



Absorption spectroscopic studies on gamma irradiated bismuth borosilicate glasses



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HIGHLIGHTS

- Gamma ray shielding behaviour of the bismuth based borosilicate glasses has been investigated.
- Effect of modifier Bi_2O_3 has been studied on glass composition.
- FT-IR spectroscopy has been employed to study the structural changes in glasses.
- Results indicate that the glass structure corresponds to that of radiation hard glasses.

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ABSTRACT

FT-IR spectroscopic measurements have been employed to investigate the structural changes in quaternary $x\text{Bi}_2\text{O}_3$ –15 Na_2O –(70 – x) B_2O_3 –15 SiO_2 glass system with $x = 5, 10, 15, 20$ and 25 (mol%). The effect of gamma irradiation in the dose range of 0.1–60 kGy on the infrared absorption spectra of these glasses is also reported. The IR spectra of the prepared samples show characteristic bands related to the sharing of triangular and tetrahedral borate and silicate groups together with Bi–O groups. The effect of the heavy metal oxide Bi_2O_3 on the glass composition is also studied.

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1. Introduction

Heavy metal oxide (HMO) glasses have been an area of interest for long due to their unique and characteristic properties such as thermal stability, high refractive index, chemical durability, high infrared transparency and high density which makes them suitable for varieties of applications [1]. Particularly, Bi_2O_3 -containing glasses find their use in electronic devices, ceramic materials, thermal and mechanical sensors, reflecting windows, optical fibres and radiation-shielding purposes [2]. Bi_2O_3 exists in a monoclinic form, and exhibits irregular octahedral arrangement of six oxygen atoms located at interatomic distances from 2.14 to 2.80 Å. Out of these, three are appreciably closer (2.14–2.29 Å) than the other three (2.48 to 2.80 Å) [3]. Due to its low field strength i.e. small charge to ionic ratio and high polarizability, pure Bi_2O_3 glass cannot be

obtained as compared with pure B_2O_3 glass. However, in the presence of very small additions of conventional glass formers such as P_2O_5 , B_2O_3 , and SiO_2 , it can build a glass network of $[\text{BiO}_n]$ ($n = 3, 6$) pyramids [4,5]. With larger additions, glasses over a wide range of compositions have been found. Because of its dual properties, as a modifier with $[\text{BiO}_6]$ octahedral and as glass former with $[\text{BiO}_3]$ pyramidal units, bismuth ions may influence the physical, structural, optical and electrical properties of oxide glasses [6]. Being a heavy metal oxide, the addition of Bi_2O_3 is known to increase the radiation hardness of the glasses. Such glass systems are expected to sustain or withstand the radiation-induced degradation caused by high energy radiations such as gamma and X-rays. Therefore, these glasses can be used for radiation shielding purposes and applications involving the use of ionizing radiations such as nuclear reactors, nuclear power stations, spacecrafts, satellites and military aircraft.

The use of radioactive isotopes in agriculture, industry, science, technology and medical applications has made it essential to develop a material, which can be used under harsh conditions of

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nuclear radiation exposure and simultaneously can act as a shielding material. For such a material, homogeneity of density and composition is an important requirement. Glasses are promising materials in this regard. Several glasses have been developed for nuclear engineering applications because they accomplish the double task of allowing visibility and at the same time can absorb high energy radiations like gamma-rays and neutrons [7,8]. Borosilicate glasses have been an area of interest for more than two decades due to its diversified use in the biological, electrical and pharmaceutical industries [9]. The use of these glasses has also been made for the entrapment/immobilisation of high level waste from nuclear power plants and armour industries [10–12]. Irradiation effects on the structural, mechanical and optical properties of a good shielding glass should be small. The effect of irradiation on glasses is dependent on several factors such as on the type and energy of irradiation, and glass composition [13]. It is well established that radiation damage in glass causes active defects which can be introduced by ionization or atomic displacement mechanisms or via the activation of the pre-existing defects [14–17]. The knowledge of the glass structure before and after irradiation therefore becomes extremely important for the understanding the structural evolution of glasses. In this paper we report the experimental results of the gamma irradiation on the glasses of $\text{Bi}_2\text{O}_3\text{--Na}_2\text{O--B}_2\text{O}_3\text{--SiO}_2$ system. The impact of radiation on the structural changes observed by using FTIR spectroscopy on these glasses has been described. The present work is also focussed to study the effect of bismuth oxide on the IR spectra of these glasses. We have studied the effect of gamma irradiation on $\text{BaO--Na}_2\text{O--B}_2\text{O}_3\text{--SiO}_2$ glasses earlier [18] and this work is in continuation of the previous one.

2. Experimental details

The glasses in the system $x\text{Bi}_2\text{O}_3\text{--}15\text{ Na}_2\text{O--}(70-x)\text{ B}_2\text{O}_3\text{--}15\text{ SiO}_2$ with $x = 5, 10, 15, 20$ and 25 (mol%) were prepared by conventional melt-quenching method. Analytic reagents Bi_2O_3 (99.99%), B_2O_3 (99.99%), SiO_2 (99.99%) and Na_2CO_3 (99.99%) were used as raw materials for 30 g batch. The chemical data for the constituent oxides is shown in Table 1. Melting was carried out in alumina crucibles using an electric furnace in the temperature range $1050\text{--}1150\text{ }^\circ\text{C}$ for one hour. The homogenized melts were then poured in disc shaped preheated stainless steel moulds for the required dimensions. The prepared samples were cooled at a rate of $25\text{ }^\circ\text{C h}^{-1}$ to room temperature using a muffle furnace. The glasses thus obtained were brown in colour.

All the glass samples were then finely powdered using a clean agate mortar pestle. Samples were subjected to a series of five radiation doses of 0.1 kGy , 1 kGy , 5 kGy , 15 kGy and 60 kGy using ^{60}Co source of gamma radiation at IUAC, New Delhi. The glass samples were irradiated for required time interval to achieve the desired overall dose. The dose rate was 7.27 kGy h^{-1} . All the powdered glass samples were wrapped in butter paper and then packed in polythene sachets before irradiation. The amorphous nature of these glass samples was confirmed by X-ray diffraction (XRD) using Bruker Axs diffractometer, Germany (Model D8 Advanced), with $\text{Cu K}\alpha$ radiation of wavelength $\lambda = 1.5406\text{ \AA}$. Fourier transform infrared (FT-IR) spectra of all the powdered samples before and

after irradiation were recorded at room temperature in the range $4000\text{--}400\text{ cm}^{-1}$ using a Thermo Nicolet 380 spectrometer using the conventional KBr pellet technique.

3. Results and discussion

The XRD spectra of the unirradiated glass samples Bi1-5 is shown in Fig. 1. The broad humps appearing in the spectra of all the glass samples indicate the amorphous nature of these samples. The IR absorption spectra of the bismuth borosilicate glasses i.e. Bi1 (5 mol%), Bi2 (10 mol%), Bi3 (15 mol%), Bi4 (20 mol%) and Bi5 (25 mol%) before irradiation is shown in the Fig. 2. The FTIR spectra of the glasses is studied in the range $4000\text{--}400\text{ cm}^{-1}$ and the active vibrational IR bands assigned to $[\text{BO}_n, n = 3 \text{ and/or } 4]$, $[\text{BiO}_n, n = 3 \text{ and } 6]$ and $[\text{SiO}_4]$ structural units, are located in the mid infrared region, i.e. in the spectral range $1600\text{--}400\text{ cm}^{-1}$. It is observed that all the bismuth containing glasses show the prominent absorption bands in the following three regions.

- (1) $600\text{--}800\text{ cm}^{-1}$
- (2) $800\text{--}1200\text{ cm}^{-1}$
- (3) $1200\text{--}1600\text{ cm}^{-1}$

Borosilicate glass is a composite glass which consists of structural units like trigonally coordinated boron (BO_3), tetrahedrally coordinated boron (BO_4) and silicon (SiO_4) structural units [19]. Bi_2O_3 containing glasses have four fundamental vibrations in the IR spectral regions at 830 cm^{-1} , 620 cm^{-1} , 450 and 350 cm^{-1} . The results of IR spectra in the present investigation are interpreted using the method given by Tarte [20,21] and Condrate [22,23] where the experimental datas are compared with the results obtained for the crystalline samples. To study the effect of gamma rays and effect of bismuth oxide on the glass system, the absorption spectra of each composition before and after irradiation is discussed separately below.

3.1. Bi1 glass

3.1.1. Before irradiation

The IR absorption spectrum of the unirradiated Bi1 glass containing 5 mol% Bi_2O_3 is illustrated in Fig. 2. The unirradiated glass reveals three clear and distinct regions containing several absorption bands extending from the beginning of the measurements at 400 cm^{-1} up to 4000 cm^{-1} . Bi_2O_3 in binary bismuth borate glasses can share in the structure in three different ways; it creates a four coordinated state by giving part of its oxygen to the boron,

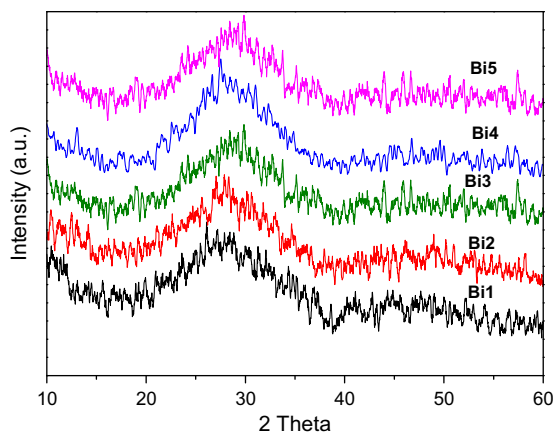


Fig. 1. XRD spectra of Bi1-5 glass samples before irradiation.

Table 1
Chemical composition of Bi1, Bi2, Bi3, Bi4 and Bi5 samples in mol%.

Composition	Bi_2O_3	Na_2O	B_2O_3	SiO_2
Bi1	5	15	65	15
Bi2	10	15	60	15
Bi3	15	15	55	15
Bi4	20	15	50	15
Bi5	25	15	45	15

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