

Middle- and high-permittivity dielectric compositions for low-temperature co-fired ceramics

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Abstract

Various middle- and high-permittivity dielectric material systems for LTCC were examined. By designing several borosilicate glass frit systems carefully, we tried to lower the temperature of densification to 875 °C in selected host dielectric materials still maintaining acceptable dielectric properties. The effects of glass addition on the densification, electrical properties, and phase changes in the host dielectric materials were examined. © 2006 Published by Elsevier Ltd.

Keyword: LTCC; Sintering; Dielectric properties; Glass ceramics

1. Introduction

Low-temperature co-fired ceramics (LTCC) technology has become essential in the development of various multifunctional modules and substrates, especially in wireless and/or high frequency applications.^{1–3} It is important to incorporate passive components such as resistors, capacitors, inductors, and other functional parts in LTCC multilayer structures in order to achieve highly integrated and multi-functional LTCC modules.^{1,4,5} Among the components which could be realized inside the LTCC packages, the resonators and the internal capacitors are most desirable in the current state of the art technology. The resonator structures, which are typical and/or important, consist of planar antenna and filters. Considering the RF frequency range in the current telecommunication system (1–30 GHz) and the typical chip sizes in the packaging technologies (2–10 mm), the most appropriate permittivity (K) range of the materials is 20–100 to realize strip or microstrip resonator structures in LTCC multilayer structures.⁶ Meanwhile, the internal capacitors are typically used to realize decoupling capacitors monolithically in the LTCC package. As for the capacitor applications, the high- K dielectric materials are desirable as long as their losses are within an acceptable range.

There have been several approaches to develop the middle- K ($K=20$ –100) and high- K ($K>500$) dielectric material systems which could be fired at below 900 °C. Although some middle- K dielectric compositions have been reported to exhibit the low densification temperature without any additives,^{7–9} it is more general and is considered an easy method to add glass frits into the host dielectric compositions to realize the acceptable densification at the low-temperature range of below 900 °C together with good electrical properties.^{10–13} The several microwave dielectric compositions including (Zr,Sn)TiO₄,¹² BaO–TiO₂–WO₃,¹³ and BaO–Re₂O₃–TiO₂ (Re: La, Sm, Nd, Eu) systems^{14,15} have been studied for the development of the middle- k dielectric compositions for LTCC by using glass frits. For examples, Takada et al.¹² reported that (Zr,Sn)TiO₄ ceramic with reactive ZnO–B₂O₃–SiO₂ glass with 5 wt.% was sintered at 1100 °C and had $k=20$ and $Q=2100$ at a frequency of 8 GHz. Cheng et al.¹⁴ reported that Ba–Nd–Sm–Bi–Ti–O ceramic was sintered at 950 °C by using the BaO–B₂O₃–SiO₂ glass with 5 wt.% and had $k=40$ and $Q \times f \sim 3000$ GHz. However, the temperature of densification was higher than 900 °C, which is still far too high for cofiring of Ag inner electrode.

In this work, we aimed at developing some middle- and high- K dielectric material systems for LTCC by mixing the host dielectric material systems and borosilicate glass frit systems which were carefully designed. We controlled the glass composition and processing parameters so as to lower the densification temperature to 875 °C, while still maintaining acceptable dielec-

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tric properties. The effects of glass addition on the densification and electrical properties were examined.

2. Experimental procedure

The glass compositions were designed with a glass simulation software (SciGlass V3.5, Scivision, USA) in consideration of physical and electrical properties. The designed glass compositions were then fabricated by a conventional glass fabrication process. Powders of SiO₂, B₂O₃, Li₂O, CaO, and Al₂O₃ were used as starting materials for glass fabrication. Through the melting of starting powders in a Pt crucible at the temperatures of 1100–1300 °C and the quenching with a quench roller, glass flakes were prepared. After the ball-milling of the flakes, the average particle size of the glass powders was about 1 μm. The physical and electrical properties of the glass frits were measured from the glass ingots quenched, as summarized in Table 1.

We selected some host dielectric materials which have been reported to exhibit good dielectric properties enough to be applied for LTCC material systems.^{7–15} They may be categorized as $K=20–100$ and $K>500$. The detailed compositions, together with their sintering and dielectric properties which are measured from our preliminary studies, are summarized in Table 2. The dielectric compositions were fabricated by a solid state sintering process from the oxide forms of reagents (BaO, TiO₂, ZnO, MgO, ZrO₂, Nb₂O₅, CaO, PbO. All >99.9%, High Purity Chem.). Some of the compositions were the formulated powders from the commercial supplier. In our preliminary experiments, all the dielectric compositions could be densified at temperatures higher than 1300 °C and exhibit the relative density of more than 98% with good dielectric properties, without the addition of glass, as shown in Table 2.

For low-temperature sintering, the host dielectric compositions and 3–25 wt.% of glass frit were mixed by ball-milling for 24 h. The slurry was dried at 90 °C for several hours and granulated with 3 wt.% of a PVA (poly-vinyl alcohol) solution.

Table 1

The physical and electrical properties of the glass frit series developed in this study

	MA series (mol%)	MB series (mol%)	HA series (mol%)
Chemical composition			
LiO ₂	33–51	7–50	45–61
B ₂ O ₃	23–43	18–30	30–49
SiO ₂	10–35	12–47	5–15
CaO + AbO ₃	–	2–10	–
Physical properties			
Density (g/cm ³)	2.23–2.68	2.31–2.52	2.35–2.52
$\alpha_{RT-400\text{ }^\circ\text{C}}$ ($\times 10^{-6}\text{ }^\circ\text{C}^{-1}$)	8.3–14.3	6.3–12.5	10.1–15.3
Glass transition point, T_g (°C)	420–503	371–575	408–450
Glass softening point, T_s (°C)	437–524	390–627	450–480
Dielectric properties			
Permittivity ^a	6.5–8.5	7.0–9.0	7.5–9.5
$Q \times f$ (GHz) ^a	1000–1600	1200–2300	1200–1900
τ_f (ppm/°C) ^a	–80 to –160	–83 to –223	–103 to –153
Tan d (%) ^b	0.4–0.6	0.2–0.5	0.6–0.9

^a Dielectric properties were measured using a network analyzer in the frequency range of 12–16 GHz.

^b Tan d were measured using an impedance/gain-phase analyzer at 1 MHz.

The granulated powders were pressed isostatically into pellets at 100 MPa and sintered in the temperature range of 800–950 °C for 2 h in order to evaluate densification behavior.

Microwave dielectric properties were measured by two kinds of methods, viz. the parallel plate resonator method and the cavity resonator method in conjunction with a network analyzer (8720C, HP, USA) in the s₂₁ transmission mode examined with a certain frequencies ranging from 8 to 12 GHz. Measurement of dielectric constants mainly hinges on the Hakki–Coleman method with a silver plate followed by an employment of the TE₀₁₁ resonance mode.¹⁶ Unloaded Q values were calculated

Table 2

The physical and electrical properties of the host dielectric materials used in this study

Permittivity range	Abbreviation	Malarial	Sintering properties			Dielectric properties		
			Optimum sintering temperature (°C)	Relative density (%)	Linear shrinkage (%)	Permittivity	$Q \times f$ (GHz)	τ_f (ppm/°C)
Middle- k ($20 < k < 100$)	MCT	(Mg, Ca)TiO ₃	1350	98.7	15.7	21.6	45000	–3
	CZ	CaZrO ₃	1500	99.5	15.8	28.5	14000	–20
	ZST	(Zr,Sn)TiO ₄	1350	99.2	16.1	38.1	52000	–11
	BT4 ^a	BaTi ₄ O ₉	1350	98.7	16.9	38.6	44500	+3
	B5N4	Ba ₅ Nb ₄ O ₁₅	1375	98.9	15.3	41.5	25000	+68
	BBNT ^b	BaO–Bi ₂ O ₃ –Nd ₂ O ₃ –TiO ₂	1300	98.8	16.3	91.2	6100	+6
	BPNT	(Ba _{0.5} Pb _{0.5})O·Nd ₂ O ₃ ·5TiO ₂	1300	99.5	15.8	94.7	5600	+8
High- k ($k > 500$)	BT ^c	BaTiO ₃	1350	99.8	15.8	>2000 ^e	1.9% ^f	–
	BCTZ ^d	(Ba _{0.95} Ca _{0.05})(Ti _{0.88} Zr _{0.12})O ₃	1260	99.7	16.5	>3000 ^e	2.5% ^f	–

^a Commercial powder (MWF-38, Hayashi Chemical Industry, Co. Ltd., Japan).

^b Commercial powder (MBRT-90, Fuji Titanium Industry, Co. Ltd., Japan).

^c Commercial powder (Ferro, Co. Ltd., USA).

^d Samsung electro-mechanics provided for scientific purposes (not commercial).

^e Permittivity was measured using an impedance/gain-phase analyzer at MHz.

^f Tan d was measured using an impedance/gain-phase analyzer at MHz.

from the loaded Q values. The loaded Q values were measured at frequency ranging from 8 to 12 GHz via the transmission open-cavity method accompanied by a copper cavity. The value of τ_f was measured at temperature ranging from +25 to +80 °C by the open-cavity method employing an invar cavity in a chamber. For the high- K dielectric compositions, the low-frequency dielectric properties were measured by an impedance analyzer (4194, HP, USA) at 1 MHz.

3. Results and discussion

3.1. The properties of glass frits

To lower the densification temperature of the host dielectric materials below 900 °C maintaining acceptable dielectric properties, it is important to design the glass compositions which have lower glass transition temperature (T_g) and dielectric loss. Throughout repeated simulations and experiments as illustrated in Fig. 1, we found that the glass composition should have T_g below 500 °C to guarantee the densification temperature of below 875 °C. Meanwhile, the dielectric loss of the glass frits should be minimized to get the acceptable dielectric properties in LTCC compositions. In these regards, SiO₂–B₂O₃–Li₂O-based glass compositions were most appropriate in our simulations and measurements. Furthermore, we modified glass compositions by adding other oxide components such as CaO and Al₂O₃ so as to control their glass properties and electrical properties.

Among those tested, we selected three typical varieties of glass frits for this study, as listed in Table 1. The features of low permittivity, high quality factor, low glass transition temperature (T_g) are desirable, in general, for the role of sintering additives for host dielectrics. In this regard, the MA series and MB series in Table 1 were the most appropriate glass frits for the densification of middle-permittivity dielectric compositions. The MB series, which have some CaO and Al₂O₃ additionally, show higher permittivity and quality factors, whereas they

exhibit a higher glass transition temperature (T_g) also. Thus, the MA series displays a higher capacity for low-temperature sintering, whereas the MB series exhibits better electrical properties. The HA series, which consists of higher Li₂O content compared to the MA and MB series, were proven to have the highest capacity for low-temperature sintering. However, this series exhibit rather high dielectric losses and large negative values of τ_f . The added amount of frits should be minimized in the sintering of high- K dielectrics to prevent the degradation of permittivity. Thus, this HA series were most appropriate for sintering of high- K dielectrics.

3.2. The properties of host dielectric materials

For the development of the middle- K and high- K LTCC material systems, it is necessary that the materials should be densified at below 900 °C. In this study, we tested a good number of microwave dielectric materials and ferroelectric materials which have permittivities of 20–2000 for the middle- K and high- K LTCC material systems. We found that the dielectric materials listed in Table 2 were appropriate for low-temperature sintering with a small amount of glass frits developed in this work. The host dielectric materials could be categorized as $K=20$ –100 for resonator applications and $K>500$ for capacitor applications. The detailed compositions and their sintering and dielectric properties, which are measured directly from our preliminary studies, are summarized also in Table 2.

MCT, ZST, and BT4 are the microwave dielectric compositions^{17–19} which have a permittivity of 20–40 with nearly zero τ_f . BPNT and BBNT are the microwave materials which have a rather high permittivity.^{18,20,21} BT and BCTZ are the materials which have a permittivity higher than 500 and are typically used for MLCC materials showing X7R characteristics.²² As the BT4, BBNT, BT, and BCTZ are commercial powders, their exact compositions include some additives together with main phases listed in Table 2.

At the optimum firing temperatures of the compositions, we observed the relative densities which were higher than 98% and acceptable microwave dielectric properties as summarized in Table 2.

3.3. Low-temperature sintering of middle- K and high- K compositions with glass frits

The effects of glass addition on the sintering behavior of the selected host dielectric materials were examined. As a representative case, the sintering behavior and microwave dielectric properties of BT4 (BaTi₄O₉-based composition) with addition of MA05 glass frits are shown in Figs. 2 and 3. As the amount of glass frit increases, both the linear shrinkage and bulk density increases at the temperature range of 850–950 °C as shown in Fig. 2. When 10 wt.% of the frit was added, significant densification was achieved at 875 °C. As the amount of added frit increases, higher densification was achieved at below 875 °C, whereas the final density of the sintered body decreases at temperatures higher than 900 °C. When 10 wt.% of the glass frit was added at 875 °C, a density of 4.11 g/cm³ (relative density 98%),

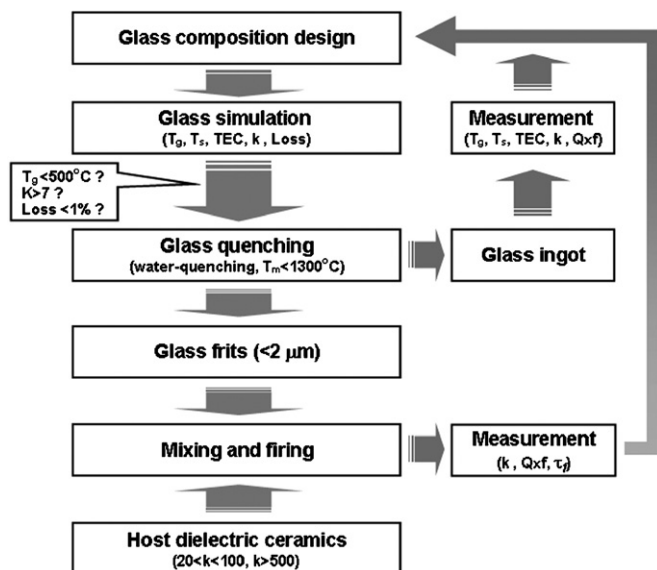


Fig. 1. A flow chart of the glass composition design.

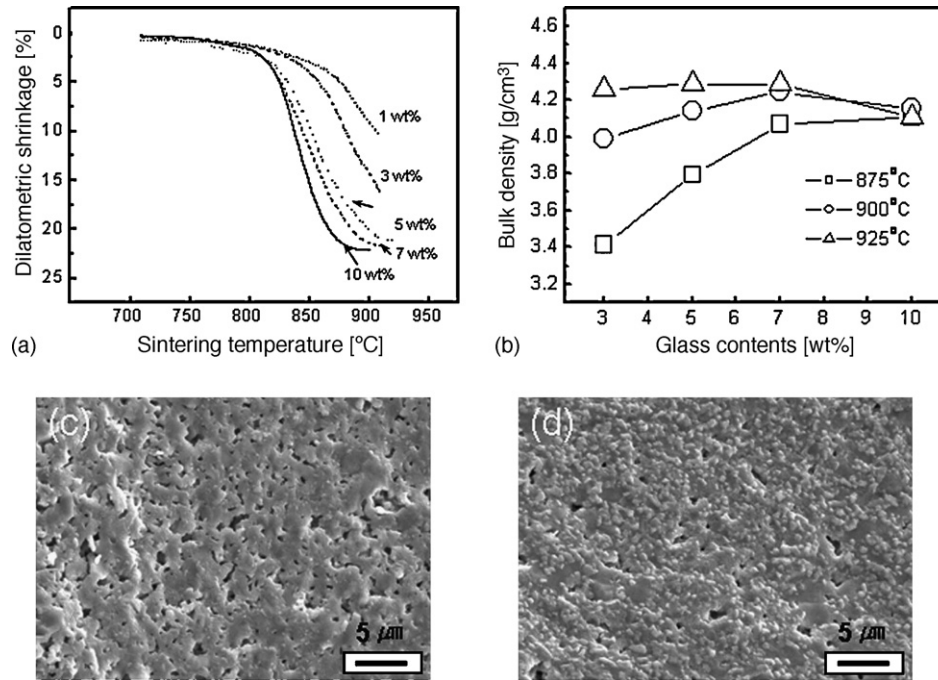


Fig. 2. The effects of glass content and processing temperature on the sintering behaviors in BT4-based ceramics. The dilatometric shrinkage (a) and bulk density (b) of the samples with variation of MA05 glass content and sintering temperature. SEM images of BT4 ceramics with 5 wt.% MA05 glass (c) and 10 wt.% MA05 glass (d) sintered at 875 °C for 2 h.

linear shrinkage 16.1%, and open porosity 0.17% were obtained. The fact that the density decreases in proportion to the amount of frit added in the temperature range of 900–925 °C could be ascribed to the low density of glass itself. As the firing temperature of middle-*k* LTCC composition should be below 900 °C to be compatible with the Ag inner electrode, the added amount of glass frit used in this study should be higher than 7 wt.%. As shown in the scanning electron micrography (SEM) images, the densification was accelerated with increasing glass frits.

Table 3 summarizes all the crystalline phases in BT4 ceramics with variation of the amount of glass frit and firing temperature. As the amount of glass frit increases, the on-set temperature of decomposition from the BaTi₄O₉ phase to other second phases decreases. For examples, BT4 composition exhibits only the BaTi₄O₉ phase up to the temperature of 1350 °C whereas the BaTi₄O₉ phase in the BT4 composition with 10 wt.% of glass frit start to decompose to BaTi₅O₁₁ and Ba₄Ti₁₃O₃₀ phases at the temperature of 700 °C. As the EPMA analysis (Fig. 3(d)) indicates that the local composition of the grains and grain-boundaries are different slightly from point to point, it is believed that the phases co-exist at a certain experimental condition as

listed in Table 3 are not always thermodynamically mixed phase. They could rather result from the compositional fluctuations according to the local inhomogeneity of the mixing ration of BT4 and glass frit. Every sequence in the phase change with increasing the amount of glass frit at a certain firing temperature, in Table 3, indicates that Ti-rich phases evolve with increasing the amount of glass frit. From the fact that the mole percentage of Ti becomes richer along this sequence, it seems that glass frit in which alkali is rich, absorbs Ba more preferably than Ti.

Fig. 3(a)–(c) shows the effects of glass addition on the microwave dielectric properties in BT4 ceramics. As the added amount of the frit increases, the magnitude of permittivity (*K*) increases at the temperature range of 850–900 °C. This could be attributed to the densification with glass addition as discussed earlier. However, at the temperature range of 900–950 °C, *K* decreases as the amount of glass frit is increased. This is related to the relatively low permittivity of glass frit itself (~7.5), considering that the relative densities of the samples are similar in this temperature range. In the case of the quality factor (*Q* × *f*), the increase of *Q* × *f* with increasing the firing temperature should be ascribed to the acceleration of densification at the

Table 3

A summarized list of the crystalline phases in BT4 ceramics with variation of the amount of MA05 glass

	RT	600 °C	700 °C	800 °C	875 °C	950 °C	1350 °C
BT4	BT ₄	BT ₄	BT ₄	BT ₄	BT ₄	BT ₄	BT ₄
BT4 + 1 wt.% frit	BT ₄	BT ₄	BT ₄	BT ₄	BT ₄ + BT ₅ + B ₂ T ₉	B ₂ T ₉	–
BT4 + 3 wt.% frit	BT ₄	BT ₄	BT ₄	BT ₄ + BT ₅	BT ₅ + B ₂ T ₉	BT ₅ + B ₂ T ₉	–
BT4 + 5 wt.% frit	BT ₄	BT ₄	BT ₄ + BT ₅	BT ₅	BT ₅	BT ₅	–
BT4 + 7 wt.% frit	BT ₄	BT ₄	BT ₄ + BT ₅	BT ₅	BT ₅	BT ₅	–
BT4 + 10 wt.% frit	BT ₄	BT ₄	BT ₄ + BT ₅ + B ₄ T ₁₃	BT ₅ + BT ₄ + B ₄ T ₁₃	BT ₅ + B ₄ T ₁₃	BT ₅ + B ₄ T ₁₃	–

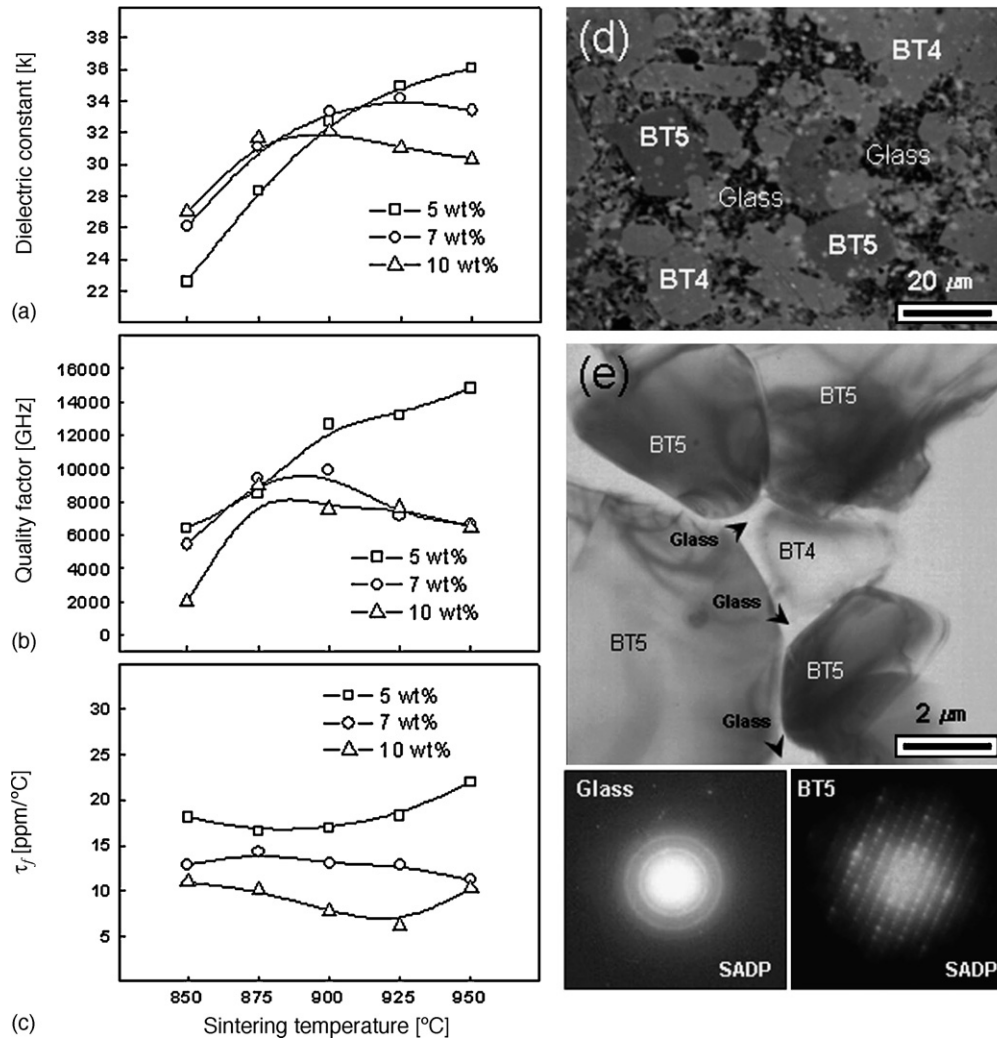


Fig. 3. (a)–(c) The effects of MA05 glass content and sintering temperature on the dielectric properties of BT4 ceramics. Backscattered electron image (d) and typical TEM image (e) of BT4 ceramics with 10 wt.% MA05 glass sintered at 875 °C for 2 h.

elevated temperatures. As the added amount of glass frit composition increases, the magnitude of $Q \times f$ decreases due to the low $Q \times f$ of the glass frits composition. As a minor contribution to the decrease in the value of $Q \times f$, the formation of second phases generated from the reaction between dielectric composition and glass frits could lead to such a decrease.

As for K , there are no abrupt changes with the main phase change because the magnitude of K in all the BaO–TiO₂ phases, including BaTi₅O₁₁, Ba₂Ti₉O₂₀, and Ba₄Ti₁₃O₃₀, are in a similar range.^{23–25} When the amount of glass frit increases, τ_f decreases monotonically. This decrease in τ_f with increasing glass frit composition can be ascribed to the negative τ_f of glass frits. Typically, the glass compositions in this study exhibit the τ_f of -80 to -220 ppm/°C, as summarized in Table 1. In the development of middle- K and high- K LTCC compositions, a sufficient amount of densification could be obtained with the addition of a considerable amount of lithium borosilicate glass frit composition. However, undesirable decomposition of the host dielectric materials and formation of the second phases may occur. Thus, one should determine the composition and the amount of glass frit composition so that the second

phase formation could be minimized, maintaining the sufficient densification.

Fig. 3(d) and (e) shows an EPMA and TEM analysis of the BT4 ceramics sintered at 875 °C with 10 wt.% glass frit composition. The EPMA analysis indicates that the local composition of the grains and grain-boundaries differ slightly from point to point. This may be attributed to the phase changes from BaTi₄O₉ to BaTi₅O₁₁ and Ba₄Ti₁₃O₃₀ due to the reaction between BaTi₄O₉ and glass frits, which result in the modification of microwave dielectric properties. When the amount of glass frit increases up to 7 wt.%, the main phase changes from BaTi₄O₉ to BaTi₅O₁₁, which results in a decrease of $Q \times f$ value due to the complex phases and microstructures. From TEM analysis, the amorphous diffraction patterns for the glass phases were observed in the grain-boundaries whereas the crystalline patterns for BaTi₄O₉ or BaTi₅O₁₁ were observed in the grains.

Fig. 4 shows the sintering behavior and dielectric properties of the other host dielectric materials with the addition of MA and MB glass frit composition. CZ (CaZrO₃-based) and B5N4 (Ba₅Nb₄O₁₅) were tested as $K=20$ – 40 compositions, whereas BBNT (BaO–Bi₂O₃–Nd₂O₃–TiO₂) and BPNT

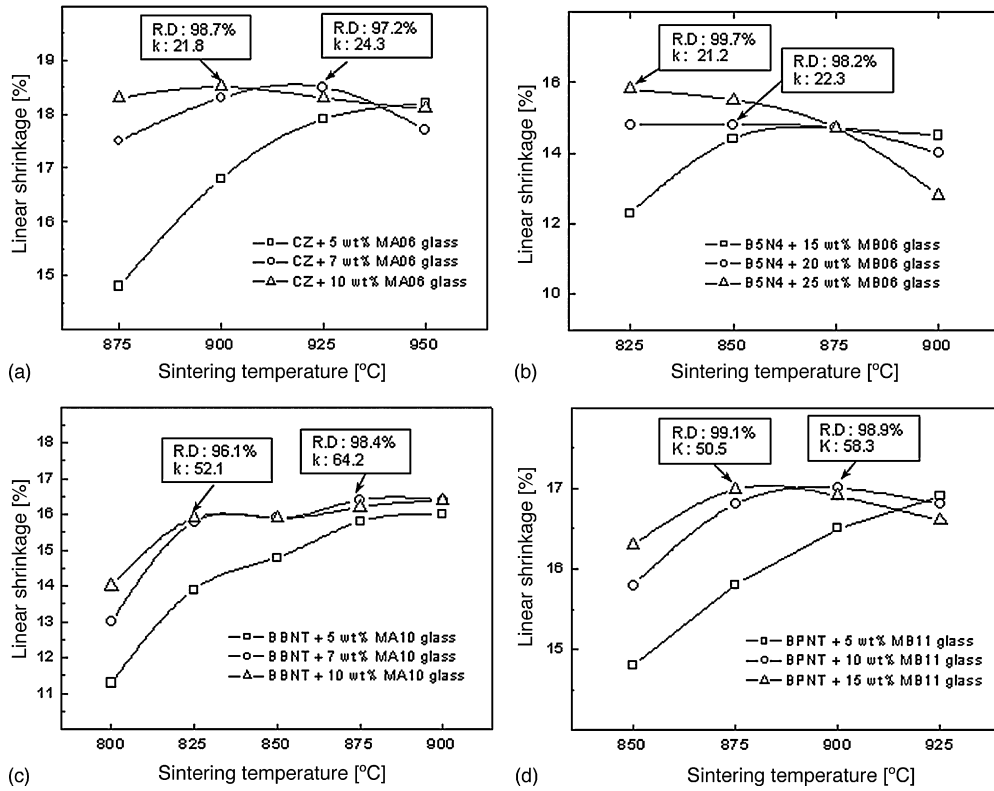


Fig. 4. The effects of glass contents and sintering temperature on the sintering behaviors and dielectric properties of the various middle-K LTCC materials. (a) CZ, (b) B5N4, (c) BBNT, and (d) BPNT compositions. The relative densities and permittivities of the selected samples were shown in inset boxes.

Table 4a

A summarized table of the sintering behaviors and the electrical properties of the host dielectric materials with glass frits

Ceramics composition		Glass frit		Firing temp. (°C)	Relative density (%)	Permittivity	$Q \times f$ (at GHz)	τ_f (ppm/°C)
Type	Contents (wt.%)	Type	Contents (wt.%)					
MCT	90	MA06	10	950	96.0	19.5	26700	-12
B5N4	85	MB22	15	875	96.5	21.5	3400	-15
CZ	90	MA05	10	875	98.5	21.9	4700	-58
ZST	90	MA05	10	875	95.9	27.5	9000	+14
BT4	85	MB22	15	875	98.2	26.0	10200	0
BT4	90	MA05	10	875	98.2	31.7	9000	+10
BT4	93	MA00	7	875	99.8	33.8	12700	+25
BBNT	90	MA10	10	875	97.5	53.3	2400	+3
BPNT	85	MB22	15	875	97.9	50.5	2500	+17
BT	90	HA06	10	900	95.4	510 ^a	2.2% ^b	-
BCTZ	90	HA11	10	875	96.3	730 ^a	2.3% ^b	-

^a Permittivity was measured using an impedance/gain-phase analyzer at 1 MHz.

^b Tan d was measured using an impedance/gain-phase analyzer at 1 MHz.

Table 4b

Selected physical and electrical properties of the glass frits used in this study

Glass code	Density (g/cm ³)	Permittivity	$Q \times f$ (at GHz)	τ_f (ppm/°C)	T_g (°C)	T_s (°C)
MA00	2.38	7.7	1200	-153	403	422
MA05	2.40	8.1	1200	-86	412	433
MA06	2.37	7.5	1500	-111	433	464
MA10	2.23	8.5	1000	-142	359	377
MB22	2.31	8.1	1100	-158	371	390
HA06	2.52	8.7	1500	-124	428	461
HA11	2.49	8.6	1300	-98	431	472

$(\text{Ba}_{0.5}\text{Pb}_{0.5}\text{O})\cdot\text{Nd}_2\text{O}_3\cdot 5\text{TiO}_2$) were tested as $K=40\text{--}100$ compositions. At all cases, the relative densities higher than 98% were obtained at sintering temperatures of below 875°C with the addition of appropriate glass frit composition. The detailed sintering behavior and dielectric properties are summarized in Tables 4a and 4b. On the whole, relative densities higher than 98% were obtained at sintering temperatures below 875°C , with the addition of 5–15 wt.% of glass frit composition. As for the $K=20\text{--}40$ composition, the microwave quality factors higher than 10,000 were obtained in several compositions. The τ_f of the samples were typically -60 to $+30$ ppm/ $^\circ\text{C}$. As for $K=40\text{--}90$ compositions, microwave quality factors higher than 2500 were obtained on the whole and the τ_f of the samples were 0 to $+30$ ppm/ $^\circ\text{C}$.

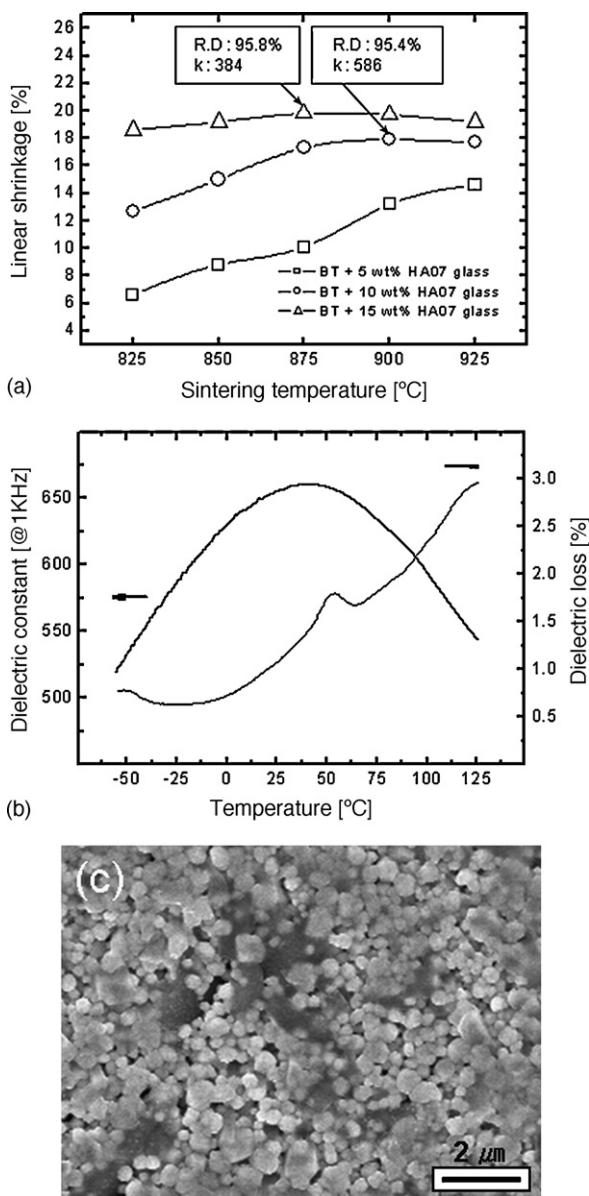


Fig. 5. The effects of glass content and sintering temperature on the sintering behaviors (a) and the dielectric properties (b) of BT with addition of HA06 glass frit. (c) SEM image of the BT ceramics with 10 wt.% HA07 glass sintered at 875°C for 2h.

As for high- K dielectric compositions, BT (BaTiO_3 -based) and BCTZ ($(\text{Ba}_{0.95}\text{Ca}_{0.05})(\text{Ti}_{0.88}\text{Zr}_{0.12})\text{O}_3$) were tested to realize high- K ($K > 500$) LTCC compositions. Fig. 5(a) shows the sintering behavior of the BT with the addition of HA06 glass frit composition. When 15 wt.% of HA06 glass frit was added to BT, relative densities higher than 95% and permittivity of 650 were obtained at sintering temperatures of 875°C . To improve the densification and dielectric properties, we examined BCTZ composition, which has a lower optimum sintering temperature and higher permittivity as shown in Table 2. In the case of BCTZ, the permittivity of 800–900 was obtained at the temperature of ca. $850\text{--}875^\circ\text{C}$ and the overall temperature characteristics were Y5S. In Tables 4a and 4b, all the detailed sintering behavior and dielectric properties are summarized. On the whole, the microstructures of BT and BCTZ samples show acceptable densifications and this result could be attributed to the sufficient liquid phase resulting from glass frits having low T_g .

4. Conclusions

In this study, middle- and high-permittivity LTCC dielectric compositions were studied. By designing a $\text{SiO}_2\text{--B}_2\text{O}_3\text{--Li}_2\text{O}$ -based glass frit system, we could reduce the temperature of densification to 875°C , while still maintaining acceptable microwave dielectric properties in selected middle- and high- K host dielectrics.

As for $K=20\text{--}40$ compositions, microwave quality factors higher than 10,000 were obtained in several compositions. The τ_f of the samples were -60 to $+30$ ppm/ $^\circ\text{C}$. As for $K=40\text{--}100$ compositions, microwave quality factors higher than 2500 were obtained in several compositions. The τ_f of the samples were 0 to $+30$ ppm/ $^\circ\text{C}$. As for $K > 500$ compositions, dielectric loss factors of less than 3% were obtained in several compositions. All these material systems could be sintered at below 875°C and hence could be applied to middle- and high- K LTCC material systems.

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