Comparative Study of Lead Borate and Lead Silicate Glass Systems Doped with Aluminum Oxide as Gamma-Ray Shielding Materials

Sandeep Kaur, K.J. Singh

Abstract*—*Gamma ray shielding properties of PbO-Al_2O_3-$B_2O_3$and PbO-Al_2O_3-SiO_2 glass systems have been evaluated in terms of mass attenuation coefficient, half value layer, mean free path and effective atomic number parameters. Structural information of both the glass systems has been obtained by using density, XRD, DSC and ultrasonic measurements. It has been inferred that addition of PbO improve the gamma ray shielding properties and simultaneously decrease the rigidity of the glass systems due to formation of non bridging oxygen. Gamma ray shielding properties of our glass systems have been compared with standard nuclear radiation shielding concretes.

***Index Terms*—Attenuation coefficients, DSC studies, Glasses,Ultrasonic measurements.**

I. INTRODUCTION

The most conventional material used for the purpose of radiation shielding for the nuclear reactors is concrete. It is a mixture of hydrogen and other light nuclei and nuclei of fairly high atomic number. One of the major drawbacks associated with the usage of concrete as radiation shielding material lies in the variability in its composition and water content. This variation causes uncertainty in prediction of the radiation distribution and attenuation in the shield. Density and the structural strength of concrete as the radiation shielding material is significantly decreased in case of usage of high water content in concrete. Also continuous exposure to the nuclear radiations results in the absorption of energy from radiations which makes concrete hot causing loss of water. Another drawback of concretes is that one cannot see through the concrete based designs because concretes are not transparent to the visible light.

Glasses can be transparent to the visible light and the fact that their properties can be modified to a large extent by changing the composition and adopting variations in the preparation techniques, these materials can be one of the best possible alternatives to concrete and can be used as gamma ray shielding materials [1], [2], [3].

Glasses containing heavy metal oxide such as PbO have many advantages due to their high density and high refractive index. Thesefeatures makethem importantmaterials for development of advanced optical telecommunication and processing devices and gamma-ray shielding materials [4], [5]. On the other hand, aluminum is a good modifier. Aluminum can increase the glass formation ability, mechanical strength and chemical durability of glasses [6], [7]. Silicate is a very well established network former.

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In the light of this situation, authors have made comparison of two series to explore the possibility of using PbO-B_2O_3-Al_2O_3andPbO-SiO_2-Al_2O_3 glass systems as alternatives to concretes for gamma-ray shielding applications. At low concentration of PbO which varies from 25 to 45 mole %, it behaves as network modifier and at higher content of PbO which varies from 50 to 80 mole %, it acts as glass former. Pb is the most important shielding material which has better gamma ray properties because it has higher atomic number.Moreover, PbO containing glasses have good mechanical, thermal and electrical properties [8]. Authors have characterized PbO-SiO_2-Al_2O_3 and PbO-B_2O_3-Al_2O_3 glass systems for gamma ray shielding properties in terms of mass attenuation coefficients, half value layer (HVL), mean free path (MFP) and effective atomic number (Zeff) values. The density, molar volume, X-ray diffraction, DSC investigations and ultrasonic measurements have been performed to extract the structural information for both glass systems. DSC studieshas been used to evaluate the glass transition temperature of the glass systems. The propagation of ultrasonic waves in solids such as glasses gives useful information regarding the solid state motion in the material. Longitudinal moduli(L) can be obtained from ultrasonic velocity and density values.

II. THEORETICAL BACKGROUND

Mass attenuation coefficient has been computed at photon energy 662keV using the WinXCOM computer software developed by National Institute of Standards and Technology (NIST) [9], [10]. It has been verified already that the aforesaid software can provide authentic values at 662keV gamma energy for glass systems. The mass attenuation coefficient(μ/ρ) is given by the relation;

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

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

HVL can be calculated by using the relation;

$$HVL=0.693/\mu, \qquad (2)$$

Here, μ is linear attenuation coefficient.

Zeff can be computed from the formula;

$$Z_{\text{eff}} = \frac{\sum_i f_i A_i (\mu/\rho)_i}{\sum_i f_i \frac{A_i}{Z_i} (\mu/\rho)_i} \qquad (3)$$

Wheref_i is the molar fraction of the ith constituent element (normalized so that $\sum_i f_i = 1$); A_i is the atomic mass, Z_i is the atomic number, and $(\mu/\rho)_i$ is the mass attenuation coefficient of i^{th} element.

MFP is calculated as [11];

MFP= $\int_0^\infty x\, e^{-\mu x} d$ / $\int_0^\infty e^{-\mu x}\, dx$ =1/μ,(4)

The molar volume V_g is evaluated from;

V_g=M/ρ, (5)

here ρ is the density and M is the molar mass.

III. EXPERIMENTAL DETAILS

A. Glass sample preparation

Cylindrical shaped glass samples of the systems xPbO,(0.90-x)B_2O_3.0.10Al_2O_3in the interval (x = 0.25 up to 0.45) and xPbO.(0.90-x)SiO_2.0.10Al_2O_3(x varies from 0.50 up to 0.80) were prepared by using melt and quenching technique. For the preparation of glass samples, appropriate amounts of PbO, B_2O_3, SiO_2 and Al_2O_3 (AR grade) were weighed using an electronic balance with an accuracy of 0.001g. 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 900^0 up to 1000^0C. Dry oxygen was bubbled through melts using quartz tube in order to ensure homogeneity of the glass melt. The melt was poured into a pre-heated copper mould. The glass samples were then annealed in a separate annealing furnace. All samples were annealed around 300°C and slowly cooled to room temperature. Annealing temperature has been kept same for all samples so that have similar thermal history for all the samples. The prepared glass samples were grounded and polished with different grades of silicon carbides and aluminum paper. Densities of all samples was measured by using Archimedes's principle with benzene as the immersion liquid.

B. XRD studies

X-ray diffraction studies shows that prepared samples are amorphous. A Philips PW 1710 diffractrometer was used and radiation used was CuKα. The values was recorded at angular range (2θ) of 10-70^0. Absence of crystallization peak in XRD data was observed.

C. Ultrasonic measurements

A Matec set-up (imported from USA) was used for the measurements of ultrasonic velocity. The setup utilizes Matec SR-9010 Digitizer and Matec SR-9000 synthesizer. The time of flight measurements was carried out by pulsing and receiving ultrasonic waves through cylindrical glass samples. Pulse-echo mode has been used to measure the time of flight between two echoes at frequency of 5MHz. Ultrasonic jelly was used for making an contact between samples and transducer.

Longitudinal modulus (L) is given by;

L = ρV_L^2 (6)

Where ρ is the density of sample and V_L is longitudinal ultrasonic velocity.

D. DSC measurements

Perkin Elmer differential scanning calorimeter was employed with the heating rate of 20^0C/min in nitrogen atmosphere. Sample amounts of 10-20 mg were used to obtain DSC measurements.

IV. RESULTS AND DISCUSSIONS

The studied chemical composition, density and molar volume of PbO-B_2O_3-Al_2O_3 and PbO-SiO_2-Al_2O_3 glass systems are given in Table 1.

Table 1: Chemical compositions (in mole fractions), Densities and Molar volumes and Glass Transition Temperature of PbO-B_2O_3-$Al_2O_3$and PbO-SiO_2-Al_2O_3glasssystems.

Sample No.	Composition (Mole Fraction)				Density (g/cm^3)	Molar Volume (cm^3/mol)
	PbO	B_2O_3	SiO_2	Al_2O_3		
PbBAlG1	0.25	0.65	-	0.10	3.406	32.66
PbBAlG2	0.30	0.60	-	0.10	3.807	31.24
PbBAlG3	0.35	0.55	-	0.10	4.152	30.49
PbBAlG4	0.40	0.50	-	0.10	4.453	30.15
PbBAlG5	0.45	0.45	-	0.10	4.711	30.13
PbSAlG1	0.50	-	0.40	0.10	5.023	29.03
PbSAlG2	0.55	-	0.35	0.10	5.351	29.78
PbSAlG3	0.60	-	0.30	0.10	5.429	29.92
PbSAlG4	0.65	-	0.25	0.10	5.462	31.17
PbSAlG5	0.70	-	0.20	0.10	5.502	32.43
PbSAlG6	0.75	-	0.15	0.10	5.567	33.52
PbSAlG7	0.80	-	0.10	0.10	5.786	33.66

Thedensity values increase of both glass systems with increase in the mole concentration of PbO which may be contributed to higher atomic weight of Pb. The molar volume is calculated with the help of density values and are shown in table 1. The(μ/ρ) and HVL at photon energy 662keV of both the glass systems along with barite and ferrite concretes are shown in Figs 1 and 2.

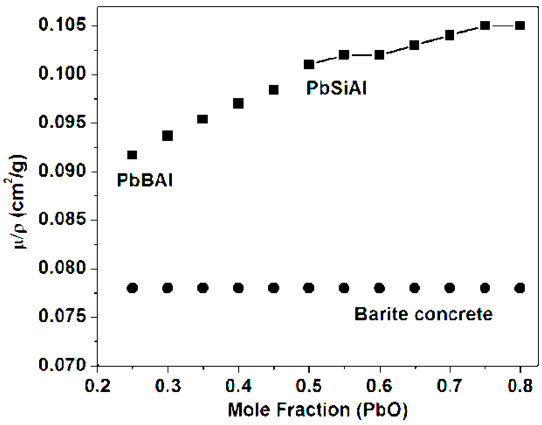

Figure 1.Variation of (μ/ρ) versus mole fraction of PbOat photon energy 662keV in thePbO-B_2O_3-Al_2O_3 (■)and PbO-SiO_2-Al_2O_3(■) glass systems.Theoreticalvalues at same energy for Bariteconcrete(●)isincluded forcomparison.

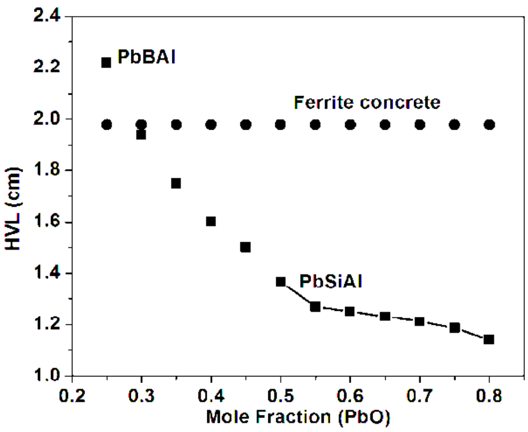

Figure 2.Variation of HVL versus mole fraction of PbOat photon energy 662keV in the PbO-B_2O_3-Al_2O_3 (■)and PbO-SiO_2-Al_2O_3 (■) glass systems. Theoreticalvalues atsame energy for Ferrite concrete(●) isincluded for comparison.

The data of both concretes have been already published [12-13]. Barite concrete is best in terms of mass attenuation coefficient and ferrite concrete is best in terms of HVL values among concretes. In the light of this situation, authors have chosen barite and ferrite concretes for comparison for mass attenuation coefficient and HVL values respectively. Both glass systems have higher values of mass attenuation coefficients and lower values of HVL than existing concretes for most of the composition range. Lower HVL values indicate better gamma radiation shielding materials due to lesser volume requirements of the material during the design of nuclear reactors. It has been seen as we increase the PbO contents in glass samples, gamma-ray parameters improves.Fig. 3 shows the mean free path versus mole fraction of PbO. Mean free path decreases with the increase in PbO concentration which denotes that both systems have better gamma-ray shielding properties with addition of PbO[14].

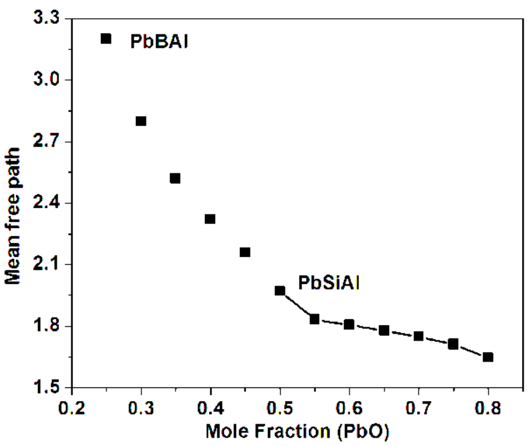

Figure 3.Variation of Zeff versus mole fraction of PbO in the PbO-B_2O_3-$Al_2O_3$and PbO-SiO_2-Al_2O_3glass systems.

Zeffversus mole fraction PbO has been evaluated for all glass samples of both systems and it has been shown in Fig. 4. It has been found that effective atomic number increases with the increase in mole fraction of PbO [15].

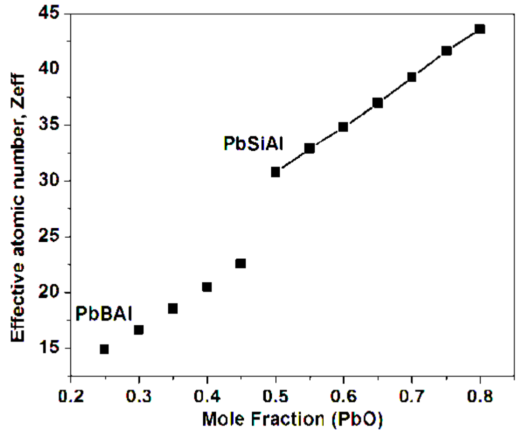

Figure 4.Variation of Zeff versus mole fraction of PbO in the PbO-B_2O_3-$Al_2O_3$and PbO-SiO_2-Al_2O_3glass systems.

It can be concluded from the results that our glass system represents comparable or better values of mass attenuation coefficient and HVL parameters to conventional concretes at energy 662keV. In the light of this situation, it is estimated that 0.80PbO. O.10SiO_2.0.10Al_2O_3 represents the best glass sample in terms of mass attenuation coefficient, MFP, HVL and Zeff parameters for gamma-ray shielding applications. Moreover, it can be interpreted from the analysis of the data of Figs 1-4 that PbO-SiO_2-Al_2O_3 glass system is better than PbO-B_2O_3-Al_2O_3 glass system for gamma ray shielding applications.

X-ray diffraction studies of both glass systems denotes the amorphous nature and shows broad halo around $2\theta = 30^0$ which indicates the absence of long range order.The glass transition temperature values is shown in Fig.5. The variation of glass transition temperature Tg decreases with the addition of PbO which gives rise to increase in the number of tetrahedral boron units [16]. Decrease in the Tg values and growth of borons with non bridging oxygen(NBO) are correlated.to each other [17].

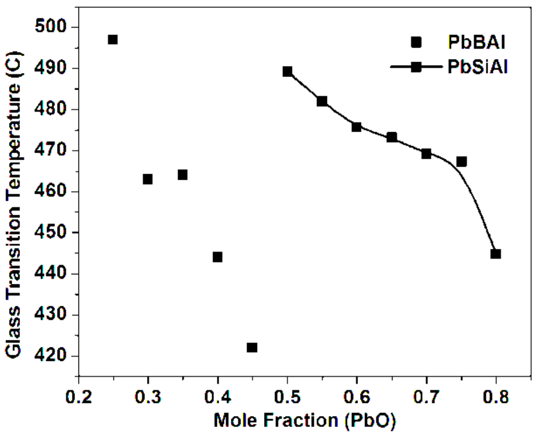

Figure 5.Variation ofGlass transitiontemperature versus mole fraction of PbO inthe PbO-B_2O_3-Al_2O_3 and PbO-SiO_2-Al_2O_3 glass systems.

Longitudinal modulus is calculated by using the values of ultrasonic velocity. The trends of longitudinal modulus are shown in Fig 6.

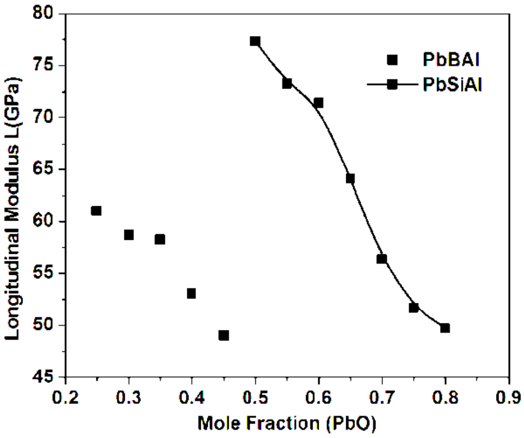

Figure 6.Variation of Longitudinal modulus versusmole fraction of PbO in the $PbO-B_2O_3-Al_2O_3$ and $PbO-SiO_2-Al_2O_3$ glass systems.

The longitudinal modulus values of both glass systems decrease with the mole fraction of PbO contents which shows the glasses become less rigid probably due to formation of NBOs. In the light of this situation, it is estimated that there is formation of NBOs in both the glass systems with the addition of PbO and the results of DSC and ultrasonic measurements support this estimation.

V. CONCLUSIONS

$PbO-B_2O_3-Al_2O_3$ and $PbO-SiO_2-Al_2O_3$ glass systems can be the potential candidates for gamma-ray shielding applications. Higher mass attenuation coefficients and lower HVL values of our glass samples for most of the composition range show that most of our prepared glass samples are better than existing concretes. It is estimated that these glasses have smaller volume requirements as gamma ray shielding materials in nuclear reactors. Results of DSC studies and longitudinal modulus of these glass systems indicate the formation of non bridgingoxygens at higher contents of PbO.

REFERENCES

1. R.S. Kaundal, SandeepKaur, Narveer Singh and K.J. Singh, Investigation of structural properties of lead strontium borate glasses for gamma-ray shielding applications.Journal of Physics andChemistry of Solids, 71, 2010, 1191-1195.
2. Chang-Min Lee, Yoon Hee Lee and Kun Jai Lee, Cracking effect on gamma-ray shielding performance in concrete structure. Programme in Nuclear Energy, 49, 2007, 303–312.
3. D. Rezaei-Ochbelaghand S.Azimkhani,Investigationof gamma-ray shielding properties of concrete containing of different percentages of lead. Applied Radiation and Isotopes,70, 2012, 2282-2286.
4. P. Limkitjaroenporn, J. Kaewkhao, P. Limsuwanand W. Chewpraditkul,Physical,Optical, Structural and gamma-ray shielding properties of lead sodium borate glasses. Journal of Physics and Chemistry of Solids,72, 2011, 245–251.
5. M. Kurudirek, Y. Ozdemir, O. Simsekand R. Durak, Comparison of some lead andnon-lead based glass systems, standard shielding concretes and commercial window glassesin terms of shielding parameters in the energy region of 1 keV-100 GeV: A comparative study. Journal of Nuclear Materials, 407 (2), 2010, 10-115.
6. Andriy O. Mylyanyh, Mykola A. Sheredko and Svyatoslav K. Melnyk, study of glass structures and crustalline phases in the $PbO-Al_2O_3-SiO_2$ system. Journal of Analytical Atomic Spectrometry, 14, 1999, 513-521.
7. AmanpreetSaini, AtulKhanna,Vladimir K. Michaelis, Scott Kroeker, Fernando Gonzalez andDavid Hernandez,Structure-property correlations in lead borate and borosilicate glasses doped with aluminium oxide. Journal of Non-crystalline Solids, 355, 2009, 2323-2332.
8. Joanna Pisarska, Luminescence behaviour of Dy3+ ions in lead borate glasses. Journal of Optical Matter,(31),2009, 1784–1786.
9. K.J. Singh, N. Singh, R.S. Kaundal, andK. Singh, Gamma-ray shielding and structural properties of $PbO-SiO_2$ glasses. Nuclear Instrruments and Methods in Physics Research B, Vol. 266, 2008, 944-948.
10. S. Tuscharoen , J. Kaewkhao, P. Limkitjaroenporn, P. Limsuwan and W. Chewpraditkul, Improvement of $BaO:B_2O_3$:Fly ash glasses: radiation shielding, physical and optical properties.Annals of Nuclear Energy, 49, 2012, 109-113.
11. N. Tsoulfaniidis, Measurements and Detection of Radiation", Mcraw-Hill Book Company, Washington DC, 571P, 1983..
12. N. Singh, K.J. Singh, K. Singh and H. Singh, Comparative study of lead borate and bismuth lead borate glass system as gamma-radiation shielding materials. Nuclear Instruments and Methods In Physics Research B, 225, (3), 2004, 305-309.
13. N. Singh, K.J. Singh and H. Singh, Gamma-ray attenuation studies of $PbO-BaO-B_2O_3$ glass system. Radiation Measurements, 41, (1), 2006, 84-88.
14. N. Chanthima and J. Kaewkhao, Investigation on radiation shielding parameters of bismuth borosilicate glass from 1keV to 100GeV. Annals of Nuclear Energy, 12:00, 2012, 429.
15. S.R. Manohara, S.M. Hanagodimath and L. Gerward, Photon interaction and energy absorption in glass: A transparent gamma ray shield. Journal of Nuclear Materials, 393, 2009, 465-472.
16. D.P. Button, R. Tandon, C. King, M.H. Velez, H.L. Tuller and D.R. Uhlmann, Insights into the structure of alkali borate glasses. Journal of Non-Crystalline of Solids, 49, 1982, 129–142.
17. S.W. Martin & C.A. Angell, Glass Formation and Transition Temperatures in Sodiumand Lithium Borate and Aluminoborate melts up to 72 mol% Alkali", Journal of Non-Crystalline of. Solids, 66, 1984, 429-442.

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