Investigation of Structural Properties of Bismuth Silicate Glass System as Gamma ray Shielding Materials

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ABSTRACT— ***Mass attenuation coefficient, half value layer and mean free path parameters of $xBi_2O_3.(1-x)SiO_2$ (x=0.50 up to 0.75) glass system have been calculated theoretically at photon energies varying from 1keV to 100GeV. The obtained results have been compared with standard radiation shielding concretes at same energies. The density, molar volume, UV-Visible, DSC and acoustical investigations have been used to study the structural properties of the prepared samples. Reported glass samples can find applications in the areas of nuclear reactors and nuclear waste storage containers.***

Keywords—Glasses, Radiation shielding materials, UV-Visible studies, Ultrasonic measurements.

1. INTRODUCTION

Bismuth silicate glasses are employed in large number of applications because these glasses exhibit high refractive index, low glass transition temperature and high atomic weight. Currently, bismuth is playing an important role of radiation shielding glass material and replacing lead as radiation protection shield. Moreover, Bi is much more environment friendly than Pb and therefore, Bi-based materials have been explored as substitutes for the Pb-based materials and the resultant glass systems have been used in several glass industries. Heavy metal oxide glasses have better gamma-ray shielding properties as compared to existing concretes as gamma ray shielding materials [1-2]. Concretes have many limitations. For example; (1) Water content in concretes decreases density and structural strength of concretes. (2) Concretes are opaque to the visible light so it is impossible to see through concrete material. In the light of this situation, it can be interpreted that an alternate to conventional gamma ray shielding material 'concrete' is desired. One of possible alternatives for concrete can be heavy metal glasses. Glasses can be transparent and their properties can be varied by changing composition and preparation techniques. Bismuth being the heavier and useful element, authors have explored the possibility of using it as gamma ray shielding material in terms of bismuth silicate glasses in this research article.

Authors have carried out investigation of Bi_2O_3-SiO_2 glass system for gamma ray shielding properties in terms of mass attenuation coefficient, half value layer (HVL) and mean free path (MFP) values. Theoretical values of mass attenuation coefficient of bismuth silicate glasses and concretes have been calculated and compared by using WinXCOM computer software developed by National Institute of Standards and Technology (NIST) at photon energies varying from 1keV to 100GeV [3-5]. Silicate glasses are commonly available glasses and bismuth is speculated to enhance the gamma ray attenuation due to higher atomic number. The structural properties of bismuth silicate glasses are obtained in terms of density, molar volume UV-visible, DSC and acoustical investigations. Information obtained from structural investigations can be used for checking the possibility of applicability of the bismuth silicate glasses as non-conventional gamma-ray shielding materials for commercial applications.

2. THEORETICAL AND EXPERIMENTAL TECHNIQUES

Six glass samples of the system xBi_2O_3. (1-x) SiO2 (x=0.50 up to 0.75) were prepared by melt quenching technique in our laboratory. Appropriate amounts of PbO and SiO_2 were weighted with an accuracy of 0.001g. Chemicals of AR-grade were mixed thoroughly. Melts of the aforesaid systems with different compositions were obtained in electrically heated furnace (at around 1050-1100^0 C). Dry Oxygen was bubbled through melt using quartz tube to ensure homogeneity. The melt was annealed in another furnace at 270^{0}C in preheated copper mould. Samples were grounded and polished by using different grades of silicon carbide and aluminium paper respectively. Density of these samples was measured by Archimedes's principle using benzene as the immersion liquid. Molar volume data has been obtained with the help of density values (Table 1). X-ray diffraction studies were carried out in order to study the amorphous nature of the prepared samples. Absence of any sharp peak shows that the prepared samples are amorphous.

Table 1: Chemical composition, Density, Molar volume and Acoustic Impedance of Bi_2O_3-SiO_2 glass samples.

Sample No.	Composition (Mole Fraction)		Density (g/cm^3) (±0.0002)	Molar Volume (cm^3/mol)	Acoustic Impedance (10^{-6} kg m^{-2} s^{-1})
	Bi_2O_3	SiO_2			
BiSiG1	0.50	0.50	5.617	46.82	19.54
BiSiG2	0.55	0.45	5.630	50.32	19.14
BiSiG3	0.60	0.40	5.661	53.63	18.96
BiSiG4	0.65	0.35	5.727	56.54	18.49
BiSiG5	0.70	0.30	5.787	59.47	18.38
BiSiG6	0.75	0.25	5.804	62.79	17.70

Mass attenuation coefficient has been calculated theoretically by using WinXCOM computer software. By using the values of gamma-ray attenuation coefficient, the values of HVL and MFP have been calculated. Authenticity of WinXCOM software for evaluating the gamma ray shielding parameters has been checked by several authors and it has been established that WinXCOM software can be used as a tool to evaluate the gamma-ray shielding parameters [5,6].

Ultrasonic measurements were carried out in our laboratory for cylindrical samples having opposite parallel faces. A Matec set-up (imported from USA) was used for ultrasonic measurements. Pulse-Echo mode was used to measure the time of flight between two successive echoes. All measurements were carried out at 5MHz with time resolution of order of 5×10^{-9}s. Ultrasonic jelly was used for the sample and the transducer contact. The longitudinal sound velocity (V_L) is related to the longitudinal modulus (L) [7]. DSC measurements of the prepared samples were carried out by using Perkin Elmer differential scanning calorimeter with the heating rate of 20^0C/min in nitrogen atmosphere. Sample amounts of 10-20 mg were used to perform the DSC measurements. UV-Visible absorption spectra were recorded on polished disc shaped glass samples in the wavelength range of 200-1100 nm on a Shimadzu double-beam spectrophotometer (Model 1601).

3. RESULTS AND DISCUSSIONS

3.1 Gamma-ray shielding properties

Mass attenuation coefficient, half value layer and mean free path values of prepared glass samples at photon energies varying from 1keV to 100GeV are shown in figures 1, 2 and 3 respectively. Mass attenuation coefficient increases (figure 1) and HVL values decreases (figure 2) with the increase in mole fraction of Bi_2O_3.

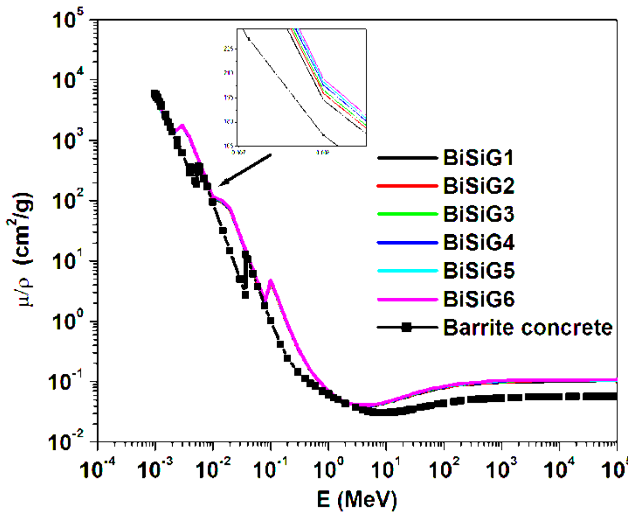

Figure 1. Variation of mass attenuation coefficient as a function of photon energy (1keV to 100GeV) in the Bi_2O_3-SiO_2 glass systems and barite concrete.

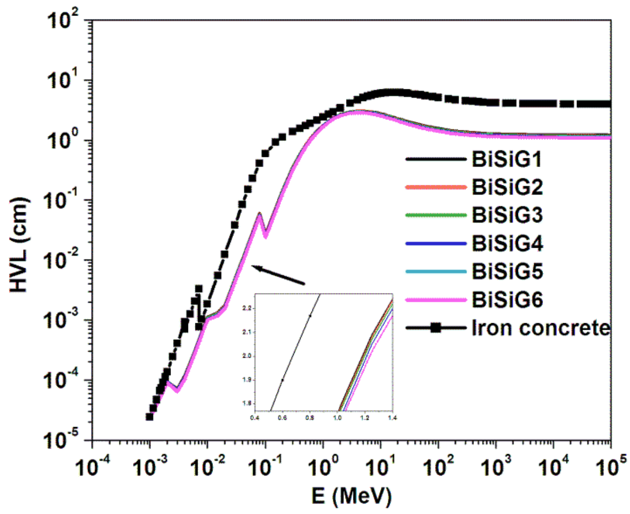

Figure 2. Variation of half value layer as a function of photon energy (1keVto100GeV) in the Bi_2O_3-SiO_2 glass system and iron concrete.

This can be attributed to increasing values of Bi which has higher atomic number as compared to other elements. It is estimated that $0.75Bi_2O_3.0.25SiO_2$ represents best glass sample in terms of mass attenuation coefficient and HVL parameters for gamma-ray shielding applications. Our glass samples show comparable or better mass attenuation coefficient and half value layer parameters than existing concretes (barite and ferrite concretes). The mass attenuation parameters are further used to calculate the mean free path. It has been found that MFP (figure 3) varies with photon energy. The MFP values increases with the increasing composition of bismuth oxide. In the light of this situation, it can be interpreted that increasing the content of Bi_2O_3 in the Bi_2O_3-SiO_2 glass system improves the radiation shielding properties in terms of mass attenuation coefficient, HVL and MFP parameters.

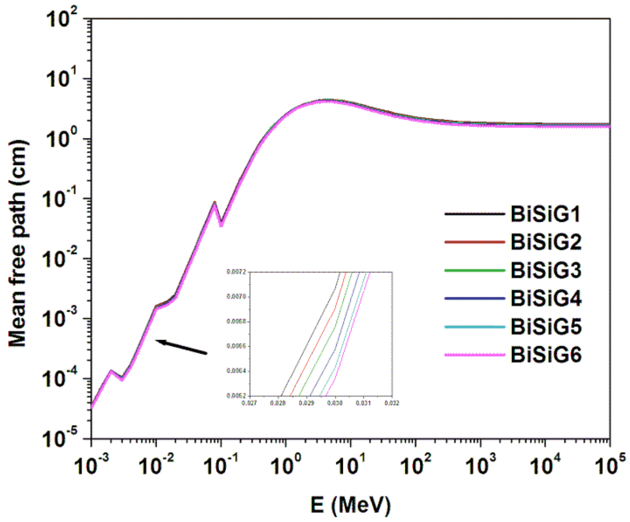

Figure 3. Variation of mean free path as a function of photon energy in the Bi_2O_3-SiO_2glass systems.

3.1.1 Structural properties

The composition, density, molar volume and acoustic impedance of bismuth silicate glasses are given in table 1. It can be seen in table 1 that the density values of our glass system increases with the increase in the content of Bi_2O_3. This can be attributed to higher atomic mass of bismuth as compared to silicate. The molar volume was calculated by using density values. It can be observed that the density value and molar volume values increases with the increasing concentration of Bi_2O_3. It is speculated that addition of bismuth oxide in bismuth silicate glasses leads to formation of open structure.

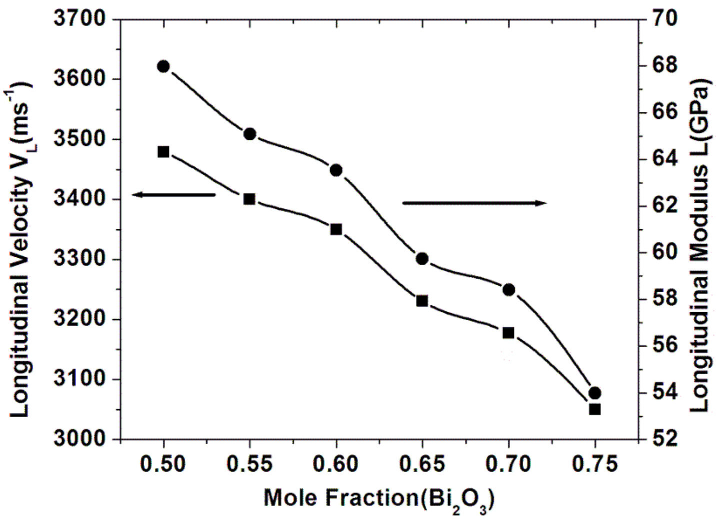

Figure 4. Variation of ultrasonic velocity and longitudinal modulus with mole fraction of Bi_2O_3 in the Bi_2O_3-SiO_2 glass systems.

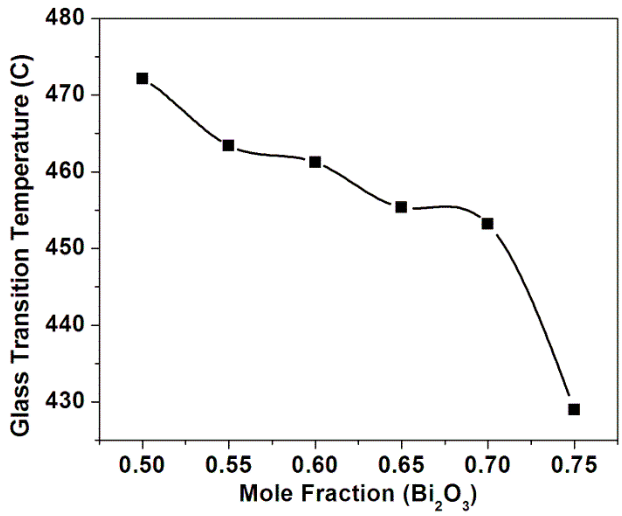

Figure 5. Variation of glass transition temperature with mole fraction of Bi_2O_3 in the Bi_2O_3-SiO_2 glass systems.

Figure 4 shows the variation of longitudinal ultrasonic velocities and longitudinal modulus of prepared glass samples as function of composition. Both quantities decrease with the increase in mole fraction of Bi_2O_3. It is speculated that longitudinal modulus decreases because oxygen bond is destroyed by the bismuth atoms which gives rise to the loose structure. The acoustic impedance also decreases as mole fraction of Bi_2O_3 increases [8]. Increase in bismuth concentration leads to increase in the formation of non-bridging oxygen atoms which causes formation of loose structure. Figure 5 shows the variation of glass transition temperature (Tg) with the mole fraction of Bi_2O_3. The values of glass transition temperature decreases from 472 to 428 ^{0}C with the increase in mole fraction of Bi_2O_3. Martin and Angell [9] had quantitatively related glass transition temperature with number of non-bridging oxygens (NBOs). As mole fraction of bismuth is increased, the decrease in values of glass transition temperature may be related to continuous increase in non-bridging oxygen atoms, which gives lesser stability of glass network.

The optical energy band gap and Urbach energies have been evaluated from UV-Visible spectra (see Table 2). The absorption coefficient as a function of wavelength, α(λ), was calculated by dividing the measured absorbance by sample thickness, the band gap (Eg) and Urbach energy (ΔE) was calculated [10]. Band gap decreases from 2.49 to 2.44 eV with the increase in mole fraction of Bi_2O_3 whereas, Urbach energy increases from 0.21 to 0.25eV with the rise in content of Bi_2O_3. The calculated results of band gap and Urbach energy supports the interpretation of our data for the molar volume, DSC and ultrasonic investigations as follows. UV- Visible light absorption in oxide glasses is due to the excitation of electrons associated with NBOs. Lesser is the concentration of NBOs in the glass network, the higher is the band gap and the lesser are the Urbach energy values [11]. Therefore, it can be concluded that increase in the content of bismuth oxide in bismuth silicate glasses leads to formation of more NBOs. Therefore, UV-Visible and DSC measurements support the conclusions of results of ultrasonic data. Refractive index, band gap and molar refraction are the important properties of glasses and their values are shown in table 2. The refractive index and molar refraction values increase with increase in mole fraction of bismuth oxide.

Table 2: Mole Fraction, Energy Band Gap, Urbach Energy, Refractive Index and Molar Refraction of Bi_2O_3-SiO_2 glass system.

Sample No.	Mole Fraction (Bi_2O_3)	Energy Band Gap Eg (eV)	Urbach Energy ΔE (eV)	Refractive index (n)	Molar Refraction (R_m)(cm^3)
BiSiG1	0.50	2.49	0.21	2.549	30.303
BiSiG2	0.55	2.48	0.22	2.553	32.601
BiSiG3	0.60	2.47	0.23	2.556	34.783
BiSiG4	0.65	2.47	0.24	2.556	36.681
BiSiG5	0.70	2.46	0.24	2.560	38.617
BiSiG6	0.75	2.44	0.25	2.566	40.864

4. CONCLUSIONS

Bi_2O_3-SiO_2 glasses are the potential candidates for non-conventional gamma-ray shielding material applications. Higher values of mass attenuation coefficient and lower values of HVL of our glass system for most of the energy range as compared to concrete shows that volume requirements for shielding with bismuth silicate glass system will be lesser. Results of ultrasonic velocity, DSC measurements and UV-Visible studies indicate the formation of non-bridging oxygens with the increase in the mole fraction of Bi_2O_3.

5. REFERENCES

[1] J. Kaewkhao, A. Pokaipisit, P.Limsuwan, "Study on Borate Glass system containingwith Bi_2O_3 and BaO for Gamma rays Shielding materials: Comparison with PbO", Nucl. Mat., 399, pp.38–40, 2010.
[2] R.S. Kaundal, Sandeep Kaur, Narveer Singh, K.J. Singh, "Investigation of structuralproperties of lead strontium borate glasses for gamma ray shielding applications", Phy.Chem. of solids, 71, pp.1191-1195, 2010.
[3] Chang-Min Lee, Yoon Hee Lee, Kun Jai Lee, "Cracking effect on Gamma-ray Shielding performance in Concrete Structure", Prog. Nucl. Energy, 49, pp. 303–312, 2007.
[4] S. R. Manohara, S. M. Hanagodimath, L. Greward, "Photon interaction and Energy Absorption in glass: A transparent Gamma ray Shield, Nucl. Mat. 393, pp. 465-472, 2009.
[5] N. Chanthima, J. Kaewkhao, "Investigation on Radiation Shielding Parameters of Bismuth Borosilicate Glasses from 1keV to 100GeV", Ann. Nucl. Energy, 55, pp. 23-28, 2013.
[6] C. Bootjomchai, J. Laopaiboon, C. Yenchai, R. Laopaiboon, "Gamma-ray Shielding and Structural Properties of Barium-Bismuth-Borosilicate Glasses", Radiat. Phys. Chem., 81, pp. 785-790, 2012.
[7] K.J. Singh, N. Singh, R.S. Kaundal, K. Singh,Gamma-ray Shielding and Structural Properties of PbO–SiO2 Glasses", Nucl. Instr. Meth. (B), 266, pp. 944-948, 2008.
[8] R.S. Kaundal, S. Kaur, N. Singh, K. J. Singh, Ultrasonic and FTIR investigationsof PbO-Bi_2O_3-SiO_2 Glass System", J. Pure Appl. Ultrason., 31, pp.76, 2009.
[9] S. W. Martin, C. A. Angell,"Glass Formation and Transition Temperatures in Sodium and Lithium Borate and Aluminoborate melts up to 72 mol% alkali",J. Non-Cryst. Solids,, 66, pp. 429–442, 1984. .

[10] G. Sharma, K.S. Thind, Manupriya, H.S. Klare, S.B. Narang, L. Gerward, V.K. Dangwal, "Effects of Gamma ray Irradiationon optical properties of ZnO-PbO-B_2O_3 glasses", Nucl. Instr. Meth. (B), 243, pp. 345-348, 2006.
[11] K. Singh, H. Singh, V. Sharma, R. Nathuram, A. Khanna, R. Kumar, S.S. Bhatti, H.S. Sahota,"Gamma-ray Attenuation Coefficients in Bismuth Borate Glasses", Nucl. Instr. Meth. (B), 194, pp. 1-6, 2002.