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Research Article

STRUCTURE INVESTIGATION OF Bi_2O_3–SiO_2 GLASS EMPLOYING ULTRASONIC STUDY

Sumathi T*[1] and Anandhi S[2]

[1]Department of Physics (DDE WING), Annamalai University
[2]Department of Physics, Annamalai University, Annamalai Nagar-608 002, Tamil Nadu, India

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ABSTRACT

Glasses with composition (100–x) Bi_2O_3–xSiO_2 (where x = 38, 41, 44, 47 and 50 mol %) have been prepared using the melt quench technique. The ultrasonic velocities (both longitudinal and transverse) were measured at 10 MHz at room temperature using the pulse echo overlap method. The physical parameter such as density, elastic constants and other parameters like Poisson's ratio, acoustic impedance, microhardness, Debye temperature and thermal expansion coefficient have been evaluated. The variations in these parameters are analyzed in terms of structural changes in the glass network with composition. The amorphous nature of the glass samples are studied using XRD in the glass samples.

INTRODUCTION

Research in the field of glass and crystal using ultrasonic methods has been carried out for many years. Ultrasonic non-destructive character on materials is a versatile tool for investigating the change in microstructure, deformation process and mechanical properties of materials. This is possible due to the close association of the ultrasonic waves with elastic and inelastic properties of the materials (Kuttruff, 1991). In recent years, the study of glasses has rapidly increased because of their diverse applications in electronics, nuclear and solar energy technologies and acousto-optic devices. In a study of the elastic constant of materials using ultrasonic methods, the main point to note is the ultrasonic wave propagation velocity and the density. Elastic properties of glasses can be obtained non-destructively by measuring ultrasonic velocity in both longitudinal and shear modes. Sound velocity is related to elastic moduli whose changes can indicate variation in rigidity and structural stability (Azanty *et al,* 2012). Heavy metal oxide (HMO) glasses containing bismuth have been investigated for possible use in high-energy of physics, or may be employed in non-linear optical use (Yasser B. Saddeek *et al,* 2004). Bismuth based oxide glasses possess high polarisability, long infrared cut-off and high non-linear optical susceptibility, which makes them promising materials for technological applications such as optical switching, photonic devices, broadband amplification devices and optical waveguides due to their low phonon energy. SiO_2 is a traditional glass former and has an extremely wide spectrum of industrial applications in its various amorphous forms (Beall *et al,* 1992).On the other hand, Bi_2O_3 is not a classical glass former but in the presence of conventional glass formers like SiO_2, PbO and B_2O_3, a glass network of BiO_3 and BiO_6 units may be built (Rajni Bala *et al,* 2015). Thus the present paper aim to throw light on the structure of bismuth glasses containing silicon oxide in terms of the density, molar volume, elastic moduli and Debye temperature.

MATERIAL AND METHODS

A series of Bismuth Silicate (BS) glass samples of formula (100-x) Bi_2O_3- *x* SiO_2 (with x varying from 38 to 50 mol%) composition were prepared by melting appropriate mixture of Bi_2O_3 and SiO_2, using Analar grade (minimum assay 99.9%) chemicals. The compositions in mol% of different glass specimen are listed in Table 1. The required amount (approximately 20g) in mol% of different chemicals in powder form were weighed using single pan balance (Model SHIMAD2U AX 200) having an accuracy of ±0.0001g.The raw materials of bismuth trioxide and silicon dioxide (SiO_2) were mixed together by grinding to obtain a fine powder. The obtained mixture was melted in a silica crucible for three hours in a muffle furnace at temperature of 850 – 1030 C until a bubble free liquid is formed. The melt was poured into a brass mould to form samples of dimensions 10 mm diameters and 6mm thickness. Glass samples were annealed at 400 ºC for 2 hours to avoid the mechanical strain developed during the

*Corresponding author: **Sumathi T***
Department of Physics (DDE WING), Annamalai University

quench process. Then the oven was switched off and glass was allowed to cool gradually to room temperature. Diamond disc and diamond powder were used to smoothen the prepared glass samples and to keep their surfaces perfectly plane. The density of the glass samples were measured using Archimede's principle. Ultrasonic longitudinal and shear velocities of the specimen were determined by using Pulse-Echo method by using X-cut and Y-cut quartz transducers having the fundamental frequency of 10 MHz. The ultrasonic longitudinal and shear velocities of the specimen were determined by using the pulse-Echo method. From XRD it was found that the nature of the sample was amorphous.

Theory and Calculation

Various parameters of the glass specimen are calculated using the measured density (ρ), longitudinal velocity (U_l) and shear velocity (U_s) using the standard expressions given below:

Longitudinal Modulus $(L) = \rho U_l^2$ (1)

Shear Modulus $(G) = \rho U_s^2$ (2)

Bulk Modulus $(K) = L - \left(\frac{4}{3}\right) G$ (3)

Young's Modulus $(E) = (1 + \sigma)\, 2G$ (4)

Poisson's ratio $(\sigma) = \left[\frac{L-2G}{2(L-G)}\right]$ (5)

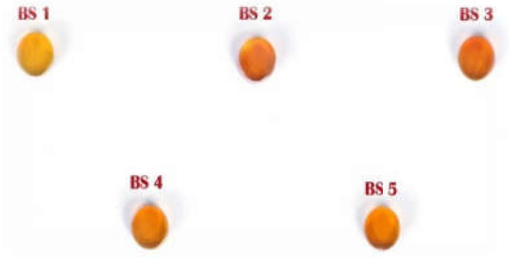
Acoustic impedance $Z) = \rho U_l$ (6)

Micro hardness $(H) = (1-2\sigma)\, \frac{E}{6(1+\sigma)}$ (7)

Debye temperature $(\theta_D) = \frac{h}{k}\left(\frac{9N}{4\pi V_m}\right)^{1/3} U_m$ (8)

Thermal expansion coefficient $(\alpha_p) = 23.2\,(U_l - 0.57457)$ (9)

Where h, k, N and Vm are the Planck's constant, the Boltzmann's constant, the Avogadro's number and the molar volume of the sample respectively.



Photograph of Bismuth based Silicate glass (BS) specimen

RESULTS AND DISCUSSION

The XRD pattern of the BS is shown in Fig. 1. XRD patterns of the prepared glass samples show no sharp Bragg's peak, but only a broad diffuse hump around low angle region. This is the clear indication of amorphous nature within the resolution limit of XRD instrument.

The experimental values of density (ρ), longitudinal ultrasonic velocity (U_l), and shear ultrasonic velocity (Us) of the different glass specimens with respect to the change in the mol% of silicon dioxide (SiO_2) are revealed in Table 2. The calculated longitudinal modulus (L), shear modulus (G), bulk modulus (K), Young's modulus (E) and Poisson's ratio (σ) are reported in Table. 3.

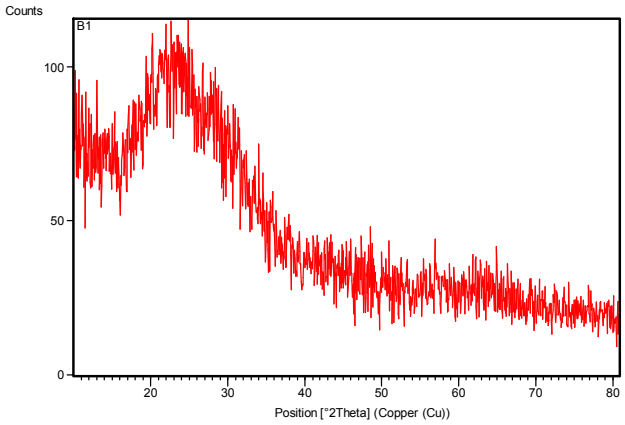


Fig. 1 XRD pattern of Bismuth based Silicate glass (BS1) Specimen

Table 1 Nomenclature and composition of glass sample

Name of the Specimen	Nomenclature	Composition in mol %
	SiO_2	Bi_2o_3
BS1	38	62
BS2	41	59
BS3	44	56
BS4	47	53
BS5	50	50

Table 2 Values of density (ρ), longitudinal velocity (U_l), shear velocity (U_s) and molar volume (V_m) of BS glass systems

Name of the Specimen	Density $\rho(10^3\ kgm^{-3})$	Molar volume V_m (cm^3/mol)	Ultrasonic velocity (ms^{-1})	
			Longitudinal velocity (U_l)	Shear velocity (U_s)
BS1	7.399	42.13	2611.97	1386.67
BS2	6.810	43.98	2901.53	1475.71
BS3	5.891	48.78	3378.63	1661.01
BS4	5.540	49.67	3608.32	1763.83
BS5	5.291	49.72	3793.85	1834.21

Table 3 Values of longitudinal, shear, bulk and Young's moduli and Poisson's ratio of BS glass systems

Name of the Specimen	Longitudinal modulus $L/(x10^9 Nm^{-2})$	Shear modulus $G/(x10^9 Nm^{-2})$	Bulk modulus $K(x10^9 Nm^{-2})$	Young's modulus $E/(x10^9 Nm^{-2})$	Poisson's ratio (σ)
BS1	50.48	14.23	28.55	32.51	0.3037
BS2	57.33	14.83	35.33	39.32	0.3255
BS3	67.24	16.25	45.01	43.57	0.3406
BS4	72.13	17.24	49.64	46.29	0.3430
BS5	76.14	17.80	53.65	47.96	0.3474

The acoustic impedance (Z), microhardness (H), Debye temperature (θ_D) and thermal expansion coefficient (α_p) are presented in the five BS glasses with composition of SiO_2 (mol%) content are presented in Table. 4

Table 4 Values of acoustic impedance (Z), micro hardness (H), Debye temperature (θ_D) and thermal expansion coefficient (α_p) of BS glass system

Name of the Specimen	Acoustic impedance $Z/(x10^7\ kgm^{-2}\ s^{-1})$	Micro hardness $H/(x10^9\ Nm^{-2})$	Debye temperature θ_D (K)	Thermal expansion coefficient $\alpha_p\ (K^{-1})$
BS1	1.9325	1.6312	162.001	60584.373
BS2	1.9759	1.7249	170.003	67302.165
BS3	1.9900	1.7264	185.028	78370.885
BS4	1.9985	1.8039	196.372	83697.373
BS5	2.0069	1.8096	203.010	88003.989

For BS glass system, the values of density (ρ) shows a continuous decrease (Table 2) with increase in molar volume as well as mol% of SiO_2 content. The decrease in density and increase in molar volume with concurrent increase in SiO_2 may be due to the partial replacement of lighter SiO_2 atom (60.08 g/mol) by heavier Bi_2O_3 atom (465.96 g/mol). Similar results were observed by Sidkey *et al*, 2004. From the Table 2 it is observed that the ultrasonic velocities (both longitudinal and shear) increase with increasing SiO_2 content. The increase of ultrasonic velocity both longitudinal and shear is attributed to increase in the rigidity of the glass network structure (Yasser B. Saddeek *et al,* 2004).

The longitudinal, shear, bulk and young's moduli (Table 3) increase in entire range composition of SiO_2 in binary BS system. The increase of elastic moduli is due to the increase in creating bridging oxygen atoms and the bond strength of Si-O of the silicate network that cause the formation of a compacted structure (Saddeek *et al,* 2014).

The Poisson's ratio (σ) increases with increase in SiO_2 content and almost remains constant above 44 mol% of SiO_2 (Table 3). The increase in poisson's ratio with increasing Bi_2O_3 content was attributed to the increase in the average cross – link density of the glass as proposed by Higazy and Bridge (1986). The acoustic impedance Z increases (Table 4) with increase in mol% of SiO_2 content in binary BS system. The acoustic impedance increases in the glass system attributed to the presence of bridging oxygen and increase in rigidity of the glass structure (Vasantharani and Sangeetha 2013).

It is observed from the Table 4, the microhardness increases with increase in mol% of SiO_2. The continuous increase in microhardness as well as Poisson's ratio reveals the absence of non bridging oxygen ion (NBO) and this cause the formation of glassy network (Sumathi and Kannappan 2012). The Debye temperature extracted from measured ultrasonic velocities increase from 162K to 203K by increasing of SiO_2 content (Table 4) and its increase implies the increase of compactness in the glass structure(Hamid –Reza *et al.,* 2013).

From the Table 4, it is observed that thermal expansion co-efficient (α_p) increase with increase in SiO_2 content. The increasing thermal expansion co-efficient (α_p) reveals an increasing number of bonds per unit volume and account for enhancement of rigidity of glass structure.

CONCLUSION

Based on the results obtained using density and velocity the following conclusions have been made on the Bismuth Silicate Glass System.

- The gradual decrease in density with mol % of SiO_2 content of the glass sample indicates the dependence of density on weight of the metal atom in the network modifier (NWM).
- The increase in ultrasonic velocity both longitudinal and shear is attributed to increase in the rigidity of the glass network structure.
- The elastic moduli, Debye temperature and other acoustical parameters increase with increase in mol% of SiO_2 in Bismuth Silicate system.
- The increase in acoustical, elastic and mechanical properties of BS glasses throw light on the rigidity and compactness in structure network.

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