

Effect of B₂O₃ on the Structure and Electrical Properties of High-Temperature Sealing Glass

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Abstract

To develop an electrical connector sealing glass suitable for stable high-temperature operation, a glass based on BaO-SiO₂-CaO-Al₂O₃-B₂O₃ (BSCA) system was studied. The coefficient of thermal expansion (CTE), glass transition temperature (T_g) and softening point temperature (T_f) of specimens were determined by means of dilatometer analysis, and the microscopic network structure of the glass powder was studied with Fourier-transform infrared spectroscopy (FTIR). The content of [BO₃] was increased with the increase in B₂O₃ content. The effect of the content of B₂O₃ on the electrical properties of the glass was discussed. This discussion shows that, as the temperature increases, the high-temperature insulation resistivity of the material decreases gradually. At the same temperature, with the increase of B₂O₃, the resistance value has a maximum value. At 500 °C, the resistance of BSCA4 is $6.19 \times 10^{-2} \text{ M}\Omega$, and its (RT ~ 300 °C) CTE is $9.2 \times 10^{-6} / \text{K}$.

Keywords: B₂O₃, sealing glass, high-temperature, electrical properties

I. Introduction

Up to the present, three main types of sealing materials, i.e. braze, mica and glass (or glass-ceramic), have been investigated in literature^{1–4}. Of these three materials, glass is the main sealant and has been extensively investigated. There is a huge demand for glass-sealed electrical connectors that can work stably in high-temperature and high-spin environments in aerospace and precision machinery manufacturing. The requirements for the sealant are stringent due to the high operating temperature (approximately $\geq 485 \text{ }^\circ\text{C}$) and the very harsh environments (oxidizing, reducing and humid). However, the thermal stability of state-of-the-art sealing glass/glass-ceramics has not yet reached a practically acceptable level. There is therefore still a need for further exploration of glass-based seals with good thermal stability in order to improve the high-temperature working stability of connectors, for example, good insulation performance at high temperature.

The barium-rich borosilicate glass system BaO-SiO₂-CaO-Al₂O₃-B₂O₃ has low cost, a high expansion coefficient and adjustable thermal expansion, which has attracted extensive attention of researchers. At present, the operating temperature range of high-temperature electrical connectors is narrow, the sealing glass is molten at high temperature, and the sealing components are prone to failure. However, research on the application of the glass in the field of high-temperature electrical connectors has been very limited. It is commonly believed that B₂O₃ is one of the most important glass components as it exhibits

strong glass-forming properties. It is often introduced into the chemical composition of silicate glasses in order to lower their melting temperature and improve their properties; at the same time, it can improve the wettability of glass and metal, so as to achieve the purpose of sealing. On the other hand, boron atoms in borate crystals and glasses usually coordinate with either three or four oxygen atoms, forming [BO₃] or [BO₄] structural units. These two fundamental units can be arbitrarily combined to form either “super-structural” units or different B_xO_y groups (e.g. boroxol rings, etc.)⁵, resulting in changes in glass properties.

The properties of barium-rich silicate glass can be optimized with B₂O₃. This can improve the wettability and thermal expansion coefficient of glass, reduce the softening point and improve the high-temperature resistance of glass. Peng *et al.*³ investigated the influence of glass compositions on the thermal stability and the leak rates of the BaO-B₂O₃-SiO₂ system. Yang *et al.*⁶ investigated the chemical interactions of SiO₂-CaO-BaO-B₂O₃-Al₂O₃-based sealing glasses with oxidation-resistant alloys³. Wei *et al.*⁶ investigated the effects of B₂O₃ addition on both the sintering behavior and microwave dielectric properties of CaO-B₂O₃-SiO₂ glass ceramics.

In this paper, the BaO-SiO₂-CaO-Al₂O₃-B₂O₃ (BSCA) system is used as the basic glass system, and B₂O₃ is used to replace SiO₂ in order to adjust the glass composition. The influence of B₂O₃ content on the structure, thermal and electrical properties of barium-rich borosilicate glass is discussed. It provides a theoretical basis for the prepara-

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tion of sealing glass with excellent high-temperature insulation performance.

II. Experimental

The chemical composition of laboratory-prepared glass is shown in Table 1. Five raw powders, BaCO₃ (99.7 %), SiO₂ (99.7 %), Al₂O₃ (99.7 %), H₃BO₃ (99.7 %), CaCO₃ (99.7 %), are used for the synthesis of the BaO-SiO₂-CaO-Al₂O₃-B₂O₃ glasses.

Table 1: Batch composition (in mole %) of sealing glasses.

Glass no.	Composition of glasses / mol%				
	SiO ₂	BaO	CaO	Al ₂ O ₃	B ₂ O ₃
BSCA1	37.0	38.6	15.5	8.0	1.0
BSCA2	35.0	38.6	15.5	8.0	3.0
BSCA3	33.0	38.6	15.5	8.0	5.0
BSCA4	31.0	38.6	15.5	8.0	7.0
BSCA5	29.0	38.6	15.5	8.0	9.0

(1) Synthesis of glass powder, glass beads, and cylinder

According to the ratio shown in Table 1, the corresponding raw materials were accurately weighed, dry-mixed in a V-type mixer for 30 min, then placed into a 500 mL corundum crucible, and melted at 1 650 °C for 2 h. Part of the melt was poured onto a graphite mold preheated to ~650 °C. The glass chips prepared in this way were subsequently annealed at 650 °C for 2 h to release residual thermal stress. The remaining melt was quenched by pouring it into de-ionized water at room temperature. The glass powder was obtained by means of ball-milling for 70 min and passed through a 200-mesh sieve.

The glass “cylinder” was cut from the bulky glass chips with a precision cutting machine.

(2) Characterization

The glass was ground into 200-mesh powder, the structure of the glass was measured with a Thermo Scientific Nicolet iS20 Fourier-transform infrared spectrometer, the reference sample was KBr, and the test range was 400 ~ 4 000 cm⁻¹.

The bulk density of sintered samples was determined according to Archimedeian immersion method using water as a medium.

Thermal expansion measurements along with determination of the glass transition temperature (*T_g*), dilatometric softening temperature (*T_f*), and coefficient of thermal expansion (CTE) of the annealed sample cut into 5 mm × 5 mm × 20 mm, were performed using a thermal mechanical analyzer and pushrod dilatometer (NET-ZSCH DIL402) at a heating rate of 5 K/min.

To confirm the glassy nature of all the samples, X-ray diffraction (Japanese Rigaku Smartlab CuKα radiation, λ = 1.542 Å) measurement was performed, the scanning speed was 10°/min, and the range was 5 °~ 90 °.

A high-temperature insulator resistance tester (RMS1000I) was used to measure the body resistance of the annealed glass sample at 100 °C ~ 600 °C.

For measuring the capacitance and dielectric loss, the glass samples were cut and optically polished. Measurements were carried out using a computer-controlled LCR HiTester (HP4294A) at 25 °C. The dielectric constant ε was calculated from the relation

$$\epsilon = \frac{Ch}{S\epsilon_0}$$

where *h* and *S* are the thickness and area of samples respectively, *C* is the measured electric capacity and ε₀ is the permittivity of free space (ε₀ = 8.854 × 10⁻¹² F/m).

III. Results and Discussion

(1) Phase characterization

Fig. 1 shows the XRD pattern of all prepared glass samples. The absence of a sharp Bragg's peak confirmed the amorphous and homogeneous nature of the glass samples. The broad hump observed in the XRD pattern of all the samples indicates the lack of a long-range order.

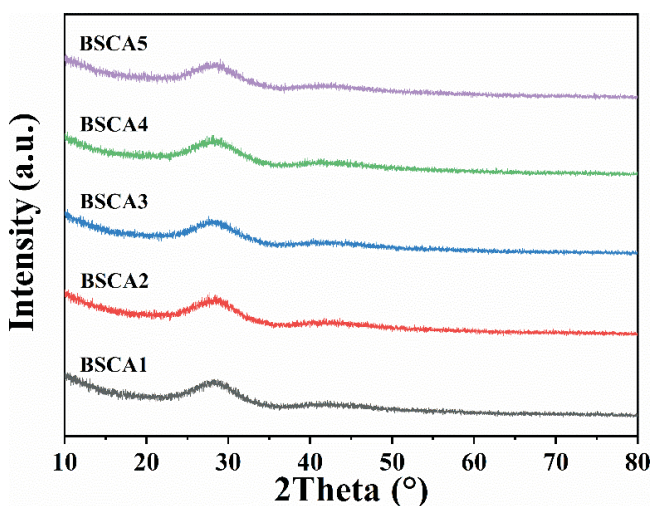


Fig. 1: XRD of BSCA glass samples.

(2) Spectroscopic study FTIR

Fig. 2 presents the FTIR spectra of the investigated glasses. The IR spectrum of BSCA1 showed four main bands. Five main bands appeared in the other four glasses. The wavenumbers and chemical bond vibration mode of the corresponding main bands are shown in Table 2. As the content of SiO₂ replaced by B₂O₃ in the glass changes, the bands located at 1 200 ~ 1 400 cm⁻¹ in BSCA1-BSCA5 gradually become obvious. Unlike the alkaline earth metal silicates, in the alkaline earth metal silicate glasses containing B₂O₃, the characteristic peaks of borates appear at 1 200 ~ 1 400 cm⁻¹; these are caused by the B-O-B asymmetric stretching vibrations of [BO₃] in different boroxol groups. It can be seen from the figure that the amount of [BO₃] in the glass increases with the increase in B₂O₃ content.

All the bands in the infrared spectrum curve in Fig. 2 show diffuse broad peaks, which indicates the structural change in the short-range region of the glass, and the fre-

quency intensity and position of each main peak are different in a certain range, which shows that various bonds and bonds in the glass have superimposed properties. The Gaussian function was used to fit the spectrum into peaks⁹ as shown in Fig. 3(a) – (e), and the proportional relationship between the chemical bonds corresponding to each absorption peak was determined. BSCA1 has five absorption peaks, and BSCA2-BSCA5 has six absorption peaks. The fitted six absorption peaks are divided into five types according to different energies, which are P1: the low wavenumber interval peak (478 cm^{-1}) caused by the combined action of bending vibrations of Si-O-Si of $[SiO_4]$ and B-O-B of $[BO_4]$, P2: the B-O-Si, B-O-B bending vibration in $[BO_3]$ and stretching vibration of Si-O-Al in glass (673 cm^{-1}), P3: O-Si-O non-bridge oxygen symmetric-stretching and anti-symmetric-stretching vibration in $[SiO_4]$ ($\sim 895\text{ cm}^{-1}$), P4: bridging oxygen stretching vibrations of Si-O-Si and $[BO_4]$ stretching vibration ($\sim 1040\text{ cm}^{-1}$), and P5: stretching vibrations of B-O in $[BO_3]$ ($\sim 1200 \sim 1400\text{ cm}^{-1}$).

The areas corresponding to the five types of absorption peaks can be known by integration, which are marked as S1, S2, S3, S4 and S5, respectively. The content of each type of bond in the glass is calculated by $Si/(S1+S2+S3+S4+S5)$. The percentage of the system (Si is the area corresponding to each absorption peak), so the histogram shown in Fig. 3(f) can be obtained⁹. It can be seen from the figure that with the increase in B_2O_3 content, the content of the Si-O-Si bridge oxygen in the glass keeps decreasing, while the content of the $[BO_3]$ triangle keeps increasing.

(3) Density

Fig. 4 shows how the density of sealing glass changes with the content of boron oxide. In terms of glass composition, compared with most borosilicate glasses, the content of barium oxide in barium-rich borosilicate glass is higher, and the density of BaO is very high, at 5.72 g/cm^3 , which

leads to the higher density of the whole sealing glass. The minimum value of BSCA5 was 3.73 g/cm^3 , and the maximum value of BSCA2 was 3.88 g/cm^3 .

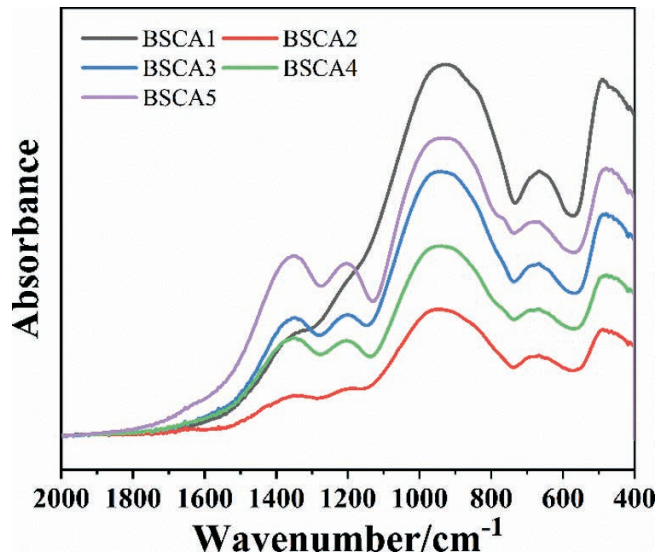


Fig. 2: FTIR spectra of BSCA glass powder at room temperature.

It can be seen that the density of the glass first increases and then decreases. The relative molecular mass of SiO_2 is 60.08, and the relative molecular mass of B_2O_3 is 69.62. An increase in density was expected when the SiO_2 was replaced with B_2O_3 . As the B_2O_3 content increases, the density of the glass gradually decreases. The reason is that the increase in B_2O_3 content will reduce the tightness of the network structure in the sealed glass. The density of glass, in general, is explained in terms of a competition between the masses and sizes of the various structural groups present in glass; the density is related to how tightly the ions and ionic groups are packed together in the substructure¹⁰.

Table 2: Spectral peaks in FTIR spectra and vibration bands of BSCA glasses^{7, 8}.

Wave number (cm^{-1})	Corresponding characteristic vibration attribution
~ 478	Si-O-Si and B-O-B bond bending vibration in $[BO_4]$
~ 673	Si-O-Al stretching vibration and B-O-Si, B-O-B bond bending vibration in $[BO_3]$
~ 895	O-Si-O non-bridge oxygen symmetric-stretching and anti-symmetric-stretching vibration in $[SiO_4]$
~ 1040	Si-O-Si anti-symmetric-stretching vibration in $[SiO_4]$ and $[BO_4]$ stretching vibration
$1200 \sim 1400$	B-O-B asymmetric stretching vibration in $[BO_3]$ triangle and $[BO_4]$ tetrahedron and other borates ($[BO_3]$ stretching), B-O vibrations of various borate groups ($[BO_3]$ stretching)

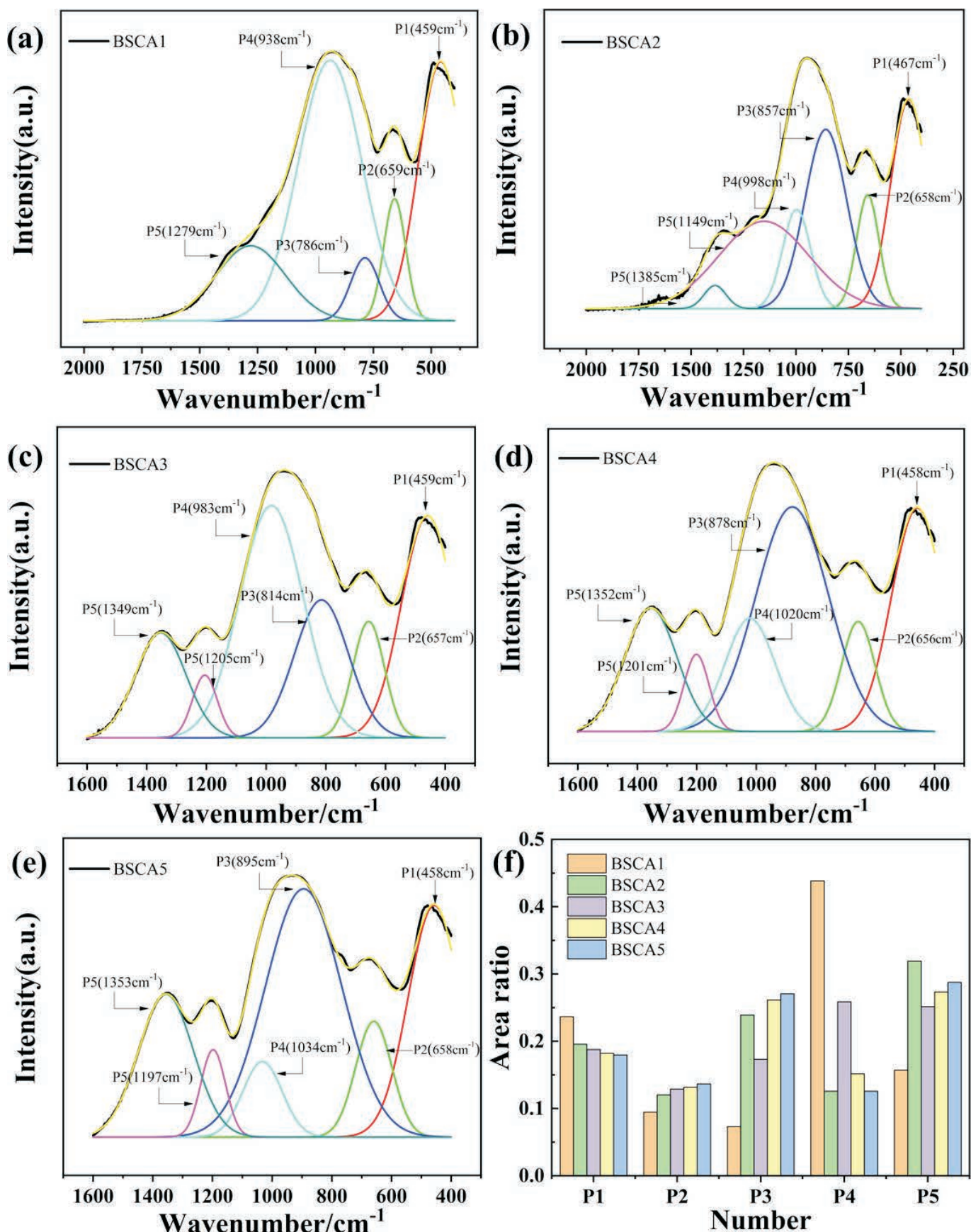


Fig. 3: BSCA peak fitting of the FTIR spectra: (a) BSCA1, (b) BSCA2, (c) BSCA3, (d) BSCA4, (e) BSCA5, (f) Area percentage of absorption peak.

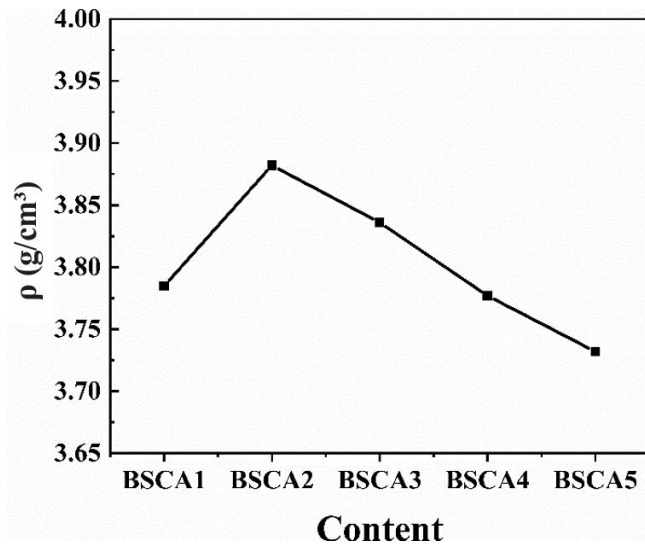


Fig. 4: Change of density ρ as function of B_2O_3 content.

(4) Thermal expansion analysis

Fig. 5 shows the relationship between the thermal expansion coefficient (CTE) and temperature of five kinds of glass. As shown in Fig. 6, combined with Table 3, the softening point (T_f) of BSCA decreased from 833.0 °C to 755.2 °C. T_f and glass transition point (T_g) of the glass decreased gradually with the increase in B_2O_3 content. The main reason is that B_2O_3 is a good flux.

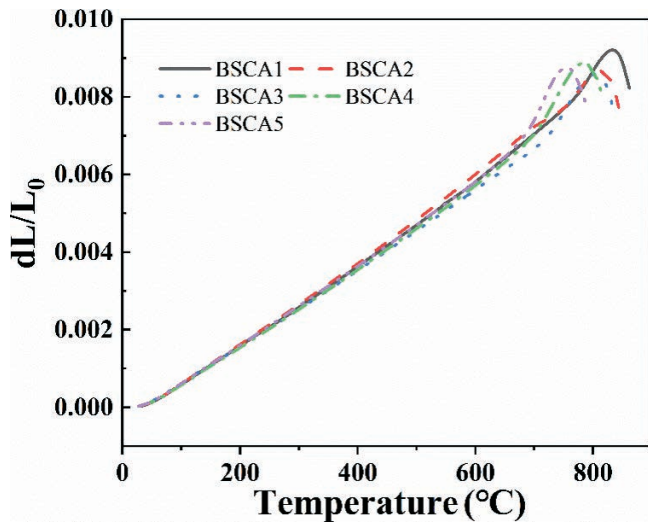


Fig. 5: Variation of thermal expansion coefficient of BSCA glass with temperature.

It can be seen from Fig. 6 that the CTE first increases, then decreases and finally increases. We believe that the first increase in the CTE is due to the fact that the addition of B_2O_3 reduces the content of SiO_2 , the number of non-bridging oxygen in the glass increases, and the bond strength of Si-O decreases. The degree of connection between $[SiO_4]$ is weakened, resulting in a loose glass network structure; secondly, the decrease is due to the continuous increase in B_2O_3 content. Due to the presence of alkaline earth metals to form $[BO_4]$, $[BO_4]$ and $[BO_3]$ maintain a relative balance, and the network structure of the glass is denser. Then the content of B_2O_3 may continue

to increase, there are more boron-oxygen triangles $[BO_3]$, and the network structure tends to be unstable.

The thermal expansion coefficient of BSCA2 is the largest, which is $9.81 \times 10^{-6} / K$, but the softening point temperature is higher than 800 °C. The thermal expansion coefficient of BSCA5 glass is $9.65 \times 10^{-6} / K$, moreover, the softening point and transition temperature are the lowest, and the softening point is 755.2 °C. For glass metal sealing, a low T_g is advantageous because below this temperature, the stress due to thermal expansion mismatch begins to develop. Therefore, in order to minimize the total stress produced, the T_g of the sealing assembly should be as low as possible in the operating temperature range.

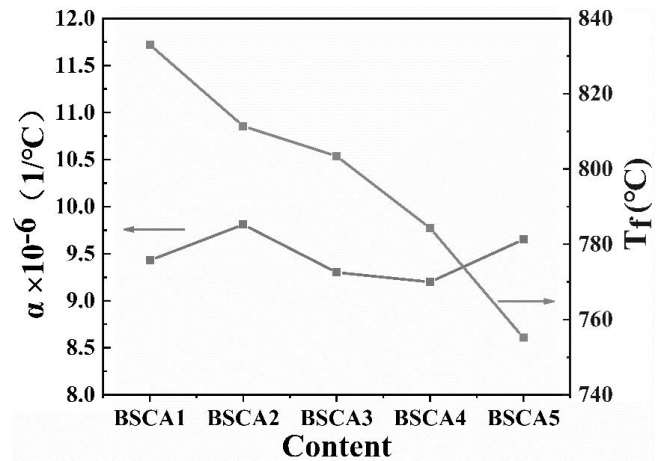


Fig. 6: Change of CTE and T_f as function of B_2O_3 content.

(5) Electrical properties

Fig. 7 shows the variation of the volume resistivity of each component with temperatures at 100 °C to 600 °C. The electrical resistivity of a seal glass depends on the composition and the devitrification (phase separation and crystallization) of the glass as well as the operating temperature¹¹. Seal glass electrical resistivity generally decreases with increasing temperature because the diffusion of ions increases with increasing temperature. Generally, conductivity is controlled by ionic charges in alkaline- and alkaline-earth-oxide-containing glasses and by

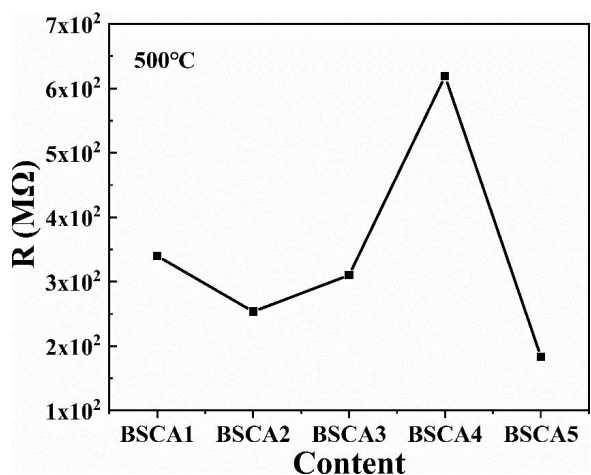


Fig. 7: Variation of high-temperature volume resistivity of BSCA glass with temperature.

Table 3: Thermal performance parameters of BSCA glass.

Glass no.	BSCA1	BSCA2	BSCA3	BSCA4	BSCA5
$T_g/^\circ\text{C}$	767.6	755.0	725.2	698.0	678.6
$T_f/^\circ\text{C}$	833.0	811.4	803.4	784.3	755.2
$\alpha(300^\circ\text{C}) \times 10^{-6}/^\circ\text{C}$	9.43	9.81	9.30	9.20	9.65

electrons and holes in transition-metal-containing glasses¹². Both types of conductivity compromise seal glass resistivity. In the barium-containing systems, the decline in resistivity caused by temperature is because of Ba^{4+} ion migration¹². In addition, it can be seen from Fig. 8 that the bulk resistance of the sample first decreased, then increased and then decreased with the increase in B_2O_3 content. Firstly, since the addition of B_2O_3 will reduce the tightness of the network structure in the sealing glass, it is beneficial to the migration of ions. Secondly, with the increase in B_2O_3 content, the content of $[\text{BO}_4]$ and $[\text{BO}_3]$ in the material tends to balance, the structure is relatively stable, and the ion-oriented migration is bound. Finally, the content of B_2O_3 continues to increase, $[\text{BO}_3]$ increases, the glass network structure becomes looser, ion migration is accelerated, and resistance decreases.

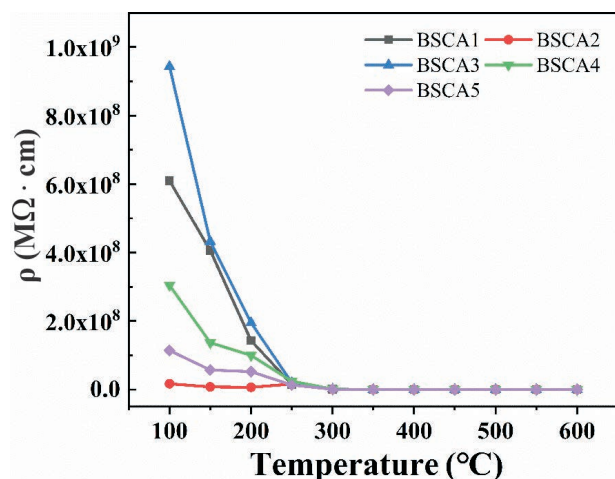
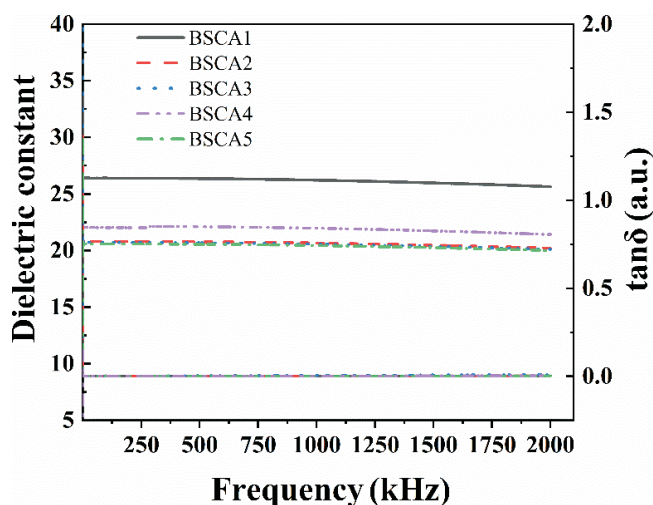
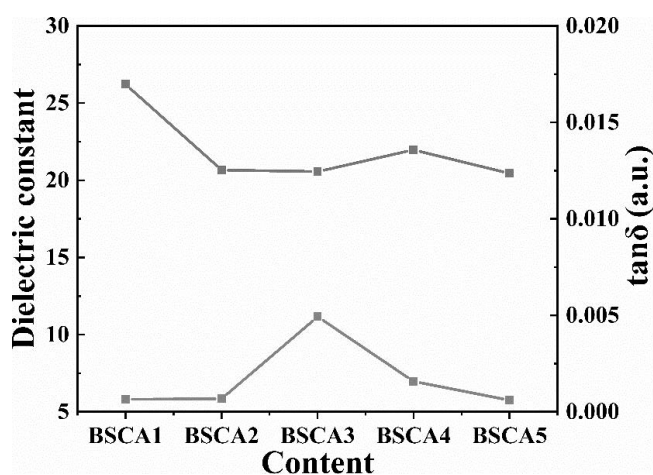
**Fig. 8:** High-temperature bulk resistance of different glasses at 500 °C.

Fig. 10 shows the frequency dependence of the dielectric constant (ϵ) and dielectric loss of BSCA glass. It is evident that value of ϵ decreases with increasing frequency and attains a constant limiting value ϵ_∞ which is not related to the hopping dynamics of mobile ions and may be due to a rapid polarization process occurring in the samples. The high value of ϵ in low-frequency region is a bulk phenomenon that may be due to the presence of metallic or blocking electrodes that do not permit the mobile ions to transfer into the external circuit, i.e. charge accumulation at the electrode-sample interface, as a result, mobile ions are piled up near the electrodes and give a large bulk polarization in the materials. In the high-frequency region, the well-known non-Debye behaviour is observed for low temperatures^{13, 14}. The dielectric loss factor ($\tan\delta$) reflects the phase difference due to energy loss within the sample at a specific frequency, it can be seen that at lower frequencies, $\tan\delta$ increases with frequency and shows a max-

imum at particular frequencies because the active component of the current is increasing more rapidly than its reactive component. It should be noted that the frequency measurement range of this study is the low-frequency microwave band, where conductivity is dominated by the DC conductivity. This frequency dependence of $\tan\delta$ is usually related to conduction losses¹⁵.

**Fig. 9:** Variation of dielectric constant and dielectric loss with frequency.**Fig. 10:** Variation of dielectric constant and dielectric loss with different B_2O_3 .

The dielectric properties of glass are also significantly dependent on chemical composition. The spatial arrangement of network-forming units, the voids and cavities, and the “hopping” of migrating ions between different sites are varied based on composition change¹⁰. Fig. 3 shows the dielectric constant and loss as a function of B_2O_3 content at 1 MHz. It can be seen from the figure that the dielectric constant of the glass sample first decreases, then increases

and then decreases with the addition of B_2O_3 . Electronic, ionic and dipole polarization effects contribute to the dielectric constant ϵ in the investigated frequency range. The dielectric constant ϵ of glass are determined based on the total polarizability of the glass system, and the dielectric constant ϵ increases with the total polarizability. The ionic polarizability of B^{3+} is $0.003 \times 10^{-30} \text{ m}^3$, and that of Si^{4+} is $0.333 \times 10^{-30} \text{ m}^3$, the total polarizability of the glass system decreases, which could be the reason for the decrease in the dielectric constant of BSCA1-BSCA3 glasses. With the change of network structure, as shown in the FTIR spectrum, the $[BO_3]$ unit increases, the ionic polarizability of B^3 is $0.009 \times 10^{-30} \text{ m}^3$, and the dielectric constant of BSCA4 is greater than that of BSCA5. The dielectric constant of glass is proportional to the rate of cation mobility, and the slower the cation mobility, the lower the dielectric constant. As the content of $[BO_3]$ continued to increase, the number of broken bonds increased, the migration of cations was hindered, and the dielectric constant decreased accordingly.

The dielectric loss is mainly affected by the frequency, and the loss exists in different forms at different frequencies. For 1 ~ 10 MHz, the structural loss and resonance loss are the main ones. When the frequency is 1 MHz, it is mainly due to the structural loss, and there are loose voids in the glass. When metal ions with weak binding force penetrate into them, under the action of the external electric field, a part of the ions can be separated from the original thermal motion bit, moving in the direction of the electric field. When the frequency is 10 MHz, it shows obvious resonance loss. With the increase of boron oxide content, $\tan\delta$ first increased and then decreased, which may be due to the increase in $[BO_3]$ content, the loose glass network structure and the increase in structural loss, BSCA3 has the highest dielectric loss, $\tan\delta = 0.00493$. At the same time, the mass fraction of B_2O_3 increases, and more bridge-oxygen bonds Si-O-B are formed. Since the length of the B-O bond is smaller than that of the Si-O bond, the voids in the glass network are smaller, and the movement range of metal ions in the network is limited, which reduces the dielectric loss.

IV. Conclusions

In this paper, different glass formulations for the BaO-SiO₂-CaO-Al₂O₃-B₂O₃ system are investigated in an attempt to develop a sealing glass that can operate at temperatures above 500 °C.

The thermal properties of the glass showed that with the increase in the B_2O_3 content, T_g and T_f decreased gradually.

FTIR shows that when B_2O_3 is added to alkaline earth metal silicate glass, the B-O-B asymmetric stretching vibration of borate glass appears in B_2O_3 -containing silicate glass, and the content of $[BO_3]$ in the glass increases with the increase of B_2O_3 content.

At 500 °C, the maximum resistance of BSCA4 is $6.19 \times 10^{-2} \text{ M}\Omega$, of which the CTE is $9.2 \times 10^{-6} / \text{K}$, the density is 3.78 g/cm^3 , the dielectric constant is 21.97, and the dielectric loss is 0.00158.

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