Sintering BaTi$_4$O$_9$/Ba$_2$Ti$_9$O$_{20}$-based ceramics by glass addition

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Abstract

After calcining BaCO$_3$+4TiO$_2$ mixed powder at 1150°C for 2 h, BaTi$_4$O$_9$ is the major phase, Ba$_4$Ti$_{13}$O$_{30}$ and BaTi$_5$O$_{11}$ are minor phases. After calcining 2BaCO$_3$+9TiO$_2$ mixed powder at 1200°C for 10 h, Ba$_2$Ti$_9$O$_{20}$ is the major phase, BaTi$_4$O$_9$ and BaTi$_5$O$_{11}$ are the minor phases. The calcining powders are used as the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ precursors. MgO–CaO–SiO$_2$–Al$_2$O$_3$ (MCAS) composite glass powder, fabricated by sol–gel method, is used as the low melting addition to lower the sintering temperatures of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ dielectric. The addition of MCAS glass does not inhibit the residual satellite phases to transform into BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$. For MCAS-doped BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics the major phases are BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$, cordierite is observed as minor phase. The temperatures needed to densify the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics are lowered down with the increase amount of MCAS glass addition. For BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics, both the densities and the dielectric constants (at 1 MHz) increase with the sintering temperature and decrease with the amount of MCAS glass addition.

Keywords: BaTi$_4$O$_9$; Ba$_2$Ti$_9$O$_{20}$; Calcination; Glass sintering aid; Sintering

1. Introduction

BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ were the two most common high-Q dielectric materials used in the microwave range. BaTi$_4$O$_9$ was first reported by Roy, and Ba$_2$Ti$_9$O$_{20}$ by Jonker, and they were investigated as microwave materials by O’Bryan and Plourde et al. The BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramic had superior dielectric properties for microwave resonator applications. For the preparation of monophase samples of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ from BaCO$_3$ and TiO$_2$ by conventional solid-state reaction, the stoichiometry must be precisely controlled, since there are various thermodynamically stable compounds in the vicinity of the desired composition of TiO$_2$-rich BaO–TiO$_2$ system: BaTi$_3$O$_7$, Ba$_2$Ti$_4$O$_9$, Ba$_3$Ti$_6$O$_{11}$, and Ba$_4$Ti$_7$O$_{10}$. In the study reported by Ritter et al., the pattern of BaO:TiO$_2$=1:4 was not completely formed BaTi$_4$O$_9$ until ~1300°C. Lu et al. also reported that the BaO:TiO$_2$=2:9 powder, obtained by sol–gel method, must be calcined at 1200°C for 110 h to produce Ba$_2$Ti$_9$O$_{20}$ sole phase. In this research, the solid-state reaction processes of BaCO$_3$ and TiO$_2$ to form BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ phase at different temperatures and times are developed, and our finding on the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ compounds have shown some degree of difference from those in the literature. In addition, a very high sintering temperature of ~1400°C was required to achieve densification of the ceramics. For that, it is imperative to lower the sintering temperatures of the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics in order to avoid the compositional fluctuation. Low melting glass additions, chemical processing, and smaller particle sizes of starting materials are three of the methods used to reduce the sintering temperature of dielectric. However, there are few references in the literature reporting the influence of glass addition on microwave properties of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ dielectric. Because of these reasons, it drew our attention to, and provided motivation to study, the effects of glass flux on the sintering behavior and properties of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ dielectric. MgO–CaO–SiO$_2$–Al$_2$O$_3$ (MCAS) composite glass, which is fabricated by sol–gel method, is used as the low melting glass addition to lower the sintering temperature of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ dielectric. The BaO:TiO$_2$=1:4 and
BaO:TiO$_2$ = 2:9 powders used in this study do not form BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ completely, and the satellite phases of BaTi$_3$O$_6$ and Ba$_2$Ti$_5$O$_{11}$ are still residual in the using powders. It is found that the addition of MCAS glass does not inhibit the residual satellite phases to form the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$. Relationships among the sintering temperatures, phase formation, and the dielectric properties of MCAS-fluxed BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics are presented.

2. Experimental procedure

In the present investigation, a homogeneous glass of composition containing (in wt%) MgO5%, CaO19%, Al$_2$O$_3$26%, and SiO$_2$50% (abbreviated as MCAS) was prepared by the sol–gel method. In a typical laboratory scale synthesis using the nitrates, 40% colloidal silica was dispersed in 600 ml of deionized water, and concentrated nitric acid was also added into the solution. To this acidic suspension we added magnesium nitrate hexahydrate, calcium nitrate hexahydrate, and aluminum nitrate hexahydrate. The subsequent addition of ammonium hydroxide resulted in the quantitative precipitation of magnesium, calcium, and aluminum hydroxides. Then the glass powder was formed in the solide format. The solids were collected by filtration and calcined at 300°C for 1 h. The calcination step was desirable to convert any ammonium nitrate present to oxides of nitrogen and water. The resulting material was the MCAS glass precursor.

Proportionate amounts of reagent-grade starting materials of BaCO$_3$ and TiO$_2$ were mixed, according to the composition 2BaCO$_3$+9TiO$_2$ and BaCO$_3$+4TiO$_2$ and ball-milled for 5 h with deionized water. After drying, the reagent was ground with an agate mortar for 1 h. Then the powders were calcined at different temperatures and times. The crystal structures of calcined powders were examined by using an X-ray powder diffractometer. The Ba$_2$Ti$_5$O$_{20}$ precursor (calcined at 1200°C for 10 h) and BaTi$_4$O$_9$ precursor (calcined at 1150°C for 2 h) were mixed with 5 wt% or 10 wt% MCAS glass by ball milling with deionized water for (1 h. After drying, the powder was pressed into pellets uniaxially in a steel die. Typical dimensions of the pellets were 15mm in diameter and 1.5mm in thickness. Sintering of these Ba$_2$Ti$_5$O$_{20}$ pellets was carried out at a temperature between 1160 and 1360°C and BaTi$_4$O$_9$ pellets between 1150 and 1350°C under ambient conditions for a duration of 4 h. Crystallization of the sintered MCAS–Ba$_2$Ti$_5$O$_{20}$ and MCAS–BaTi$_4$O$_9$ ceramics were also investigated by X-ray diffraction patterns. Density of the sintered specimens as a function of sintering temperature was measured by the liquid displacement method using deionized water as a liquid (Archimedes method). After painting silver paste on both sides of the specimens and fired at 700°C for about 20min, dielectric constants were measured with HP4192a impedance analyzer at 1 MHz.

3. Results and discussion

The X-ray diffraction patterns of calcined BaO:TiO$_2$ = 1:4 powders are shown in Fig. 1. Various phases were observed in BaO:TiO$_2$ = 1:4 oxide system. The conventional solid-state-reaction process for synthesizing BaTi$_4$O$_9$ compounds was commonly based on the following reaction:

$$\text{BaCO}_3 + 4\text{TiO}_2 \rightarrow \text{BaTi}_4\text{O}_9 + \text{CO}_2$$ (1)

However, the real solid reaction processes were usually more complex, and some satellite reactions might occur. Calcining at 1100°C for 2 h [Fig. 1(a)] the BaTi$_4$O$_9$ phase was formed, but the TiO$_2$ and satellite phases of BaTi$_5$O$_{11}$ and Ba$_4$Ti$_{13}$O$_{30}$ were also revealed. Proceeding calcination at 1150°C for 2 h [Fig. 1(b)] and 10 h [Fig. 1(c)] the BaTi$_4$O$_9$ was not the only phase, because the satellite phases of Ba$_4$Ti$_{13}$O$_{30}$ and BaTi$_5$O$_{11}$ were residual. Even though 1200°C and 10 h [Fig. 1(d)] were used as the calcining condition, the Ba$_4$Ti$_{13}$O$_{30}$ and BaTi$_5$O$_{11}$ phases were still residual. This suggests that the satellite phases of Ba$_4$Ti$_{13}$O$_{30}$ and Ba$_4$Ti$_{13}$O$_{30}$ are formed before the BaTi$_4$O$_9$ phase.

$$\text{BaCO}_3 + 5\text{TiO}_2 \rightarrow \text{BaTi}_5\text{O}_{11} + \text{CO}_3$$ (2)

$$4\text{BaCO}_3 + 13\text{TiO}_3 \rightarrow \text{Ba}_4\text{Ti}_{13}\text{O}_{30} + 4\text{CO}_3$$ (3)

![Fig. 1. The X-ray patterns of BaTi$_4$O$_9$ calcined powders (a) 1100°C for 2 h, (b) 1150°C for 2 h, (c) 1150°C for 10h, (d) 1200°C for 10 h, and (e) 1200°C for 70h (A: BaTi$_4$O$_9$, D:BaTi$_5$O$_{11}$, E :Ba$_4$Ti$_{13}$O$_{30}$, T : TiO$_2$).](image-url)
Prolonging the calcining time at 1200°C for 70 h [Fig. 1(e)], only the BaTi₄O₉ was revealed. Ritter et al. reported that the BaO:TiO₂ = 1:4 revealed a two phase mixture of Ba₂Ti₉O₂₀ and BaTi₅O₁₁ at 700°C and a two phase mixture of Ba₄Ti₁₃O₃₀ and BaTi₅O₁₁ at 1000°C. Javadpour et al. reported the X-ray data obtained on the BaCO₃ + 4TiO₂ compound heated at 700°C for 4 h showed the presence of BaTi₅O₁₁ in addition to the lines for the BaTi₄O₉ phase. However, the small amount of BaTi₅O₁₁ phase observed by Raman spectroscopy was not detectable by X-ray powder diffraction experiments carried out on samples heated above 1000°C. Our finding on this compound showed some degree of difference from those reported by Ritter et al. and Javadpour et al.

These reports and the results in this study suggest that reaction Eq. (1) and the following reaction may be processed together:

\[
\begin{align*}
\text{BaTi₅O₁₁} & \rightarrow \text{BaTi₄O₉} + \text{TiO}_2 \\
\text{Ba₄Ti₁₃O₃₀} + 3\text{TiO}_2 & \rightarrow 4\text{BaTi₄O₉} \\
or \quad 3\text{BaTi₅O₁₁} + 4\text{Ba₄Ti₁₃O₃₀} & \rightarrow 4\text{BaTi₄O₉}
\end{align*}
\]

In this study, the powder calcined at 1150°C for 2 h was used as the initial BaTi₄O₉ powder. The X-ray diffraction patterns from the as-sintered surface of MCAS–BaTi₄O₉ ceramics as a function of sintering temperature and MCAS content are shown in Fig. 2. It has been reported that in the sintered MCAS glass existed the anorthite and cordierite as the two main crystalline structures. As 0 wt%-MCAS-added BaTi₄O₉ ceramics was sintered at 1200°C [Fig. 2(a)] and 1300°C [Fig. 2(b)], the major phase was the BaTi₄O₉. For 5 and 10 wt%-MCAS-added BaTi₄O₉ ceramics and sintered at 1200°C, the major phase was BaTi₄O₉, and the minor phase was cordierite [Fig. 2(c) and (d)]. The residual Ba₄Ti₁₃O₃₀ did not appear in the sintered MCAS–BaTi₄O₉ ceramics. These results suggest that MCAS does not inhibit the residual BaTi₅O₁₁ and Ba₄Ti₁₃O₃₀ to form BaTi₄O₉. Comparing Fig. 2(c) and (d), the diffraction intensity of cordierite increased and the diffraction intensity of BaTi₄O₉ decreased slightly as the amount of MCAS addition increased. These results also suggest that MCAS glass can really be used as a sintering aid of BaTi₄O₉ ceramics, because the MCAS glass improves the residual satellite phases to transform into BaTi₅O₁₁.

The X-ray diffraction patterns of calcined Ba₂Ti₉O₂₀ powders are shown in Fig. 3. Furthermore, various phases were also observed in BaO–TiO₂ rare earth oxide system. The conventional solid-state-reaction process for synthesizing Ba₂Ti₉O₂₀ compounds were commonly based on the following reaction:

\[
\begin{align*}
2\text{BaCO}_3 + 9\text{TiO}_2 & \rightarrow \text{Ba}_2\text{Ti}_9\text{O}_{20} + 2\text{CO}_2
\end{align*}
\]

However, the real solid reaction processes are usually more complex, and some satellite reactions might also occur. As indicated in Fig. 3(a) (1150°C, 2 h) and Fig. 3(b) (1200°C, 2 h), at shorter calcining time BaTi₅O₁₁ is detected as the major phase, BaTi₄O₉ and TiO₂ as minor phases. At longer calcining time, as indicated in Fig. 3(c) (1150°C, 10 h) and Fig. 3(d) (1200°C, 10 h), the Ba₂Ti₉O₂₀ phase and satellite reaction phases of BaTi₄O₉ and BaTi₅O₁₁ coexist. This suggests that the BaTi₄O₉ and BaTi₅O₁₁ phases are formed before the Ba₂Ti₉O₂₀ phase.

\[
\begin{align*}
\text{BaCO}_3 + 4\text{TiO}_2 & \rightarrow \text{Ba}_2\text{Ti}_9\text{O}_{20} + \text{CO}_2
\end{align*}
\]
BaCO₃ + 5TiO₂ → BaTi₅O₁₁ + CO₂

O’Bryan and Thomson prepared BaTi₅O₁₁ as the major phase in a mixture of BaTi₄O₉ and rutile TiO₂. Ritter et al. reported that the synthesis of Ba₂Ti₉O₂₀ did not form until BaTi₅O₁₁ began to decompose. They also reported that if a mixture of barium and titanium species was hydrolyzed within 5 min after mixing, a precursor was produced which was disposed to form Ba₂Ti₉O₂₀, BaTi₅O₁₁, and BaTi₄O₉ when heated at 1100°C; prolonged heating can convert all of this material to Ba₂Ti₉O₂₀. Javadpour and Eror found that at 1200°C, the Ba:Ti = 1:5 powders converted to BaTi₅O₁₁ and BaTi₄O₉ only after extensive heat treatment at 1200°C. Ritter et al. also reported that if a mixture of barium and titanium alkoxide precursors that Ba₂Ti₉O₂₀ did not begin to form until BaTi₅O₁₁ began to decompose. However, prolonged calcining time, as Fig. 3(e) (1200°C, 70 h) indicates, converts all of this material to Ba₂Ti₉O₂₀. These reports and the results in this study suggest that the following reaction may be processed together:

2BaTi₅O₁₁ → Ba₂Ti₉O₂₀ + TiO₂

2BaTi₄O₉ + TiO₂ → Ba₂Ti₉O₂₀

or BaTi₄O₉ + BaTi₅O₁₁ → Ba₂Ti₉O₂₀

In order to eliminate such satellite BaTi₄O₉ and BaTi₅O₁₁ phases from the Ba₂Ti₉O₂₀ calcination, a higher calcining temperature (≥1200°C) and longer calcining time (≥ 10 h) were necessary. The calcining time was more important than the calcining temperature.

The X-ray diffraction patterns from the as-sintered surface of MCAS–Ba₂Ti₉O₂₀ ceramics as a function of sintering temperature and MCAS content are shown in Fig. 4. In MCAS glass the anorthite and cordierite existed as the two main crystalline structures and the used Ba₂Ti₉O₂₀ precursor contained the satellite phases of BaTi₄O₁₁ and BaTi₄O₉, therefore, phase control was expected to be extremely difficult in such a system. As undoped Ba₂Ti₉O₂₀ precursor was sintered at 1260°C [Fig. 4(a)], the major phase was the Ba₂Ti₉O₂₀ and the BaTi₅O₁₁ phase is still residual. As 1340°C is used as the sintering temperature, the only crystalline phase is Ba₂Ti₉O₂₀ [Fig. 4(b)]. The MCAS–Ba₂Ti₉O₂₀ system prepared here was composed of one mainly crystalline phase, Ba₂Ti₉O₂₀. Except Ba₂Ti₉O₂₀ the cordierite was observed as minor phase [sintered at 1260°C, Fig. 4(c) and 4(d) for 5 wt% and 10 wt% MCAS addition, respectively]. Comparing Fig. 4(c) and 4(d), the diffraction intensity of cordierite increased and the diffraction intensity of Ba₂Ti₉O₂₀ decreased slightly as the amount of MCAS addition increased. As Fig. 4(a)–(d) shows, the satellite phases of BaTi₄O₉ and BaTi₅O₁₁ did not appear in the undoped and MCAS-added Ba₂Ti₉O₂₀ ceramics. These results suggest that MCAS glass does not inhibit the proceeding of reactions Eqs. (10)–(12).

The X-ray diffraction patterns showed that cordierite phase was independent to the Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ phase, the influences of MCAS glass on the lattice constant and dielectric constant of Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ ceramics were unavoidable. The lattice constants of undoped and MCAS-added Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ are shown in Fig. 5. and plotted versus the sintering temperature. The plot shows that the lattice parameters of the undoped Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ ceramics were almost unchanged with sintering temperature. The lattice parameters of Ba₂Ti₉O₂₀ ceramics were calculated to give the following values: a = 1.453 nm, b = 0.380 nm, and c = 0.630 nm, and the lattice parameters of Ba₂Ti₉O₂₀ ceramics were calculated to give the following values: a = 1.4815 nm, b = 1.4285 nm, and c = 0.7107 nm. The MCAS-added Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ ceramics were isostructural with undoped Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ ceramics but had a slight difference in lattice parameters. The a and b parameters of MCAS-added Ba₂Ti₉O₂₀ and Ba₂Ti₉O₂₀ ceramics increase but c parameters slightly decreased with the sintering temperature and amount of MCAS glass addition.

SEM micrographs of MCAS–Ba₂Ti₉O₂₀ (sintering at 1200°C) and MCAS–Ba₂Ti₉O₂₀ ceramics (sintering at 1260°C) are shown in Fig. 6. Without the addition of MCAS glass, pores are easily observed in Fig. 6(a) for Ba₂Ti₉O₂₀ ceramics and Fig. 6(d) for Ba₂Ti₉O₂₀ ceramics. As 5 and 10 wt% MCAS glass are added in Ba₂Ti₉O₂₀ ceramics, almost no pores are observed.
The variation of the density of the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics against the sintering temperature and glass content are shown in Table 1. As Table 1 shows, the densities of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics increase with the increase of sintering temperature. The theoretical densities for the MCAS–BaTi$_4$O$_9$ and MCAS–Ba$_2$Ti$_9$O$_{20}$ ceramics were calculated using Eq. (13):

\[ D = \frac{W_1}{D_1} + \frac{W_2}{D_2} \]

where $W_1$ was the wt% of the BaTi$_4$O$_9$ or Ba$_2$Ti$_9$O$_{20}$ and $W_2$ was the wt% of MCAS glass, respectively; $D_1$ was the density of the BaTi$_4$O$_9$ or Ba$_2$Ti$_9$O$_{20}$ and $D_2$ was the density of MCAS glass, respectively. In estimating the density of MCAS-added Ba$_2$Ti$_9$O$_{20}$ ceramics, measured values for MCAS = 2.58 g/cm$^3$, $^{11}$ BaTi$_4$O$_9$ = 4.55 g/cm$^3$, $^{13}$ and Ba$_2$Ti$_9$O$_{20}$ = 4.58 g/cm$^3$ were used. If the MCAS glass was continuous rather than dispersed in the BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics, Eq. (11) could be used to predict a composite density. The estimated theoretical densities were 4.390 and 37.8 for undoped Ba$_2$Ti$_9$O$_{20}$ (sintered at 1340 or 1380°C) and BaTi$_4$O$_9$ ceramics (sintered at 1300°C). Several investigators had studied the electrical properties of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics, $^{10,13}$ there was little variation for undoped BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics in the $K$ values. The temperatures needed to obtain saturation dielectric constants were shifted to lower temperatures and the saturation values were decreased as the amount of MCAS glass increased. MCAS glass addition was found to have an effect on the dielectric constant. The dielectric constant of MCAS glass was measured to between 5.8 and 6.1 (at 1 MHz), the addition of MCAS glass will lower the dielectric constant of BaTi$_4$O$_9$ and Ba$_2$Ti$_9$O$_{20}$ ceramics in the $K$ values. The temperatures needed to obtain saturation dielectric constants were shifted to lower temperatures and the saturation values were decreased as the amount of MCAS glass increased.
Fig. 6. The micrographs for BaTi₄O₉ sintered at 1200°C with (a) 0 wt%, (b) 5 wt%, and (c) 10 wt% MCAS glass added and for Ba₂Ti₉O₂₀ sintered at 1260°C with (d) 0 wt%, (e) 5 wt%, and (f) 10 wt% MCAS glass added.
about 39.2 for undoped \( \text{Ba}_2\text{Ti}_9\text{O}_{20} \) dielectric to about 32.2 for \( \text{Ba}_2\text{Ti}_9\text{O}_{20} \) ceramics with 10wt% MCAS glass added. The dielectric constant of densified MCAS–Ba\(_2\)Ti\(_4\)O\(_9\) ceramics decreased from 37.8 for Ba\(_2\)Ti\(_4\)O\(_9\) ceramics to 30.5 for 10wt%-MCAS-added Ba\(_2\)Ti\(_4\)O\(_9\) ceramics.

4. Conclusions

The reaction process of BaO:Ti\(_2\)O\(_3\) = 1:4 and BaO:Ti\(_2\)O\(_3\) = 2:9 powders and the sintering characteristics of Ba\(_2\)Ti\(_4\)O\(_9\) and Ba\(_2\)Ti\(_9\)O\(_{20}\) ceramics with different amounts of MCAS glass addition have been developed in this study, and several conclusions are deduced as follows:

1. When BaO:Ti\(_2\)O\(_3\) = 1:4 at is calcined at 1150°C for 2 h and BaO:Ti\(_2\)O\(_3\) = 2:9 at 1200°C for 10h, the Ba\(_2\)Ti\(_4\)O\(_9\) and Ba\(_2\)Ti\(_9\)O\(_{20}\) are not formed completely, and the satellite phases are residual. The addition of MCAS composite glass does not inhibit the residual satellite phases to transform into Ba\(_2\)Ti\(_4\)O\(_9\) or Ba\(_2\)Ti\(_9\)O\(_{20}\).

2. The densities of MCAS-Ba\(_2\)Ti\(_9\)O\(_{20}\) and MCAS-Ba\(_2\)Ti\(_4\)O\(_9\) ceramics increase with sintering temperature but decrease with the amount of MCAS glass addition. The density curves shift to lower temperature as the amount of MCAS glass addition increases.

3. The dielectric constants of Ba\(_2\)Ti\(_4\)O\(_9\) and Ba\(_2\)Ti\(_9\)O\(_{20}\) ceramics increase with the sintering temperature and decrease with the increased amount of MCAS glass.

References