Pb-free Radiation Shielding Glass Using Coal Fly Ash

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Abstract. In this work, Pb-free shielding glass samples were prepared by the melt quenching technique using subbituminous fly ash (SFA) composed of xBi_2O_3 : $(60-x)B_2O_3$: $10Na_2O$: 30SFA (where x = 10, 15, 20, 25, 30 and 35 by wt%). The samples were investigated for their physical and radiation shielding properties. The density and hardness were measured. The results showed that the density increased with the increase of Bi_2O_3 content. The highest value of hardness was observed for glass sample with 30 wt% of Bi_2O_3 concentration. The samples were investigated under 662 keV gamma ray and the results were compared with theoretical calculations. The values of the mass attenuation coefficient (μ_m), the atomic cross section (σ_e) and the effective atomic number (Z_{eff}) were found to increase with an increase of the Bi_2O_3 concentration and were in good agreement with the theoretical calculations. The best results for the half-value layer (HVL) were observed in the sample with 35 wt% of Bi_2O_3 concentration, better than the values of barite concrete. These results demonstrate the viability of using coal fly ash waste for radiation shielding glass without PbO in the glass matrices.

Keywords: *effective atomic number; fly ash; half-value layer; radiation shielding; subbituminous.*

1 Introduction

Fly ash is a grey powder consisting of many trace elements that are produced as a by-product in coal-fired thermal power plants. During the combustion process, coal undergoes some physical and chemical changes. This accounts for 5-20 wt% of feed coal and is typically found in the form of bottom ash and fly ash, which account for 15-30 and 70-85 wt% of the total ash created, respectively [1,2]. The environmental impact of coal ashes now cause great concern because they consist of many oxides of heavy metal elements. The majority of ash is currently dumped in landfills or ash ponds. Because the particles size of fly ash

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is very small, it easily pollutes local soil, air and water. This also causes hazards to human health through the food chain.

In Thailand, the annual coal consumption is around 35 million ton. About 81% of the coal is used to generate electricity and 19% is used by the industrial sector for the production of cement, fiber, food, lime, tobacco, metals, etc. [3]. Coal usage in Thailand causes over 3 million tons of solid waste in the form of coal fly ash every year, which is increasing over time. Although coal fly ash is classified as a hazardous waste, it does have some benefits. The chemical composition of fly ash generally comprises large amounts of SiO_2, Al_2O_3 and CaO. In the past, small amounts of fly ash were used as additive in concrete products. Now, new techniques can make use of it in glass and ceramic materials since coal fly ash is rich in oxides of silicon aluminum and calcium, which are the most common components of glassy systems [4].

The environmental toxicity of lead (Pb) has inspired research interest in finding replacements for it in the field of radiation shielding materials. Bismuth (Bi) is the replacement material that has been most extensively explored and now plays an important role in radiation shielding glass [5-9]. All these investigations have affirmed that bismuth is suitable for use in shielding glass. This paper presents the use of glass from subbituminous fly ash (SFA) with addition of Bi_2O_3 as an alternative radiation shielding material. Glass samples were prepared according to the formula $xBi_2O_3 : (60-x)B_2O_3 : 10Na_2O : 30SFA$ (where x = 10, 15, 20, 25, 30 and 35 by wt%). They were investigated on physical and radiation shielding properties at 662 keV. All glass samples were measured and compared under the same preparation conditions.

2 Experimental Setup

The six rectangular glass samples were prepared according to the formula $xBi_2O_3 : (60-x)B_2O_3 : 10Na_2O : 30SFA$ (where x = 10, 15, 20, 25, 30 and 35 by wt%) by using the melt quenching technique. The oxides of bismuth, boron and sodium were analytical reagent grade. The subbituminous fly ash was obtained from a coal-fire thermal power plant in Rayong province, Thailand. The components of the subbituminous fly ash are shown in Table 1. The chemical components of each sample (20 g) were mixed and contained in a high alumina crucible. The mixtures were placed in an electrical furnace and melted at 1,100°C for 3 hours under normal atmospheric conditions. Then the melt was poured into a preheated rectangular stainless steel mold for forming the glass samples and then annealed at 500°C for 3h. After that, the glass samples were slowly cooled down to room temperature.

Table 1 Chemical Components by Weight of Subbituminous Fly Ash.

Compounds	% weight												
	Al_2O_3	SiO_2	P_2O_5	SO_3	K_2O	CaO	TiO_2	MnO	Fe_2O_3	NiO	CuO	ZnO	[a]LOI
Subbitumious fly ash	28.99	49.61	0.77	0.46	1.08	2.24	1.98	0.14	14.61	0.01	0.02	0.04	0.05

[a] Loss on ignition

By applying Archimedes' principle, the weight of each prepared glass samples was measured in air (Wa) and in xylene (Wb) using a sensitive 4-digit microbalance (AND, HR200). Then the density (ρ) was determined with the following formula:

$$\rho = \frac{W_a}{W_a - W_b} \times \rho_b \tag{1}$$

where ρb is the density of xylene (ρb = 0.863 g/cm^3). Vickers hardness measurement was performed with a DHV-1000 Micro Vickers Hardness Tester (Enkay).

The experimental setup used to study the radiation shielding performance is shown in Figure 1. We used a Cs-137 radioactive source (obtained from the Office of Atoms for Peace (OAP), Bangkok, Thailand) was used and placed in a Pb container with 3 mm of collimator. The glass samples were placed at 140 mm from the source. The distance between the glass and the NaI(Tl) detector was 190 nm. An NaI(Tl) scintillation detector with 8% of energy resolution (at 662 keV) was used to detect the gamma-ray intensity [10].

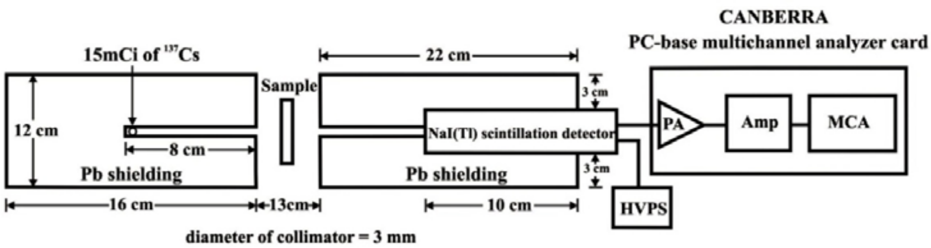


Figure 1 Experimental setup for mass attenuation coefficient determination [11-13].

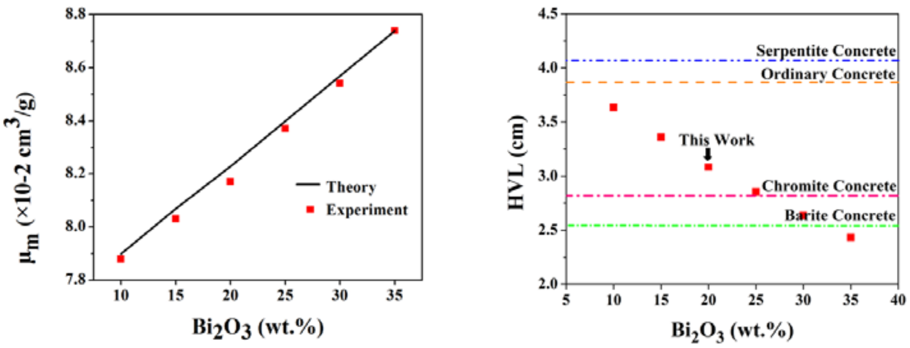
3 Result and Discussion

The density variation of the glass along with the Bi_2O_3 concentrations are shown in Table 2. The densities were seen to increase with the addition of Bi_2O_3 content into the network. They were in the range of 2.4-3.3 g/cm^3. This shows that the replacement of B_2O_3 by Bi_2O_3 caused the average density of the glass samples to increase as a result of the higher molecular weight of Bi_2O_3 in the glass network matrix. The Vickers micro hardness values in the glass products with fly ash are shown in Table 2. Initially, the hardness increased with the Bi_2O_3 content and reached 437.52 MPa at 30 wt% of Bi_2O_3. However, with further increases in the Bi_2O_3 content, the hardness decreased and attained 383.46MPa at 35 wt% of Bi_2O_3. This change may have resulted from the boron anomaly and alkali (Na_2O) content, known widely as a typical property of alkali borosilicate glass [14].

Table 2 Chemical Components by Weight of Glass and Physical Properties.

Components (wt%)				Density	Vickers
Bi_2O_3	B_2O_3	Na_2O	SFA	(g/cm$_3$)	Hardness
10	50	10	30	2.42±0.0004	310.50
15	45	10	30	2.57±0.0009	379.90
20	40	10	30	2.73±0.0004	405.02
25	35	10	30	2.90±0.0014	415.80
30	30	10	30	3.08±0.0043	437.52
35	25	10	30	3.26±0.0016	383.46

The mass attenuation coefficient and related shielding properties were calculated from experimental data [15-18]. The experimental values of the mass attenuation coefficients of the glass samples were evaluated using intensities of incident (I_0) and transmitted (I) gamma ray, while the theoretical values were calculated with the WinXCom program.



(a) (b)

Figure 2 The varying radiation properties of the glass samples: (a) mass attenuation coefficient and (b) half-value layer.

In Figure 2(a), the total mass attenuation coefficients of the glass samples are shown over Bi_2O_3 concentrations at photon energy 662 keV. It was found that when the Bi_2O_3 concentration increased, the total mass attenuation coefficient increased too.

In Table 3, good agreement can be seen between the experimental and the theoretical values of the total atomic cross-section (σ_t), the electronic cross-section (σ_e), and the effective atomic number (Z_{eff}). It was found that the effective atomic number increased with Bi_2O_3 content. The higher values of Z_{eff} represent the higher number of electrons per atom for the interaction with gamma-rays and can reduce gamma-ray intensity after travelling through the glass samples [19]. The value differences between the experimental and the theoretical results suggest systematic errors; these may be due to the non-stoichiometry ratio of the glass formula after melting at high temperature.

Table 3 Chemical Composition by Weight of Glass and Physical Properties.

Bi_2O_3 (wt%)	μ_m ($\times 10^{-2}$ cm^2/g)		σ_t (b/atom)		Z_{eff} (electron/atom)		σ_e (b/electron)	HVL (cm)
	Theory	Exp.	Theory	Exp.	Theory	Exp.		
10	7.90	7.88	2.36	2.40	7.77	7.90	0.303	3.55
15	8.07	8.03	2.55	2.52	8.27	8.18	0.308	3.36
20	8.23	8.17	2.76	2.74	8.82	8.75	0.312	3.09
25	8.40	8.37	3.00	3.06	9.41	9.60	0.318	2.78
30	8.57	8.54	3.28	3.20	10.08	9.83	0.325	2.68
35	8.74	8.74	3.60	3.60	10.80	10.80	0.330	2.42

The half-value layers of the glass samples with Bi_2O_3 content were compared with serpentite concrete, ordinary concrete, chromite concrete and barite concrete at 662 keV [6,13]; the results are shown in Figure 2(b). It was found that at 35wt% of Bi_2O_3, the half-value layer of the glass samples was shorter than that of all the standard shielding concretes, indicating that the glass in the present study may be developed as radiation shielding material.

4 Conclusion

Glass samples were prepared according to the formula Bi_2O_3-B_2O_3-Na_2O-SFA to characterize their physical and radiation shielding properties. The following conclusions can be drawn from the results:

1. The density of the glass samples increased with an increase of the concentration of Bi_2O_3. Meanwhile, no relation was found between the hardness of the glass and the Bi_2O_3 concentration. The highest hardness value was achieved at 30 wt% of Bi_2O_3.
2. The experimental values of the mass attenuation coefficient, atomic cross section and effective atomic number increased with an increase of Bi_2O_3. The results showed good agreement between the experimental and the theoretical values.
3. All glass samples showed better shielding properties than serpentite and ordinary concrete. The glass exhibited better shielding than chromite

concrete from 25 to 30 wt% of Bi_2O_3. The glass sample containing wt% of Bi_2O_3 35 showed the best shielding properties in this experiment, i.e. better than those of barite concrete.

The results show that the glass samples prepared in this work can be utilized as radiation shielding material, recycling a hazardous waste material and without Pb component in glass production.

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