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Characterization and dielectric behavior of B_2O_3-doped $0.9Mg_{0.95}Co_{0.05}TiO_3$–$0.1Ca_{0.6}La_{0.8/3}TiO_3$ ceramic system at microwave frequency

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ABSTRACT

The microwave dielectric properties and the microstructures of $(1-x)Mg_{0.95}Co_{0.05}TiO_3$–$xCa_{0.6}La_{0.8/3}TiO_3$ ceramic system, prepared by a mixed oxide route, have been investigated. The sintered $0.9Mg_{0.95}Co_{0.05}TiO_3$–$0.1Ca_{0.6}La_{0.8/3}TiO_3$ ceramics with B_2O_3 addition were characterized using X-ray diffraction spectra and scanning electron microscopy (SEM). The microwave dielectric properties are found to strongly correlate with the sintering temperature as well as the amount of B_2O_3 addition. The $Q \times f$ value of $0.9Mg_{0.95}Co_{0.05}TiO_3$–$0.1Ca_{0.6}La_{0.8/3}TiO_3$ ceramic increased with increasing temperature to 1225 °C/4 h and decreased thereafter. The decrease in $Q \times f$ value was coincident with the abnormal grain growth. A maximum $Q \times f$ value of 76,000 GHz associated with an dielectric constant (ε_r) of 22.67 and a temperature coefficient (τ_f) of −12 ppm/°C, was achieved for samples with 0.5 wt.% B_2O_3 additions sintering at 1225 °C/4 h. By appropriately adjusting the x value in the $(1-x)Mg_{0.95}Co_{0.05}TiO_3$–$xCa_{0.6}La_{0.8/3}TiO_3$ ceramic system, zero τ_f value can be obtained. A parallel-coupled line band-pass filter is designed and simulated using the proposed dielectric to study its performance.

1. Introduction

Recently, microwave dielectric devices are widely used in important applications such as resonator, filters and oscillators in microwave communication system [1]. The dielectric characteristics required for microwave resonator are high dielectric constant ($\varepsilon_r > 15$) to reduce the size of resonators (size of a dielectric resonator is $1/\sqrt{\varepsilon_r}$), high quality factor ($Q \times f$) for achieving prominent frequency selectivity and stability, and a near-zero temperature coefficient of resonant frequency (τ_f) for temperature stable circuits [2–24]. Moreover, low sintering temperature is also required to match with low-loss and low-melting point conductors (such as Ag or Cu) in fabrication of microwave dielectric devices. Several compounds such as (Zr, Sn)TiO_4, $Ba(Mg_{1/3}Ta_{1/3})O_3$, and (Mg, Ca)TiO_3 have been developed [25–28].

$MgTiO_3$-based ceramics have wide applications as dielectric in resonators, filters and antennas for communication system, radar and global positioning systems operating at microwave frequency. With partial replacement of Mg^{2+} by Co^{2+}, the $Mg_{0.95}Co_{0.05}TiO_3$ ceramic with an ilmenite-type structure was reported to possess excellent dielectric properties with an dielectric constant (ε_r) of 16.8, a quality factor ($Q \times f$) of 230,000 GHz and a temperature coefficient (τ_f) of −54 ppm/°C [29]. However, it required sintering temperature as high as 1400–1450 °C.

Chemical processing and small particle sizes of the starting materials are generally advantageous to reduce the sintering temperature of dielectric materials [30–33]. However, they required a flexible procedure, which increased the cost and time to fabricate a dielectric resonator. The liquid-phase sintering, by which adding glass or other low-melting point materials, was found to effectively lower the firing temperature of ceramics [31,32]. The microwave dielectric properties of dielectric resonators were also deeply affected by the liquid sintering temperature due to the development of microstructure at low sintering temperature or reaction between host material and addition.

The $(1-x)CaTiO_3$–$xLa_{2/3}TiO_3$ solid–solution system was first investigated by Kim et al. [34]. After sintering the specimens in air, they found that the solid–solution limit for the composition was $x = 0.96$. The superstructure reflection lines were detected for the composition $x = 0.8$. The microwave dielectric properties were also characterized at 10 GHz. The permittivity and the temperature coefficient of resonant frequency (τ_f) decreased with an increase in the amount of La substituted. The $Q \times f$ value increased rapidly at first, then increased steadily, and was almost linear for $x > 0.3$. The solid–solution limit of the composition $x = 0.96$ had the highest Q in the system and a high

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permittivity of 90. However, a poor τ_f value of +190 ppm/°C was obtained, which is not satisfactory for practical applications. Huang et al. reported that the $(1-y)\mathrm{CaTiO_3}$–$y\mathrm{La_{2/3}TiO_3}$ solid–solution system exhibited excellent microwave dielectric properties when the y value was 0.4 [35]. In our previous study, $\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ was added to $\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$ as a ceramic system of $(1-x)\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$–$x\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ (MCT–CLT hereafter), which demonstrated an effective compensation in its temperature coefficient (τ_f) [3]. As $x = 0.1$, the $0.9(\mathrm{Mg_{0.95}Co_{0.05}})\mathrm{TiO_3}$–$0.1\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ (9MCT–CLT hereafter) ceramic sintered at 1350 °C/4 h exhibited excellent microwave dielectric properties: a dielectric constant (ε_r) of 21.8, a $Q \times f$ value of 131,000 GHz and a temperature coefficient (τ_f) of −15.5 ppm/°C. In this paper, $\mathrm{B_2O_3}$ was chosen as a sintering aid to lower the sintering temperature of MCT–CLT ceramics. The measured microwave dielectric properties were discussed from the results based upon the X-ray diffraction patterns, the densification and the microstructures of ceramics. Low temperature sintering and microwave dielectric properties of 9MCT–CLT ceramics with addition $\mathrm{B_2O_3}$ were reported.

2. Experimental procedures

Samples of $\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$ and $\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ were individually synthesized by conventional solid-state methods from high-purity oxide powders (>99.9%): MgO, CoO, $\mathrm{CaCO_3}$, $\mathrm{La_2O_3}$ and $\mathrm{TiO_2}$. The starting materials were mixed according to the stoichiometry: $\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$ and $\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ and ground in distilled water for 10 h and 16 h in a ball mill with agate balls. Both mixtures were dried and calcined at 1100 °C for 4 h. The calcined reagents were mixed as desired composition $(1-x)\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$–$x\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ with the additions of 0.5 and 1 wt.% of $\mathrm{B_2O_3}$ as sintered aid and remilled for 24 h. The fine powder together with the organic binder were forced through a 100-mesh sieve and pressed into pellets with dimensions of 11 mm in diameter and 5 mm in thickness. These pellets were sintered at temperatures of 1175–1275 °C for 4 h in air. The heating rate and the cooling rate were both set at 10 °C/min.

The powder and bulk X-ray diffraction (XRD, Rigaku D/Max III.V) spectra were collected using Cu Kα radiation (at 30 kV and 20 mA) and a graphite monochromator in the 2θ range of 20–60°. The microstructural observations and analysis of sintered surface were performed by a scanning electron microscopy (SEM, Philips XL-40FEG). The bulk densities of the sintered pellets were measured by the Archimedes method. The dielectric constant ε_r and the $Q \times f$ value at microwave frequencies were determined using the Hakki–Coleman dielectric resonator method as modified and improved by Courtney [36,37]. The disks were, in turn, placed between two parallel conductor plates. Then, the $\mathrm{TE_{011}}$ and $\mathrm{TE_{012}}$ modes were measured using a system combined with a HP8757D network analyzer and a HP8350B sweep oscillator. The dielectric constant was calculated from the resonant frequency of the $\mathrm{TE_{011}}$ mode of the cylindrical disk. For the Q_d measurement, the conductor loss resulting from the eddy currents around the conductive plate surfaces must be subtracted so as to obtain the dielectric quality factor. For this purpose, two disks with different height were prepared, one is for $\mathrm{TE_{011}}$ mode and the other is for $\mathrm{TE_{012}}$ mode, where the disk for the $\mathrm{TE_{012}}$ mode measurement has 2 times height as that of the disk for the $\mathrm{TE_{011}}$ mode. Identical technique was applied in measuring the temperature coefficient of resonant frequency (τ_f). The test set was placed over a thermostat in the temperature range from 25 to 80 °C. The τ_f value (ppm/°C) can be calculated by noting the change in resonant frequency (Δf),

$$\tau_f = \frac{f_2 - f_1}{f_1(T_2 - T_1)} \tag{1}$$

where f_1 and f_2 represents the resonant frequencies at T_1 and T_2, respectively.

A parallel-coupled line band-pass filter with a center frequency of 3.0 GHz was designed. The filter was simulated by the EM simulator (IE3D) to determine its physical dimensions and performance on different substrate materials (such as FR4, alumina, and 9MCT–CLT–B).

3. Results and discussion

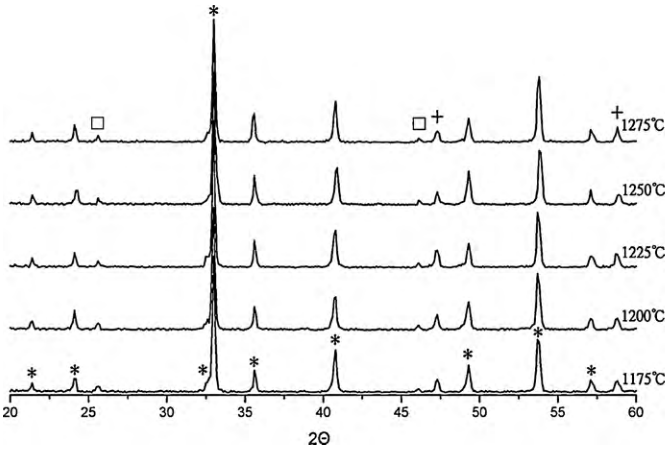

Fig. 1. X-ray diffraction patterns of 9MCT–CLT ceramic with 0.5 wt.% $\mathrm{B_2O_3}$ additions at different sintering temperatures for 4 h.

Fig. 1 shows the X-ray diffraction patterns of 0.5 wt.% $\mathrm{B_2O_3}$-doped 9MCT–CLT (9MCT–CLT–B hereafter) ceramics at different sintering temperatures for 4 h. The XRD patterns showed that peaks indicating the presence of $\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$ as the main crystalline phase, in association with $\mathrm{Ca_{0.6}La_{0.8/3}TiO_3}$ and $\mathrm{Mg_{0.95}Co_{0.05}Ti_2O_5}$ as minor phases. It is understood that crystal structures of $\mathrm{MgTiO_3}$ and $\mathrm{CaTiO_3}$ are trigonal (JCPDS #06-0494) and orthorhombic (JCPDS #22-0153), respectively. The formation of mixed phases in the MCT–CLT ceramic system was due to structural differences and because the average ionic radii of $\mathrm{Ca^{2+}}$ (0.99 nm) and $\mathrm{La^{3+}}$ (1.15 nm) were larger than $\mathrm{Mg^{2+}}$(0.72 nm); therefore, a solid solution could not be obtained; The X-ray diffraction patterns of 9MCT–CLT ceramics have no significant change with 0.5 wt.% $\mathrm{B_2O_3}$ addition at sintering temperature of 1175–1275 °C. According to the XRD patterns of $\mathrm{Mg_{0.95}Co_{0.05}Ti_2O_5}$ phases with orthorhombic crystal structure (JCPDS #09-0016), usually formed as an intermediate phase, was identified and difficult to completely eliminate from the sample prepared by mixed oxide route. The forming of the second phase $\mathrm{Mg_{0.95}Co_{0.05}Ti_2O_5}$ was mainly due to the LOI of the starting powder MgO. It is also possibly a result from the following reaction:

$$2\mathrm{Mg_{0.95}Co_{0.05}TiO_3} \rightarrow \mathrm{Mg_{0.95}Co_{0.05}Ti_2O_5} + \mathrm{Mg_{0.95}Co_{0.05}O} \tag{2}$$

The formation of second phase $\mathrm{Mg_{0.95}Co_{0.05}Ti_2O_5}$ in the $\mathrm{Mg_{0.95}Co_{0.05}TiO_3}$ ceramics would cause a serious drop in the quality factor of 9MCT–CLT ceramics [38]. Similar result of the X-ray diffraction patterns of the 9MCT–CLT ceramics with various amounts of $\mathrm{B_2O_3}$ addition sintered at 1225 °C for 4 h have not changed significantly as shown in Fig. 2.

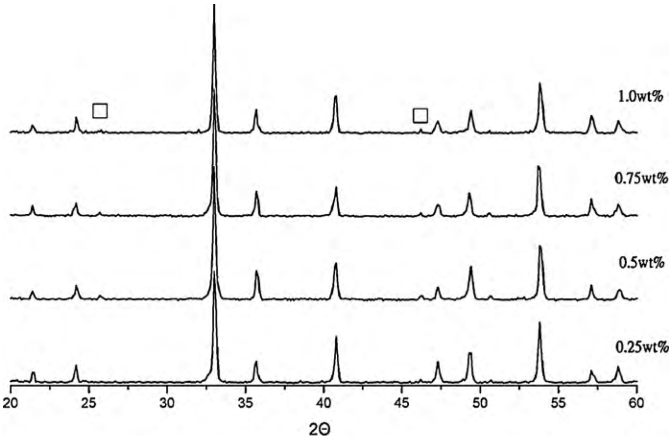

Fig. 2. X-ray diffraction patterns of MCT–CLT ceramic with different amount of $\mathrm{B_2O_3}$ additions sintering at 1225 °C/4 h.

The lattice parameters of 9MCT–CLT ceramics with different amount of $\mathrm{B_2O_3}$ addition sintering at 1225 °C/4 h were also measured in this study as shown in Table 1. It was found that $\mathrm{MgTiO_3}$ has a hexagonal structure with the following lattice parameters: $a = b = 0.5054$ nm, $c = 1.3898$ nm (ICDD-PDF#00-006-0494). The results illustrate that the replacement of $\mathrm{Mg^{2+}}$ by 0.05 moles

Table 1
The lattice parameter of 9MCT–CLT ceramic sintered at 1225 °C/4 h with different amount of B_2O_3 addition.

B_2O_3 additions (wt.%)	a (nm)	c (nm)
0	0.5054	1.3899
0.25	0.50532 ± 0.00014	1.39575 ± 0.00057
0.5	0.5045 ± 0.00072	1.38693 ± 0.00291
0.75	0.50341 ± 0.00097	1.389 ± 0.00399
1.0	0.50653 ± 0.00056	1.3928 ± 0.00226

of Co^{2+} leads to a variation in the lattice parameters. This is because the relatively small Co^{2+} ions (radii = 0.082 nm; Mg^{2+} ions radii = 0.078 nm) are added to $MgTiO_3$, making the lattice of $Mg_{0.95}Co_{0.05}TiO_3$ locally distorted. When $Ca_{0.6}La_{0.8/3}TiO_3$ was added to $Mg_{0.95}Co_{0.05}TiO_3$ to form a ceramic system of MCT–CLT, the lattice parameters of $Mg_{0.95}Co_{0.05}TiO_3$ did not change with $Ca_{0.6}La_{0.8/3}TiO_3$ content. Furthermore, the formation of mixed phases in the 9MCT–CLT ceramics system was due to structural differences and because the average ionic radii of Ca^{2+} (0.106 nm) and La^{3+} (0.122 nm) were larger than these of Mg^{2+} (0.078 nm) and Co^{2+} (radii = 0.082 nm). This confirms the existence of a two-phase ceramic system of MCT–CLT, with $Mg_{0.95}Co_{0.05}TiO_3$ as the main crystalline phase and $Ca_{0.6}La_{0.8/3}TiO_3$ as the minor phase. These results are in agreement with XRD patterns.

The microstructures of 0.5 wt.% B_2O_3-doped 9MCT–CLT ceramic under different sintering condition are demonstrated in Fig. 3a–e. The B_2O_3-doped samples were not dense, and grains did not grow after it was sintered at 1175 and 1200 °C for 4 h. Furthermore, these samples exhibited very low apparent density, as shown in Figs. 3 and 4. It may directly affect the microwave dielectric properties of the ceramic samples. As the sintering temperatures increased from 1225 to 1250 °C the pores eliminated and densification enhanced. The pores of 9MCT–CLT–B ceramics were almost disappeared sintering at 1225 °C for 4 h. Comparing the microstructures of 9MCT–CLT–B ceramics sintered at different conditions, the grain size increased with the increasing of sintering temperature. However, non-uniform grain growth will be happened at high sintering temperature (1250 and 1275 °C) which might degrade the microwave dielectric properties of the 9MCT–CLT–B ceramics. Furthermore, as shown in Fig. 3f–h illustrates the SEM photographs of

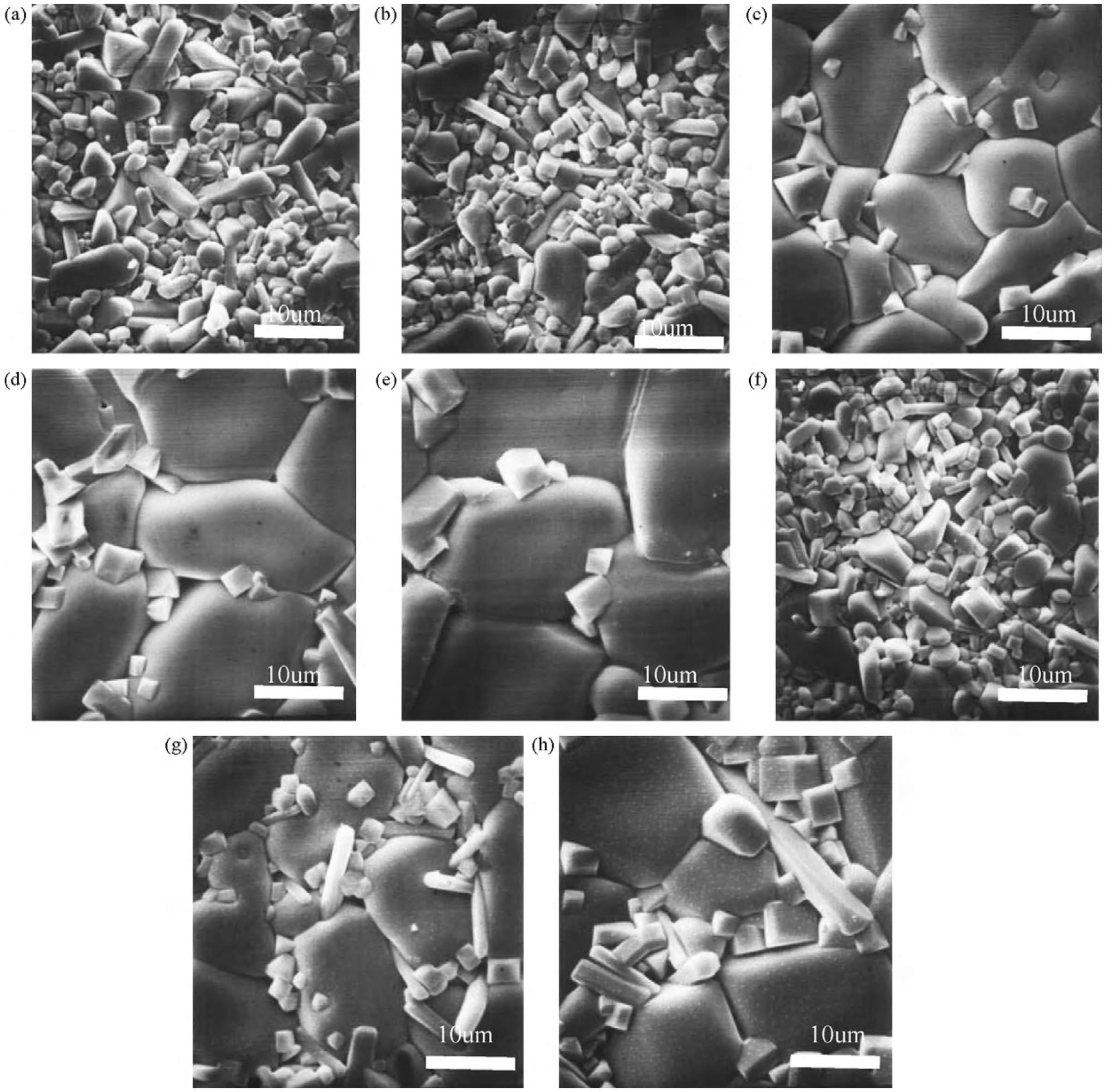

Fig. 3. SEM images of 0.5 wt.% B_2O_3-doped 9MCT–CLT ceramics sintered at (a) 1175 °C, (b) 1200 °C, (c) 1225 °C, (d) 1250 °C, (e) 1275 °C for 4 h, (f) 0.25 wt.%, (g) 0.75 wt.%, (h) 1.0 wt.% B_2O_3-doped/1225 °C for 4 h.

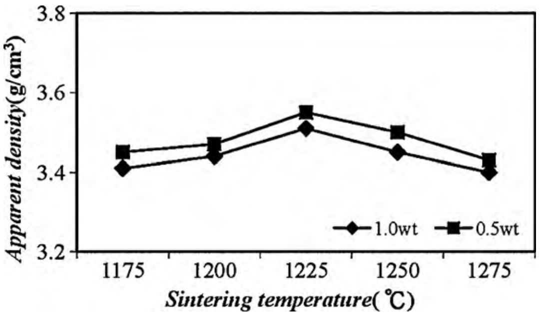

Fig. 4. Apparent density of 9MCT–CLT ceramics with various B_2O_3 additions sintered at different temperature.

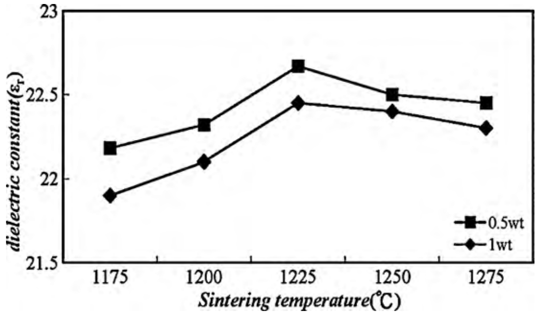

Fig. 5. Dependence of sintering temperature of 9MCT–CLT ceramics on dielectric constants with various B_2O_3 additions.

the different amount of B_2O_3 additions sintering at 1225 °C/4 h. The grain growth of the 9MCT–CLT ceramics rapidly increased above 0.5 wt.% B_2O_3 additions. The average grain size increased with increasing B_2O_3 content since the liquid-phase sintering promotes the densification of ceramics. The liquid disappears or is retained in the final stage of liquid-phase sintering.

Fig. 4 shows the bulk densities of 9MCT–CLT ceramics with variation of B_2O_3 additions at different sintering temperature for 4 h. With increasing sintering temperature, the bulk density was found to increase to a maximum value of at 1225 °C and thereafter decreased. When 0.5 wt.% of B_2O_3 added into the system, the apparent density significantly increased from 3.45 to 3.55 g/cm^3. The obtained apparent density of 3.55 g/cm^3 corresponded to the apparent density of 9MCT–CLT ceramics sintered at 1225 °C. It can be considered that liquid-phase sintering is occurred by addition of B_2O_3. As increasing B_2O_3 addition over 0.5 wt.%, the apparent density turned to decrease slightly. In case of liquid-phase sintering, generally, sintering aids with appropriate content can reduce sintering temperature due to enhanced sinter-ability. When sintering aids are excessive, however, the bulk density and microwave dielectric properties were degraded as shown in Table 2.

Fig. 5 demonstrates the dielectric constant (ε_r) of 9MCT–CLT ceramics with different amount of B_2O_3 additions as a function of their sintering temperature. The dielectric constant (ε_r) slightly increased with an increased sintering temperature, which reveals the same trend with those between densities and sintering temperature, since higher density means lower porosity ($\varepsilon_r = 1$). The increase of dielectric constant (ε_r) could be explained as owing to higher densities. The dielectric constant (ε_r) rapidly increased from 1175 to 1225 °C and saturated at 22–23 for the well-sintered ceramics. With 0.5 wt.% B_2O_3 addition, a dielectric constant (ε_r) of 22.67 was obtained for 9MCT–CLT ceramics sintered at 1225 °C/4 h.

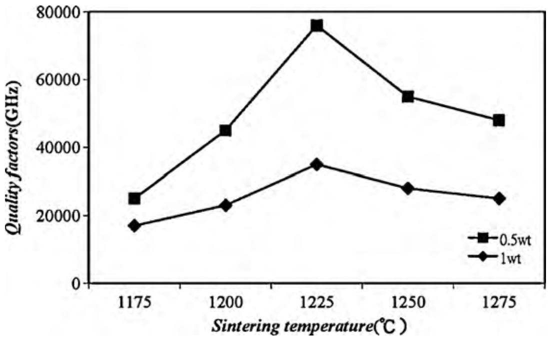

Fig. 6. Dependence of sintering temperature of 9MCT–CLT ceramics on quality factor ($Q \times f$) with various B_2O_3 additions.

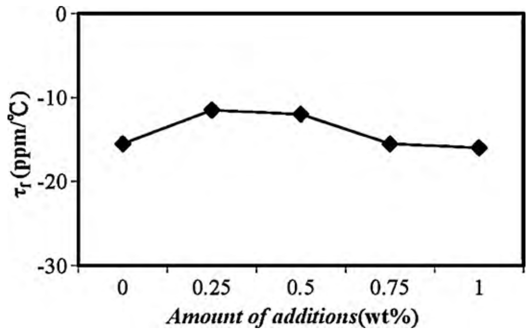

Fig. 7. Dependence of the amount of B_2O_3 additions of 9MCT–CLT ceramics on temperature coefficient sintered at 1225 °C for 4 h.

Fig. 6 shows the quality factors ($Q \times f$) of 9MCT–CLT ceramics with various B_2O_3 additions at different sintering temperature. The $Q \times f$ value increased with the sintering temperature from 1175 to 1225 °C. After reaching a maximum at 1225 °C, it decreased. At 1225 °C, the maximum $Q \times f$ value of 76,000 GHz was obtained for the 9MCT–CLT ceramics with 0.5 wt.% B_2O_3 addition. The trend for the density was not completely identical. It was reported previously that the microwave dielectric loss is caused not only by the lattice vibration modes, but also by the pores, the grain morphology and the second phases [39]. Density also plays an important role in controlling the dielectric loss and this has been shown for other microwave dielectric materials. The increase in $Q \times f$ value at low temperatures was due to the increase in density as well as the uniformity of grain growth as observed in Fig. 1. As shown in Fig. 1, second phase $Mg_{0.95}Co_{0.05}Ti_2O_5$ appeared at the levels of 0.5 and 1 wt.% B_2O_3 additions. That would explain the slight decrease in $Q \times f$ value for 9MCT–CLT ceramics with 0.5 and 1 wt.% B_2O_3 addition. The quality factors of B_2O_3-doped 9MCT–CLT ceramics were relatively lower than that of purity 9MCT–CLT ceramics due to the pores, the grain boundary phase, and the second phase $Mg_{0.95}Co_{0.05}Ti_2O_5$.

Table 2
Microwave dielectric properties of 9MCT–CLT with different amount of B_2O_3 addition sintered at 1225 °C for 4 h.

B_2O_3 (wt.%)	Density (g/cm^3)	Dielectric constant (ε_r)	Quality factor ($Q \times f$)	Temperature coefficient (τ_f)
0.25	3.61	22.78	69,000	−11.5
0.5	3.55	22.67	76,000	−12
0.75	3.53	22.56	46,000	−15.5
1	3.51	22.45	35,000	−16

Fig. 8. Physical layout of the parallel-coupled line band-pass filter.

Table 3
Simulation results of the band-pass filters using different dielectrics.

	FR4	Alumina	9MCT–CLT-B
Dielectric constant (ε_r)	4.7	9.7	22.67
Tan δ	0.015	0.0003	0.0000329
Central frequency (GHz)	2.94	2.97	2.98
Insertion loss (dB)	−3.53	−0.8658	−0.7298
Return loss (dB)	−18.35	−28.42	−28.52
Fractional bandwidth (MHz)	310	320	315
Efficacious dimensions (mm^2)	8.8 × 53.27	4.85 × 38.77	4.98 × 26.09

9MCT–CLT–B: $0.9Mg_{0.95}Co_{0.05}TiO_3$–$0.1Ca_{0.6}La_{0.8/3}TiO_3$ + 0.5 wt.% B_2O_3.

Fig. 7 illustrates the temperature coefficients of resonant frequency (τ_f) of 9MCT–CLT ceramics with various amounts of B_2O_3 added at 1225 °C for 4 h. The temperature coefficient of resonant frequency (τ_f) is mainly related to the composition, the additive and the second phase of the material. The temperature coefficient of resonant frequency (τ_f) of MCT–CLT ceramics rapidly increased with increasing x value due to a large positive τ_f of $Ca_{0.6}La_{0.8/3}TiO_3$ (τ_f = 213 ppm/°C). Although significant change in the τ_f value was not observed for specimens at different amount of B_2O_3 additions, it slightly shifted toward negative as higher B_2O_3-doped. This might be a result from $Mg_{0.95}Co_{0.05}TiO_3$-preferred phase formation as observed a rapid grain growth of $Mg_{0.95}Co_{0.05}TiO_3$ at high B_2O_3-doped in Fig. 3f–h. It varied from −11.5 to −16 ppm/°C as the amount of B_2O_3 addition increased from 0.25 to 1 wt.% as shown in Table 2.

To verify the performance of the proposed material, a parallel-coupled line band-pass filter with a central frequency of 3.0 GHz was designed and simulated on various dielectric substrates: FR4, alumina, and 9MCT–CLT–B ceramic. Fig. 8 shows the physical layout of the designed filter with a central frequency of 3.0 GHz. The simulation results are listed in Table 3. Compared to FR4 and alumina, the filter using the 9MCT–CLT–B ceramic shows a tremendous reduction in the insertion loss and demonstrates a large reduction in its size.

4. Conclusions

The effect of B_2O_3 addition on the microwave dielectric properties and the microstructures of $(1-x)Mg_{0.95}Co_{0.05}TiO_3$–$xCa_{0.6}La_{0.8/3}TiO_3$ ceramics was investigated. B_2O_3 additions could not only effectively lower the sintering temperature but also improve the microwave dielectric properties of $(1-x)Mg_{0.95}Co_{0.05}TiO_3$–$xCa_{0.6}La_{0.8/3}TiO_3$ ceramics. With increasing B_2O_3 addition, the density of $0.9Mg_{0.95}Co_{0.05}TiO_3$–$0.1Ca_{0.6}La_{0.8/3}TiO_3$ ceramics increased slightly. A maximum quality factor ($Q \times f$) of 76,000 GHz associated with an dielectric constant (ε_r) of 22.67 and a temperature coefficient (τ_f) of −11.55 ppm/°C, was achieved for 0.5 wt.% B_2O_3-doped samples at 1225 °C/4 h. However, degradation in the $Q \times f$ value was observed at highly doped specimen due to the existing of more B_2O_3 liquid phase. These results indicated that large amount of B_2O_3 addition is not suggested. Zero τ_f value can be obtained by properly adjusting the x value of $(1-x)Mg_{0.95}Co_{0.05}TiO_3$–$xCa_{0.6}La_{0.8/3}TiO_3$ ceramics. Therefore, the 9MCT–CLT ceramic with 0.5 wt.% B_2O_3 is suitable for applications in microwave dielectric resonators and filters because of its excellent microwave dielectric properties. Compared to FR4 and alumina, the filter using 9MCT–CLT ceramic with 0.5 wt.% B_2O_3 shows a tremendous reduction in the insertion loss and demonstrates a large reduction in its size.

References

[1] K. Wakino, T. Nishikawa, Y. Ishikawa, Br. Ceram. Trans. J. 89 (2) (1990) 39.
[2] C.L. Huang, J.J. Wang, Y.P. Chang, J. Am. Ceram. Soc. 90 (2007) 858.
[3] C.L. Huang, C.L. Pan, J.F. Hsu, J. Alloys Compd. 461 (2008) 521–526.
[4] C.L. Huang, S.S. Liu, J. Alloys Compd. 468 (2009) L13–L16.
[5] C.L. Huang, C.H. Shen, J. Alloys Compd. 468 (2009) 516–521.
[6] C.L. Huang, Y.B. Chen, J. Alloys Compd. 469 (2009) 357–361.
[7] C.L. Huang, S.S. Liu, J. Alloys Compd. 471 (2009) L9–L12.
[8] C.H. Shen, C.L. Huang, J. Alloys Compd. 472 (2009) 451–455.
[9] C.H. Shen, C.L. Huang, J. Alloys Compd. 475 (2009) 391–395.
[10] C.L. Huang, Y.B. Chen, J. Alloys Compd. 477 (2009) 712–715.
[11] C.H. Shen, C.L. Huang, J. Alloys Compd. 477 (2009) 720–725.
[12] C.L. Huang, J.Y. Chen, J. Alloys Compd. 478 (2009) 554–558.
[13] C.L. Huang, J.Y. Chen, J. Alloys Compd. 478 (2009) 842–846.
[14] C.L. Huang, S.S. Liu, J. Alloys Compd. 480 (2009) 794–797.
[15] C.L. Huang, J.Y. Chen, J. Alloys Compd. 484 (2009) 494–497.
[16] C.L. Huang, J.Y. Chen, J. Alloys Compd. 485 (2009) 706–710.
[17] C.L. Huang, J.Y. Chen, J. Alloys Compd. 487 (2009) 420–424.
[18] C.H. Shen, C.L. Huang, J. Alloys Compd. 489 (2010) 170–174.
[19] C.L. Huang, S.H. Lin, J. Alloys Compd. 489 (2010) 541–544.
[20] C.L. Huang, S.H. Lin, J. Alloys Compd. 489 (2010) 719–721.
[21] J.Y. Chen, C.L. Huang, J. Alloys Compd. 494 (2010) 205–209.
[22] C.L. Huang, W.R. Yang, J. Alloys Compd. 495 (2010) L5–L7.
[23] J.Y. Chen, C.L. Huang, J. Alloys Compd. 496 (2010) L10–L13.
[24] C.L. Huang, J.Y. Chen, J. Alloys Compd. 499 (2010) 48–52.
[25] S. Nomura, K. Toyama, K. Kaneta, Jpn. J. Appl. Phys. 21 (1982) 624.
[26] G. Wolfram, H.E. Gobel, Mater. Res. Bull. 16 (1981) 1455.
[27] C.L. Huang, C.F. Tasi, Y.B. Chen, J. Alloys Compd. 453 (2008) 337.
[28] C.L. Huang, C.F. Tseng, C.H. Hsu, J. Am. Ceram. Soc. 90 (2007) 1619.
[29] J.H. Sohn, Y. Inaguma, S.O. Yoon, Jpn. J. Appl. Phys. 33 (1994) 5466.
[30] T. Kakada, S.F. Wang, Syoshikawa, J. Am. Ceram. Soc. 77 (1944) 1909.
[31] T. Kakada, S.F. Wang, Syoshikawa, S.T. Jang, J. Am. Ceram. Soc. 77 (1944) 2485.
[32] S.I. Hirno, Taashi, Hayashi, A. Hattori, J. Am. Ceram. Soc. 74 (1991) 1320.
[33] V. Tolmer, G. dedqardin, J. Am. Ceram. Soc. 80 (1997) 1981.
[34] I.S. Kim, W.H. Jung, Y. Inaguma, Mater. Res. Bull. 30 (1995) 307.
[35] C.L. Huang, J.T. Tsai, Y.B. Chen, Mater. Res. Bull. 36 (2001) 547.
[36] B.W. Hakki, P.D. Coleman, IEEE Trans. Microwave Theory Technol. 8 (1960) 402.
[37] Y. Kobayashi, M. Katoh, IEEE Trans. Microwave Theory Technol. 33 (1985) 586.
[38] C.L. Huang, C.H. Shen, J. Am. Ceram. Soc. 92 (2009) 384.
[39] B.D. Silverman, Phys. Rev. 125 (1962) 1921.