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Shielding Properties of Lead Zinc Borate Glasses

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ABSTRACT

In the present work, $ZnO \cdot PbO \cdot B_2O_3$ glasses are prepared to be used as γ-ray shielding materials. The attenuation properties of these glasses with different concentrations of PbO (20% - 70%) have been investigated at photon energies 662, 1173, 1332 and 2614 keV. The optical absorption spectra of some glass samples have been measured from 200 to 1100 nm before and after γ-ray irradiation. The analyses of these spectra have been interpreted and discussed.

Keywords: Glasses-Ray Shielding; Attenuation Coefficient

1. Introduction

With increasing use of gamma rays in different purposes such as medicine and industry along with the wide spread of nuclear power plants, it has now become necessary to study attenuation properties of various materials and compounds.

Major mass of nuclear radiation shield consists of different concretes with different densities, but water contents in concretes add uncertainty in calculation of shielding properties and moreover they are also opaque to visible light [1]. Glass materials are one of the possible alternatives because they are transparent to visible light and their properties can be adjusted by preparation techniques [1,2]. Borate glasses containing heavy metal oxides are thoroughly studied and the results show that they have potential applications in radiation shielding [3-6].

Many publications compiled the data of the mass attenuation parameter for different materials [7-10]. Berger and Hubbell [11] developed a computer program called the XCOM to evaluate cross-sections and attenuation coefficients for elements, compounds and mixtures for photon energies from 1 keV to 100 GeV. This program was transformed to the Windows platform by Gerwed *et al.* [12,13]. This Windows version is called winXCOM. In a previous work, Singh *et al.* [2] have determined the attenuation properties of $xZnO \cdot 2xPbO \cdot (1 - 3x)B_2O_3$ (x = 0.1 - 0.26) glasses at photon energies 511, 662, 1173 and 1332 keV. It is pointed out that these glasses deserve great interest in radiation shielding applications.

The aim of the present work is to investigate gamma interactions with $ZnO\text{-}PbO\text{-}B_2O_3$ glasses by measuring the mass attenuation coefficient in a broad energy range making use of the 662, 1173, 1332 and 2614 keV gamma lines. The attenuation properties of the hard 2614 keV gamma line are of special importance because the total γ-rays emitted from the interaction of the reactor collimated fast and thermal neutrons beam with the ordinary shielding material such as magnetite concretes lies between 2 - 8 MeV and γ-rays of energy within 2 - 4 MeV are the most predominant. From the experimental results of the attenuation coefficients, the values of the mean free path (MFP) and half value layer (HVL) have been obtained. A comparison between the mass attenuation coefficients at different gamma ray energies with the corresponding predictions of the winXCOM computer program has been performed.

The relation between the increase of the PbO weight fraction and hardness of glass has been tested. Additionally, the present work includes a preliminary experimental analysis of the UV absorption bands in terms of intensity and position.

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2. Theoretical Aspects

It is well known that a parallel beam of monoenergetic gamma-ray or X-ray photons is attenuated in matter according to the Lambert-Beer law:

$$I = I_o e^{-\mu t} \quad (1)$$

where I_o and I are the incident and transmitted intensities, t is the thickness of the absorbing medium and μ is the linear attenuation coefficient.

For photons in an attenuating medium, the mass attenuation coefficient (μ/ρ) is given by

$$\frac{\mu}{\rho} = \ln\left(\frac{I_o}{I}\right)\Big/\rho t \quad (2)$$

where ρ is the density of the absorbing material.

The mean free path (MFP) and the half value layer (HVL) can be evaluated using the following relations:

$$\text{MFP}(\lambda) = 1/\mu \quad (3)$$

$$\text{HVL} = 0.693/\mu \quad (4)$$

3. Experimental Procedure

Glass batches were prepared from chemically pure raw materials. The orthoboric acid (H_3BO_3) was used to obtain boron oxide B_2O_3. The chemicals were thoroughly mixed and porcelain crucibles containing the batch were placed in electrically-heated furnaces and kept at a temperature of 1050˚C for two hours under normal atmospheric conditions. The crucibles were removed from the furnaces and rotate through to produce homogeneous glass.

The glass was cast at a preheated stainless steel molds with dimensions 8 × 6 × 1 cm. All the glasses were properly annealed at 400˚C temperature in a muffle furnace. Then the muffle furnace was left to cool at a rate of 30˚C/hours down to room temperature. Annealing process was done to avoid and minimize the stresses and strains, which may be found in the final glass product.

The density of the prepared glasses was measured by indirect method based on Archimedes' principle using xylene as immersion liquid.

The hardness was measured by a shore durometer (Instron Company United States). The samples were ground and highly polished before absorption measurements. Optical absorption was measured using a Shimadzu spectrophotometer (Japan) covering the wavelength range 200 - 1100 nm.

A ^{60}CO gamma cell was used as a gamma-ray source for irradiation at room temperature with a dose rate of 1.5 Gy/sec. The glass samples were placed in the gamma cell in the manner that each sample was subjected to the same gamma dose. Attenuation measurements were performed using a narrow-beam transmission method.

The experimental arrangement consists of Hyper Pure Germanium (HPGe) detector with relative efficiency ≈10% relative to a 3"× 3" NaL (TI) detector, active volume 62.3 cm^3 and resolution 1.8 keV at 1.33 MeV γ-line. The detector was coupled through an amplifier to the computer with MCA plug-in card. Because of the sensitivity of HPGe detector, it is usual to shield them from the environment. Therefore, lead shield of thickness 5 cm is used in this study. The energy calibration of the spectrometer was performed using the well-known standard sources (^{22}Na, ^{60}CO, ^{57}Co, ^{232}Th and ^{241}Am).

4. Results and Discussion

Chemical composition, densities and hardness for the prepared glass samples are given in **Table 1**. From this table, it is shown that by increasing the lead oxide content instead of boron oxide, the density of the glass batch increases. This may be attributed to the similarity of the position on the glass matrix. *i.e.* lead, which is the most absorbing component in the compound, takes the same positions in the glass network as boron; not in the intersects of the network as modifiers [14]. Moreover, it is obvious from the data that the hardness increases monotonously with decreasing B_2O_3 and replacing it by PbO. This may be due to that the strength of B-O bonds in these glasses is considerably weak compared with Pb-O bonds.

Experimental and theoretical values of mass attenuation coefficients are shown in **Figure 1**, where a good agreement between them is obviously noticed. The theoretical values of mass attenuation coefficients have been calculated using winXcom computer program [11]. Also, **Figure 1** shows that, mass attenuation coefficients increase linearly with the weight fraction of PbO and decrease by increasing gamma-ray energy. In all experimental results, the estimated experimental uncertainties are about ±3%.

Table 2 shows the MFP and the HVL as a function of PbO content, at different γ-ray energies, for the lead borate glasses. As expected, the MFP decreases with increasing PbO content, which is the most absorbing one of the three compounds. Similarly, the HVL decreases with increasing weight fractions of PbO in this glass system. This may be due to the increase in densities of glass samples and the higher values of mass attenuation coefficients. Iron con-

Table 1. Chemical compositions of glass samples.

S. No.	Compositions (wt%)			Density (gm/cm^3)	The hardness (in shore)
	B_2O_3	ZnO	PbO		
1	70	10	20	3.675	94
2	50	10	40	5.088	95
3	25	25	50	5.917	96
4	30	10	60	6.212	96
5	25	5	70	6.650	98

Table 2. Mean free path (MFP) and half value layer (HVL) for the glass samples at different gamma ray energies.

Lead concentration %	662 keV			1173 keV			1332 keV			2614 keV		
	μ	MFP (cm)	HVL (cm)	μ	MFP (cm)	HVL (cm)	μ	MFP (cm)	HVL (cm)	μ	MFP (cm)	HVL (cm)
20	0.3072	3.2552	2.2559	0.2161	4.6275	3.2069	0.2049	4.8804	3.3821	0.1497	6.6764	4.6267
40	0.4536	2.2046	1.5278	0.3082	3.2447	2.2485	0.2845	3.5149	2.4356	0.2143	4.666	3.2335
50	0.5399	1.8522	1.2836	0.3616	2.7655	1.9165	0.3283	3.046	2.1109	0.2612	3.8277	2.6526
60	0.5918	1.6898	1.171	0.3850	2.5974	1.80	0.3468	2.8835	1.9983	0.2627	3.8065	2.6379
70	0.6582	1.5193	1.0529	0.4274	2.3397	1.6214	0.3787	2.6406	1.8299	0.2851	3.507	2.4303

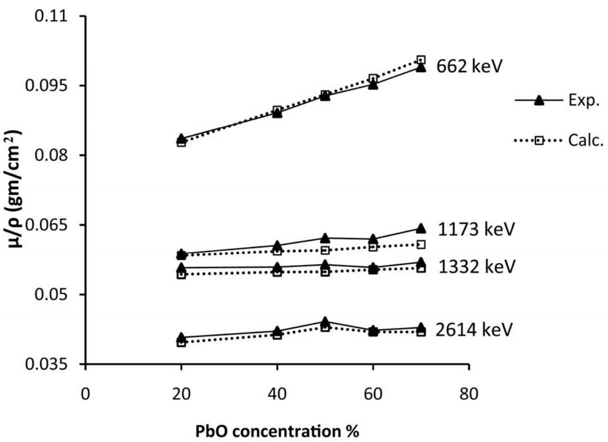

Figure 1. Mass attenuation coefficient for different lead concentration of the glass batches.

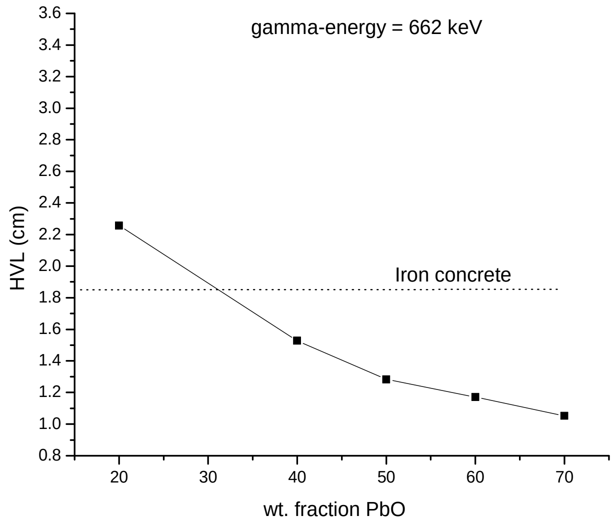

Figure 2. Dependence of half value layer (HVL) on the weight fraction of PbO in B_2O_3-ZnO-PbO glass system. Value of HVL for iorn concrete at 662 keV [17] is included for comparison.

crete's HVL values have been included in **Figure 2** for comparison. The results indicate that lead glasses have higher values of mass attenuation coefficients and smaller values of HVL than standard shielding concretes.

Figure 3 shows the ultraviolet (UV)-visible absorption spectra of the undoped base triborate glass before and after different irradiation doses. The absorption spectra of the unirradiated base reveal an UV absorption peaks at about 235 and 280 nm with no visible bands. Irradiation of this glass to a dose 2 MR of gamma rays causes an increase in the UV bands intensity and the first band splits to two peaks. The visible region shows a broad band at about 540 nm. By increasing the gamma irradiation dose the UV bands intensities grow where the first band increase is larger than that of the second UV band. It is also noticed that the intensity of the visible band grow as a direct consequence to the irradiation dose. The present absorption spectra of the undoped base triborate glass is similar to that obtained previously by El-Batal *et al.* [15]. In their work, the UV-visible absorption spectrum belongs to the undoped base lithium diborate. Therefore, this similarity means that Li contribution to the fore mentioned absorption spectra is negligible. As a matter of fact, "Li" is the smallest metal element that characterized by strong co-

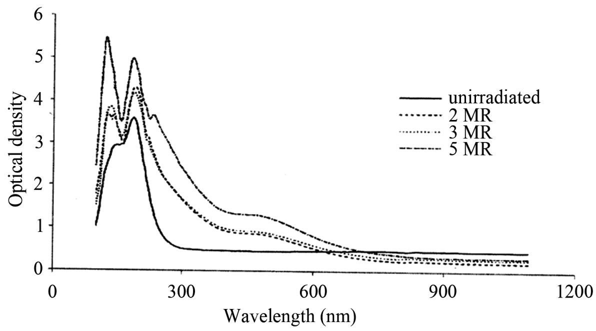

Figure 3. UV-visible absorption spectra of the glass sample no. (1) before and after successive gamma irradiation.

valent bonds and has an infinitesimally small ionic behavior. Also, preliminary investigation of the different doped glass samples specified in **Table 1** via UV-visible absorption spectra before and after different gamma ray irradiation doses indicates that there is an apparent increase in the intensities of both UV and visible absorption bands as the irradiation dose increases. Moreover, it is

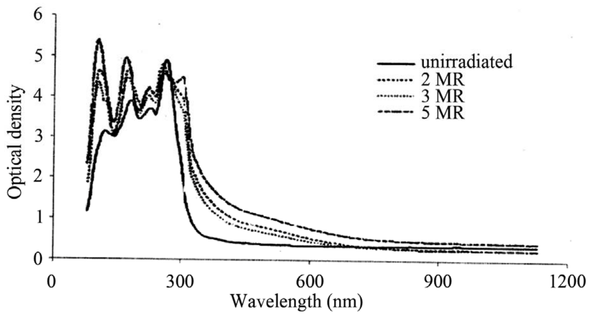

Figure 4. UV-visible absorption spectra of the glass sample no. (5) before and after successive gamma irradiation.

observed that, addition of lead oxide shifts the UV cut-off towards higher values of the wavelength.

In a previous work [16], there is an experimental evidence that lead phosphate possesses an important property concerning its potential resistance against gamma irradiation. Therefore, the formation of color centers as a direct result of gamma irradiation necessitates further study based on the addition of selected materials to avoid formation of those centers and keep the transparency property of the ZnO-PbO-B_2O_3 glass to a suitable extent.

In **Figure 4** the unirradiated glass (sample no 5) reveals strong UV absorption spectrum. This spectrum consists of four consecutive absorption bands at about 210, 275, 310 and 350 nm. With progressive gamma irradiation the intensities of the UV peaks grow and the increase is faster in the first and second peaks and a further new UV peak is resolved at about 380 nm. Also, it is apparent in the figure that there are two visible bands and in general the growth of the bands becomes slower reaching almost saturation.

5. Conclusions

In the present work, the ZnO·PbO·B_2O_3 glass has been observed to be promising gamma-ray shielding material capable to minimize the nuclear radiation hazards below the permissible dose. The dependence of the radiation shielding properties of the related glass on the percentage concentrations of PbO, B_2O_3 and ZnO combinations has been experimentally undertaken. Both linear and mass attenuation coefficients increase with increasing the lead concentration in glass systems for photon energies 662, 1173, 1332 and 2614 keV. The high values of mass attenuation coefficients and low values of half value layer (HVL) of glasses containing lead in comparison to the standard shielding materials indicate that the volume required for shield design will be less than them and the investigated glasses possess the shielding advantages for different nuclear technology applications. It has been also observed that higher percentage PbO content in glass system improves its hardness.

The ultraviolet-visible absorption spectra before and after successive gamma irradiation indicate that possible rate of induced color centers formation is fast at first and then becomes slower or reaches saturation. Therefore, a future work has to be undertaken to set up the material such as P_2O_3 that can be added to help in avoiding the formation of these color centers and keep the transparent property of glass for a suitable long time.

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