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Research Article

INFRARED SPECTROSCOPIC ANALYSIS OF γ -IRRADIATED ZnO-BaO-B₂O₃-SiO₂ GLASSES

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ABSTRACT

New series of borosilicate glasses containing ZnO and BaO as network modifiers with compositions of $x\text{ZnO}-2x\text{BaO}-1/2(1-3x)\text{B}_2\text{O}_3-1/2(1-3x)\text{SiO}_2$ (x varying from 0.10 to 0.21 mole fraction) were prepared by the conventional melt quench technique. Structural properties have been determined before and after successive γ -ray irradiation at 2.5 kGy dose using, Fourier Transform Infrared (FTIR) spectroscopic techniques to elucidate the effect of γ -irradiation on these glasses.

Key Words:

Glasses, FTIR Spectroscopy, Borosilicate, Gamma Irradiation.

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INTRODUCTION

In modern world of technology, borosilicate oxide glasses have wide range of applications in the field of optics, electronic industry, glass solders for sealing CTV bulbs, IC packages, nuclear waste management, shielding materials, etc.¹⁻⁴. Recently, glasses containing barium oxide have attracted considerable attention due to their interesting properties leading to different applications. Oxide glasses containing significant concentrations of transition metal ions or low mobility metal ions such as Zn²⁺ or Ba²⁺ exhibit electronic conductivity and may be regarded as high resistivity semiconductors⁵. Borosilicate glass formulations adopted worldwide for immobilization of high-level radioactive liquid waste (HLW). Prado *et al.*⁶ subjected the glasses used for nuclear waste immobilization to high levels of radiation in order to study the effect on their physicochemical properties. Glasses containing barium are also proved to be good radiation shielders^{7, 8}. The radiation shielding properties of barium-borate and barium borate fly-ash glasses have recently been reported by Singh *et al.*⁹. Hamilton *et al.*¹⁰ considered borosilicate glasses containing barium to be the base glasses for the large family of barium crown optical glasses. The knowledge of the glass structure before and after irradiation is a prerequisite for understanding the structural evaluation of glasses under long term irradiation¹¹. The investigation of radiation effects leads

to a better understanding of the intrinsic structure as well as the alterations, which results from the interaction with radiation.

Experimental Details

Glass samples of compositions $x\text{ZnO}-2x\text{BaO}-(1-3x)/2\text{B}_2\text{O}_3-(1-3x)/2\text{SiO}_2$ with $x=0.1 - 0.21$ mole fraction prepared by melt quench technique at temperature range 1000-1250^oC are shown in Table 1. The procedure for the preparation of glass samples has been reported elsewhere¹². The raw materials lithium oxide, barium oxide, boric oxide and silicon oxide of analar grade were obtained from Aldrich Chemical Company.

Table 1 Nominal composition (mole fraction) of glass samples.

Sample No.	ZnO	BaO	B ₂ O ₃	SiO ₂
BBSi1	0.100	0.200	0.350	0.350
BBSi2	0.130	0.260	0.305	0.305
BBSi3	0.170	0.340	0.245	0.245
BBSi4	0.210	0.420	0.185	0.185

The Infrared Spectra of the glasses were investigated between 400 and 4000 cm^{-1} by the KBr technique using a Shimadzu FTIR – 8700 spectrometer.

All the samples were irradiated using ⁶⁰Co radioisotope. The dose rate was 1.77 Gy/min. The glass samples were irradiated for the necessary time interval to achieve the desired overall dose. Total dose was 2.5 kGy.

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RESULT AND DISCUSSION

Effect of glass composition

FTIR Spectroscopy

It is generally accepted that the building blocks of the silicate glasses the SiO₄ tetrahedra are linked at the vertices to form a network, while various modifiers such as alkali and alkaline earth ions occupy interstices in the networks. Some oxygen atoms are bonded to two silicon atoms and are called bridging oxygen atoms and some are bonded to one silicon only. Vitreous pure borate consists of only boroxol rings in its structure forming *sp*² trigonally coordinated structure. However, the addition of modifiers changes the coordination. Borosilicate glasses therefore assumed to have SiO₄ tetrahedral and BO₃ trigonal units in its structure. Addition of modifiers further adds the complexity in its network structure. FTIR spectra for zinc barium borosilicate glasses are shown in Fig. 1. The assignment of different peaks as studied in the literature¹⁵ for silicate and borate structural chains are listed in Table 2.

For qualitative analysis, the spectra are divided into following three regions:

1. 600 – 800 cm⁻¹
2. 800 – 1200 cm⁻¹
3. 1200 – 1600 cm⁻¹

Table 2 Positions and assignments of the observed infrared absorption bands.

Peak Position (cm ⁻¹)	Assignment
Borate Chains	
A – BO ₃ stretching	
1420-1550	B-O bond vibrations
1550-1400, 1250	B-O vibrations of various borate groups
1220-1250	Boroxol rings, tri-, tetra- and pentaborate groups pyro- and other borates
B – BO ₄ stretching	
1050	Tri-, tetra- and pentaborate
900-1000	Diborate
880	Tri-, tetra-, and pentaborate
760-770, 690-730	Oxygen bridging between tetra- and trigonal boron atoms oxygen bridges between trigonal atoms
550	In-plane bending vibrations of orthoborate units
3200-3700	Presence of molecular water and B-OH in the sample.
Silicate Chain	
1065-1095	Si-O-Si anti-symmetric stretching of bridging oxygen within the tetrahedral
940-970	Si-O ⁻ stretching with 1 non-bridging oxygen
860-940	Si-O ⁻ stretching with 2 non-bridging oxygens
750-820	Si-O-Si symmetric stretching of bridging oxygen
460-510, 600	Si-O-Si and O-Si-O bending modes (V ₄)
Other groups	
1640	Molecular water
3000-3750	Hydrogen, molecular water, silane group (SiOH)

The spectra in region 1 has small band at about 700 cm⁻¹ which is more prominent in sample 3 (Fig. 1), the region 2 is characterized by intense absorption band centered around 1000cm⁻¹ while region 3 has a band centered at around 1460cm⁻¹. Fig. 1 shows the change in the position and intensity of these bands with composition. The band at 1020 cm⁻¹ is a characteristic absorption of BO₄ structural units and the band observed at 1460 cm⁻¹ is a characteristic absorption of BO₃ structural units in the glass. The bands appearing in the region 800 – 1100 cm⁻¹ are due to B-O bond stretching of tetrahedral BO₄ units, which are overlapped with silicate bands. These bands are assigned to the vibration of tetraborate (triborate and

pentaborate) groups of BO₄ units^{13, 14}.

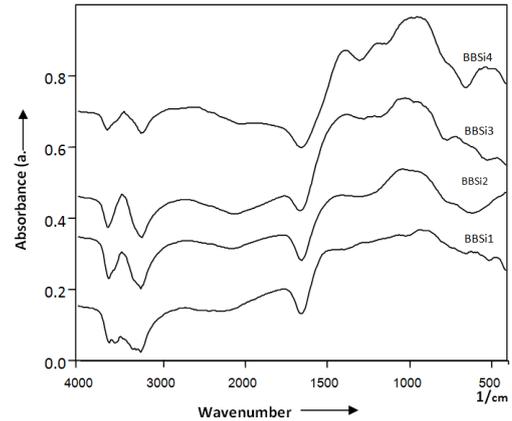


Figure 1 The IR spectra of ZnO-BaO-B₂O₃-SiO₂ before irradiation

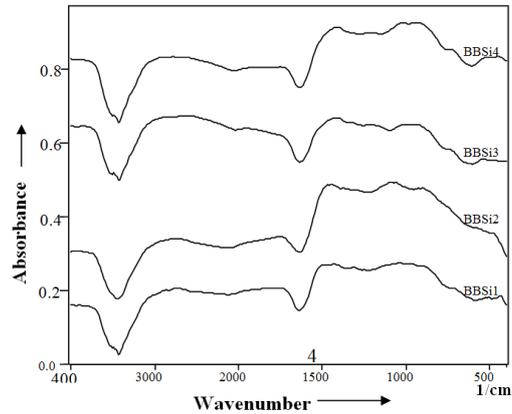


Figure 2 The IR spectra of ZnO-BaO-B₂O₃-SiO₂ after irradiation

In the present system of zinc barium borosilicate glasses, the sub band analysis is not reliable to separate these bands as they are too overlapped. However, in view of the nominal compositions of the glasses, the fraction of fourfold coordinated boron can be supposed to correspond with its maximum value. According to literature, by ¹¹B NMR spectroscopy the addition of SiO₂ in borate glass stabilize the formation of the BO₄ groups, as a result all the negative charges balancing the cations are mainly associated with BO₄ groups while SiO₄ groups have four bridging oxygen atoms^{16, 17}. In the studied glasses, the presence of such SiO₄ units is confirmed by the band at about 1200 cm⁻¹, assigned to Si-O stretching of the SiO_{4/2} units¹⁸.

Irradiation effects

A number of fundamental problems of radiation physics of glasses are not yet solved and mostly exist in the experimental phases. Silicate, borate and phosphate glasses have a certain level of radiation-optical stability (patents: East Germany No. 2559263, U. S. No. 5057464, Japan Nos. 45-27426 and 55-429646, Bulgaria No. 25961) along with high linear attenuation factors^{19, 20}. The effects of radiation on the glasses have been summarized by Friebele⁴⁴ to fall into three categories:

1. Atomic displacement by momentum and energy transfer
2. Ionization and charge trapping and
3. Radiolytic or photochemical effects.

However, the relative contribution to the net damage depends on the type and energy of the radiation, as well as on the total dose.

FTIR Spectroscopy

FITR spectra of zinc barium borosilicate glasses after irradiation are shown in FIG. 2. By comparing the IR spectra of before and after irradiation of samples, it is quite clear that the band centered at around 1000 cm^{-1} has been reduced in its intensity to very large extent and there is an increase in the intensity of band centered at around 1400 cm^{-1} which clearly indicates the transfer of tetrahedrally coordinated to trigonally coordinated network structure which is also supported by the density results.

CONCLUSIONS

Stable glasses using B_2O_3 and SiO_2 as network former, ZnO and BaO as network modifiers have been achieved by the melt-quench technique. A significant change in the structural properties has been observed with systematic variation in composition and with the effect of γ -irradiation on the prepared glasses. Analysis of compositional dependence revealed that the addition of ZnO and BaO transfers the network structure from the trigonal to tetrahedral coordination and hence more stable structure formation to a particular extent of MO. Beyond this extent (at $x=21$ mole fraction), formation of NBOs have been observed. With γ -irradiation, weakening of all the bands observed shows the effect of these radiations on glass structure. However, the variation of all the parameters for sample 2 (in both the series) is very less which shows the stability of this composition to γ -rays of present dose.

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