



Low temperature sintering and dielectric properties of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3\text{-MgO}$ composite ceramics with $\text{CaO-B}_2\text{O}_3\text{-SiO}_2$ glass addition

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ABSTRACT

Self-made glass frits (calcium borosilicate glasses) were employed as the sintering aids to reduce the sintering temperatures of $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ (for short BST)-MgO composite ceramics. The effects of $\text{CaO-B}_2\text{O}_3\text{-SiO}_2$ (for short CBS) glass on the sintering behavior, microstructures and dielectric properties of BST-MgO ceramics have been investigated. The pure BST-MgO ceramics usually sintered at above 1350°C . It was found in our experiment that 20 wt.% of CBS glass additives can reduce the sintering temperature to 850°C effectively. With increasing the content of CBS glasses, both of dielectric constant (ϵ_r) and dielectric loss ($\tan \delta$) were decreased. The samples sintered at 850°C for 2 h with 20 wt.% CBS glass addition consist of $\text{Ca}_4\text{Ti}_3\text{O}_{10}$, Mg_2SiO_4 , and $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$ phases, and possess good dielectric properties: moderate dielectric constant $\epsilon_r = 66$ and small dielectric loss $\tan \delta = 2.0 \times 10^{-3}$.

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1. Introduction

In recent years, with the development of advanced communication systems, multi-layer microwave devices have been widely used in microwave circuits to miniaturize related components [1]. Low temperature co-fired ceramics (LTCC) have received much attention for their use in microwave applications. In the multi-layer structures, the sintering temperature of the dielectric materials must be reduced to or below 900°C , in order to co-fire with highly conductive internal electrode metals, such as silver (the melting point of silver is about 961°C) [2]. However, conventional solid state synthesized BST ceramics, which are considered one of the most important ferro-para electric materials used in microwave applications, usually sintered at high temperatures between 1350 and 1450°C [3]. This has become an apparent barrier in developing BST devices for microwave applications.

Generally speaking, there are several methods to lower the sintering temperature of BST ceramics, such as addition of low-melting glasses or oxides as sintering aids, chemical processing and using nanosized powders as raw materials [4,5]. Among the methods mentioned above, low-melting glasses and oxides for liquid phase sintering are lower in cost and easier to process. Therefore, it has attracted much attention.

Until now, B_2O_3 and Li_2CO_3 have been widely employed as effective dopants for low temperature sintering process. However, Li-doped BST ceramics have high dielectric loss because

the introduction of Li^+ will increase the leak conductivity [6]. $\text{BaCu}(\text{B}_2\text{O}_5)$ (BCB) additions have also been commonly investigated [7,8], but the sintering temperature cannot be reduced below 900°C for BST ceramics. Many other glass additions, including $\text{B}_2\text{O}_3\text{-Bi}_2\text{O}_3\text{-SiO}_2\text{-ZnO}$ glass [9], $\text{ZnO-B}_2\text{O}_3$ glass [10], $\text{MgO-CaO-SiO}_2\text{-Al}_2\text{O}_3$ glass [11], have also been studied. However, the sintering temperatures of these glasses added ceramics were still higher than 900°C . Hsiang et al. [12] discussed the effects of $\text{BaO-B}_2\text{O}_3\text{-SiO}_2$ and $\text{PbO-B}_2\text{O}_3\text{-SiO}_2$ glass addition on BaTiO_3 -based ceramics. While the sintering temperature can be reduced to 900°C , the dielectric loss is larger than 1%. Therefore, choosing a suitable glass additive which can reduce the sintering temperature below 900°C without deterioration of dielectric properties of the host material is very important. The desirable dielectric properties in our study include a moderate dielectric constant ($\epsilon_r = 50\text{--}200$) and a low dielectric loss ($\tan \delta \leq 2.0 \times 10^{-3}$).

In order to achieve a moderate dielectric constant, it is important to introduce low dielectric constant secondary phases to the sintered body. It was found by Koh et al. [13] that MgO doping to the BST ceramics can make BST-MgO ceramics have lower dielectric constant with reasonable dielectric properties. The non-ferroelectric MgO with low dielectric constant and low dielectric loss can be used as the secondary phase to improve the dielectric properties. In addition, many researchers have reported that a little amount of Mn-doping to BST ceramics can significantly decrease the dielectric loss [14,15]. The substitution of Mn^{4+} ions to Ti^{4+} ions can prevent electrons hopping between different titanium ions, which finally decreased the dielectric loss.

In this study, we focused on the $\text{CaO-B}_2\text{O}_3\text{-SiO}_2$ (CBS) glass as low temperature sintering aids for $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ (BST)-based

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Table 1
Properties of BST ceramics with different amount of glass frit.

Content of glass frits (wt%)	Sintering temperature (°C)	Shrinkage (%)	Relative density (%)	ϵ_r (20 °C)	$\tan \delta$ (20 °C)
5	850	5.4	85	84	0.0097
	900	10.3	86	164	0.0018
	950	13.3	94	237	0.000998
10	850	7.9	89	74	0.0041
	900	11.5	93	126	0.004
	950	13.1	97	161	0.0014
15	850	9.5	93	64	0.0037
	900	11.3	95	89	0.0034
	950	12.9	98	135	0.0013
20	850	8.8	94	66	0.002
	900	11.3	96	88	0.0019
	950	12.4	95	95	0.0016

The dielectric properties were measured at 2 MHz and 20 °C.

ceramics. MgO was selected to improve the dielectric properties such as moderate dielectric constant and low dielectric loss. 1 mol% MnCO_3 was also added to the BST–MgO composites to further decrease the dielectric loss according to our previous experimental results. The relationships between the sintering temperatures, microstructure evolution and dielectric properties of BST–MgO composite ceramics with various amounts of glass additives are investigated.

2. Experimental

In this experiment, the $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ powders were firstly prepared by chemical co-precipitation method. Then the powders were calcined in alumina crucible at 750 °C for 4 h in air. For CBS glass, high purity powders of CaO, B_2O_3 , and SiO_2 were weighed according to the composition of 14% CaO–58% B_2O_3 –28% SiO_2 (in mol%). The powders were mixed, dried, and melted at 1200 °C for 1 h. The melt was quenched in water into fragments which were then pulverized and screened through an 80 mesh sieve to form glass. This glass frit has a density of 2.47 g/cm³. Judging from the high temperature microscope, the softening temperature where CBS glass starts to melt is about 560 °C. MgO powders were commercially purchased.

The composites of 55 wt% $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ –(45 wt%– x) MgO– x CBS glass ($x=5, 10, 15, 20$ wt%) with an addition of 1 mol% MnCO_3 were weighed and then ball-milled with ZrO_2 balls and alcohol for 4 h. After drying in an oven, the mixtures were granulated by adding 8 wt% methylcellulose binder. The powders were dry-pressed into pellets of 12 mm diameter and 0.5 mm height. These specimens were then debindered at 600 °C and sintered at 850–950 °C for 2 h.

The crystal phases were characterized by X-ray diffraction (XRD) via a Rigaku D/max 2500 diffractometer with Cu K α radiation. Surface morphologies and elements were analyzed by field emission scanning electron microscope (FE-SEM: Hitachi, S4800) and energy dispersive spectroscopy (EDS: EDAX Genesis XM2), respectively. The bulk densities (ρ_{bulk}) of sintered samples were measured by Archimedes method. The theoretical densities (ρ_{theo}) were calculated by the formula $1/\rho_{\text{theo}} = m\%/\rho_m + n\%/\rho_n + q\%/\rho_q$, where $m\%$, $n\%$, and $q\%$ are the mass fraction of BST, MgO, and CBS glass in the composite ceramics, ρ_m , ρ_n , and ρ_q are the theoretical densities of BST, MgO, and CBS glass, respectively. The relative densities (ρ_r) were calculated by the formula $\rho_r = \rho_{\text{bulk}}/\rho_{\text{theo}} \times 100\%$. The dielectric properties (dielectric constant and dielectric loss) of the samples were measured using Agilent E4980 LCR meter. The temperature dependences of dielectric constant were measured by high-temperature incubator tank and YY2813 LCR meter at 1 MHz. The temperature range was set from –55 to 125 °C.

3. Results and discussion

3.1. Sintering process and microstructure of BST–MgO–CBS composite ceramics

The properties of the BST–MgO–CBS composite ceramics with different amount of glass frit sintered at different temperatures are summarized in Table 1. It is clearly that for the samples added with 5 wt% CBS glass, the full densification was not obtained below 900 °C, which may be the result of insufficient liquid phase. When increasing the sintering temperature to 950 °C, full densification with the relative density of 94% was achieved. As the amount of CBS glass increased to 10 and 15 wt%, high densification was achieved at 900 °C and 850 °C, respectively. Further increasing the glass

content to 20 wt%, a moderate increase and then a slight decrease of the densities with increasing the sintering temperature was observed. The decrease of the relative density may due to the over-sintering of the composite ceramics and the formation of secondary phases. Therefore, appropriate addition of CBS glass is thought to play a crucial role in facilitating the densification of the composite ceramics. Moreover, the morphology of BST powders prepared by chemical co-precipitation method is shown in Fig. 1. The particles of BST are spherical in nature with sizes in the range of 80–100 nm, which are notably smaller than the particles prepared by conventional solid state method (the sizes are in the range of 1–2 μm). Besides, the particles of BST prepared by chemical co-precipitation method possess a large surface area of 15.32 m²/g, which is many times greater than the particles prepared by solid state method (the surface area is about 3.428 m²/g). Therefore, with high specific surface energy, the nanometer BST powders enabled the composite ceramics sintered densely at a relatively lower temperature.

Fig. 2 shows the XRD patterns of the BST–MgO–CBS composite ceramics which contain various glass contents and were sintered at 850 °C and 900 °C for 2 h. The main crystalline phases of all sintered samples are indexed to be a cubic perovskite BST phase and a cubic MgO phase. It was also found that the composites consisted of four crystalline phases: $\text{Ca}_4\text{Ti}_3\text{O}_{10}$ phase, Mg_2SiO_4 phase, $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$ phase and $\text{Ba}_2\text{TiSi}_2\text{O}_8$ phase. For 5 wt% CBS glass added composites, only the $\text{Ba}_2\text{TiSi}_2\text{O}_8$ phase and a small amount of Mg_2SiO_4 phase were detected besides the main BST and MgO crystalline phases. The formation of the $\text{Ba}_2\text{TiSi}_2\text{O}_8$ secondary phase was also reported by Divya [16] and Dou Zhang [3], which are

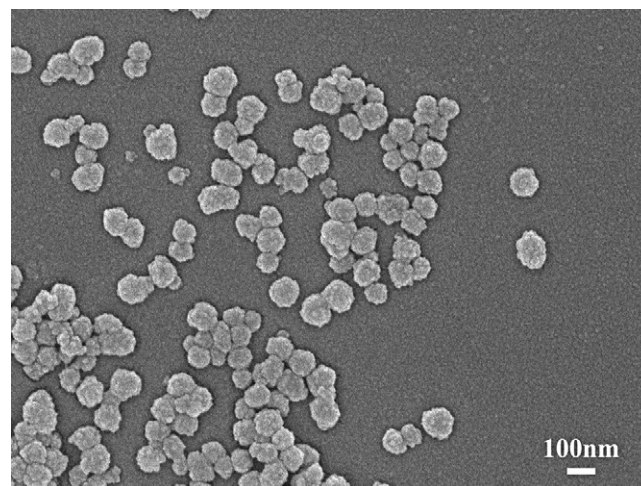


Fig. 1. SEM micrograph of BST powders prepared by chemical co-precipitation method.

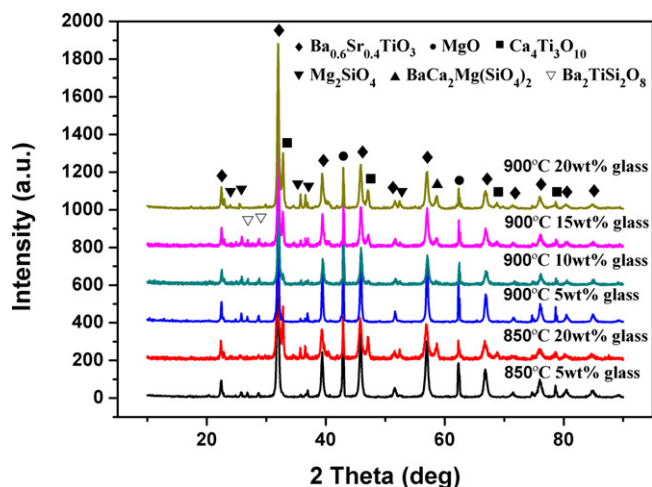


Fig. 2. XRD patterns of BST–MgO–CBS composite ceramics with different glass contents sintered at 850 °C and 900 °C for 2 h.

consistent with our results. According to their research, such phase in the BST-based ceramics would occur when borosilicate glass content was over 5 mol% and the firing temperature was above 850 °C. It is common for the formation of $\text{Ba}_2\text{TiSi}_2\text{O}_8$ secondary phase in the BST-based ceramics when introducing the glass composition containing SiO_2 [16–18]. With increasing the glass content, the secondary phases $\text{Ca}_4\text{Ti}_3\text{O}_{10}$ and $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$ were observed. The amount of the secondary phases $\text{Ca}_4\text{Ti}_3\text{O}_{10}$, $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$ and Mg_2SiO_4 increased with increasing the CBS glass content, while the amount of $\text{Ba}_2\text{TiSi}_2\text{O}_8$ phase decreased and cannot be detected at 20 wt% glass addition. Also, the X-ray diffraction intensities of BST and MgO reduced continuously. It is considered that at low glass contents, BST may firstly react with CBS glass and generate $\text{Ba}_2\text{TiSi}_2\text{O}_8$ phase. However, with further increasing the glass contents, $\text{Ca}_4\text{Ti}_3\text{O}_{10}$, $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$ and Mg_2SiO_4 secondary phases are more likely to be formed by the reaction of BST, MgO and CBS glass, which finally inhibited the formation of $\text{Ba}_2\text{TiSi}_2\text{O}_8$ phase. Besides, due to the formation of the secondary phases, the sintering process obviously belonged to reactive sintering. Therefore, it is believed that dynamic mechanism of the chemical reaction is also one of the factors in facilitating the sintering process. In addition, it was noted that the (1 1 0) diffraction peak of BST first shift slightly towards higher angle with increasing glass content to 10 wt%, and then shift back towards lower angle with further increasing the glass content to 20 wt%. The shift towards higher angle reveals that some calcium ions may enter into the BST lattice and substitute with Ba/Sr site. This is due to the smaller ionic radius of Ca^{2+} (1.340 Å, CN = 12) as compared to the ionic radius of Ba^{2+} (1.610 Å, CN = 12) and Sr^{2+} (1.440 Å, CN = 12). This substitution leads to the smaller lattice parameters of BST with the increase of the glass content to 10 wt%. However, further increase of the glass content would more likely to form the secondary phases $\text{Ca}_4\text{Ti}_3\text{O}_{10}$ and $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$, which consumed the CaO in the glass. Therefore, less Ca^{2+} will enter into the BST lattice, and finally leads to the observed shift of the (1 1 0) peak towards lower angle with the glass content varied from 10 to 20 wt%.

The SEM micrographs of the BST–MgO–CBS composite ceramics with various amounts of CBS glass sintered in the 850–950 °C temperature range are shown in Fig. 3. It is very clear that two component phases are co-existed. EDS analysis (which is not shown here) indicated that the dark grain contained primarily Mg, O elements, which means that the dark grain is MgO phase. The white grain contained primarily Ba, Sr, Ti, O elements, which means that the white grain is perovskite BST phase. With increasing the sintering temperature or the glass content, a kind of dark, columnar shape

grain can be observed. It was noted that with increasing the sintering temperature and the glass content, the amount of columnar grains increased. The element composition of the columnar grains was detected by the EDS analysis, which is shown in Fig. 4. Clearly it contained primarily Mg, O elements with a little amount of Ti, Ba elements. This implies that the columnar shape grain is mainly composed of MgO phase. In addition, B element was also detected by the EDS. Therefore, it is considered that with the introduction of B_2O_3 , MgO can orient growing to the columnar shape in the process of liquid phase sintering.

Typical microstructure evolution of the BST–MgO–CBS composite ceramics can also be seen from Fig. 3. The samples sintered at 850 °C with 5 wt% glass addition show an obvious porous microstructures. With increasing the glass contents, the amount of pores appeared to decrease. At 20 wt% glass content, a dense structure has been observed. Increasing the temperature by 50 °C greatly promoted the densification while the grain size still remained small. Further increase of the sintering temperature led to much denser microstructures and considerably increased grain sizes. This indicates the glass phase can provide the liquid when the sintering temperature is above 850 °C or the glass content is larger than 5 wt%, and enhance the densification of the composite ceramics at lower sintering temperatures through a liquid phase sintering mechanism. However, at 950 °C sintering temperature, too much glass contents may lead to the deterioration of the structures because of the formation of pores and large amount of secondary phases. Besides, the samples containing lower glass contents exhibited larger grain sizes than those of higher glass contents. This implies that the addition of CBS glass can inhibited grain growth of the composite ceramics. This is mainly because in the last period of liquid phase sintering, partial CBS liquid precipitated in the grain boundaries, which exerted pinning effect to the grain boundaries' movement, and finally inhibited the grain growth.

3.2. Dielectric properties of BST–MgO–CBS composite ceramics

The variation of dielectric constant values of BST–MgO–CBS composite ceramics with various contents of glass additives and sintering temperature is shown in Table 1. The dielectric constant of the samples exhibited similar trend of change to that of relative density with increasing sintering temperature. At 850 °C, due to the existence of pores, the composite ceramics had relatively low dielectric constant. With increasing the sintering temperature, the samples exhibited great enhancement of dielectric constant because of the increase of the relative density. At the same time, the dielectric constant decreased with increasing CBS glass content which is mainly attributed to the low dielectric constant of CBS glass and higher amount of secondary phases. For example, the dielectric constant of secondary phase Mg_2SiO_4 is 6.9, which is much smaller than the main crystalline phase. The observed secondary phase $\text{Ba}_2\text{TiSi}_2\text{O}_8$ is a low dielectric constant phase which has been discussed in many literatures [3,12,16]. Similar results have also been obtained by Qilong Zhang [5] on $\text{Ba}_2\text{Ti}_3\text{Nb}_4\text{O}_{18}$ ceramics with $\text{Li}_2\text{O}-\text{B}_2\text{O}_3-\text{SiO}_2$ glass additives and by Hsiang [19] on $\text{ZnO}-\text{B}_2\text{O}_3-\text{SiO}_2$ glass added BaTiO_3 ceramics.

The dielectric loss of BST–MgO–CBS composite ceramics with various contents of glass additives sintered at different temperatures is also shown in Table 1. For the samples sintered at 850 °C, the dielectric loss shows a dramatic decrease with increasing the glass contents, and is small at 20 wt% glass addition ($\tan \delta = 2.0 \times 10^{-3}$). This is mainly due to the absence of pores and increase of densification, which can be seen from the SEM micrographs. When increase the sintering temperature to 900 °C, the dielectric loss increased as the glass content increased from 5 to 10 wt%. This implies that the samples sintered at 900 °C can be well densified with 5 wt% glass addition. Further increase of the glass contents can lead to

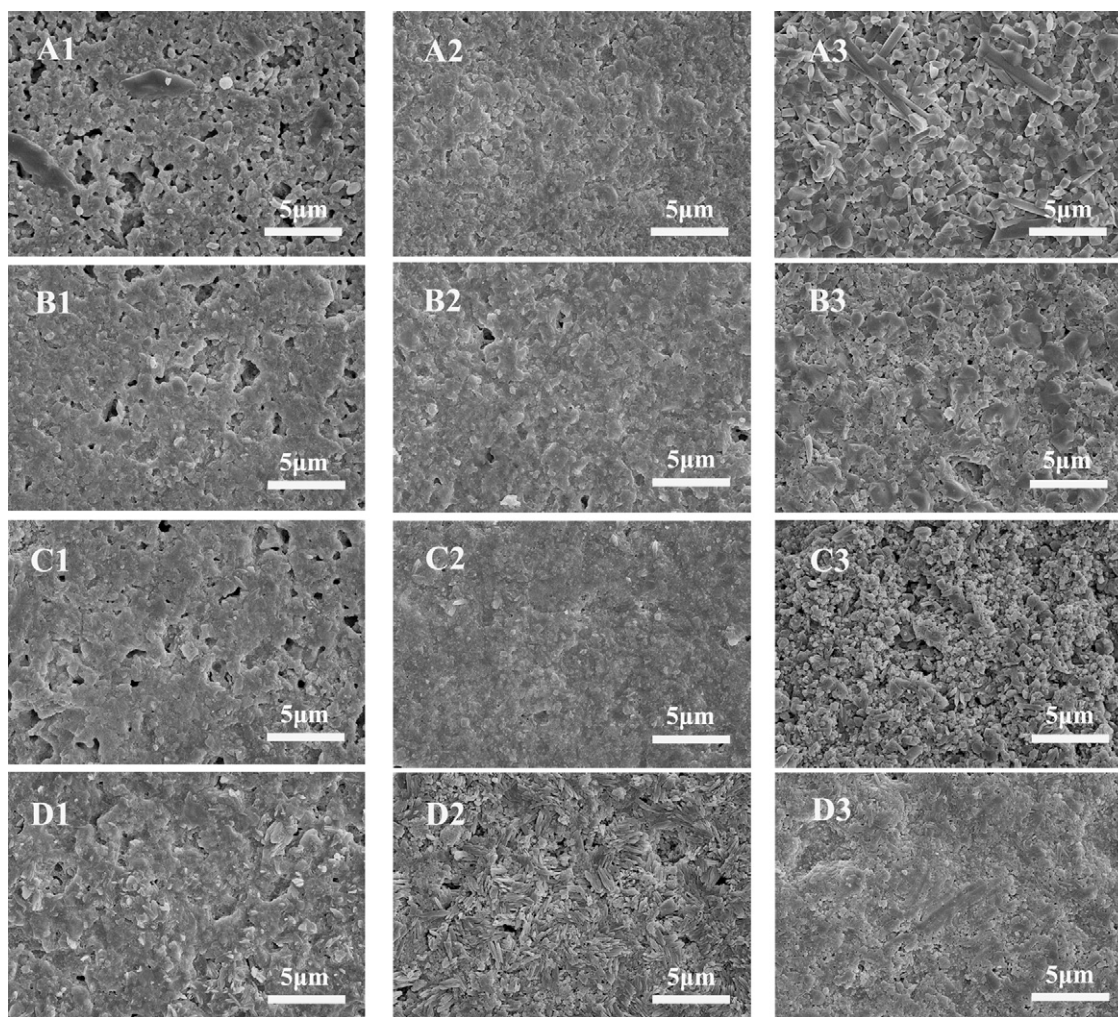


Fig. 3. SEM micrographs of the samples with various amounts of CBS glass sintered in the 850–950 °C temperature range. A1: 5 wt%, 850 °C; A2: 5 wt%, 900 °C; A3: 5 wt%, 950 °C; B1: 10 wt%, 850 °C; B2: 10 wt%, 900 °C; B3: 10 wt%, 950 °C; C1: 15 wt%, 850 °C; C2: 15 wt%, 900 °C; C3: 15 wt%, 950 °C; D1: 20 wt%, 850 °C; D2: 20 wt%, 900 °C; D3: 20 wt%, 950 °C.

the formation of pores and secondary phases, which is detrimental to the dielectric loss. On the contrary, the dielectric loss of the samples decreased with further increasing the glass contents from 10 to 20 wt%, which is probably related to the formation of a little low dielectric loss phase Mg_2SiO_4 ($\tan \delta = 2.5 \times 10^{-4}$). However, the

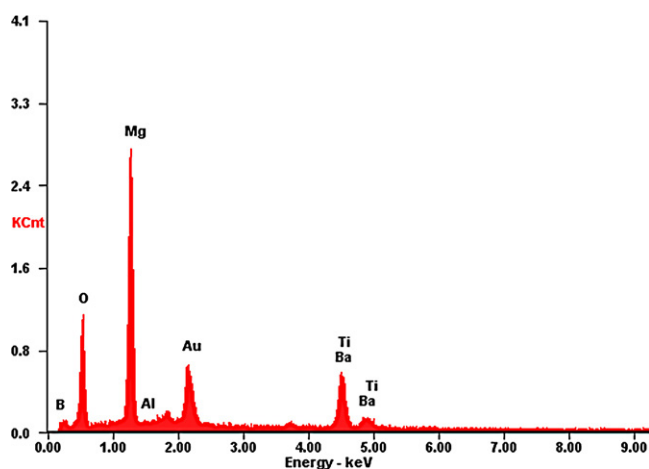


Fig. 4. EDS spectra of the samples with columnar grains.

dielectric properties of the other two secondary phases ($\text{Ca}_4\text{Ti}_3\text{O}_{10}$ and $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$) are not clear at present from the existing literature. Therefore, in order to better understand the dielectric behavior of the composite ceramic, further investigation on the nature of individual crystal phase will be necessary. In the case of the samples sintered at 950 °C, the dielectric loss revealed almost low constant value around 1.0×10^{-3} as the glass content increased from 5 to 15 wt% and then slightly increased to 1.6×10^{-3} as the glass content increased from 15 to 20 wt%. This was probably due to the increase of the secondary phases and a little over-sintering of the samples.

The temperature dependence of dielectric constant with different amount of glass frit sintered at different temperatures is shown in Fig. 5. With the increase of the glass content, the Curie temperature T_C shifted to lower temperature. The shift of T_C is attributed to the clamping effect on the ferroelectric crystallites by the glass matrix and the compositional change of the ferroelectric phases. With the addition of CaO contained glass frit, Ca^{2+} will substitute on the Ba/Sr site and result in the formation of regions of CaTiO_3 , which has lower T_C than that of BST.

The dielectric peaks of composite ceramics are suppressed and broadened with increasing the glass content. The sharp dielectric peak gradually becomes a diffuse peak with the increase of the glass content, which means that the diffuse phase transition is related strongly to the addition of the glass phase. The cations

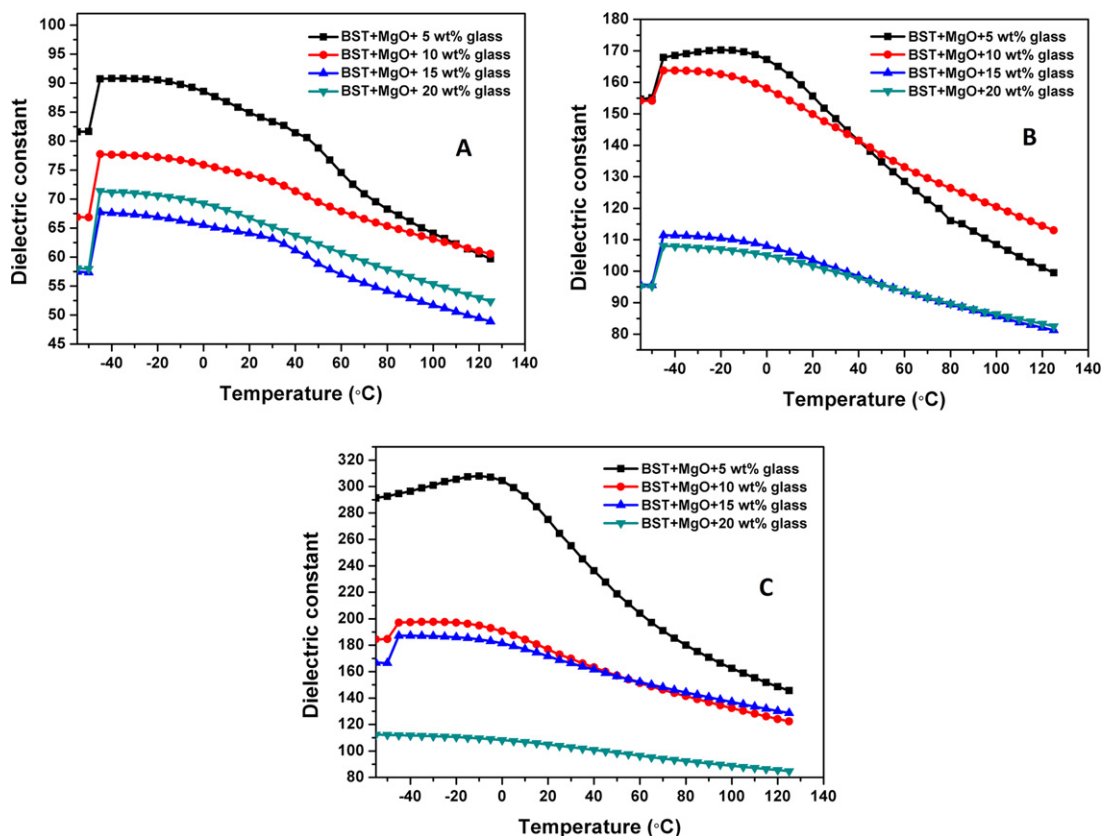


Fig. 5. Variation of dielectric constant for BST-MgO-CBS composite ceramics as a function of temperature with various contents of glass addition. Sintering temperature: A: 850 °C, B: 900 °C, C: 950 °C.

introduced with the glass phase can enter into BST lattices, leading to a disordered distribution of different ions on the equivalent crystallographic sites, and the observation of relaxor behavior and diffuse transitions. With increasing the amount of the glass content, the peaks of the secondary phases become stronger, as shown in Fig. 2 of the XRD result, and meanwhile, as shown in Fig. 3, the BST grain sizes reduced. Both the effect can lead to the decrease in dielectric constant and the broadening of the transition peak. More importantly, the substitution of Ca^{2+} in Ba/Sr site can generate an uneven distribution of BST composition, leading to an enhancement of the broadening effects.

In general, for each given composition, increasing the sintering temperature led to higher density of BST-MgO-CBS composite ceramics, and generally improved dielectric properties with higher dielectric constant and lower dielectric loss. At the same sintering temperature, higher glass contents can result in increased density of BST-MgO-CBS composite ceramics, and this effect is more obvious in the samples sintered at lower temperatures. Although higher glass contents are effective in promoting liquid phase sintering at low sintering temperatures and enhancing the densification of the BST-MgO-CBS composite ceramics, the dielectric properties deteriorate with high glass contents at high sintering temperatures. The samples with 20 wt% glass additives sintered at 850 °C show small dielectric loss $\tan \delta = 2.0 \times 10^{-3}$ and moderate dielectric constant $\epsilon_r = 66$. Its low sintering temperature and reasonable good dielectric properties make it a good candidate for LTCC applications.

4. Conclusions

In this study, 55 wt% $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ -(45 wt%- x) MgO- x CBS glass ($x=5, 10, 15, 20$ wt%) composite ceramics were prepared by solid-state method. The effects of $\text{CaO-B}_2\text{O}_3\text{-SiO}_2$ glass on the

sintering behavior, microstructures and dielectric properties of BST-MgO composite ceramics have been investigated for LTCC applications. The BST-MgO-CBS composite ceramics were sintered densely at 850 °C with glass content larger than 15 wt%, which was much lower than the sintering temperature of pure BST-MgO composite ceramics. The $\text{CaO-B}_2\text{O}_3\text{-SiO}_2$ glass played a crucial role in low temperature sintering. The ceramics obtained in this work had a very complicated phase composition. Besides the main phases $\text{Ba}_{0.6}\text{Sr}_{0.4}\text{TiO}_3$ and MgO, $\text{Ca}_4\text{Ti}_3\text{O}_{10}$, $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$, Mg_2SiO_4 and $\text{Ba}_2\text{TiSi}_2\text{O}_8$ secondary phases were also existed. The amount of the secondary phases $\text{Ca}_4\text{Ti}_3\text{O}_{10}$, $\text{BaCa}_2\text{Mg}(\text{SiO}_4)_2$ and Mg_2SiO_4 increased with increasing the CBS glass content, while the amount of $\text{Ba}_2\text{TiSi}_2\text{O}_8$ phase decreased. From the SEM images, by increasing the CBS glass content, grain sizes were decreased and crystalline properties were improved. With increasing the sintering temperature, a denser microstructure and increased grain sizes were observed. Besides, the dielectric constant and dielectric loss decreased as the amount of CBS glass increased. The strong broadening effects of the phase transition with increasing the glass contents have been detected. This is mainly due to the introduction of glass phase, the substitution of Ca^{2+} in Ba/Sr site and the formation of secondary phases. The BST-MgO composite ceramics with 20 wt% CBS glass additives sintered at 850 °C show a relative density of 94%, a moderate dielectric constant of 66, and a small dielectric loss of 2.0×10^{-3} , which made it a promising candidate for LTCC applications.

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