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Investigation of lead borate glasses doped with aluminium oxide as gamma ray shielding materials

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ABSTRACT

Gamma-ray attenuation coefficients of xPbO·$(0.90-x)B_2O_3$·$0.10Al_2O_3$ (x = 0.25, 0.30, 0.35, 0.40 and 0.45) glass system have been calculated with WinXCOM computer program developed by National Institute of Standards and Technology. Results have been further used to calculate half value layer and mean free path values. Gamma-ray shielding parameters of glass samples have been compared with standard nuclear radiation shielding concretes. The prepared glass samples have higher values of mass attenuation coefficients and lower values of half value layer as compared to concretes for most of gamma ray energy range. The density, molar volume, X-ray diffraction, UV–visible studies, ultrasonic and DSC investigations have been used to study the structural properties of the glass samples. It has been inferred that increase in the composition of PbO leads to the formation of non bridging oxygens which leads to decrease in the rigidity of the glass samples.

1. Introduction

The study of gamma-ray shielding parameters such as mass attenuation coefficient (μ/ρ), half value layer (HVL) and mean free path (MFP) play important role in the research area of radiation physics. Mass attenuation coefficient is the most commonly used parameter to study the interaction of gamma radiations. Concretes are commonly used as shielding materials in nuclear reactors for several types of nuclear radiations including α, β, γ and neutrons (Pisarska, 2009; Kaundal et al., 2010; K.J. Singh et al., 2008; D. Singh et al., 2008; Chanthima and Kaewkhao, 2012).

Concretes as shielding materials in nuclear reactors suffer from several limitations including the following; (1) Addition of moisture content continuously modify the shielding properties of the concretes. (2) They are opaque to visible light and hence, it is not possible to see through concrete based shield. (3) Crack formation occurs after long exposure to nuclear radiations and aging (Singh et al., 2006; Lee et al., 2007). (4) Loss of water occurs in the concrete based shield due to heat generated at concrete. Heat is generated due to interaction of concrete with nuclear radiations. Heavy metal oxide glasses are one of the possible alternatives of concretes for gamma ray shielding purposes. They are transparent to the visible light and their chemical composition can be varied widely to attenuate several types of the nuclear radiations originating in the nuclear reactors. It has been estimated that oxide glasses can be used as potential candidates as alternatives to concretes for gamma ray shielding applications. (Manohara et al., 2009).

In the light of this situation, authors have selected $PbO–B_2O_3–Al_2O_3$ glass system for the purpose of its study for its application as gamma-ray shielding material. $PbO–B_2O_3–Al_2O_3$ glass system is moisture resistant (Medhat, 2009; Singh et al., 2004). PbO form stable glasses due to its dual nature; one as glass modifier (at low PbO additions) and other as glass former (at higher PbO additions). Pb has higher atomic number in the periodic table which implies lead based glasses may have better gamma-ray shielding properties (Kaundal et al., 2010; Ready et al., 2001). Aluminium oxide is added for increasing the mechanical strength (Andriy et al., 1999).

Authors have carried out gamma ray shielding investigations of $PbO–B_2O_3–Al_2O_3$ glasses in terms of mass attenuation coefficient, half value layer and mean free path values. The structural properties are obtained in terms of density, molar volume UV–visible, ultrasonic and DSC measurements for checking the possibility of applicability of the aforesaid system as gamma-ray shielding materials for commercial applications. These glasses are easy to prepare because they have low melting temperatures.

2. Theoretical background

Mass attenuation coefficient has been determined theoretically using the WinXCOM computer software developed by National Institute of Standards and Technology (NIST) (Berger and Hubbel, 1987; Gerward et al., 2004). The mass attenuation coefficient is given by;

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$$\mu/\rho = \sum w_i(\mu/\rho)_i, \quad (1)$$

where w_i is weight fractions and $(\mu/\rho)_i$ mass attenuation coefficients of the several elements.

Half value layer is calculated with the help of linear attenuation coefficient (μ):

$$\text{HVL} = 0.693/\mu, \quad (2)$$

Mean free path has been calculated from the relation described by Tsoulfaniidis (1983).

The molar mass V_g is evaluated from;

$$V_g = M/\rho, \quad (3)$$

Here ρ is the density and the molar mass (M) is given by:

$$M = xM_1 + (0.90 - x)M_2 + 0.10M_3 \quad (4)$$

Here M_1, M_2 and M_3 are the molar masses of PbO, B_2O_3 and Al_2O_3 respectively.

3. Theoretical and experimental details

3.1. Glass preparation

Cylindrical shaped glass samples of composition $xPbO{\cdot}(0.90 - x)B_2O_3{\cdot}0.10Al_2O_3$ in the interval (x = 0.25–0.45) were prepared by using melt quenching technique. For the preparation of glass samples, appropriate amounts of PbO, Al_2O_3 and H_3BO_3 (AR grade) were weighed using an electronic balance. The chemicals were mixed in a pestle mortar for half an hour. Porcelain crucible was placed in an electric furnace for about one hour in temperature range from 850 to 950 °C. Dry oxygen was bubbled through melts using quartz tube in order to obtain homogeneity of the glass melt. The melt was poured into a preheated copper mould. The glass samples were then annealed in a separate annealing furnace. All samples were annealed at 270 °C for 12 h. The prepared glass samples were grinded and polished with different grades of silicon carbides and aluminium paper respectively. Densities of these samples was measured by using Archimedes's principle by using benzene as the immersion liquid (Table 1).

3.2. Gamma-ray shielding properties

For the energies varying from 1 keV to 100 GeV, it is expected that WinXCOM computer software can be used as an authentic tool to evaluate the gamma-ray shielding parameters. It has been verified experimentally that WinXCOM computer program gives the results close to experimental results (K.J. Singh et al., 2008; D. Singh et al., 2008; Singh et al., 2006). In the light of this situation, it is speculated that it is possible to obtain authentic data of mass attenuation coefficients of our glass samples and several concretes by WinXCOM computer software in the wide energy range (1 keV to 100 GeV). HVL and MFP parameters are evaluated from mass attenuation coefficients.

Table 1
Chemical composition (in mole fractions), density and molar volume of $PbO–Al_2O_3–B_2O_3$ glass samples.

Sample no.	Composition (mole fraction)			Density (g/cm^3)	Molar volume (cm^3/mol)
	PbO	Al_2O_3	B_2O_3		
PbBAlG1	0.25	0.10	0.65	3.406	32.66
PbBAlG2	0.30	0.10	0.60	3.807	31.24
PbBAlG3	0.35	0.10	0.55	4.152	30.49
PbBAlG4	0.40	0.10	0.50	4.453	30.15
PbBAlG5	0.45	0.10	0.45	4.711	30.13

3.3. XRD studies

X-ray diffraction studies shows that prepared samples are amorphous. A Philips PW 1710 diffractometer was used with Cu Kα radiation. The values was recorded at angular range (2θ) of 10–70°. Absence of crystallization peak in XRD data shows that prepared samples are amorphous.

3.4. UV–visible investigations

UV–visible absorption spectra were taken on polished disc shaped glass samples, in the wavelength range of 200–1100 nm on a Shimadzu double-beam spectrophotometer. The absorption coefficient as a function of wavelength, $\alpha(\lambda)$, was calculated by dividing the measured absorbance by sample thickness. In order to calculate the band gap (E_g), the following relation was used D. Singh et al., 2008);

$$\hbar\omega\ \alpha(\omega) = \beta[\hbar\omega - E_g]^n \quad (5)$$

where β is constant, $\hbar\omega$ is the photon energy, α is the absorption coefficient. and $n = 2$ for indirect transition. $[\hbar\omega\ \alpha(\omega)]^{1/2}$ was plotted as function of $\hbar\omega$ for each glass sample. From the linear extrapolation to zero ordinate, the value of E_g was calculated.

Urbach energy (ΔE) was calculated using the following relation (Sharma et al., 2006);

$$\ln(\alpha) = C + \hbar\omega/\Delta E \quad (6)$$

where C is a constant, ΔE is obtained from the reciprocal of the slope of the graph of logarithm of absorption coefficient, α versus the photon energy.

3.5. Ultrasonic measurements

Ulrtasonic measurements were carried out in our laboratory by using Matec equipment (Matec SR-9010 Digitizer and SR-9000 synthesizer). The cylindrical samples having opposite parallel faces were used for ultrasonic measurements. The time of flight of two successive echoes has been measured by using Pulse-Echo mode. All the measurements has been done at 5 MHz by using ultrasonic jelly between the sample and transducer contact. Longitudinal modulus (L) and ultrasonic velocity (V_L) are related to each other by the following relation;

$$L = \rho(V_L)^2 \quad (7)$$

3.6. Differential scanning calorimetry (DSC)

DSC measurements of the prepared samples were carried out by using Perkin Elmer differential scanning calorimeter in the temperature range of 200–1200 °C with the heating rate of 20 °C/min in nitrogen atmosphere. 10–20 mg of the powdered glass samples were used to perform the DSC measurements.

4. Results and discussion

4.1. Gamma-ray shielding properties

Mass attenuation coefficient, half value layer and mean free path values of prepared samples as function of energy are shown in Figs. 1–3 respectively. For a better radiation shielding material, higher mass attenuation coefficient and low HVL values are required. Mass attenuation coefficient increases with the increase in the lead composition for our prepared glass samples (Fig. 1). Moreover, there is decrease in HVL values with the increase in the content of Pb (Fig. 2). The prepared glass sample which has