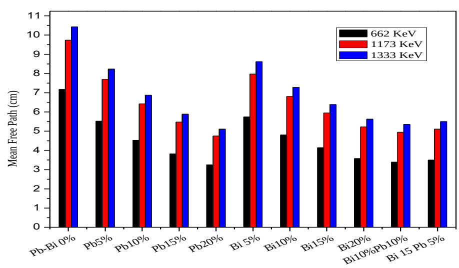

Fig. (4):- The Mean Free Path for different concentration of all studies Borosilicate glass samples at energies 662, 1173 and 1333 KeV.

Using the mass attenuation coefficient (cm^2/g), the effective atomic numbers (Z_{eff}) for glass samples at the photon energy 662 KeV have been calculated and the results have been displayed in Fig. (5) versus mass attenuation coefficients.

The calculated effective electron density (electros/g) and the effective atomic numbers (Z_{eff}) for glass samples at the photon energy 662 KeV is shown in Fig. (6).

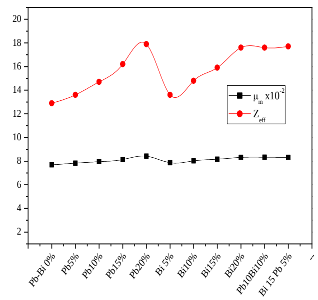

Fig. (5):-The relation between effective atomic numbers Z_{eff} with mass attenuation coefficient (μ_m) x10-2 cm2/g for all glass Samples at 662 KeV

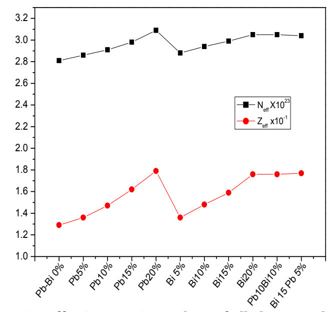

Fig.(6):- Effective atomic numbers of all glass samples versus effective electron density (electrons/g) as obtained based on the Xcom method at 661 KeV

From these figures we can conclude that values of effective atomic number (Z_{eff}) and the mass attenuation coefficient increase as the amount of Bi_2O_3 and PbO increase This is due to the ratio of high atomic number elements in the glass samples composition and indicates that the composite materials having high Z_{eff} values with effectively absorbed incoming photos. From the theoretical approach, the photoelectric absorption is related to more the atomic number than Compton scattering; which depends more on the density of the glass [27]. Moreover, the effective atomic number of lead glasses is relatively equal that of bismuth glass. It is evident from fig. (6) that the effective electron density (N_{eff}) varies in the range of (2.81-3.09) x10^{23} electron/g. The effective atomic number Z_{eff} and electron density N_{eff} are found to increase with PbO and Bi_2O_3 contents. It was found that the Z_{eff} value rather than the N_{eff} value is a better indicator for PbO and/or Bi_2O_3 contents.

The theoretical calculated of optical basicity (Λ_{th}) for all glass samples have been displayed in Fig. (7). The calculated optical basicity (Λ_{th}) values increases with increase in Bi_2O_3 and PbO content. The increased optical basicity of the glasses with large Bi_2O_3 and PbO content indicates that the acid-base properties of Bi_2O_3 and PbO have a significant effect. The low optical basicity is a reduced ability of oxide ions to transfer electrons to the surrounding cations. The barium bismuth and barium lead borosilicate glass samples with a large amount of Bi_2O_3 and PbO content possess high optical basicity. High optical basicity means high electron donor ability of the oxide ions to the cations. The understanding of optical basicity would be useful for the design of the novel optical functional materials with higher optical performances

With reference to lead, the % of heaviness of the other conventional shielding materials along with barium bismuth and barium lead borosilicate glass samples was evaluated and shown in Fig. (8). With lead at 100% heavier than other shielding materials under consideration, steel is at 69.47%, heavier of lead. In case of barium bismuth and barium lead borosilicate glasses composites, all eleven samples were lighter than lead and steel. From this figure it is observed that the glass samples having mole % of bismuth oxide are less heaviness than samples having mole % of lead oxide. These results prove that barium bismuth borosilicate glasses composites considered exhibit excellent lightness when compared to conventional radiation shielding materials such as lead and steel.

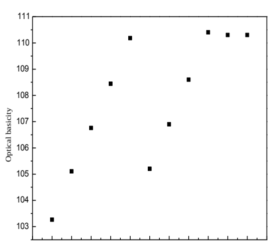

Fig. (7):- Variation of optical basicity with different concentration of lead and bithmuth in glass samples.

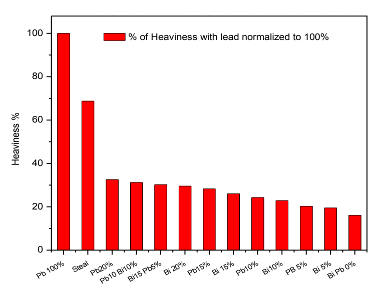

Fig. (8) : Variation of heaviness % with different materiales

5. CONCLUSIONS

In the present work, gamma ray shielding properties of barium bismuth and barium lead borosilicate glasses were studied experimentally and theoretically with Mont Carlo Method and Xcom code. The simulations show that theoretical values of gamma ray attenuation coefficients were generally in good agreement with experimental obtained value.

From this study it can be concluded that:
- Mass attenuation coefficient (μ_m) of the studied glasses sampled increase with increasing Bi_2O_3 and PbO concentration because of increasing photoelectric absorption interaction of all glass samples. The obtained result show that the values of mass attenuation coefficient (μ_m) for Bi_2O3 glasses samples at different concentration are approximately closed to that in PbO glasses samples.
- We have succeeded in developing gamma-ray shielding materials from borosilicate glass. The results show that the minimum value of the mfp is observed in glass samples Pb20% and Pb10%Bi10%. Therefore, we reported here that the lower mean free path (mfp) values of the BaO–Bi_2O_3 glasses are the better radiation shielding materials in comparison to standard shielding concretes.
- The effective atomic number of lead glasses is relatively equal that of bismuth glass.
- By comparing the variation of ratio of % heaviness of all samples with respect to lead and steel materials we conclude that, all samples showed better shielding effectiveness than lead glass. The barium bismuth borosilicate glasses samples having mole % of bismuth oxide are less heaviness than samples having mole % of lead oxide.

The Bi_2O_3 and BaO glass will open new possibility for a lead-free radiation protecting glass with non-toxicity to our environment. The current trends show that the bismuth oxide glasses are very attractive and important optical materials for various scientific as well as technological applications. It can be also concluded that modeled geometry can be used in the fields of radiation such as nuclear medicine and high energy radiation.

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