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ARTICLE

Development of BaO:B_2O_3:Flyash Glass System for Gamma-rays shielding Materials

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Glasses with composition xBaO:(80-x)B_2O_3:20Flyash (x = 45, 50, 55, 60, 65 and 70 wt.%) have been prepared using the melt quenching method and the mass attenuation coefficients of the fabricated glass system at 662 keV have been determined using the narrow beam transmission method. The results are consistent with theoretical calculation from the WinXCom software. The mean free path (mfp) result showed that the better shielding properties are achieved at higher BaO concentrations. The density measurements are reported as a function of a BaO fraction. These data should be useful for potential applications of flyash in developing of radiation shielding glasses.

KEYWORDS: mass attenuation coefficient, gamma-ray shielding, borosilicate glasses, rice husk flyash

I. Introduction

Radiations that are most important in nuclear reactor shielding are neutrons, primary gamma-rays originating within the core and the secondary gamma-rays produced by neutron interactions with materials external to the reactor core such as reflector, pressure vessel and shield, etc. Concrete is the most commonly used shield material as it is inexpensive and adaptable for any construction design. There are however several drawbacks associated with the usage of concrete, such as considerable variability in its composition and water content. This variation results in uncertainty in predictions of the radiation distribution and attenuation in the shield. A large water contents leads to decrease of both the density and structural strength of concrete, and the water is lost when concrete becomes hot by absorption of energy from radiations. Some special concretes of higher density than normal have been designed, consisting of elements like barium and iron in addition to light elements. Another drawback of concretes is that they are opaque to visible light and thus it is difficult to see through the concrete-based shields. Therefore, it is an important task to develop better radiation shielding materials in terms of size requirements and transparency to visible light1). Glass materials are one of the possible alternatives to concrete because they can be transparent to visible light and their properties can be modified by composition and preparation techniques. In nuclear industries, borosilicate glass is one of important glasses because it is mainly used as a medium for immobilizing the radioactive ions present in the waste generated in nuclear reactors2-4).

In Thailand, a rice husk is one of the major agricultural wastes. The large proportion of it has been recycled as a raw material for Portland cement and another application e.g. refractory, insulator, waterproofing chemicals etc5). Nevertheless, the usage in these applications has low value in economics. Therefore, it is necessary to search for a new option for the usage of the rice husk ash. When the rice husk ash was burnt, it has a high silica content and low transition oxide contamination. Therefore, the burnt rice husk ash can be a substitution for a silica source in the process to fabricate a borosilicate glass for gamma-rays shielding.

Barium is a good candidate for development of Ba-based radiation shielding glass owing to strong absorption of X-rays, gamma-rays and non-toxicity compared with lead. In literature, many constituents of radiation shielding glasses e.g. lead-silicate, lead borate, bismuth borate, bismuth lead borate glass system1,6-8) have been reported, but a report on barium-based glass is lacking. In the case of the flyash, some literatures have been published for barium-borate-flyash glass system taken from thermal power plants9). However, flyash from power plants has a lot of transition oxide (such as Fe_2O_3) contaminations, which produces opaque property or high intensity color in glass making the glass not suitable in practical usages. Until now, there is no report on the study of glasses production from rice husk flyash in borosilicate glass as shielding materials.

The purposes of the present work are to prepare a xBaO-B_2O_3-Flyash glass system and determine some shielding properties at 662 keV photon energy. We have measured the mass attenuation coefficient, mean free path (mfp) and compared these parameters with theoretical values. The density of the absorber is also determined because they are important for the attenuation of radiations.

II. Experiments

The experimental work has been divided into two parts in

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the following way

1. Preparation of glasses

Six rectangular shaped glass samples of composition xBaO:(80-x)B_2O_3:20Flyash (x = 45, 50, 55, 60, 65 and 70 wt.%) have been prepared by using the melt quenching technique. The oxides of barium and boron used in this work were of analytical reagent grade, and the oxide of silica was of rice husk flyash procured from the Nakorn Pathom Province, Thailand. The rice husks were placed in porcelain crucibles and then calcined at 400, 600, 800, 1000 and 1100 ^{o}C for 5 hours in a muffle electrical furnace. The chemical composition of the rice husk flyash whose composition is not known was analyzed with an energy dispersive x-ray fluorescence (EDXRF) instrument of type Panalytical, Minipal 4 spectrometer (PW 4030/45B) with Rh X-ray tube operation. For preparation of a glass sample an appropriate amounts of BaO, B_2O_3 and rice husk flyash (at highest silica content) were weighed using an electronic balance having accuracy of 0.0001 g. The chemicals were mixed and contained in a crucible to place in an electric furnace at 1,200 ^{o}C for an hour. The melt was poured into a preheated stainless steel mold. The glass sample was then annealed in a separate annealing furnace for 3 hours and then slowly cooled to room temperature.

The thickness of prepared samples was measured by a vernier calliper, which can measure down to 0.05 mm. The density of each sample was measured by the Archimedes' principle using distilled water as the immersion fluid applying the relation

$$\rho = \rho_l[\ W_a / (W_a - W_b)], \qquad (1)$$

where ρ_l is the density of the immersion liquid (density of distilled water is 1.0000 g/cm^3), W_a and W_b are the weight of samples in air and the immersion fluid, respectively. The experiment was repeated three times. The chemical composition (wt.%), average thickness and the density of prepared glass samples are listed in **Table 1**. and the chemical composition of the flyash (calcianed at 1100 ^{o}C) by weight is given in **Table 2**. In this paper we chose rice husk flyashes calcianed at 1100 ^{o}C because it has the highest silica content.

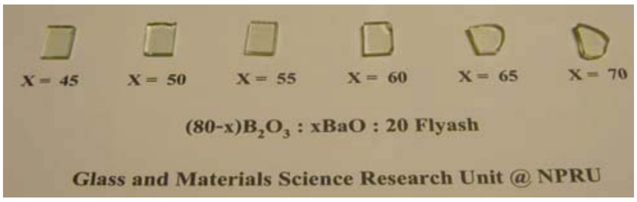

Fig. 1 The glasses sample prepared by cut and polishing

Table 1 Chemical composition (wt.%), thickness and density of barium-borate-flyash glasses

wt.%			t (cm)	ρ (g/cm^3)
(x)BaO	(80- x)B_2O_3	Flyash		
45	35	20	1.6500±0.0043	3.1230±0.0001
50	30	20	1.7325±0.0066	3.1025±0.0013
55	25	20	1.6592±0.0488	3.2471±0.0100
60	20	20	1.6500±0.0436	3.3607±0.0202
65	15	20	1.4900±0.0433	3.4493±0.1190
70	10	20	1.4117±0.0029	3.6655±0.1256

Table 2 Chemical composition by weight of rice husk flyash (calcianed at 1,100 ^{o}C)

Compound	Weight percentage					
	SiO_2	P_2O_5	K_2O	CaO	MnO	Fe_2O_3
Rice husk flyash	87.20	1.64	6.23	3.12	0.82	0.97

2. Measurement of transmitted γ - ray spectra

A narrow beam γ-ray transmission geometry was used for the attenuation measurements of prepared barium-borate-fly ash glass samples. The diagram of the geometry is shown in **Fig. 2.** The source was enclosed in a lead container with one face aperture, 3 mm in diameter. Samples were positioned on a specimen holder at 400 mm from the source. The distance between the source and the detector is 550 mm. A 2”×2” NaI (Tl) crystal detector with the energy resolution 8% at 662 keV and a Multi-Channel Analyzer (MCA) plug-in-card were used with associated electronics to record the pulse-height spectra to a ^{137}Cs radioactive source.

The radioactive sources were procured from Office of Atom for Peace (OAP), Bangkok, Thailand. The intensities of photons were measured without and with placing the sample between the source and the detector. Typical pulse-height distribution of the gamma-ray spectrometer at 662 keV is shown in **Fig. 3**. The intensities of incident and transmitted photons, I_0 and I, respectively, were measured for a fixed preset time by selecting a narrow symmetrical region with respect to the centroid of the photo peak.

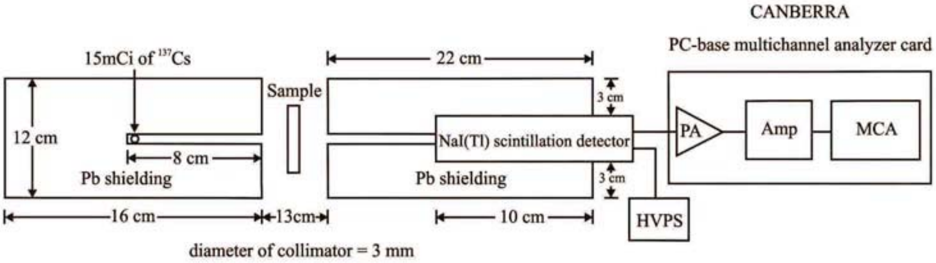

Fig. 2 Experimental setup of narrow beam transmission method

The net area under each peak gives the intensity of gamma-rays. The counting time for each measurement was chosen so that 10^5 counts were recorded under each peak giving a statistical accuracy better than 0.3 %. The statistical error in this experiment was calculated from the relative error, ratio of standard deviation to mean values in three stages (i) gamma-ray measurement, (ii) density measurement and (iii) thickness measurement. Finally, the total error has been determined by combining the errors for three stages in quadrature.

III. Result and Discussions

1. Physical properties

From the experimental values, the density of the barium-borate-flyash glass system with BaO content of 45, 50, 55, 60, 65 and 70 wt% was obtained as shown in Table 1. The replacement of of B_2O_3 and flyash with BaO leads to an increase in density from 3.1025 to 3.6655 g/cm^3 with the increase of BaO content from 45 to 70 wt.%.

2. Gamma –ray shielding properties

The thickness and density of each sample glasses which is necessary in order to derive the mass attenuation coefficient from the measured incident and transmitted gamma-ray intensities is shown in Table 1.

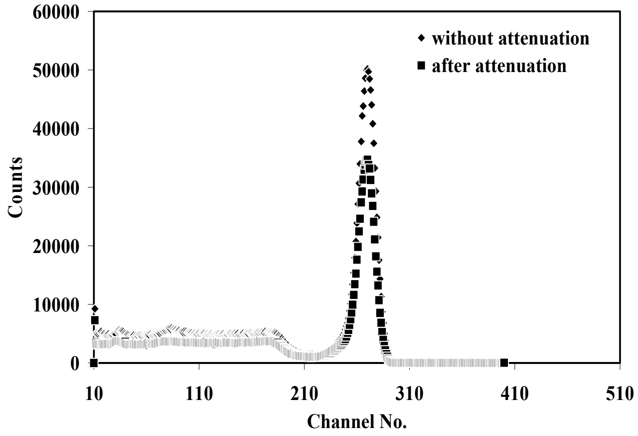

Fig. 3 Typical pulse-height distribution of the γ-ray spectrometer for the cases of without attenuated and with attenuation by glasses samples

Table 3 lists the experimental and theoretical values of the total mass attenuation coefficients of xBaO:(80-x)B_2O_3:20Flyash glass system for $45 \le x \le 70$. In general, the experimental values agree with the theoretical values which are obtained from WinXCom[10)], within relative difference 0.03 – 1.42 %.

The mean free path of 662 keV gamma-rays the prepared glasses are shown in **Table 4**, and compared with those in some conventional radiation shielding concretes. The reference data for conventional shielding concrete were selected from published literature by Bashter[11)].

Table 3 Experimental and theoretical mass attenuation coefficients of barium-borate-flyash glasses

xBaO:(80-x)B_2O_3:20Flyash				
Sample No.	wt. % Of BaO	Theoretical Value	Experimental Value	% RD.[*] of Experimental Value
1	45	0.0767	0.0756±0.0010	1.40
2	50	0.0768	0.0765±0.0010	0.42
3	55	0.0769	0.0771±0.0008	0.29
4	60	0.0770	0.0759±0.0009	1.42
5	65	0.0771	0.0771±0.0009	0.03
6	70	0.0772	0.0770±0.0008	0.25

% RD = relative difference of μ_m between experiment and theory

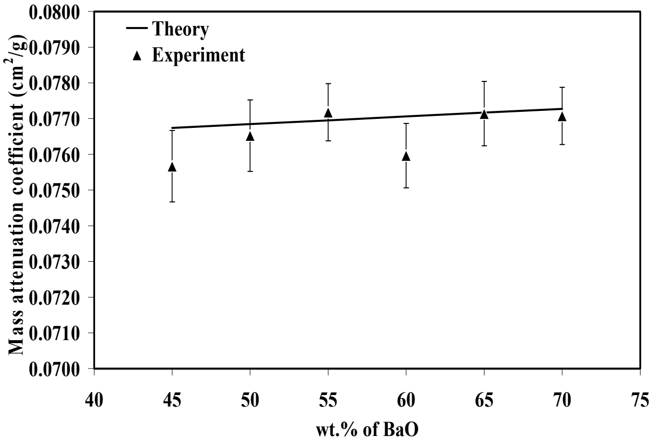

Fig. 4 Theoretical and experimental mass attenuation coefficients of xBaO:(80-x)B_2O_3:20Flyash glass system as a function of x (wt.%)

Figure 5 shows the mean free path of 662 keV gamma-rays for the sample glasses in comparison with conventional shielding concretes. The photon mean free path decreases with increasing, BaO concentration. It is observed that for the glass samples having 45-60 wt.% of BaO content the mean free paths are shorter than ordinary, hermatite-serpentine, ilmentite-limonite and basalt-magnetite concretes. For higher 65 wt%, the mean free path is shorter than all of conventional concretes. Then, we can conclude that the barium-borate-flyash glasses in this work are better shields for gamma-rays in comparison with conventional radiation shielding concretes.

For practical applications in connection with radioactive waste disposal, it would be desirable to measure the mass attenuation coefficient at higher energies but such sources were not available to us. Good agreement between the experiment and the theory in the present work, however, give some confidence that reliable attenuation data can be obtained from existing codes such as WinXCom.

Table 4 Mean free path (mfp) values of 662 keV gamma-rays for barium-borate-flyash glasses in the present study (Left column) and that of some radiation shielding concrete (Right column)[11)]

Sample No.	mfp (cm)	Type of concrete	mfp(cm)
1	4.1802	Ordinary	5.5803
2	4.2024	Hematite-serpentine	5.1948
3	4.0100	Ilmentite-limonite	5.5662
4	3.8744	Basalt-magnetite	4.2753
5	3.7602	Ilmenite	3.7950
6	3.5339	-	-

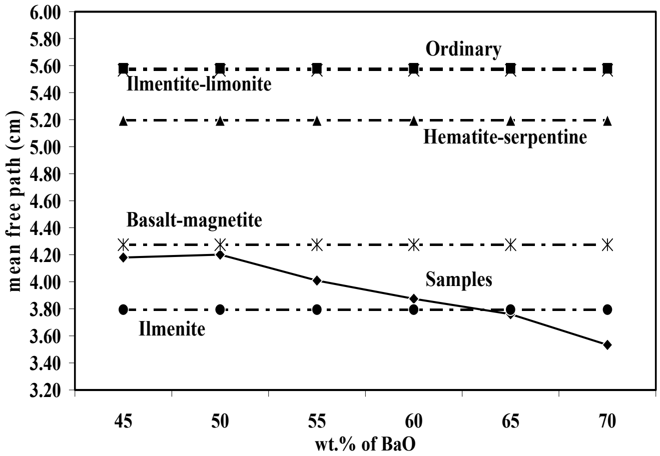

Fig. 5 Variation of photon mean free path (mfp) values for barium-borate-flyash as a function of the BaO concentration at photon energy of 662 keV together with those for some standard radiation shielding concretes.

IV. Conclusion

In conclusion, we prepared xBaO:(80-x)B_2O_3:20Flyash glass system for x=45-70 wt.% and obtained experimental data for the radiation shielding parameters of them at 662 keV photon energy. The mass attenuation coefficient for the given photon energy varies only slowly with composition but increase with increasing fraction of BaO. The glass samples having 45-60 wt.% of BaO content showed the value of mean free path shorter than ordinary hematite-serpentine, ilmentite-limonite and basalt-magnetite concretes, and the values are shorter than all of conventional concretes for higher 65 wt.%. The data should be helpful in potential applications in gamma-ray shielding.

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