

## Research Article

# Preparation and Optical Investigations of $[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-[2\text{SiO}_2\text{B}_2\text{O}_3]-[\text{CeO}_2]$ Glasses

Chandkiram Gautam, Anod Kumar Singh, and Abhishek Madheshiya

*Advanced Glass and Glass Ceramic Research Laboratory, Department of Physics, University of Lucknow, Lucknow 226007, India*

Correspondence should be addressed to Chandkiram Gautam; [gautam\\_ceramic@yahoo.com](mailto:gautam_ceramic@yahoo.com)

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We are reporting synthesis and structural and optical investigation of strontium bismuth titanate borosilicate glasses with addition of one mole percent cerium oxide ( $\text{CeO}_2$ ). Glasses were synthesized by conventional rapid melt quench method. XRD studies of the glass samples confirm the amorphous nature. Infrared absorption spectra various strontium bismuth titanate borosilicate glass samples having glass system  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$  ( $x = 0.0, 0.1, 0.2, 0.4$ ) were recorded over a continuous spectral range from 400 to  $4000\text{ cm}^{-1}$ . IR spectra were analyzed to determine and differentiate of various vibrational modes in the structural change. Raman spectroscopy of all glass samples was also carried out in the wave number range from 200 to  $2000\text{ cm}^{-1}$ .

## 1. Introduction

Glasses are defined as inorganic product of fusion which has been cooled to a rigid condition without crystallization [1]. The main distinction between glass and crystal is the presence of long range order in the crystal structure. For many years glasses containing transition metal ions have attracted attention because of their potential applications in electrochemical, electronic, and electrooptics devices [2]. The most important fact about the glass is that it is amorphous, transparent, and brittle in nature. The glass has been used as an engineering material since ancient time. But because of rapid progress made in the glass industry in recent time, the glass has come out as the most versatile engineering material of the modern time. With help of techniques developed in the glass industry, the glass of any type and quality can be produced to suit the requirements of different industries. Glass has become established as commercially important material in the field of consumer product, vacuum tube envelopes, telescope mirror blanks, radomes for the aerospace industry, and protective coating for metal [3].

The study of various oxide glasses has received considerable attention due to their structural property [4, 5].

These glasses have wide application in the field of electronics, nuclear and solar energy technologies, and acoustic-optics device [6–10]. The IR spectroscopy is one of the most common spectroscopic techniques used by organic and inorganic chemists. Simply it is the absorption measurement of different IR frequencies by sample positioned in the path of an IR beam. The main goal of IR spectroscopic analysis is to determine the chemical functional group of the samples. Using various samples accessories, IR spectrometer can accept a range of samples such as gasses, liquids, and solids. The IR spectroscopy is an important and popular tool for structural elucidation and compound identification [11]. Raman spectroscopy is also an essential tool for characterization of structure, environment, and dynamics of glassy materials. In the Raman spectroscopy, the nature of light matter interaction is not the same as IR spectroscopy and fundamental difference between the two processes determines the selection rule, which control Raman or IR activity of normal mode of the vibrations. Interaction of IR radiation with normal mode of vibration only occurs when the electric field radiation oscillates with the same frequency as instant dipole caused by atomic vibration. A normal vibration can be IR active only if change in the dipole moment of the vibration occurs

and is one photon process, as only photon is absorbed [12]. Therefore, IR spectra give more additional information than Raman spectra by which the symmetry of normal mode of vibration of the molecules and crystal lattice are determined [13–15]. More recently, a lot of work has been done by Gautam et al. on the optical properties of lead strontium/barium strontium titanate borosilicate glasses [16–20]. But the glass ceramics of these glasses have potential applications in the designing and manufacturing of microelectronic components such as barrier layer capacitors to store high energy, cryogenic temperature sensors, capacitance thermometer, and dielectric bolometer [21].

In this research paper we are reporting our results on  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$  ( $x = 0.0, 0.1, 0.2, 0.4$ ) glass samples doped with one mole percent of  $\text{CeO}_2$  for the characterization of XRD, infrared, and Raman spectroscopy. These characterizations give the valuable information about the formation of bonding network between different oxide groups. These results are not reported anywhere so far. Therefore, authors have been taking an attempt to explore the above mentioned characterization on this glass system.

## 2. Experimental Methods

**2.1. Sample Preparation.** High purity analytical reagent grade chemicals  $\text{SrCO}_3$  (Himedia 99%),  $\text{Bi}_2\text{O}_3$  (Himedia 99.999%),  $\text{TiO}_2$  (Himedia 99%),  $\text{H}_3\text{BO}_3$  (Himedia 99.8%),  $\text{SiO}_2$  (Himedia 99.5%), and  $\text{CeO}_2$  (Himedia 99%) were used for the preparation of various glass samples in the glass system  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$  ( $x = 0.0, 0.1, 0.2, 0.4$ ). Appropriate amounts of raw materials, as per the composition of glasses, were properly weighed and mixed in an agate mortar using acetone as mixing medium and dried the powder in an oven at  $100^\circ\text{C}$  for 20 minutes. The glass batches of 20 grams were melted in high grade alumina crucible in open air atmosphere using a programmable electric furnace having silicon carbide heating elements. The melting temperatures for different compositions were in the range from  $1250$  to  $1400^\circ\text{C}$ . The melt was maintained at the casting temperature  $1400^\circ\text{C}$  in the furnace for 30 minutes for refining and homogenization. The melt was poured into a steel mould and pressed by thick aluminum plate and then immediately transferred into preheated muffle furnace for annealing at temperature  $450^\circ\text{C}$  for 4 hours to remove the residual stresses due to temperature gradient, which is produced by rapid cooling of the glass melt. The glasses were cooled to room temperature within the furnace after annealing. Nomenclature of prepared glass samples contains 4 letters represented as SBTC; first three letters refer to strontium titanate borosilicate and C indicates the content of cerium oxide used as dopant.

**2.2. X-Rays Diffraction Measurements.** XRD of powder glass samples was carried out using a Rigaku Miniflex-II X-ray diffractometer using  $\text{Cu-K}_\alpha$  radiation to check the amorphous state of the prepared glass samples.

**2.3. Density Measurements.** The density strongly depends on the compositions and structure of glass samples. Density of glass samples was determined by liquid displacement method by using Archimedes principle [22]. Distilled water was used as the liquid medium. The density of glass samples was calculated using the formula

$$\rho = \frac{(W_2 - W_1)}{(W_4 - W_1) - (W_3 - W_2)}, \quad (1)$$

where  $\rho$  = density (gram/cc),  $W_1$  = weight of empty specific gravity bottle (gram),  $W_2$  = weight of specific gravity bottle with sample (gram),  $W_3$  = weight of specific gravity bottle with sample and distill water (gram), and  $W_4$  = weight of specific gravity bottle with distill water (gram).

**2.4. Molar Volume Calculation.** Molar volume was calculated using the following relation:

$$V_M = \frac{\sum X_i M_i}{\rho}, \quad (2)$$

where  $M_i$  is the molecular weight of the  $i$ th component and  $X_i$  is the molar fraction of the  $i$ th component [5].

**2.5. Infrared Spectroscopy.** The powder glass samples were mixed with KBr powder and pressed as pellets. Then, these pellets are used as samples for recording the IR spectra. The IR spectra of  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$  ( $x = 0.0, 0.1, 0.2, 0.4$ ) glass samples are carried out using JASCO FT/IR-5300 in the wave number range  $400-4000\text{ cm}^{-1}$  at room temperature.

**2.6. Raman Spectroscopy.** The powdered glass samples are used as in the wave number range from  $200$  to  $1500\text{ cm}^{-1}$ . Micro-Raman setup (Renishaw, UK) equipped with grating of 1800 lines/mm and Olympus (Model MX-50) A/T was attached with spectrometer which focuses the laser light on the sample and collect the scattered laser light at  $1800$  by scattering geometry. The  $15.4\text{ nm Ar}^+$  laser was used as an excitation source and GRAM-32 software was used for data collection.

## 3. Results and Discussion

**3.1. XRD Analysis of Glass Samples.** The XRD patterns of both glass samples SBTC0.0 and SBTC0.1 are shown in Figure 1. Transparent and thin glasses of good quality in the composition  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1\text{CeO}_2$ . ( $0.0 \leq x \leq 0.4$ ) in molar ratio were synthesized by a conventional melt quench technique. These XRD patterns exhibit a broad diffuse scattering at different angles instead of crystalline peaks, confirming a long range structural disorder characteristic of amorphous glassy network.

**3.2. Density and Molar Volume Analysis of Glass Samples.** The variation in the values of density and molar volume of the glass samples has been listed in Table 1. It is well

TABLE I: Density and molar volume of glass samples in the system  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$ .

Composition $x$	Glass sample code	Glass composition (mole %)						Density (gm/cc)	Molar volume ( $\text{cm}^3/\text{mole}$ )
		$\text{SrCO}_3$	$\text{Bi}_2\text{O}_3$	$\text{TiO}_2$	$\text{SiO}_2$	$\text{B}_2\text{O}_3$	$\text{CeO}_2$		
0.0	SBTC0.0	38.93	0	21.07	19.25	19.75	1	2.51	39.761
0.1	SBTC0.1	30.79	10.69	18.52	19.25	19.75	1	2.85	48.781
0.1	SBT0.1	30.80	10.70	18.51	19.75	20.25	1	2.69	50.639
0.2	SBTC0.2	24.42	19.07	16.51	19.25	19.75	1	2.98	55.325
0.4	SBTC0.4	15.06	31.37	13.58	19.25	19.75	1	3.10	66.05

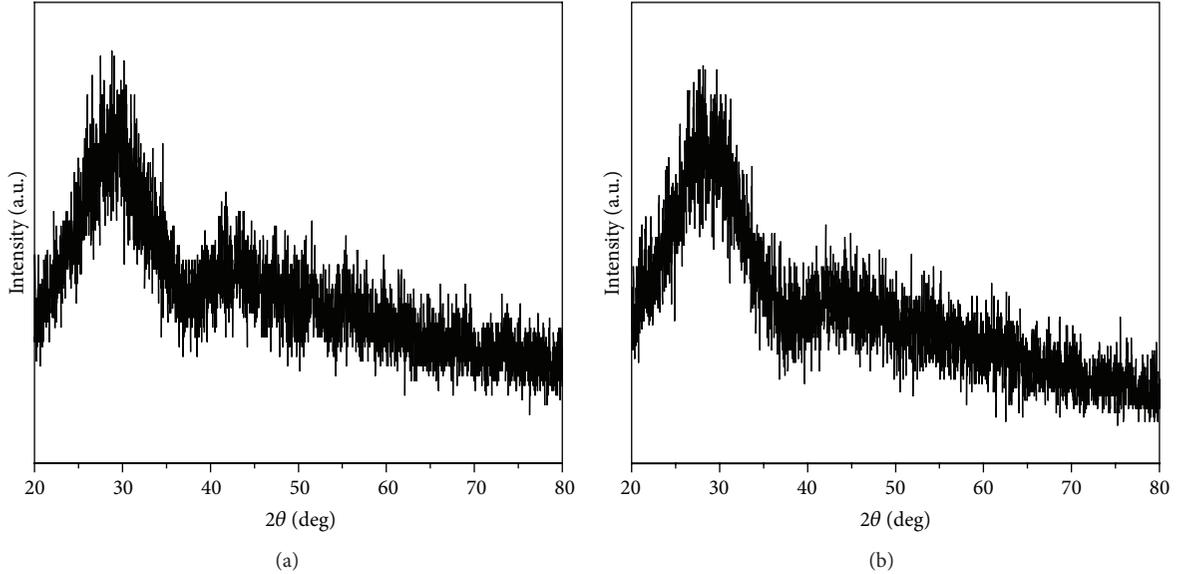


FIGURE 1: XRD patterns of the glass samples (a) SBTC0.0 and (b) SBTC0.1.

known that density is an important tool to explore the structural compactness/softening, the change in geometrical configurations, coordination number, cross-link density, and so forth [23]. Thus it helps to reveal the degree of change in the structure with change in composition in any glass systems. The value of density of strontium bismuth titanate borosilicate glass samples has been found between 2.51 and 3.10 gm/cc. It increases with increasing concentration of  $\text{Bi}_2\text{O}_3$  and also with the doping of  $\text{CeO}_2$  in the strontium bismuth titanate borosilicate glasses. This may be due to high density of  $\text{Bi}_2\text{O}_3$  (8.90 gm/cc) and  $\text{CeO}_2$  (7.65 gm/cc). The variation of density versus bismuth concentration is shown in Figure 2 and the variation of the molar volume with  $\text{Bi}_2\text{O}_3$  mole percent is shown in Figure 3. From Figure 3, it is clear that, by increasing  $\text{Bi}_2\text{O}_3$ , the density and molar volume are increasing. This is due to the replacement of all  $\text{SiO}_2$  and  $\text{B}_2\text{O}_3$  (molecular weight is 60.084 and 61.83 g/mol, resp.) with  $\text{Bi}_2\text{O}_3$  (molecular weight is 465.959 gm/mol) [24]. The molar volume which is defined as the volume occupied by the unit mass of glass increases from 39.761 to 55.325 as  $\text{Bi}_2\text{O}_3$  content increases from 0 to 19.07 mol% [25]. It is reported that the molar volume depends on ionic radius of the glass modifier. If the ionic radius of modifier ions (ionic radius of  $\text{Bi}^{3+}$  is  $1.03 \text{ \AA}$ ) is larger than the interstices of the glass network former

(ionic radius of  $\text{Si}^{4+}$  and  $\text{B}^{3+}$  is  $0.40$  and  $0.27 \text{ \AA}$ , resp.), their attraction to the oxygen ion can lead to an augment in the size of the interstices and result in an enhancement of molar volume. Density and molar volume of strontium bismuth titanate borosilicate glass samples almost linearly vary with the variation of  $\text{Bi}_2\text{O}_3$ .

**3.3. Infrared Spectroscopy Analysis.** IR spectra occur due to change in dipole moment of the molecules. It involves the twisting, bending, rotating, and vibrational motion in molecules. In the borate glasses,  $\text{B}_2\text{O}_3$  is the basic glass former because of its higher bond strength, lower cation size, and small heat of fusion. In these glasses,  $\text{B}^{3+}$  ions are triangularly coordinated by oxygen and  $\text{B}_2\text{O}_3$  units are corner bonded in a random configuration [26]. The structure of borate glasses heavily depends on the cooling rate of the melt through the range of glass transition temperature [27]. Bismuth oxide and strontium oxide can enter the glass both as a network former and also as a network modifier. Due to this nature of  $\text{BiO}$  and  $\text{SrO}$ , the structure of these glasses is expected to be different from that of alkali borate, phosphate, and silicate glasses. The IR spectra of various glass samples of strontium bismuth titanate borosilicate glass doped with  $\text{CeO}_2$  are shown in Figure 4. IR spectra of glass sample consist

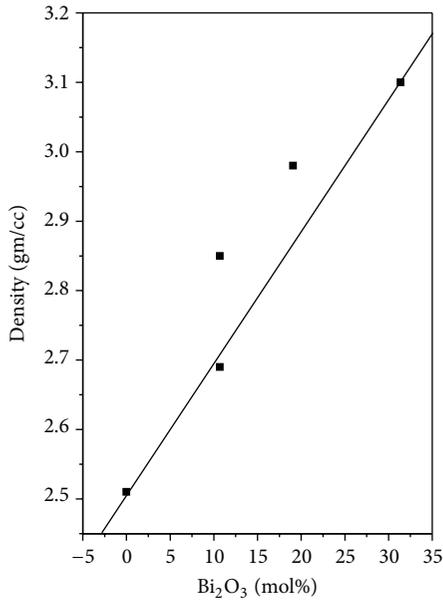


FIGURE 2: Variation of density versus Bi content.

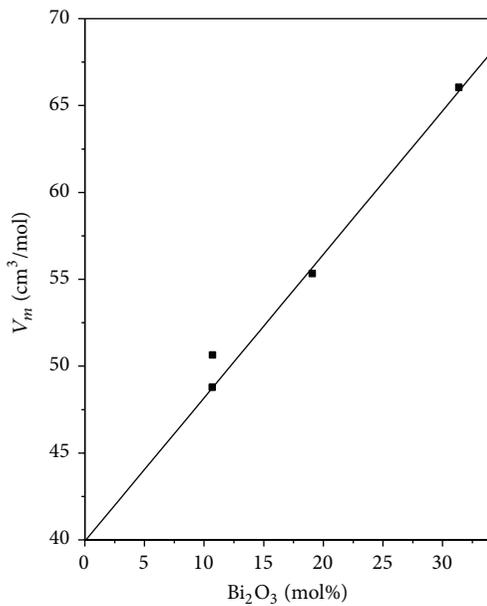
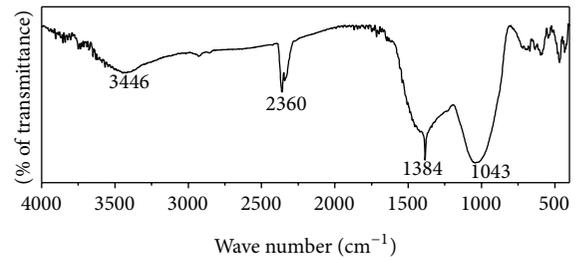
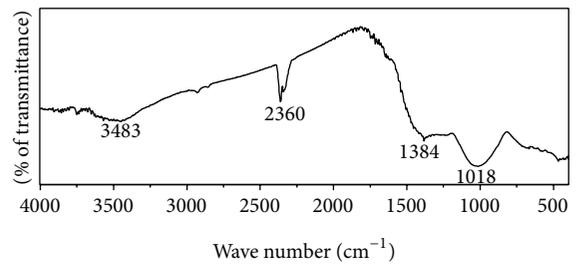


FIGURE 3: Variation of molar volume versus content of Bi.

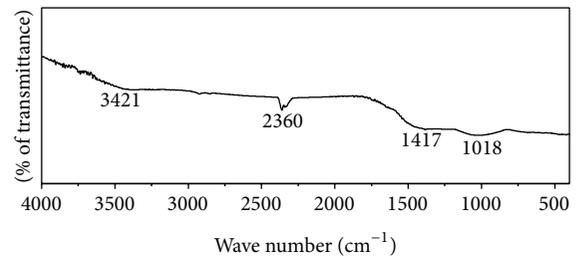
of broad, medium, and sharp absorption bands in different region which lie between 400 and 4000  $\text{cm}^{-1}$ . These bands are strongly affected due to the variation of compositional change because the positions of some absorption bands are shifted due to the compositional variations. Wave numbers and percentage of transmittance of different absorption peaks for all the glass samples have been listed in Table 2. The broad bands are exhibited in the oxide spectra, most probably due to the combination of high degeneracy of vibrational state, thermal broadening of the lattice dispersion bands, and mechanical scattering from powder samples. It is known that boron



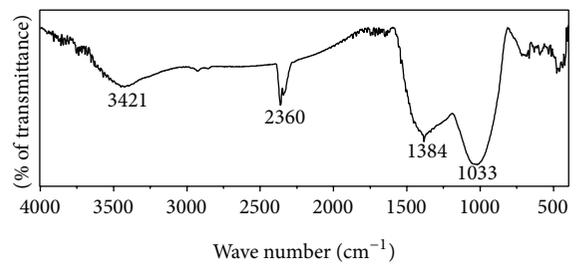
(a)



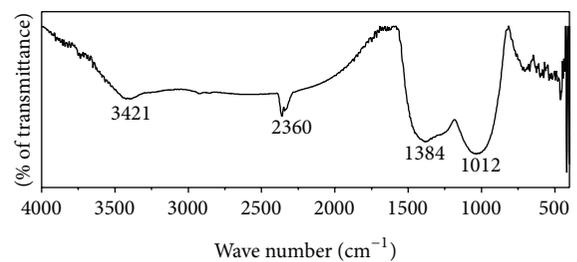
(b)



(c)



(d)



(e)

FIGURE 4: IR pattern of glass samples (a) SBTc0.0, (b) SBTc0.1, (c) SBTc0.2, (d) SBTc0.4, and (e) SBTc0.4.

exhibits more than one stable configuration. The addition of alkali or alkaline earth oxides to the borate network changes the boron coordination from three to four. This results in the formation of di-, tri-, tetra-, and pentaborate groupings.

TABLE 2: Peak positions in IR spectra of different glass samples in the system  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$ .

Glass sample code	Wave number of different absorption peaks ( $\text{cm}^{-1}$ )				
	1	2	3	4	5
SBTC0.0	3446	2923	2360	1384	1043
SBTC0.1	3483	2927	2360	1384	1018
SBT0.1	3421	—	2360	1417	1018
SBTC0.2	3421	2927	2360	1384	1033
SBTC0.4	3421	2923	2360	1384	1012

TABLE 3: Peak positions in Raman spectra of different glass samples in the glass system  $[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$ .

Glass sample code	Raman position band ( $\text{cm}^{-1}$ )	
	1	2
SBTC0.0	348	1713
SBTC0.1	288	—
SBT0.1	347	—
SBTC0.2	250	1701
SBTC0.4	348	1747

Due to boron anomaly, addition of modifier oxides forms  $\text{BO}_4$  units. The first absorption peak lies in the wavenumber range  $3421\text{--}3483\text{ cm}^{-1}$ . This peak is broad in bismuth rich glass samples while it is sharp in strontium rich glass samples, which is mainly due to hydroxyl or water group present in the glass samples and attributed to the O–H stretching vibration. Absorption peak number 2 is absent in cerium free composition while it is present in all other compositions and these absorption bands are attributed to the formation of hydrogen bonding [28–30]. Absorption peak number 3 is constant in all compositions. The absorption bands in the wave number range from  $1417$  to  $1384\text{ cm}^{-1}$  are observed due to asymmetric stretching relaxation of B–O bonds of trigonal  $\text{BO}_3$  units [31, 32]. The bands around  $1000\text{ cm}^{-1}$  (peak number 4) are attributed due to stretching vibration of B–O–Si linkage [33].

**3.4. Raman Spectroscopy Analysis.** Raman spectroscopy is one of the powerful techniques to investigate the structure of the material. Raman spectra of various glass samples in glass system  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]-39[2\text{SiO}_2\text{B}_2\text{O}_3]-1[\text{CeO}_2]$  ( $x = 0.0, 0.1, 0.2, 0.4$ ) doped with cerium oxide are shown in Figures 5(a)–5(e). Peaks positions of Raman bands have been listed in Table 3. Raman spectra of these glasses indicate that two peaks at different position lie at the wave number range from  $250$  to  $348\text{ cm}^{-1}$  and from  $1701$  to  $1747\text{ cm}^{-1}$ . Figure 5(a) shows Raman spectra of bismuth free glass sample SBTC0.0 ( $x = 0$ ). Band formation at lower wave number side ranging from  $250$  to  $348\text{ cm}^{-1}$  in all Raman spectra of the glass samples is attributed to metaborate groups and symmetric

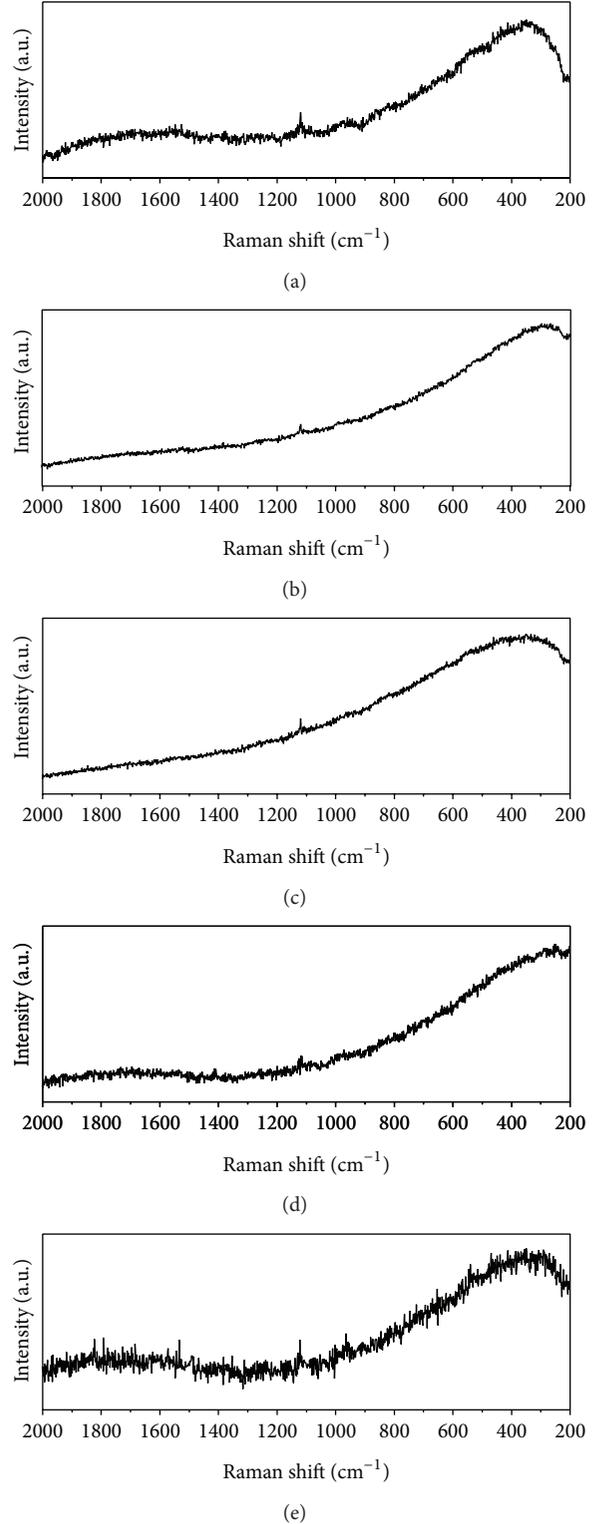


FIGURE 5: Raman spectra of glass samples (a) SBTC0.0, (b) SBTC0.1 (c) SBT0.1, (d) SBTC0.2, and (e) SBTC0.4.

breathing vibration of six member rings with one or two  $\text{BO}_3$  triangles replaced by  $\text{BO}_4$  tetrahedra [34]. These bands also occurred due to bending vibrations of planar  $\text{BO}_3$  triangles in random network [35]. The positions of the bands have

little bit changed with the variation of compositions. Peak at low wave number  $250\text{ cm}^{-1}$  was observed due to  $\text{CeO}_2$  and  $\text{KO}_2$  (metallic cation) bonding vibration in bismuth free glass sample. The high intensity peaks are observed in the wave number range  $1701\text{--}1747\text{ cm}^{-1}$ . This peak is absent in the glass compositions SBTC0.1 and SBT0.1. These peaks occur due to symmetric mode, where there is no change in dipole moment; therefore polarizability fluctuates significantly.

#### 4. Conclusion

Bulk transparent glasses were prepared by melt quench technique. IR spectra of  $60[(\text{Sr}_{1-x}\text{Bi}_x)\text{TiO}_3]\text{--}39[2\text{SiO}_2\text{B}_2\text{O}_3]\text{--}1[\text{CeO}_2]$  ( $x = 0.0, 0.1, 0.2, 0.4$ ) glasses occur due to vibrational mode of the borate network of asymmetric stretching vibration B–O bond of trigonal  $\text{BO}_3$  units, molecular water, and OH bonding, B–O–B, B–O–Si linkages. The addition of  $\text{CeO}_2$  decreases the sharpness of the peaks and increases the broadness of the absorption peaks in their IR spectra. Raman spectra of all glass samples are attributed to the metaborate groups and symmetric breathing vibration of six member rings with one or two  $\text{BO}_3$  triangles replaced by  $\text{BO}_4$  tetrahedra which give the best agreement with IR spectra. Density of the glass samples increases with increasing the concentration of  $\text{Bi}_2\text{O}_3$  and also with the doping of  $\text{CeO}_2$  in the SBT borosilicate glasses.

#### Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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