



Conductivity, dielectric, and structural studies of (30-x) SrO-xBaO-10Al₂O₃-45SiO₂-5B₂O₃-10Y₂O₃ (5 ≤ x ≤ 25) glasses

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Abstract

Composition of (30-x) SrO-xBaO-10Al₂O₃-45SiO₂-5B₂O₃-10Y₂O₃ (mol%) (5 ≤ x ≤ 25, in step of 5) has been synthesized using melt-quench route. The structural aspects of glasses were investigated by Fourier transform infrared (FTIR) spectroscopy. Electrical conductivity and dielectric properties of the glasses were also investigated with the help of impedance analyzer. With the increase of BaO content in these glasses, density has been found to increase due to higher density of BaO (5.72 g cm⁻³) than SrO (4.70 g cm⁻³). The highest value of electrical conductivity is observed 3.34 × 10⁻⁶ S cm⁻¹, where BaO and SrO are in the same ratio (15:15) mol%, i.e., 15BS glass. The conductivity curves show two distinct slopes except 10BS glass, indicating change in hopping mechanism. 10BS glass shows the highest thermal and frequency-independent dielectric constant, which could be used in microelectronic applications.

Keywords Glasses · Impedance analyzer · Dielectric properties · Electrical conductivity · Jonscher's power law

Introduction

Glasses emerge as an important class of materials due to their potential applications in the various fields such as electronic devices, electro-optic devices, thermo-mechanical sensors, telecommunications, and solid oxide fuel cells, reflecting windows and medical and aerospace industry applications. Glasses and glass ceramics are the most promising candidates due to their low dielectric constant, losses, and conducting properties as actuators and sensors. Many oxide glasses show semiconducting behavior endowing it with important technological applications such as memory optical switching devices as well as telecommunications.

The most important aspect of glasses is tailoring properties according to need and application with ease as compared to crystalline system. Basically, the variation in initial ingredient introduces considerable changes in the local structure of glasses due to the variation of inter-atomic forces bond angles and connectivity among various units. The degree of their

local structural units in the glasses highly depends upon the chemical nature as well as concentration of modifiers, former and intermediate oxides [1, 2]. Higher chemical/thermal stability, network connectivity, bond strength, smaller heat of fusion, and lower cation size make SiO₂ and B₂O₃ as excellent candidate for glass former compounds [3, 4]. On the other hand, intermediate oxides such as Al₂O₃, Y₂O₃, Cr₂O₃, ZrO₂, and TiO₂ can play significant dual role and thereby enhance optical and chemical properties of the glasses. The intermediate oxide can act as a glass modifier and glass former depending upon the glass composition, their coordination number, and field strength [5–7]. In our earlier study, Al₂O₃ that contained 40SiO₂-30SrO-20B₂O₃ glass showed higher thermal and chemical stability [2]. In addition to this, Al₂O₃ is also known for delaying devitrification of glasses. On the other hand, Y₂O₃ increases the glass transition temperature and thermal expansion of the glasses [6, 8].

Lara et al. [9] discussed the electrical behavior of glass ceramics in the system RO-BaO-SiO₂ (R = Mg, Zn). The electrical conductivity for Zn and Mg doped glasses was found to be (5.6 ± 0.3) × 10⁻⁹ and (6.3 ± 0.3) × 10⁻⁹ S cm⁻¹ with corresponding activation energies of 0.86 and 0.9 eV, respectively. The similar composition was examined by Pascual et al. [10] for the high temperature seal applications for SOFCs, and the electrical conductivity was obtained in the range (1.4–7.8) × 10⁻⁶ S cm⁻¹ at 950 °C along with the

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activation energies of 122–154 kJ mol⁻¹. Wang et al. [11] obtained high value of electrical resistivity (10^{12–13} Ω cm), low dielectric constant (5–6), and dielectric loss (10^{-2–10}-3) for the borosilicate glasses. Recently, our group also reported the dielectric and conduction behavior agriculture waste-derived glass and glass ceramics with low dielectric constant [12]. The low dielectric constant has also been observed for the B₂O₃-Li₂O-ZnO-TiO₂-V₂O₅ glass system with wide range independency on frequency and temperature [13]. Literature validates the presence of many reports based on the electrical conductivity and dielectric properties of the borosilicate glasses. However, limited reports are available, which examine the effect of ionic radii and field strength of Ba²⁺/Sr²⁺ on the electrical and dielectric properties of SrO/BaO yttrium aluminoborosilicate glasses. In the present study, a modifier (SrO) is replaced by another modifier (BaO), which is having higher polarizability and lower field strength than SrO. In addition to this, the presence of two modifiers gives unusual properties due to competition between them. Moreover, the uniqueness of the present study is to have two glass formers (SiO₂ + B₂O₃), two intermediate oxides (Al₂O₃ + Y₂O₃), and variable two modifiers (SrO + BaO). Lower dielectric constant with lower electric conductivity glasses can be used as substrate and in microwaves and microelectronics. Thus, it is worthwhile to investigate the physical, structural, electrical, and dielectric properties of the yttrium aluminoborosilicate based glasses for possible application in microelectronics.

Therefore, a glass series (30-*x*) SrO-*x*BaO-45SiO₂-10Al₂O₃-10Y₂O₃-5B₂O₃ (mol%) [$5 \leq x \leq 25$, in steps of 5] has been synthesized to study the effect of radii size differences as well as field strength of alkaline earth metal BaO as modifier on the electrical and dielectric properties of these glasses.

Experimental details

The glass composition chosen for the present study is given in the Table 1. Glass samples with composition (30-*x*) SrO-*x*BaO-45SiO₂-10Al₂O₃-10Y₂O₃-5B₂O₃ (mol%) [$5 \leq x \leq 25$, in steps of 5] were prepared using conventional melt-quench techniques. Batches (50 g) of the samples were prepared by taking required stoichiometric amounts of high purity (99.9%)

constituent oxides. The stoichiometric amounts of these constituents were mixed homogeneously in agate mortar pestle for 2–3 h and then melted at 1550 °C in recrystallized alumina crucible in high resistance furnace. The molten glass was quenched in air between the thick copper plates.

FTIR spectra of the glass powder were recorded at room temperature in the region 500–3500 cm⁻¹ using PerkinElmer-Spectrum-RF-1 FTIR spectrometer. For FTIR measurements, the 5 mg of glass powder was mixed with 20 mg of KBr in an agate mortar pestle. The spectrum of each glass sample was normalized to the spectrum of the blank KBr. By Archimedes principle, the density (ρ) of prepared glass samples was measured at room temperature with xylene as a buoyant. For the conductivity and dielectric measurements, the powder of glasses was pelletized. Using few drops of polyvinyl alcohol (PVA) as a binder were used, thereby compacting these powders in the hydraulic press at a pressure ~11 kN cm⁻¹. The pellets of 20 mm diameter with thickness 1–2 mm were prepared and sintered at 850 °C for 12 h at heating rate of 5 °C min⁻¹ in the pit furnace. The XRD pattern of sintered pellets was taken between angular ranges 20°–80° (at 2° min⁻¹) with Cu K α radiations ($\lambda = 1.54$ Å) using PANalytical X'Pert Pro MPD diffractometer.

DSC/TG (NETZSCH STA 449F3) analysis was done at a heating rate of 10 °C min⁻¹ up to 1100 °C in the nitrogen (N₂) environment to have an idea about the crystallization of the samples. The electrical conductivity and dielectric measurements of the platinum (Pt) coated pellets were taken with Solarton Impedance Analyzer (SI-1260). These pellets were coated by Pt on both the sides using JEOL Auto Fine Coater (JEC-3000FC) at 20 Å for 3 min. The measurements are performed at the heating rate of 10 °C min⁻¹ with temperature stability ± 1 °C, in the frequency and temperature range of 10 Hz to 2 MHz and 50–600 °C, respectively.

Results and discussion

Density and molar volume analysis

The densities, ionic concentration of alkaline earth ions, inter ionic concentration, and molar volume of the as-quenched samples are given in Table 2. The molar volume (V_m) of the samples is calculated by the following relation:

$$V_m = \frac{M}{\rho} \quad (1)$$

where M is molar weight, and ρ is density of the glass. Ionic concentration of alkali earth metal ion (N) and their inter-ionic distance (R) can be calculated by the following relations:

$$N = (6.023 \times \text{mol\% of cation} \times \text{valency}) / V_m \quad (2)$$

Table 1 Glass compositions (mol%) with their labels

Sample label	BaO	SrO	SiO ₂	B ₂ O ₃	Al ₂ O ₃	Y ₂ O ₃
5BS	5	25	45	5	10	10
10BS	10	20	45	5	10	10
15BS	15	15	45	5	10	10
20BS	20	10	45	5	10	10
25BS	25	5	45	5	10	10

Table 2 Densities, molar mass, molar volume of the present glasses, and the ionic concentration of alkaline earth ions along with their ionic distance

Sample label	Density, ρ (g cm ⁻³)	Molar mass (M)	Molar volume, V_m (cm ³ mol ⁻¹)	Ionic conc., N ($\times 10^{21}$ cm ⁻³)		Inter-ionic distance, R (Å)		T_c (°C)
				N_{Ba}	N_{Sr}	R_{Ba}	R_{Sr}	
5BS	3.308	96.866	29.27	2.05	10.28	0.786	0.459	955
10BS	3.550	99.351	27.98	4.41	8.83	0.609	0.483	
15BS	3.558	101.836	28.62	6.23	6.23	0.543	0.543	964
20BS	3.566	104.321	29.25	8.23	4.11	0.494	0.623	
25BS	4.701	106.807	22.72	10.60	2.65	0.455	0.722	974

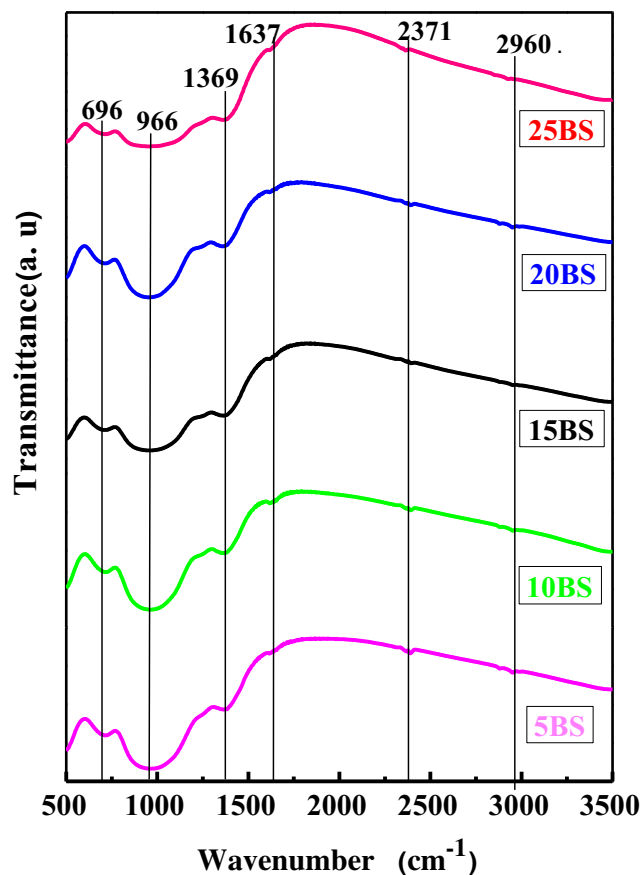
$$R = (1/N)^{1/3} \quad (3)$$

The density of the present glasses increases with the increase of BaO content in the glass composition. The 25BS glass with highest BaO content of 25 mol% shows the higher density (4.7 g cm⁻³), which is attributed to the higher atomic weight of BaO as compared to SrO. The density of multicomponent system usually follows the additive law. Hence, the replacement of lighter SrO (4.70 g cm⁻³) with denser BaO (5.72 g cm⁻³) results in the increase of the density for glasses. In general, the molar volume should decrease with increase in the density being inversely proportional to each other. So, 5BS sample has the highest value of molar volume due to its lowest density among all the glasses. Molar volume all prepared samples does not follow any trend, which could be seen from the Table 2. It might be associated with mixed effect of SrO and BaO, which is well reported in case of mixed alkali metal oxides glasses [14]. It might be associated with the affinity of individual constituents in multi-component glass composition. The increasing trend of ionic concentration of particular cation, i.e., Ba²⁺ and Sr²⁺ is obvious due to their increasing concentration in the composition. Similarly, inter-ionic distance decreases with high number of ions present per unit volume in the matrix. Barium ionic concentration is highest for the glasses, which contain highest amount of Ba²⁺, i.e., for 25BS sample, ionic concentration is 10.60×10^{21} cm⁻³ and its inter-ionic distance is also lowest. In addition to this, highest ionic concentration was found in 25BS glass followed by 10BS glass. These parameters are responsible to the hopping mechanism and predict the ionic conductivity of the glasses as discuss in the “Dielectric losses” section.

FTIR analysis

The FTIR spectra of all prepared glass samples are shown in Fig. 1. Four major IR bands at 696, 966, 1369, and 1637 cm⁻¹ are exhibited by all the glasses. The slight shifting of the peaks to the lower wavenumber is attributed to the change in strength of chemical bonds in the glasses caused by variation in modifier concentration [15]. The field strength of Ba²⁺ is lower than Sr²⁺

so, as BaO content increases, the FTIR bands shifted to lower wave number. The IR band at 696 cm⁻¹ may be attributed to Si-O-Si stretching vibrations of bridging oxygen between tetrahedron, i.e., SiO₄ [16, 17]. The bands within 650–800 cm⁻¹ can also be assigned to the stretching vibrations of Al-O bands ions in four fold coordination, i.e., AlO₄. Band at 966 cm⁻¹ is assigned to the stretching vibrations of the tetrahedral SiO₄ units with different numbers of bridging oxygen atoms [18]. As content of BaO increases in glass composition, particularly 25BS glass, this band becomes swallowing. It could be related to change in Q₄ to Q₃, Q₂, or Q₁ units of SiO₂. Moreover, this band is more symmetric in 10BS and 20BS glasses. Two IR bands are observed around ~1369 and ~1637 cm⁻¹. A small kink is

**Fig. 1** FTIR spectra for all the prepared glass samples

observed at $\sim 2300\text{ cm}^{-1}$. These bands occurred due to the vibrational mode of the borate network and asymmetric stretching relaxation of the B-O bonds of trigonal BO_3 units [13]. Another small kink at ~ 2960 is observed, which is assigned to hydrogen bonding [19–22]. According to central force model, the shifting of Si-O-Si stretching vibrational frequency can be attributed to the structural changes in the present glass network, which are related with the variation of inter tetrahedral Si-O-Si bond angles and number of connecting oxygen anions with Si cations [2].

X-ray diffraction

The heat treated samples exhibit the broad peaks as shown in Fig. 2. These peaks are not fully resolved indicated that $850\text{ }^\circ\text{C}$ temperature is not sufficient for crystallization of these glasses. The broadness of the peaks clearly indicates that these formed phases are not fully grown and might be in the metastable phase or nanocrystalline phases embedded in glass matrix. This phenomenon has been reported in many glass systems [23]. The broad peaks are index with SiO_2 (ICDD card no.-00-002-458). In addition to SiO_2 phase, another crystalline phase (Al_2SiO_5) might be present, particularly in 10BS glass sample. Interestingly, this glass shows higher tendency of crystallization as it exhibited more resolved XRD peaks than other glasses (Fig. 2). It is well reported in the literature that in the silicate based glasses, initially, SiO_2 starts to nucleate followed by the dissolution of other cations to form the stable crystalline phase [24].

Thermal analysis

Figure 3 represents differential thermal calorimetry (DSC) thermograph of 5BS, 15BS, and 25BS as-quenched samples.

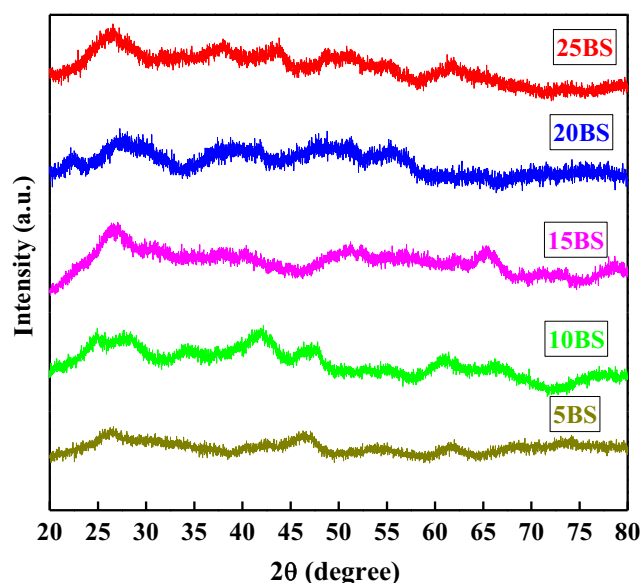


Fig. 2 XRD pattern of all sintered glass samples at $850\text{ }^\circ\text{C}$ for 12 h

These samples exhibit a clear exothermic peak corresponds to the glass crystallization temperature (T_c). The obtained values of T_c from DSC thermograph of 5BS, 15BS, and 25BS as-quenched samples are shown in Table 2. It is observed that the T_c value of the as-quenched glasses increases with the increasing content of BaO, which is due to the high bond length, cross-linking density, and closeness of packing of present glasses. 25BS glass exhibits highest T_c , which suggested that 25BS glass has high glass forming ability and also rigidity as compared to other glasses [25].

Real and imaginary dielectric constant analysis

The real part of dielectric permittivity (also called dielectric constant) (ϵ') is the measurement of the energy storage capability of the materials, whereas imaginary permittivity (ϵ'') measures the dissipation ability of the materials when an external field is applied, i.e., the dielectric loss. The ratio of ϵ' and ϵ'' is relative losses for a material and is given by $\tan \delta$. Dielectric constant of materials is directly related to the electronic, ionic, dipole, and space charge polarizations phenomena. In general, dielectric constant decreases with increasing the frequency due to slow orientation of molecules and ions than applied time period of electric field. On the other hand, it increases with the temperature if space charge polarization is dominating in the system.

The dielectric characteristics at different temperatures (250 and $600\text{ }^\circ\text{C}$) with respect to frequency for all the heat treated samples are shown in Fig. 4a, b. It is clearly observed that at temperature $250\text{ }^\circ\text{C}$, dielectric constant is almost constant at higher frequencies ($\sim 10^5\text{ Hz}$), which results from rapid polarization processes induced under applied field [26, 27]. In case of 5BS glass, the dielectric

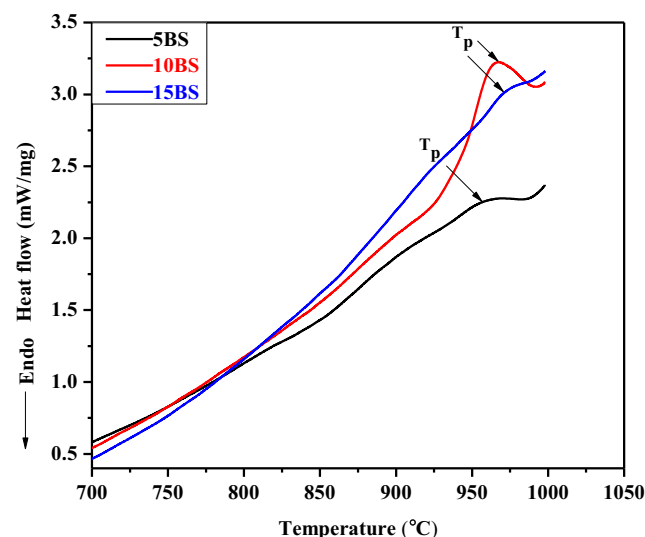


Fig. 3 DSC thermograph of 5BS, 15BS, and 25BS as-quenched glasses heat treated at $10\text{ }^\circ\text{C min}^{-1}$ in N_2 atmosphere

constant decreases within frequency regions, i.e., 10^2 – 10^4 Hz, thereafter becoming constant for higher frequency range of 10^5 – 10^6 Hz. The increment in dielectric constant at lower frequencies and high temperature is attributed to the space charge polarization dominance in the system [28, 29]. On the other hand, the decrement in dielectric constant with frequency can be associated with rapid and random motion of ions, which hinders their motion and obstructs the accumulation of charges [30, 31]. The same trend of the dielectric constant with respect to the frequency has also been reported in many glasses and glass ceramics [32, 33]. 10BS glass shows the higher degree of temperature and frequency independency followed by 20BS sample. The total ionic concentration and total inter-ionic distance (as given in Table 2) are higher and lower side, respectively, for 10BS glass, which could be used as a substrate in microelectronic applications.

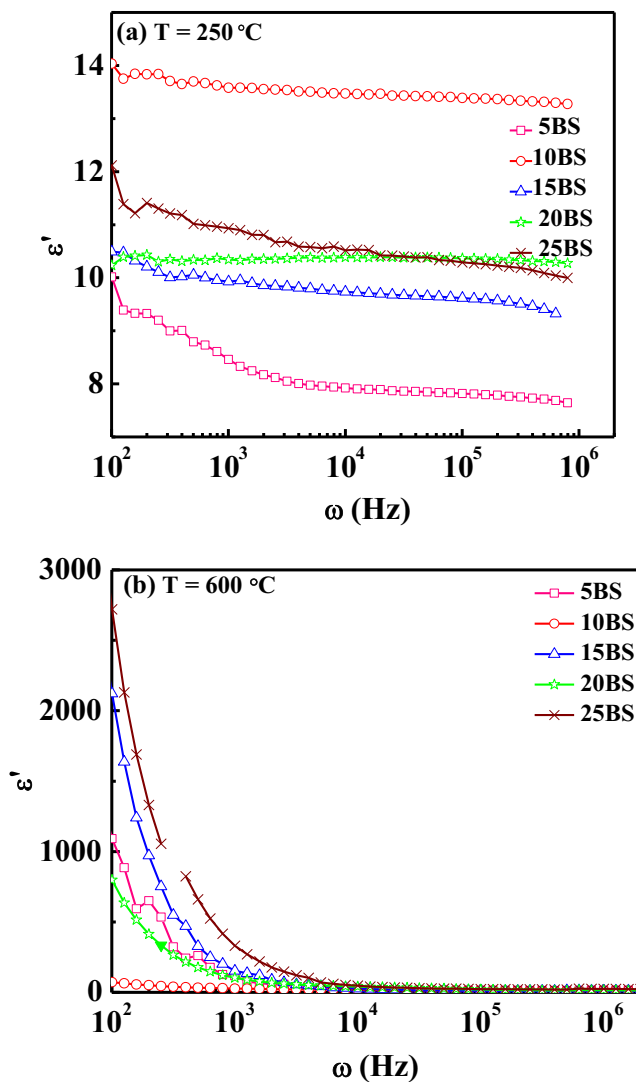


Fig. 4 Variation of dielectric constant as a function of frequency for all the glasses at **a** 250 °C and **b** 600 °C, respectively

Figure 5a, b represents variation of ε' as a function of temperature for all the glasses at selected frequencies, i.e., 10 Hz and 2 MHz. It is observed that the dielectric constant is almost constant below ~ 300 °C at selected frequencies for all the glasses. It is related to polarization which decreases with decreasing temperature. The value of dielectric constant increases with increasing temperature for the 15BS glass. The higher ionic conductivity is also responsible for increasing the dielectric constant [30]. At 10 Hz, the highest value of dielectric constant of the order of 10^4 could be attained for 15BS glass sample, indicating its high tendency for charge storage. 10BS glass exhibits the lower losses in dielectric value than the other samples with higher dielectric constant stability. It might be associated to starting of the nucleation of Al_2SiO_5 phase in the glass matrix. Moreover, this particular sample also exhibits one activation region (as discussed in the “Electrical conductivity analysis” section).

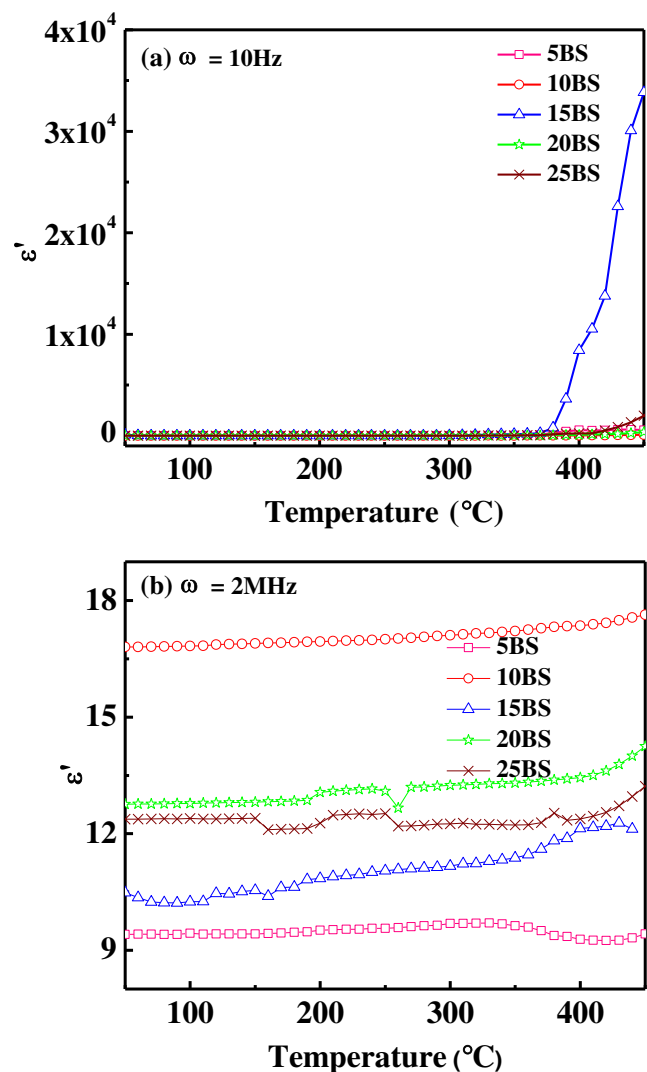


Fig. 5 Variation of dielectric constant as a function of temperature for all the glasses at **a** 10 Hz and **b** 2 MHz, respectively

Dielectric losses

In general, dielectric losses can be divided into three parts: conduction losses, dipole losses, and vibration losses. The losses due to conduction involve the migration of ions over large distances. The increase of the dielectric loss at low frequency is due to the dipole losses. Figure 6a, b represents the variation in the dielectric loss (ϵ'') versus frequency at selected temperatures 250 and 600 °C. At temperature 250 °C, dielectric loss rapidly decreases with increasing the frequency till $\sim 10^4$ Hz than its value increases in the frequency region (10^5 – 10^6 Hz) for all the glasses. At higher temperature (600 °C), the dielectric losses for all the samples decrease with increasing frequency up to a limit $\sim 10^4$ Hz after that remains almost constant, whereas for higher temperature, the conduction loss increases.

Figure 7a, b represents the variation in dielectric loss with the temperature at two different frequencies (10 Hz and 2 MHz). It can be observed that for all the glasses, dielectric

losses are almost constant below 300 °C at these frequencies. The value of the dielectric losses rapidly increases above 400 °C, particularly in 15BS glass followed by 20BS glass. As the temperature increases, the electrical conduction losses also increases, which may be responsible for the higher value of the dielectric loss for all the samples [12]. At low frequency (10 Hz), the highest value of dielectric loss of an order of 10^5 is observed for the 15BS glass sample. The dielectric losses decrease at higher frequency (2 MHz) as compared to lower frequency (10 Hz) with respect to temperature. At frequency of 2 MHz, the value of dielectric losses ~ 8 is observed for the 15BS sample, which is too low as compared to the values of dielectric loss obtained at low frequency (10 Hz). As discussed in X-ray diffraction, this glass shows poor tendency for crystallization due to higher competition between BaO/SrO (1:1). For the 15BS glass with BaO/SrO = 1:1, the charge storage capacity and the dissipation ability are both high. The variation of tangent of loss with frequency at different temperatures (250 and 600 °C) is represented in the Fig. 8a, b. Tan δ

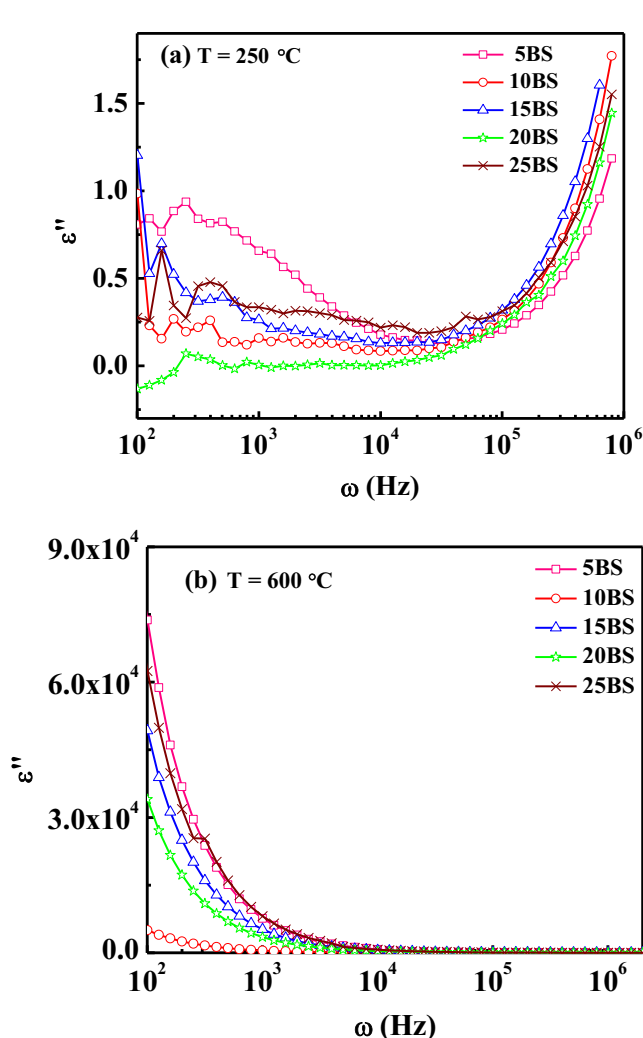


Fig. 6 Variation in dielectric loss with frequency at **a** 250 °C and **b** 600 °C, respectively

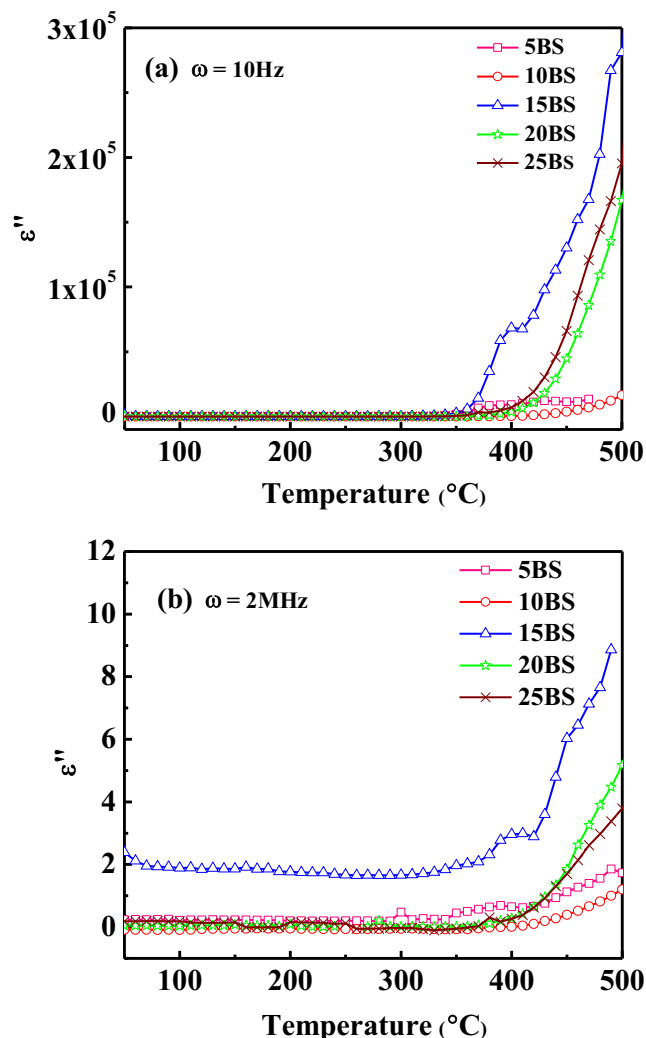


Fig. 7 Variation in dielectric loss with the temperature at **a** 10 Hz and **b** 2 MHz, respectively

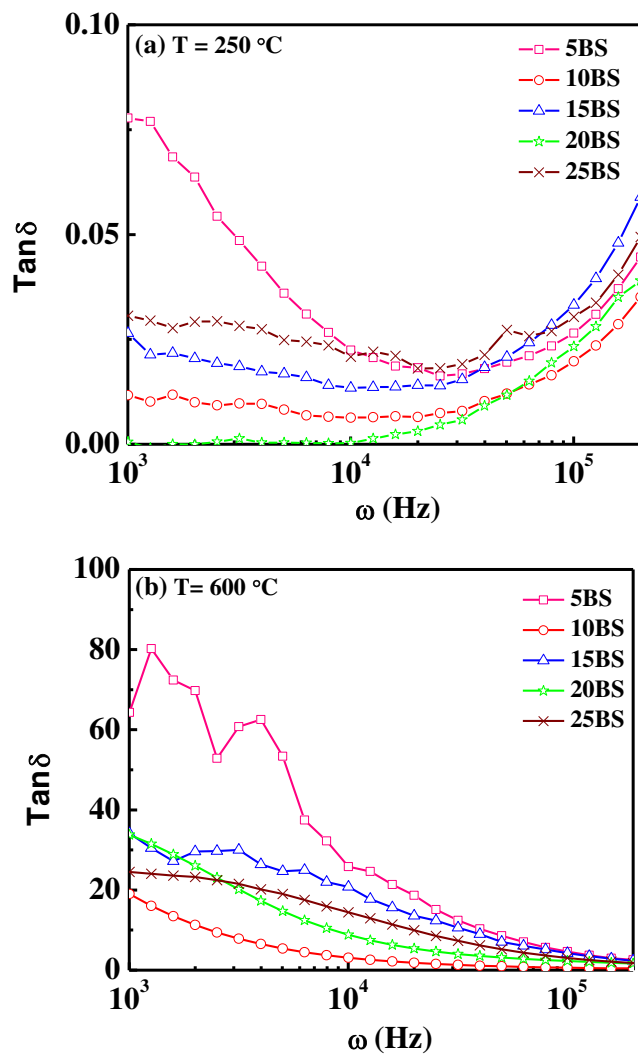


Fig. 8 Dependence of $\tan \delta$ on frequency for all glasses at **a** 250 °C and **b** 600 °C, respectively

represents the dissipation of energy due to various physical relaxation processes in the glass system. These losses are related to porosity, defects, and ion hopping between various sites inside the materials. In the present samples, at 250 °C, $\tan \delta$ decreases with frequencies up to 10^3 – 10^4 Hz for all the samples and then increases slightly at higher frequency region (10^4 – 10^5 Hz). However, at higher temperature (600 °C), $\tan \delta$ rapidly decreased with the increasing frequency. Moreover, at 600 °C, highest values of tangent of losses are observed as compared to the lower temperatures. Glass materials have always higher values of losses as compared to the crystalline counterparts of similar materials due to absence of any symmetry [12, 34].

Figure 9a, b represents the variation in tangent loss with respect to temperature at constant frequencies (1 kHz and 1 MHz). It is evident that tangent of loss exhibits strong temperature dependence at higher temperature and lower frequencies. The highest value of tangent of loss ~ 8 is obtained at

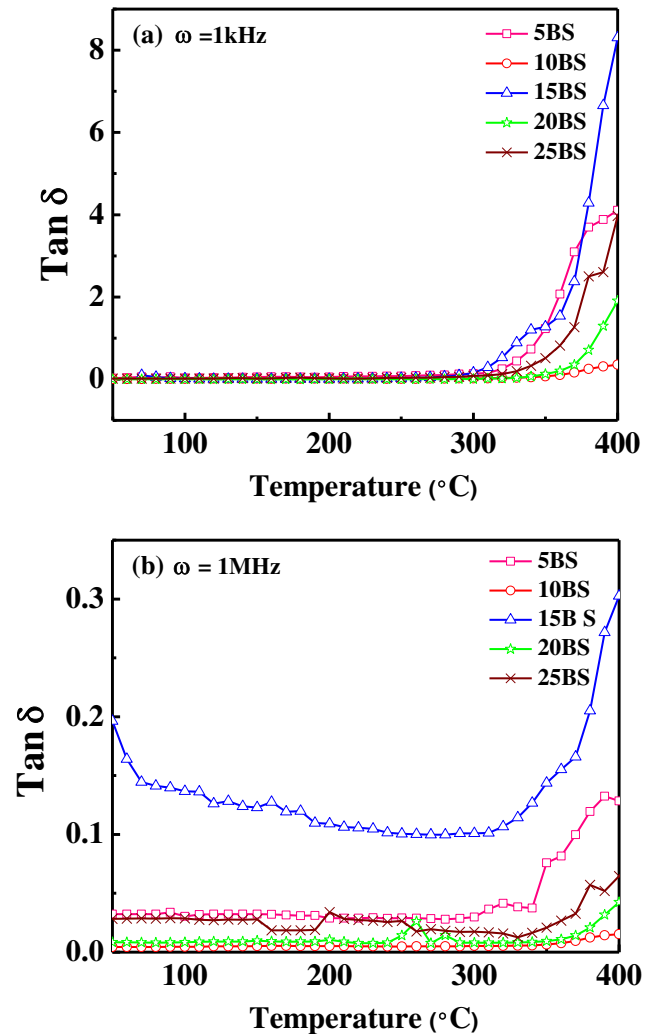


Fig. 9 Dependence of $\tan \delta$ on temperature for all the glass samples at **a** 1 kHz and **b** 1 MHz, respectively

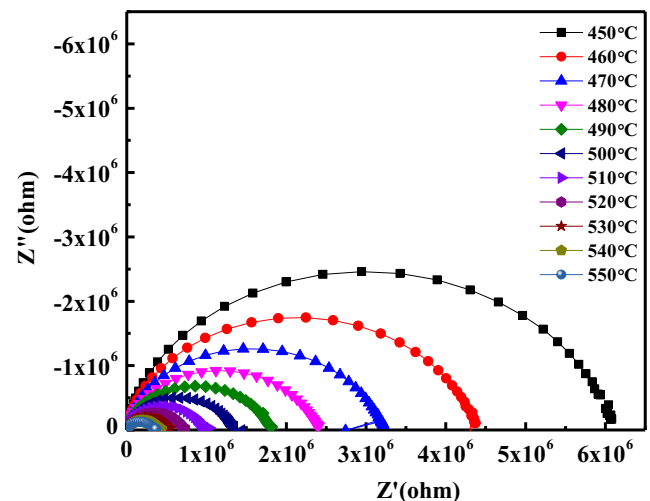


Fig. 10 Representative Cole-Cole plots of 10BS glass at different temperatures

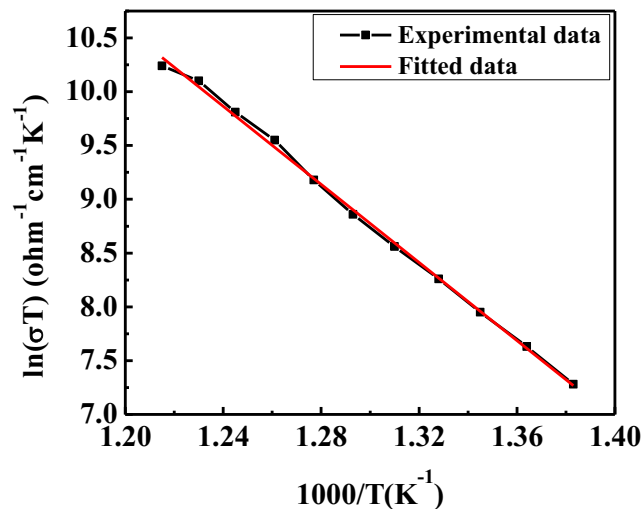


Fig. 11 Arrhenius plots of conductivity for the 10BS glass sample

1 kHz frequency for the 15BS sample as compared to the value obtained at frequency 1 MHz. The compositions with BaO/SrO = 1:1 yield highest value of $\tan \delta$ and dielectric constant among the series. This indicates that the concentration of Ba^{2+} and Sr^{2+} plays an important role in the variation of $\tan \delta$ with temperature as well as crystallization and conductivity as discussed in following section.

Electrical conductivity analysis

The Pt-coated glasses were characterized by two probe ac conductivity as a function of frequency and temperature. No semicircle was observed below 350 °C and above 550 °C for any glass. As the temperature increased, the complete semicircle was fully resolved for all the glasses. The radius of the semicircle decreased with increasing temperature as shown in the representative Fig. 10. The Arrhenius plots represent variation of $\ln(\sigma T)$ versus $1000/T$ for all the glasses and are shown in Fig. 11. The conductivity was calculated using the following relation:

$$\sigma = l/RA \quad (4)$$

where l is thickness, A is cross-sectional area of Pt coated pellet, and R is the resistance of glass calculated from the point of intersection of Cole-Cole semicircle plot on the X -axis. It can be observed that the curves show two regions separated by a characteristic point of inflection, where the activation energy for conduction of the glass samples changes. These regions were fitted to calculate the activation energies using the following relation:

$$\sigma T = \sigma_0 \exp\left(\frac{-E_a}{kT}\right) \quad (5)$$

where σ_0 is pre-exponential factor, E_a is activation energy for conduction, k is Boltzmann constant, and T is the absolute temperature. The electrical conductivity as well as activation energies is listed in the Table 3.

It can be observed from Fig. 11 that for representative 10BS glass samples, conductivity increases with increasing temperature, indicating predominant role of ionic conduction in the present glasses [12, 35]. The activation energy is associated with the reorientation of ionic hopping within the glasses. In the lower temperature region, the activation energy for conduction is higher in all the glass samples. The nonlinearity of the Arrhenius plots indicates that the activation energy is temperature dependent, which is associated with the small polaron hopping conduction mechanisms. The conduction is associated to the migration of either Sr^{2+} or Ba^{2+} or both the ions within the glass matrix [36]. It is also observed that as BaO content increases down the series (from 5BS to 25BS), conductivity also increases up to 15BS after that again decreases. In the 15BS glass sample, highest value of conductivity is observed $3.34 \times 10^{-6} \text{ S cm}^{-1}$. In general, conductivity increases with decreases activation energy. However, in the present, glass samples could not show any trend. It is associated to mixed alkaline earth metal oxide (modifier) effect. Only 10BS glass exhibits single activation energy as shown in Table 3. Probably all the present glasses are phase

Table 3 Activation energy and conductivity of the prepared glass samples

Sample ID	Activation energy (eV)		Conductivity at 450 °C (S cm^{-1})	Relaxation time at 450 °C (s)
5BS	$\Delta E_a = 0.55$	(452–474 °C)	6.31×10^{-8}	2.47×10^{-5}
	$\Delta E_a = 1.5$	(398–431 °C)		
10BS	$\Delta E_a = 1.5$	(450–550 °C)	2.02×10^{-8}	9.55×10^{-5}
15BS	$\Delta E_a = 0.61$	(431–403 °C)	3.34×10^{-6}	3.21×10^{-5}
20BS	$\Delta E_a = 2.66$	(394–352 °C)	2.57×10^{-7}	0.89×10^{-5}
	$\Delta E_a = 0.95$	(473–502 °C)		
25BS	$\Delta E_a = 2.2$	(417–457 °C)	3.9×10^{-7}	0.39×10^{-5}
	$\Delta E_a = 0.90$	(473–502 °C)		
	$\Delta E_a = 1.82$	(421–462 °C)		

separated. However, lower content of BaO may prevent this tendency up to 10 mol% of BaO in the present glasses.

Figure 12a, b indicates the frequency dependent A.C. conductivity plots of the samples at different temperatures 50 and 250 °C, respectively. The value of frequency-dependent conductivity was analyzed by using Jonscher's power law as follows:

$$\sigma(\omega) = \sigma_{dc} + A\omega^s \quad (6)$$

where σ_{dc} is the D.C. conductivity, A is a constant and ω is the angular frequency of the applied electric field, and the s is frequency exponent factor, which lies in the range of 0 to 1. When $s = 1$, $\sigma(\omega)$ is proportional to ω and becomes frequency independent.

$$\sigma(\omega) = \varepsilon_0 \omega \varepsilon'' \quad (7)$$

where ω , ε'' , and ε_0 are angular frequency, imaginary permittivity, and permittivity of free space, respectively. This region is known as the constant loss regime. When s lies between 1.0 and 0.6, the region is known as the Jonscher regime. The

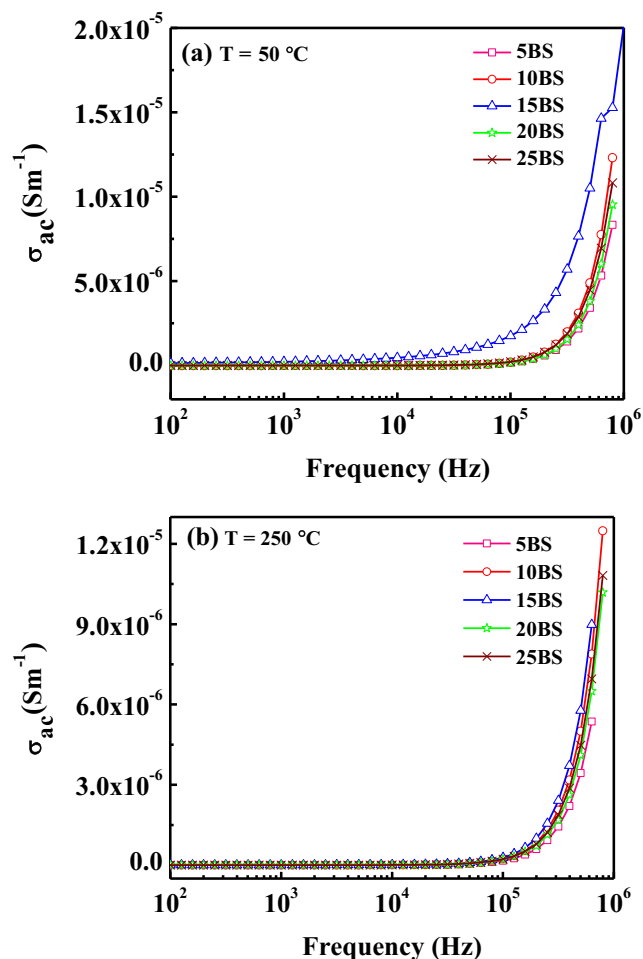


Fig. 12 A.C. conductivity versus frequency curves for all the samples at a 50 °C and b 250 °C, respectively

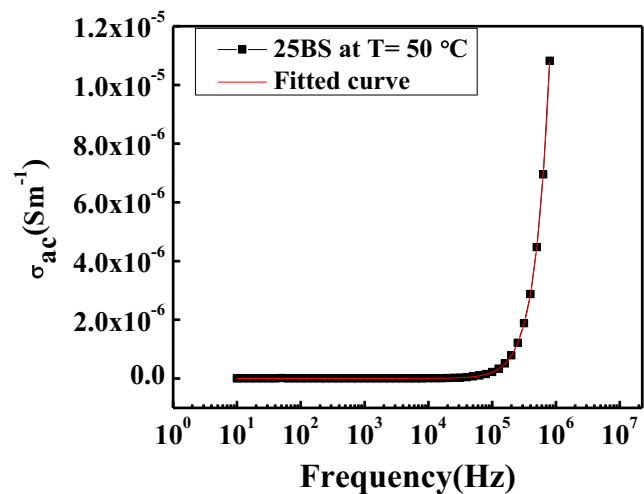


Fig. 13 A representative curve of 25BS glass sample at 50 °C fitted as per super linear power law $\sigma(\omega) = A\omega^s$

frequency-dependent conductivity behavior is nearly constant for all the glass samples as shown in Fig. 12. At temperatures 50 and 250 °C, the conductivity increases sharply with increasing frequency and follows the universal super linear power law ($\sigma(\omega) = A\omega^s$). The conductivity followed the super linear power law for higher frequencies as shown in a representative curve (Fig. 13) for the 25BS glass at 50 °C. In the present glasses, conductivity shows both temperature and frequency dependent behavior. This indicates that A.C. conductivity is attributed to the thermally activated processes [37]. Relaxation time (τ_m) for all the glasses is calculated with the help of imaginary impedance (Z'') versus frequency (f) plots, and the values are given in Table 3. It is not single valued but distributed continuously/discrete around a mean, $\tau_m = \omega_m^{-1}$ ($\omega_m = 2\pi f_{max}$) [38]. In general, lower relaxation time leads to higher conductivity and vice versa. In present case, 10BS glass with highest relaxation time (9.55×10^{-5} s) exhibits lowest conductivity, i.e., 2.02×10^{-8} S cm⁻¹. However, no clear trend is observed in the relaxation time and conductivity values with respect to the composition of the present glass samples.

Conclusion

The BaO content increases the density of all the glasses. The FTIR confirms the presence of SiO₄, AlO₄, and BO₃, structural units in the present glasses. The highest conductivity (3.34×10^{-6} S cm⁻¹), dielectric constant, and dielectric losses are observed in 15BS glass due to mixed alkaline earth metal oxides (modifier) effect. For all the glass samples, conductivity follows the super linear power law for higher frequencies and low temperature region. 15BS glass may be used as optical switches due to its good capacity of dissipation of heat along with higher dielectric constant. The dielectric constant

of 10BS glass exhibits temperature and frequency independent behavior. This particular glass can be useful for micro-electronic applications.

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