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## Review Article

### ULTRASONIC AND STRUCTURAL PROPERTIES OF $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$ GLASSES FOR GAMMA RAY SHIELDING PROPERTIES

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#### ABSTRACT

The elastic properties of the system of the glass system  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  have been calculated by measuring the ultrasonic wave velocities by pulse echo technique at a frequency of 5MHz at room temperature. Our aim is to improve the brittleness of the heavy metal oxide glasses by elastic properties. The Structural properties are characterized by X-ray diffraction, Density, Molar Volume, UV-Visible technique, DSC measurements and Gamma-ray shielding applications. All the valuable data may provide the information which may improve the mechanical properties of the prepared glass system.

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#### INTRODUCTION

The heavy metal oxide glasses are commonly used because of their diverse applications in the field of electronics, nuclear and solar energy technologies, and acoustic-optic devices [1]. The propagation of ultrasonic waves in solids such as glass provides valuable information regarding the solid state motion in the material [2]. The longitudinal velocity, longitudinal modulus and acoustic impedance are the suitable parameters which can be studied by measuring ultrasonic velocities. The composition of the bismuth silicate glasses doped with aluminum oxide give information about the microstructure and dynamics of the glasses. Elastic properties combined with acoustic properties are quite significant because of their applications in certain devices such as delay lines, light modulators and solid state sensors[3-6].

Glasses containing  $\text{Bi}_2\text{O}_3$  has many advantages due to high density and high refractive index and this feature is very important for advanced optical tele-communication and processing device [2,5].  $\text{Bi}_2\text{O}_3$  is known as a conditional glass former. Bi-based materials have been expected as substitute for the Pb-containing materials because Bi is much safer than Pb [7]. These heavy metal oxide based glasses are used as alternatives of concretes for Gamma-ray shielding applications. Glasses have the advantages over concretes because they can be prepared in wide range of composition [8]. It has been found

that the bismuth silicate glasses doped with aluminum oxide plays an important role in observing the structure of glasses.

#### Experimental and Theoretical Details

Six cylindrical shaped glass samples of the system  $x\text{Bi}_2\text{O}_3\text{.}(0.90-x)\text{SiO}_2\text{.}0.10\text{Al}_2\text{O}_3$  in the interval of ( $x=0.50$  up to  $0.75$ ) were prepared by using melt and quenching technique. For the preparation of glass samples, appropriate amounts of  $\text{Bi}_2\text{O}_3$ ,  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  (AR grade) were weighted 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 1100 up to 1150<sup>o</sup>C. Dry oxygen was bubbled through melts using quartz tube in order to ensure 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 275<sup>o</sup>C and slowly cooled to room temperature. Annealing temperature has been kept same for all six samples so that have similar thermal history for all the samples. The prepared glass samples were grinded and polished with different grades of silicon carbides and aluminum paper respectively.

The compositions of all the samples are given in Table 1. Densities of all samples was measured by using Archimede's principle with benzene as the immersion liquid.

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### XRD studies

X-ray diffraction studies shows that prepared samples are amorphous. A Philips PW 1710 diffractometer was used and radiation used was  $\text{CuK}\alpha$ . The values was recorded at angular range ( $2\theta$ ) of  $10\text{-}70^\circ$ . Absence of crystallization peak in XRD data shows that prepared samples are amorphous.

### Ultrasonic measurements

Ultrasonic studies were carried out in our laboratory for the cylindrical samples having opposite parallel faces. A Matec setup (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 the samples and the transducer contact.

Ultrasonic velocity (U) was calculated using the relation,

$$U = \frac{2d}{t} \quad (1)$$

where d is the thickness of the specimen and t is the time of flight between two echoes.

Longitudinal modulus (L) is given by

$$L = \rho U^2 \quad (2)$$

Where  $\rho$  is the density of samples and U is the ultrasonic velocity.

Acoustic impedance

$$Z = U \rho \quad (3)$$

### Diffraction scanning calorimetry (DSC)

DSC measurements of the prepared samples were carried out by using Perkin Elmer differential scanning calorimeter with the heating rate of  $20^\circ\text{C}/\text{min}$  in nitrogen atmosphere. Sample amounts of 10-20 mg were used to perform the DSC measurements.

### UV-Visible spectroscopic studies

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). The absorption coefficient as a function of wavelength,  $\alpha(\lambda)$ , was calculated by dividing the measured absorbance by sample thickness.

### Gamma ray shielding properties

Mass attenuation coefficient and half value layer (HVL) has been theoretically calculated by using XCOM computer program developed by National institute of standards and technology (NIST) [5]. The mass attenuation coefficient ( $\mu/\rho$ ) is given by;

$$\mu/\rho = \sum w_i (\mu/\rho)_i \quad (4)$$

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

The following relation is used to calculate the HVL,

$$\text{HVL} = 0.693/\mu, \quad (5)$$

where  $\mu$  is the linear attenuation coefficient.

## RESULTS AND DISCUSSION

### Density measurements

Glass samples of  $x\text{Bi}_2\text{O}_3 \cdot (0.90-x)\text{SiO}_2 \cdot 0.10\text{Al}_2\text{O}_3$  glass system were prepared for the values of  $x = 0.50, 0.55, 0.60, 0.65, 0.70$  and  $0.75$ . It can be seen in table 1 that the density values of our glass system increases 5.41 to 6.16  $\text{gcm}^{-3}$  with the increase in the content of  $\text{Bi}_2\text{O}_3$ . The increase in density of glasses is due to a change in cross link density and coordination numbers of  $\text{Bi}^{3+}$  ions.

The molar volume was calculated by using density values. It can be observed in Table 1 that molar volume value also increases from 47.71 to 65.62  $\text{cm}^3 \text{mole}^{-1}$  with the increasing concentration of  $\text{Bi}_2\text{O}_3$ .

### Structural Properties

Fig. 1 shows the variation of longitudinal ultrasonic velocity and longitudinal modulus of  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  glass samples. It is seen that the longitudinal velocity decreases from 4191 to 3550  $\text{ms}^{-1}$  and longitudinal modulus also decreases from 95.05 to 77.73 Gpa. The formation of non-bridging oxygens (NBOs) at higher  $\text{Bi}_2\text{O}_3$  contents may be responsible for the for the behaviour of the sound velocity. Table 1 shows the acoustic impedance also decreases from 95.06 to 77.73 ( $10^6 \text{kg m}^{-2} \text{s}^{-1}$ ). As mole fraction of  $\text{Bi}_2\text{O}_3$  is increased the formation of non-bridging oxygen atoms start taking place causing loose structure. Fig 2 shows DSC pattern of  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$ . The glass transition temperature decreases with the increase in mole fraction of  $\text{Bi}_2\text{O}_3$ . Button et al. [9] had undertaken qualitative analysis between increasing transition temperatures (Tg) and increase in the number of tetrahedral borate units.

**Table 1** Chemical composition, Density and Molar volume of  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  glass samples.

Sample No.	Composition (Mole Fraction)			Density ( $\text{g}/\text{cm}^3$ ) ( $\pm 0.0002$ )	Molar Volume ( $\text{cm}^3/\text{mol}$ )	Acoustic Impedance ( $10^6 \text{kg m}^{-2} \text{s}^{-1}$ )
	$\text{Bi}_2\text{O}_3$	$\text{SiO}_2$	$\text{Al}_2\text{O}_3$			
BiSiAlG1	0.50	0.40	0.10	5.4121	47.71	95.06
BiSiAlG2	0.55	0.35	0.10	5.5676	51.64	91.94
BiSiAlG3	0.60	0.30	0.10	5.7216	53.80	84.10
BiSiAlG4	0.65	0.25	0.10	5.7681	56.88	81.11
BiSiAlG5	0.70	0.20	0.10	6.1030	57.08	79.09
BiSiAlG6	0.75	0.15	0.10	6.1681	65.62	77.73

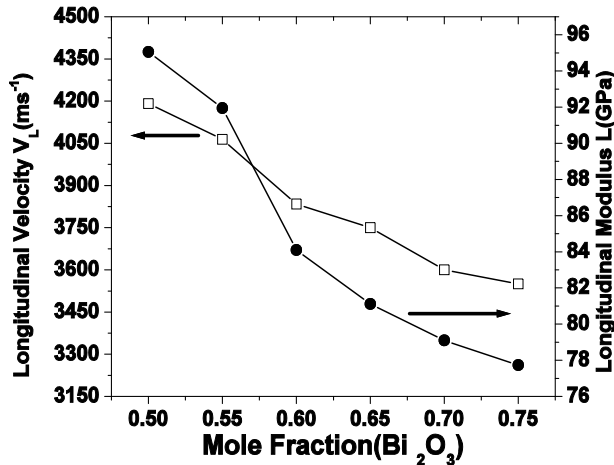


Figure 1 Variation of longitudinal ultrasonic velocity and longitudinal modulus with mole fraction of  $\text{Bi}_2\text{O}_3$ .

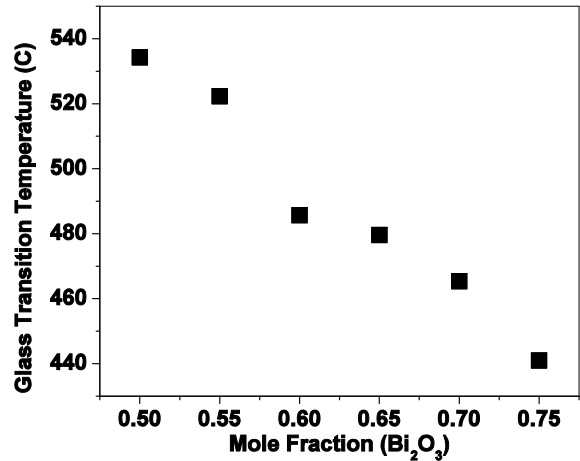


Figure 2 Variation of glass transition temperature with mole fraction of  $\text{Bi}_2\text{O}_3$  in  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  glass system.

Table 2 Mole Fraction, Energy Band Gap and Urbach energy of  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  glass system.

Sample No	Mole Fraction ( $\text{Bi}_2\text{O}_3$ )	Energy Band Gap $E_g$ (eV)	Urbach Energy $\Delta E$ (eV)
BiSiAlG1	0.50	2.45	0.22
BiSiAlG2	0.55	2.43	0.23
BiSiAlG3	0.60	2.42	0.25
BiSiAlG4	0.65	2.42	0.25
BiSiAlG5	0.70	2.42	0.25
BiSiAlG6	0.75	2.40	0.26

They had suggested that the decrease in  $T_g$  and growth of borons with non-bridging oxygens are correlated especially, in the higher alkali region. Martin and Angell [10] had quantitatively related glass transition temperature with number of non-bridging oxygens (NBOs). Band Gap and Urbach Energy have been calculated from UV-Visible spectra (see Table 2). The Band Gap decreases and Urbach energy increases with the rise in content of  $\text{Bi}_2\text{O}_3$ . 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 optical band gap and lesser is the Urbach energy values in the borate glasses and vice-versa. Therefore DSC spectra and UV-Visible data supports the conclusion of results of the ultrasonic data.

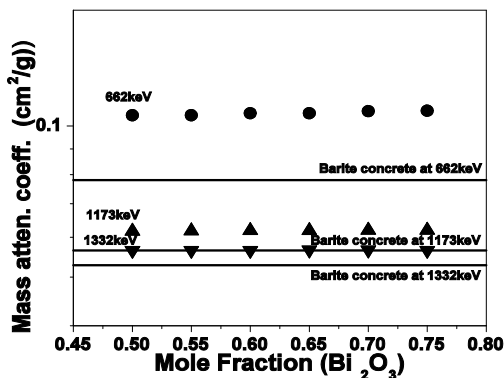


Figure 3 Variation of mass attenuation coefficient with mole fraction of  $\text{Bi}_2\text{O}_3$  at 662, 1173 and 1332keV. Theoretical values (—) for Barite concretes at same energies are included for comparison.

### Gamma-ray shielding properties

Mass attenuation coefficient and HVL values of prepared samples at different energies (662, 1173 and 1332 keV) are shown in figures 3 and 4 respectively. For a better shielding material, higher mass attenuation and lower HVL parameter is desired. In  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  mass attenuation coefficient increases and HVL values decreases with the mole fraction of  $\text{Bi}_2\text{O}_3$ . Mass attenuation coefficient and HVL values for different concretes used in nuclear reactors are evaluated using XCOM software at 662, 1173 and 1332 keV energies [3,11]. It can be concluded from the results that our glass system represents better mass attenuation coefficient and HVL parameters than concretes.

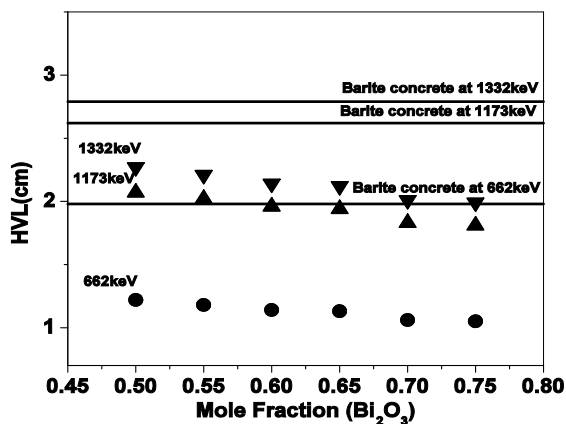


Figure 4 Variation of HVL with mole fraction of  $\text{Bi}_2\text{O}_3$  at 662, 1173 and 1332keV. Theoretical values (—) for Ferrite concretes at same energies are included for comparison.

### CONCLUSIONS

Glasses of the system  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  with different composition of  $\text{Bi}_2\text{O}_3$  were prepared. Their Ultrasonic velocity and Gamma-ray attenuation studies have been analysed. It has been observed that prepared glass samples provides best results of sound velocity and there is also formation of more number of NBO's with the increasing content of  $\text{Bi}_2\text{O}_3$ . This glass system also has lower values of HVL parameters and higher values of mass attenuation coefficient so volume requirements

for shielding with  $\text{Bi}_2\text{O}_3\text{-SiO}_2\text{-Al}_2\text{O}_3$  glass system will be lesser.

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