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ARTICLE

Study on Interaction of Bi_2O_3, PbO and BaO in Silicate Glass System at 662 keV for Development of Gamma-Rays Shielding Materials

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The mass attenuation coefficients and partial interaction of $x\mathrm{R_mO_n}$:(100-x)SiO_2 glass system (where R_mO_n are Bi_2O_3, PbO and BaO, with $30 \leq x \leq 70$ % by weight) have been investigated at 662 keV on the basis of experiments and calculation. The theoretical values of total and partial interaction were obtained by the WinXCom software. The glass system was prepared by the melt-quenching method following the theoretically investigated compositions and its radiation shielding properties were measured. The experimental results showed good agreement with the theoretical ones. Total mass attenuation coefficients of glasses increased with the increases of Bi_2O_3 and PbO component, because of the increase of photoelectric absorption. On the contrary, there was no significant change in mass attenuation coefficient when the fraction of BaO increased. These results indicated that photons were more attenuated in Bi_2O_3 and PbO glasses than the BaO glass. The half value layer (HVL) and effective atomic number results indicate that Bi can replace Pb at this energy as a gamma-ray shielding material. For BaO, HVL was better than the ordinary concrete and commercial windows. This indicates that BaO glasses can be used to shield gamma-ray in replace of both of them in this energy. The Bi_2O_3 and BaO glass will open new possibility for lead-free radiation protecting glasses with non-toxicity to our environment.

KEYWORDS: Mass attenuation coefficients, Effective atomic numbers, Interaction, Glass, Shielding Materials

I. Introduction

The interaction of high-energy photons with matter is important in radiation medicine and biology, nuclear engineering, and space technology. Glass has the double functions of being transparent to visible light and absorbing gamma rays and neutrons, thus providing a radiation shield for observers or experimenters[1)]. Silica (SiO_2) is one of the chief constituents of the earth's crust. The silicate glasses are the most commonly available commercial glasses owning to ease of fabrication and excellent transparency to visible light[2)]. In addition, the very high viscosity allows the glass to be formed, cooled and annealed without crystallizing. This makes the material particularly useful for optical windows in various industries.

Nowadays, bismuth (Bi) and barium (Ba) are playing an important role in radiation shielding glass and replacing lead (Pb) because of the environmental toxicity of Pb. Extensive research in Bi-based and Ba-based glass systems for radiation shielding materials were recently published[2-7)]. These results show that the Bi and Ba can be used in radiation shielding glass. In the present work, we have measured the total mass attenuation coefficients and derived the half value layer (HVL) and the effective atomic numbers on the candidate materials, the glasses containing Bi_2O_3 and BaO for development of radiation shielding glass in silicate glass system at 662 keV, and compared these results with that for the glass containing PbO. The experimental values were compared with theoretically calculated ones using the WinXCom program[8-9)]. Moreover, shielding parameters were also compared with those for the ordinary concrete and commercial windows for the design of non-toxic radiation shielding glass.

II. Theory

In this section, we summarize theoretical relations used in the present work. A parallel beam of mono-energetic gamma-ray is attenuated in matters according to the Lambert-Beer law[10)]:

$$\mathrm{I} = \mathrm{I}_0 \exp(-\mu_m \rho x), \qquad (1)$$

where I_0 and I are the incident and transmitted intensities of gamma radiation, respectively, μ_m is the mass attenuation coefficient of the material, x is the thickness of the absorber (cm) and ρ is the density of the target (g/cm^3).

The mass attenuation coefficient, for a compound or mixture is given by[10)]:

$$\mu_m = \sum_i w_i (\mu_m)_i, \qquad (2)$$

where w_i and $(\mu_m)_i$ are the weight fraction and mass attenuation coefficient of the i^{th} constituent element, respectively. For a chemical compound the fraction by weight (w_i) is given by; $w_i = n_i A_i / \sum_j n_j A_j$ where A_i is the atomic weight of the i^{th} element and n_i is the number of formula units. The values of the mass attenuation coefficients were then used to determine the total molecular

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*Corresponding Author, E-mail: mink110@hotmail.com cross-section ($\sigma_{t,m}$) by the following relation:

$$\sigma_{t,m} = \mu_m \frac{M}{N_A}, \tag{3}$$

where $M = \sum_i n_i A_i$ is the molecular weight of the compound, N_A is the Avogadro's number, n_i is the total number of atoms (with respect to mass number) in the molecule and A_i is the atomic weight of the i^{th} element in a molecule.

The total atomic cross-section ($\sigma_{t,a}$) can be easily determined from the following equation:

$$\sigma_{t,a} = \frac{1}{N_A} \sum_i f_i A_i (\mu_m)_i, \tag{4}$$

Similarly, effective electronic cross-section ($\sigma_{t,el}$) for the individual element is given by[11]:

$$\sigma_{t,el} = \frac{1}{N_A} \sum_i^n \frac{f_i A_i}{Z_i} (\mu_m)_i = \frac{\sigma_{t,a}}{Z_{eff}}, \tag{5}$$

where $f_i = n_i / \sum_j n_j$ and Z_i are the fractional abundance and atomic number of constituent element i, respectively, n_i is the total number of atoms of the constituent element i and $\sum_j n_j$ is the total number of atoms present in the molecular formula.

Now, the effective atomic number (Z_{eff}) can be given as

$$Z_{eff} = \frac{\sigma_{t,a}}{\sigma_{t,el}}. \tag{6}$$

III. Experiments

The glass samples $x\mathrm{R_mO_n}$:(100-x) SiO_2 were prepared by the melt-quenching technique for the composition range of x from 30 to 70 (weight %) where $\mathrm{R_mO_n}$ is the dopant either Bi_2O_3, PbO or BaO. The starting materials (Analytical Reagent Grade 99.9% purity) used in the present work were Bi_2O_3, $BaCO_3$, PbO and SiO_2. All the chemicals were weighed accurately using an electrical balance, ground to fine powder and mixed thoroughly. Each batch of about 50 g in alumina crucible was melt in an electrical furnace for one hour, at 1,250 °C for Bi_2O_3 and PbO glasses and 1,600 °C for BaO glasses. The melts were then poured between the stainless steel molds. The quenched glasses were annealed at 500 °C for 3 hours to reduce thermal stress, and cooled down to the room temperature. At the room temperature, densities (ρ) of all glass samples were measured with the Archimedes's method using xylene as an immersion liquid. The density measurement apparatus is shown in **Fig. 1**.

The diagram of experimental setup for mass attenuation coefficient determination is shown in **Fig. 2**. The source and absorber system were mounted on a composite of adjustable stands. This setup can move in the transverse direction for proper beam alignment. The ^{137}Cs radioactive source of 15 mCi (555 MBq) strength was obtained from the Office of Atom for Peace (OAP), Thailand. The incident and transmitted gamma-rays intensities were measured for a fixed preset time in each experiment by recording the corresponding counts, using the 2″×2″ NaI(Tl) detector having an energy resolution of 8% at 662 keV (BICRON model 2M2/2), with CANBERRA photomultiplier tube base model 802-5. The dead time in this experiment was 0.73%-1.37%. The pulse shaping time was 0.5 μs. An optimum sample thickness ($0.5 \le \mu x \le 5.0$) was selected in this experiment on the basis of the Nordfors criteria[9]. The statistical error in this experiment calculated from the standard error of 3 items (i) ray-sum measurement, which calculated from experiment, the ray-sum is product of linear attenuation coefficient (μ) with thickness (x), (ii) density measurement and (iii) thickness measurement. Finally, the total standard error has been determined by combining errors for the ray-sum measurement, density measurement and thickness measurement in quadrature. The spectra were recorded using a CANBERRA PC-based multi-channel analyzer. In this experiment, the validity of the mass attenuation measurement was confirmed by measuring a lead slab.

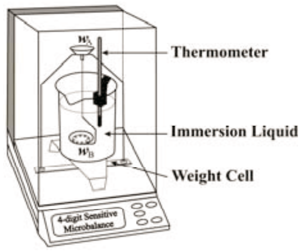

Fig. 1 The density measurement apparatus

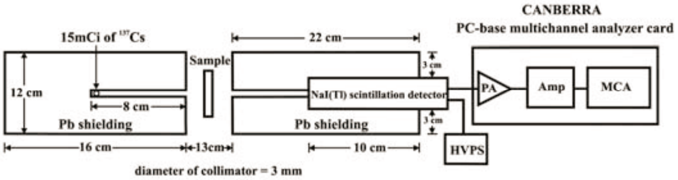

Fig. 2 Experimental setup for mass attenuation coefficient determination

IV. Results and Discussion

The chemical compositions, % by weight, and the density of the glass samples are given in **Table 1**. It is seen that the density of the glass samples increases with higher PbO and Bi_2O_3 contents, because of higher molecular weight of PbO and Bi_2O_3 in comparison to SiO_2. In the case of PbO glass, the density increases by about 60%, and the densities of glasses in ascending order are BaO, PbO and Bi_2O_3. Moreover, the effect of increasing the concentration is small for BaO compared with Bi_2O_3 and PbO glasses.

Table 1 Density of glass samples

% Weight	Density (g/cm^3)		
	BaO-glass	**Bi_2O_3-glass**	**PbO-glass**
30	3.42±0.02	4.89±0.03	3.94±0.01
40	3.45±0.01	5.05±0.05	4.37±0.01
50	3.49±0.02	5.12±0.03	4.73±0.01
60	3.49±0.04	5.48±0.01	5.02±0.01
70	3.50±0.04	5.69±0.01	4.93±0.01

Table 2 Comparison of theoretical and experimental total mass attenuation coefficients of Bi_2O_3, PbO and BaO in silicate glass system [$\times 10^{-2}$ cm^2/g]

% Weight	BaO-glass			Bi_2O_3-glass			PbO-glass		
	μ_m (theory)	μ_m (experiment)	% RD*	μ_m (theory)	μ_m (experiment)	% RD*	μ_m (theory)	μ_m (experiment)	% RD*
30	7.73	7.74±0.25	0.13	8.68	8.03±0.65	7.49	8.64	8.64±0.23	0.00
40	7.73	7.81±0.15	1.03	8.99	8.58±1.13	4.56	8.95	9.47±0.12	5.81
50	7.74	7.59±0.12	1.94	9.31	9.20±0.62	1.18	9.25	9.31±0.11	0.60
60	7.74	8.04±0.10	3.88	9.62	9.68±0.31	0.62	9.56	9.72±0.18	1.67
70	7.74	7.76±0.09	0.26	9.94	9.51±0.38	4.33	9.86	9.86±0.26	0.00

*RD = Relative difference between theory and experiment

Table 2 shows the experimental and theoretical values of total mass attenuation coefficients of glass samples. In general, the experimental values agreed with the theoretical values. In Table 2, there are differences beyond quoted errors between the experimental and theoretical values in some cases. These will be mainly attributed to the non-stoichiometry of glass formula ratio after melting at high temperature. The total mass attenuation coefficients of the Bi_2O_3 glasses and PbO glasses are comparable, and higher than that of the BaO glasses.

Table 3 Mass attenuation coefficient for partial interactions of Bi_2O_3, PbO and BaO in silicate glasses

Photoelectric Interaction ($\times 10^{-2}$ cm^2/g)			
% composition of Bi_2O_3/PbO/BaO	Bi_2O_3-glass	PbO-glass	BaO-glass
30	1.23	1.21	0.32
40	1.63	1.61	0.43
50	2.04	2.01	0.54
60	2.44	2.42	0.64
70	2.85	2.82	0.75
Compton Interaction ($\times 10^{-2}$ cm^2/g)			
% composition of Bi_2O_3/PbO/BaO	Bi_2O_3-glass	PbO-glass	BaO-glass
30	7.25	7.23	7.30
40	7.10	7.07	7.17
50	6.95	6.92	7.04
60	6.80	6.76	6.91
70	6.65	6.60	6.78
Coherent Interaction ($\times 10^{-2}$ cm^2/g)			
% composition of Bi_2O_3/PbO/BaO	Bi_2O_3-glass	PbO-glass	BaO-glass
30	0.20	0.20	0.11
40	0.26	0.26	0.14
50	0.32	0.32	0.16
60	0.38	0.38	0.19
70	0.44	0.44	0.22

The total mass attenuation coefficients of glasses increase with increasing Bi_2O_3 and PbO concentrations. In the case of BaO, however, there is only a little change in the mass attenuation coefficient with increase of concentration.

Calculated mass attenuation coefficients for the photoelectric absorption and the Compton scattering are shown in **Fig. 3** and **Fig. 4**, respectively as a function of fraction of Bi_2O_3, PbO and BaO. The numerical values are summarized in **Table 3**. In Fig. 3, the mass attenuation coefficient for photoelectric absorption increases in all glass samples when the composition of dopant rises.

In Fig. 3, the mass attenuation coefficients for photoelectric interaction of Bi_2O_3 glass are comparable to that of PbO glass and higher than that of BaO glasses. This means that there is more photon absorption in the Bi_2O_3 and PbO glasses than in the BaO glass.

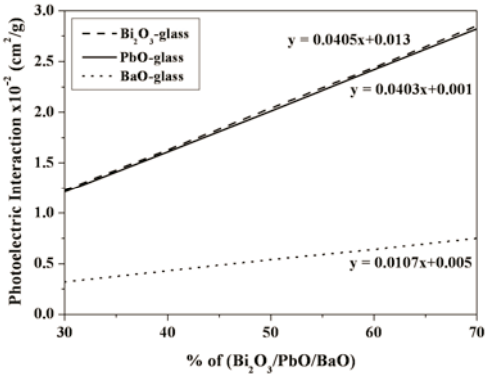

Fig. 3 The mass attenuation coefficients for the photoelectric interaction of the Bi_2O_3, PbO and BaO glasses

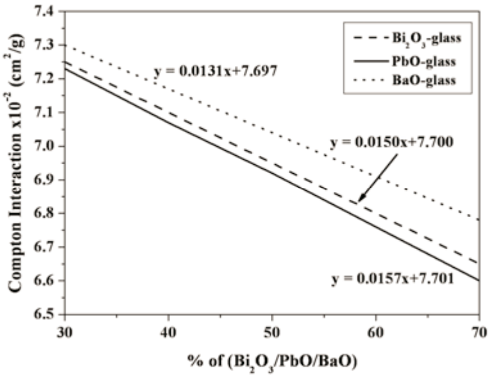

Fig. 4 The mass attenuation coefficient for the Compton scattering interaction of the Bi_2O_3, PbO and BaO glasses

Figure 4 indicates that the Compton scattering interactions of all glasses decrease with increasing dopant concentration. The mass attenuation coefficients for the

Compton interaction of BaO glass are higher than that of Bi_2O_3 and PbO glasses. This indicates that there is more photon scattering in the BaO glasses in this energy compared with the case of other two glasses.

Furthermore, the effective atomic numbers are calculated using Eq.(6) and shown in **Fig. 5**. The effective atomic numbers of PbO glasses are comparable with those of Bi_2O_3 glasses and both are greater than BaO glasses's ones. In addition, the effective atomic numbers increase with increasing Bi_2O_3, PbO and BaO concentration.

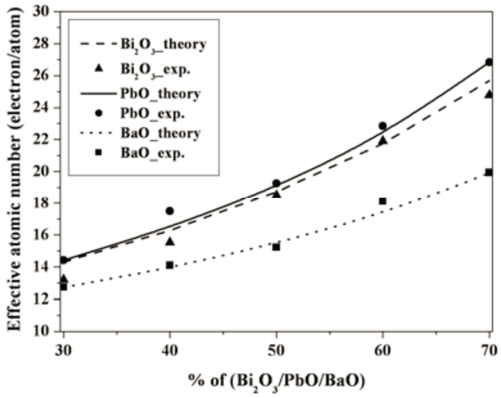

Fig. 5 Comparison of experimental values and theoretical values for the effective atomic number of the Bi_2O_3, PbO and BaO glasses

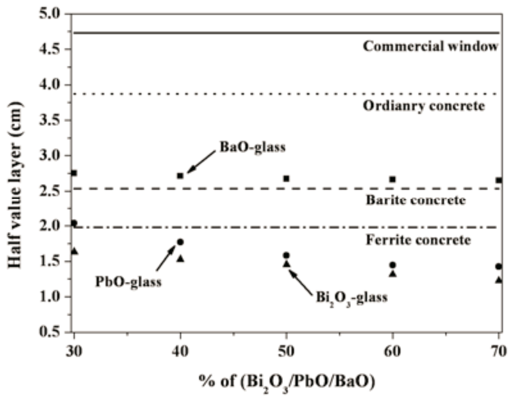

Fig. 6 Comparison of the half value layer of the Bi_2O_3, PbO and BaO glass system by this work with those of shielding concretes and commercial window

The half value layers (HVL) of all glass samples based on the present work are shown in **Fig. 6** with some standard radiation shielding concretes and commercial windows taken from literature[5,11]. It is found that the Bi_2O_3 and PbO glasses have better shielding properties than commercial windows, ferrite and barite concrete, reflecting the advantage of bismuth and lead component in radiation shielding glass. In the case of the BaO glass, the HVL values are better than ordinary concrete and commercial window, but lower than barite and ferrite concrete. This indicates that the BaO glasses can be used to shield gamma-rays in place of ordinary concretes and commercial windows in this energy.

V. Conclusion

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. For the BaO glass, although there was no significant change in the mass attenuation coefficient values with increase of BaO concentration, the glass exhibits a better shielding property like half value layer (HVL) than ordinary concretes and commercial windows. The Bi_2O_3 and BaO glass will open new possibility for a lead-free radiation protecting glass with non-toxicity to our environment.

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