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Gamma-ray mass attenuation coefficient and half value layer factor of some oxide glass shielding materials

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ABSTRACT

The variation in dosimetric parameters such as mass attenuation coefficient, half value layer factor, exposure buildup factor, and the photon mean free path for different oxide glasses for the incident gamma energy range 0.015–15 MeV has been studied using MicroShield code. It has been inferred that the addition of PbO and Bi_2O_3 improves the gamma ray shielding properties. Thus, the effect of chemical composition on these parameters is investigated in the form of six different glass compositions, which are compared with specialty concrete for nuclear radiation shielding. The composition termed 'Glass 6' in this paper has the highest mass attenuation and the smallest half value layer and may have potential applications in radiation shielding. An example dry storage cask utilizing an additional layer of Glass 6 as an intermediate shielding layer, simulated in MicroShield, is capable of reducing the exposure rate at the cask surface by over 20 orders of magnitude compared to the case without a glass layer. Based on this study, Glass 6 shows promise as a gamma-ray shielding material, particularly for dry cask storage.

1. Introduction

Due to the overwhelming concern about release of radionuclides from various sources, as well as the increasing use of gamma ray-emitting isotopes in industry, medicine, and agriculture, it has become necessary to study the shielding properties of new and improved materials. There is always a need to develop new materials that can be used under the potentially harsh conditions of radiation exposure and act as shielding materials for extended periods of time.

The most conventional material used for the purpose of radiation shielding for nuclear reactors and nuclear waste storage is concrete with various aggregates. It is a mixture of light nuclei (primarily hydrogen) and heavy nuclei, giving it the ability to be an effective shield against neutron and gamma radiation. Concrete is relatively inexpensive and easy to cast in many shapes and sizes, in addition to being strong and structurally sturdy. However, prolonged exposure to nuclear radiation results in heating of the concrete, which causes a decrease in density and a possible loss of cooling water and/or gas. Another drawback of concrete is that it is not transparent to visible light, and one cannot see through the concrete to monitor what goes on inside.

Glasses can also act as effective shielding materials as an alternative to concrete, or as an additive into concrete mixes. They are typically transparent to visible light, if used without concrete mixes, and their properties can be modified significantly by changing composition and adopting variations in preparation techniques (Kaundal et al., 2010; Rezaei-Ochbelagh and Azimkhani, 2012). During recent years, there has been increasing interest in the synthesis, structure, and physical properties of heavy metal oxide glasses due to their high refractive index, high infrared transparency, high density, and good shielding of gamma rays (Gupta and Sidhu, 2013; Kaur and Singh, 2013; Singh et al., 2013, 2004; Manohara et al., 2011; Limkitjaroenporn et al., 2011; Kumar and Singh, 2012). In particular, lead oxide and bismuth oxide have been used as additives in several silicate and borate glasses in order to achieve superior physical and shielding properties (Kharita et al., 2012; Chanthima and Kaewkhao, 2013; Bootjomchai et al., 2012).

The present work has been undertaken to evaluate different types of glass systems as gamma-ray shields, which include changes in chemical composition. These glasses will be compared to a specialty concrete composition for radiation shielding applications. Additionally, dry storage casks utilizing thin layers of these glass compositions are simulated computationally to determine changes in exposure rates.

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2. Computational methods and materials

Linear attenuation coefficients are calculated from the well-known exponential attenuation law:

$$I_x = I_0 \exp(-\mu_t x) \tag{1}$$

where I_0 is the initial intensity, I_x is the transmitted intensity, x is the penetration depth, and μ_t is the total linear attenuation coefficient at specific photon energy. The mass attenuation coefficient is defined as:

$$\mu/\rho = (1/\rho t)\ln(I_0/I) \tag{2}$$

where ρ is the mass density and t is the absorber thickness. The mass attenuation coefficient for a mixture of materials is:

$$(\mu/\rho)_{total} = \sum_i w_i(\mu/\rho)_i \tag{3}$$

where the total mass attenuation coefficient $(\mu/\rho)_{total}$ is the sum of the mass attenuation coefficient of the individual components $(\mu/\rho)_i$ multiplied by the weight fraction w_i of component i. Finally, the half value layer (HVL), which is the thickness at which the transmitted intensity is one half the initial intensity, is determined from Eq. (1) and depends only on the linear attenuation coefficient:

$$HVL = \frac{\ln(2)}{\mu} \tag{4}$$

The MicroShield v5.03 software package (GrooveSoftware, 1998) is used as the principle computational tool in this study. MicroShield® uses properties of individual materials to compute theoretical mass attenuation coefficients using Eq. (3), as well as exposure buildup factors based on input material composition and density. The program can handle many different geometries, including a cylindrical spent fuel cask, which is of primary interest. Mass attenuation coefficients and buildup factors are computed for each composite material across a photon energy range of 15 keV to 15 MeV.

Six compositions of glass for radiation shielding composed of various metal oxides are considered in this study. The relevant properties of the metal oxides used to simulate the compositions of glass are provided in Table 1. The composition of each of the six glasses is given in Table 2, along with its resulting mass density. Each of the glass compositions contains at least 25% PbO by weight, as lead absorbs photons very efficiently based on its high atomic number and density. Clearly the density of the glass increases as the content of either PbO, Bi_2O_3, or CdO increases because of the high density of each. Table 3 shows the composition of the specialty concrete termed ‘Concrete 6’, which was investigated in a previous study by the authors (Waly and Bourham, 2015), used as the overpack material in the simulation of the waste storage cask. The cement in the concrete is Portland and the aggregate is 80% SiO_2 and 20% $CaCO_3$. Densities for the materials described in Tables 1–3 are exhibited in Fig. 1.

Table 1
Physical properties of relevant metal oxides.

Glass oxide	Molar mass M (g/mol)	Mass density ρ (g/cm^3)	Molar volume $V = (M/\rho)$ (cm^3/mol)
PbO	223.20	9.53	23.421
Bi_2O_3	465.95	8.90	52.360
CdO	128.41	8.15	15.760
Al_2O_3	101.96	3.95	25.810
SiO_2	60.08	2.65	22.670
B_2O_3	69.62	2.46	28.300

Table 2
Chemical composition (% by weight) and mass density of glasses simulated in this work.

Glass type	Composition						Density (g/cm^3)
	PbO	Al_2O_3	B_2O_3	SiO_2	CdO	Bi_2O_3	
Glass 1	0.25	0.1	0.65	–	–	–	4.3765
Glass 2	0.45	0.1	0.45	–	–	–	5.7905
Glass 3	0.5	0.1	–	0.4	–	–	6.22
Glass 4	0.3	–	0.2	–	0.5	–	7.726
Glass 5	0.3	–	0.2	–	–	0.5	7.801
Glass 6	0.8	0.1	–	0.1	–	–	8.284

Table 3
Chemical composition (% by weight) of ‘Concrete 6’ (Waly and Bourham, 2015).

Concrete 6 Composition					Density (g/cm^3)	
Cement	Water	Aggregate	Additive			
13.98%	7.63%	23.517%	39.195% magnetite (Fe_3O_4) 15.678% lead oxide (PbO)		4.64	
Composition Weight Percent	CaO 8.8074	SiO_2 21.8892	Al_2O_3 0.4194	Fe_2O_3 0.699	MgO 0.4194	SO_3 0.4194
	Na_2O 0.04194	K_2O 0.09786	H_2O 7.63	$CaCO_3$ 4.7034	Fe_3O_4 39.195	PbO 15.678

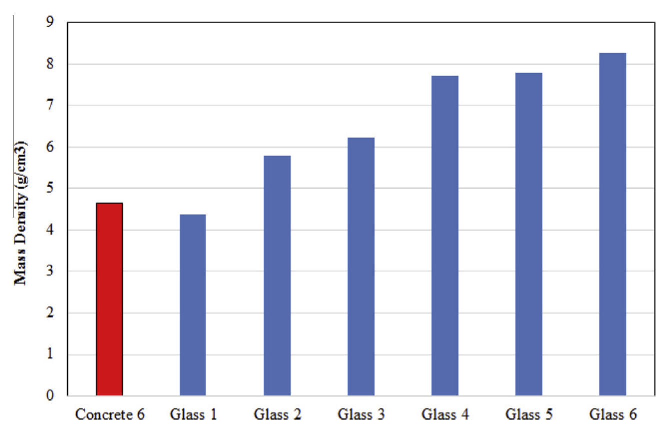

Fig. 1. Mass density of Concrete 6 and the six oxide glasses.

3. Results and discussion

3.1. Mass attenuation coefficient

The mass attenuation coefficient as a function of incident photon energy for the six compositions of glass and ‘Concrete 6’ is given in Fig. 2. The photon energy range may be divided into three regions based on the type of interaction that dominates. In the low energy region, which extends from 15 keV to several hundred keV, attenuation decreases sharply with increasing energy. Photoelectric absorption is the dominant interaction mechanism for low energy gammas, which has a strong dependence on atomic number. A higher effective atomic number of the medium means photons are more likely to be absorbed as there are more electrons with which to interact. The increase in attenuation is attributable to the Pb K-edge. The K shell binding energy of lead is approximately 88 keV (Larkins, 1997), which results in an increase in absorption efficiency for photons of incident energy slightly higher the binding energy.