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Ceramics International 40 (2014) 3895–3902

CERAMICS INTERNATIONAL

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Low temperature sintering and microwave dielectric properties of $CaSiO_3$–Al_2O_3 ceramics for LTCC applications

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Received 27 June 2013; received in revised form 3 August 2013; accepted 6 August 2013
Available online 22 August 2013

Abstract

The effects of CuO, Li_2CO_3 and $CaTiO_3$ additives on the densification, microstructure and microwave dielectric properties of $CaSiO_3$–1 wt% Al_2O_3 ceramics for low-temperature co-fired applications were investigated. With a single addition of 1 wt% Li_2CO_3, the $CaSiO_3$–1 wt% Al_2O_3 ceramic required a temperature of at least 975 °C to be dense enough. Large amount addition of Li_2CO_3 into the $CaSiO_3$–1 wt% Al_2O_3 ceramics led to the visible presence of $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ second phases. Fixing the Li_2CO_3 content at 1 wt%, a small amount of CuO addition significantly promoted the sintering process and lowered the densification temperature to 900 °C whereas its addition deteriorated the microwave dielectric properties of $CaSiO_3$–1 wt% Al_2O_3 ceramics. Based on 10 wt% $CaTiO_3$ compensation in temperature coefficient, good microwave dielectric properties of $\varepsilon_r = 8.92$, $Q \times f = 19{,}763$ GHz and $\tau_f = -1.22$ ppm/°C could be obtained for the 0.2 wt% CuO and 1.5 wt% Li_2CO_3 doped $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C. The chemical compatibility of the above ceramics with silver during the cofiring process has also been investigated, and the result showed that there was no chemical reaction between silver and ceramics, indicating that the as-prepared composite ceramics were suitable for low-temperature co-fired ceramics applications.

Keywords: Microwave dielectric properties; $CaSiO_3$ ceramic; Al_2O_3 ceramic; LTCC

1. Introduction

The rapid growth of the telecommunication and satellite broadcasting industry has created a high demand for microwave ceramic components. Multi-layer devices, which consist of alternating microwave dielectric ceramics and internal metallic electrode layers, enable the microwave components to be miniaturized and hybridized. As a metallic electrode, Ag has been widely used because of its high conductivity and low cost. Compared with the melting temperature of Ag, which is about 961 °C, the sintering temperature of the microwave dielectric ceramics is generally high, above 1300 °C. Therefore, from the point view of the fabrication of multi-layer devices, it is important to develop low temperature co-fired ceramics (LTCC) [1]. Several LTCC systems such as (Mg, Ca) TiO_3, $Ba_5Nb_4O_{15}$, $Li_2ATi_3O_8$ (A = Mg, Zn), $Ba_2Ti_9O_{20}$ and $ZnTiNb_2O_8$ have been investigated for microwave applications [2–8]. However, most of the reported LTCC microwave dielectric ceramics show a relatively high dielectric constant, which could be applied only in low frequency ranges such as 1.89 and 2.45 GHz. Comparatively, LTCC microwave dielectric ceramics with low-permittivity, which can work at a high frequency such as millimeter-wave range and are required for multilayer components particularly antennas and baluns, have not been extensively studied [9].

$CaSiO_3$ ceramic has been proved to be an excellent dielectric material with a low dielectric constant and a low dielectric loss, and it might be regarded as a suitable candidate for antennas and baluns. However, the sintering temperature range of neat $CaSiO_3$ ceramic is very narrow. Chakradhar et al. pointed out that it was difficult to obtain dense $CaSiO_3$ ceramic since its grains grew exceptionally and the bulk $CaSiO_3$ ceramic became more porous with the increase of the calcination temperature [10]. In our previous work, the sintering behavior and microwave dielectric properties of $CaSiO_3$ ceramics have been investigated by a traditional solid-state

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http://dx.doi.org/10.1016/j.ceramint.2013.08.031

process and a sol–gel method, respectively [11]. The maximum bulk density of $CaSiO_3$ ceramic sintered at 1340 °C prepared by the conventional solid-state process was 2.439 g/cm^3, and the microwave dielectric properties were $\varepsilon_r=6.59$ and $Q\times f=13{,}109$ GHz. Whereas for $CaSiO_3$ ceramic obtained by the sol–gel method, the maximum bulk density was 2.505 g/cm^3 and the microwave dielectric properties were $\varepsilon_r=6.69$ and $Q\times f=25{,}398$ GHz. The density value of the above two samples synthesized either by the traditional solid-state method or by the sol–gel method is much smaller than that of the theoretical density of the $CaSiO_3$ ceramic, which is 2.91 g/cm^3, indicating that it is difficult to obtain dense $CaSiO_3$ ceramic.

In order to improve the sintering characteristic and microwave dielectric properties, Sun and co-workers have used Mg^{2+} to substitute Ca^{2+} in the $CaSiO_3$ host to prepare the CaO–MgO–SiO_2 ceramics for LTCC applications [9,12]. In our previous work, we have found that the Al_2O_3 addition can restrict the growth of $CaSiO_3$ grains by surrounding their boundaries and also improve the bulk density of $CaSiO_3$–Al_2O_3 ceramics [13]. The optimum amount of Al_2O_3 addition was found to be 1 wt%, and the derived $CaSiO_3$–1 wt% Al_2O_3 ceramic presented improved microwave dielectric properties of $\varepsilon_r=6.66$ and $Q\times f=24{,}626$ GHz. However, the sintering temperature is high, which is about 1250 °C, so the $CaSiO_3$–Al_2O_3 ceramics cannot be co-fired with the Ag electrode.

In this work, CuO and Li_2CO_3 were added to the $CaSiO_3$–1 wt% Al_2O_3 ceramics to decrease the sintering temperature for LTCC applications, and $CaTiO_3$ was selected as compensation for a zero temperature coefficient. Furthermore, microstructure, microwave dielectric properties, green tape and co-firing with silver electrode of the CuO, Li_2CO_3 and $CaTiO_3$ added $CaSiO_3$–1 wt% Al_2O_3 ceramics were also investigated.

2. Experimental procedure

Specimen powders were prepared by a conventional solid-state method using commercial oxide powders (> 99.5%) of $CaCO_3$, SiO_2, Al_2O_3, CuO, Li_2CO_3 and TiO_2 as raw materials. Stoichiometric $CaCO_3$ and SiO_2 (TiO_2) powders were weighed and ground in ethanol for 24 h in a balling mill with ZrO_2 balls. Prepared powders were dried and calcined at 1200 °C for 2 h in air to obtain $CaSiO_3$ ($CaTiO_3$). The calcined powders were mixed as desired composition $CaSiO_3$ with 1 wt% Al_2O_3 and different amounts of CuO, Li_2CO_3 and $CaTiO_3$ additives and re-milled for 24 h. After drying and sieving, the as-prepared powders together with the organic binder (5 wt% polyvinyl alcohol) were uniaxially pressed under a pressure of 135 MPa into cylinders of 15 mm in diameter and 7–8 mm in thickness. These samples were then sintered at 875–1000 °C for 2 h in air with a heating rate of 5 °C/min.

The bulk densities of the sintered pellets were measured by the Archimedes method using distilled water as medium. The sintered bulks were broken up and ground to powders using an agate mortar. Crystal structures of the powders were performed by the X-ray diffraction (XRD, ARL XTRA) with Cu K_α radiation (36 kV, 30 mA, $2\theta=10$–$80°$). The polished surfaces of the ceramics were investigated by scanning electron microscopy (SEM, S-4800) after thermal etching. The microstructure observation of the green tape and the interface of ceramic and silver were performed by scanning electron microscopy (SEM, TM3000). The dielectric constants ε_r and the quality values $Q\times f$ at microwave frequencies were measured by Hakki–Coleman dielectric resonator method [14,15] using an Agilent 8719ET (50 MHz to 13.5 GHz) Network Analyzer. The temperature coefficient of the resonant frequency τ_f was also measured by the same method in the temperature range from 25 °C to 80 °C and calculated by the following equation:

$$\tau_f=\frac{f_{80}-f_{25}}{f_{25}\times 55}\times 10^6\ (\mathrm{ppm}/^\circ C)$$

where f_{80} and f_{25} represent the resonant frequency at 80 °C and 25 °C, respectively.

3. Results and discussion

3.1. Effects of the CuO addition on $CaSiO_3$–1 wt% Al_2O_3 ceramics

Fig. 1 shows the bulk densities of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 1 wt% Li_2CO_3 and different amounts of CuO additions. It is firstly evidenced that ceramics with the addition of CuO and Li_2CO_3 could be sintered at temperatures in the range of 900–925 °C whereas the ceramic without CuO addition requires a higher temperature, which is at least 975 °C, to be dense enough. The effect of CuO addition on crystal structure is investigated by the XRD analysis. Due to the small quantity of Al_2O_3, Li_2CO_3 and CuO, only the phase of $CaSiO_3$ (PDF 27-0088) is observed, as is shown in Fig. 2.

The SEM photographs of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 1 wt% Li_2CO_3 and different amounts of CuO sintered at 900 °C for 2 h are illustrated in Fig. 3(a)–(d).

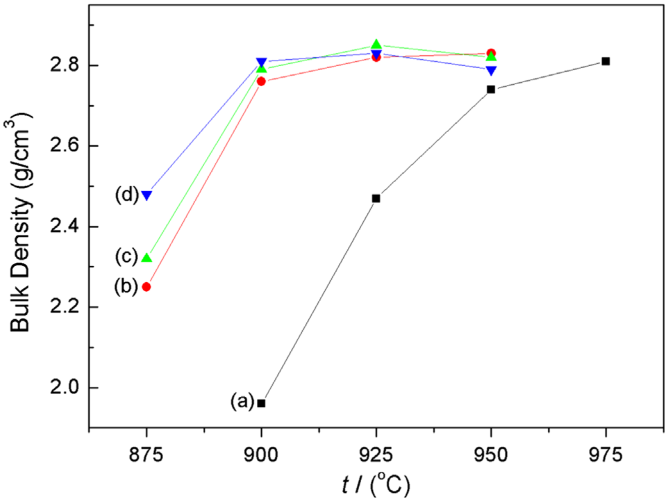


Fig. 1. Bulk densities of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 1 wt% Li_2CO_3 and (a) 0 wt%, (b) 0.2 wt%, (c) 0.4 wt% and (d) 0.8 wt% CuO additions.

Fig. 3(a) shows that the average grain size is about 0.5–1 μm and there are many pores in the bulks, which indicates that the $CaSiO_3$–1 wt% Al_2O_3 ceramic without CuO addition could be hardly sintered at 900 °C. With the addition of 0.2 wt% CuO, the grains grow and the porosity decrease, as shown in Fig. 3 (b). As CuO content increased gradually, the specimens become well-densified as well as rapid grain growth. In the case of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 1 wt % Li_2CO_3 and various amount of CuO, the volume fraction of the liquid increases with the CuO addition. The grains may dissolve into the liquid phase and rapidly rearrange, in which contact points between agglomerates will be dissolved and re-crystallized into grains. Base on this approach, the grains will grow and the pores will disappear.

The microwave dielectric properties (ε_r and $Q\times f$) of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 1 wt% Li_2CO_3 and various contents of CuO sintered at different temperatures are presented in Figs. 4 and 5. It is evident that the dielectric constants increase to a maximum value and then they are saturated (Fig. 4). With the fixed component, the relationship between dielectric constant and sintering temperature follows the same trend as that between bulk density and sintering temperature because a higher density is associated with a lower porosity and this could result in a higher dielectric constant. For quality factor value, as illustrated in Fig. 5,

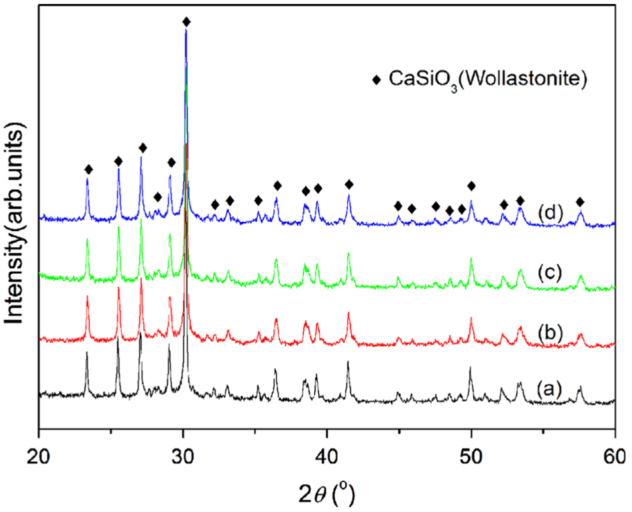


Fig. 2. XRD patterns of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C with 1 wt% Li_2CO_3 and (a) 0 wt%, (b) 0.2 wt%, (c) 0.4 wt% and (d) 0.8 wt% CuO additions.

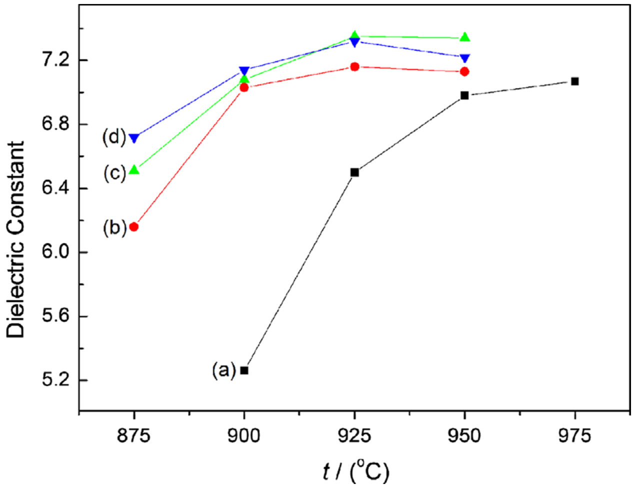


Fig. 4. Dielectric constants of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 1 wt% Li_2CO_3 and (a) 0 wt%, (b) 0.2 wt%, (c) 0.4 wt% and (d) 0.8 wt% CuO additions.

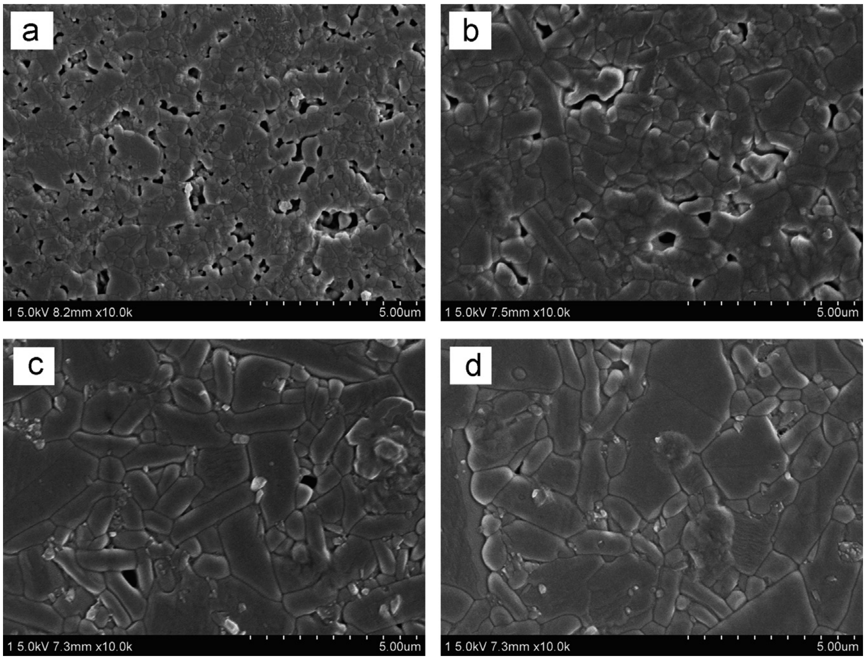


Fig. 3. SEM photographs of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C with 1 wt% Li_2CO_3 and (a) 0 wt%, (b) 0.2 wt%, (c) 0.4 wt% and (d) 0.8 wt% CuO additions.

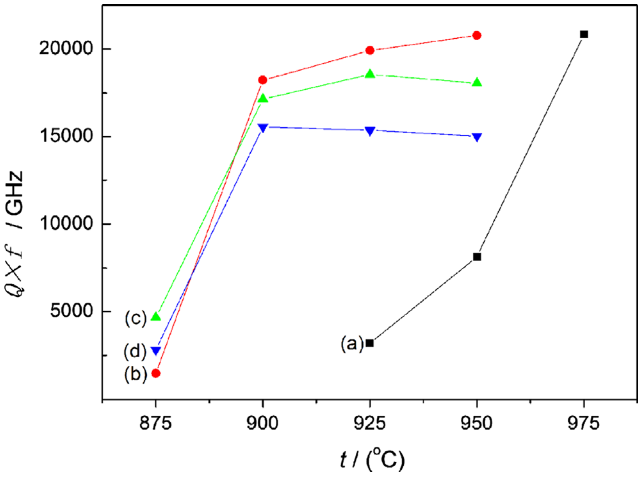


Fig. 5. $Q \times f$ values of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 1 wt% Li_2CO_3 and (a) 0 wt%, (b) 0.2 wt%, (c) 0.4 wt% and (d) 0.8 wt% CuO additions.

ceramics with a higher CuO addition exhibit lower value for the flux addition from 0 wt% to 0.8 wt%. With 0.2 wt%, 0.4 wt% and 0.8 wt% CuO additions, the $Q \times f$ value of the $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C is 18,220 GHz, 17,144 GHz and 15,548 GHz, respectively. Although a small amount of CuO addition can significantly promote the densification process and lower the sintering temperature to 900 °C, the above results indicate that CuO addition deteriorates the microwave dielectric properties of $CaSiO_3$–1 wt% Al_2O_3 ceramics.

3.2. *Effects of the Li_2CO_3 addition on $CaSiO_3$–1 wt% Al_2O_3 ceramics*

Fig. 6 shows the X-ray diffraction patterns of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 0.2 wt% CuO and different amounts of Li_2CO_3 sintered at 900 °C for 2 h. For the specimens with 0.8 wt% Li_2CO_3 content, the entire diffraction pattern correlates to the $CaSiO_3$ (PDF 27-0088) ceramic and no other phases are found. However, when the Li_2CO_3 content exceeds 1.0 wt%, peaks corresponding to $Li_2Ca_3Si_6O_{16}$ (PDF 31-0713) and $Li_2Ca_4Si_4O_{13}$ (PDF 31-0714) phases appear and the intensity of these peaks increases with the increase of Li_2CO_3 addition. From the bulk densities of $CaSiO_3$–1 wt% Al_2O_3 ceramics shown in Fig. 7, it is clear that the Li_2CO_3 addition improves the sintering process and decreases the densification temperature. With the Li_2CO_3 addition amounts of 0.4 wt%, 0.8 wt%, 1.0 wt%, 1.5 wt% and 2.0 wt%, the bulk density of the ceramics sintered at 900 °C is 2.59 g/cm^3, 2.73 g/cm^3, 2.76 g/cm^3, 2.79 g/cm^3 and 2.77 g/cm^3, respectively. According to the phase diagram of Li_2O–CaO–SiO_2 solid solution, a eutectic point at 954 °C exists between the $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ phases [16,17]. Therefore, it is considered that Li_2CO_3 will exist as liquid phase during the sintering process and assist the densification of $CaSiO_3$–1 wt% Al_2O_3 ceramics, and then react with $CaSiO_3$ to form $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ phases.

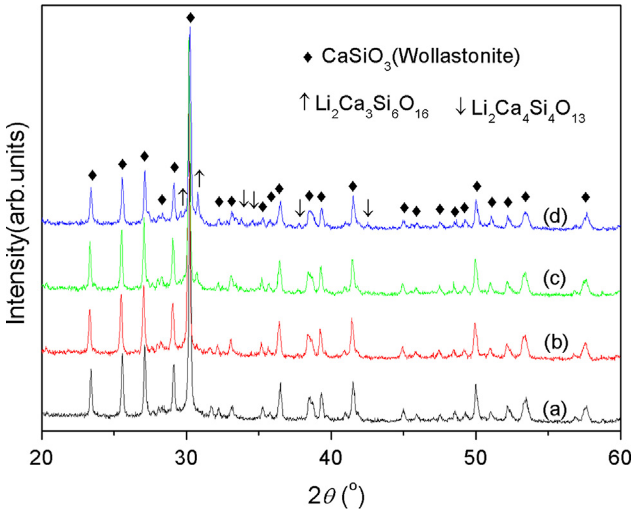


Fig. 6. XRD patterns of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C with 0.2 wt% CuO and (a) 0.8 wt%, (b) 1.0 wt%, (c) 1.5 wt% and (d) 2.0 wt% Li_2CO_3 additions.

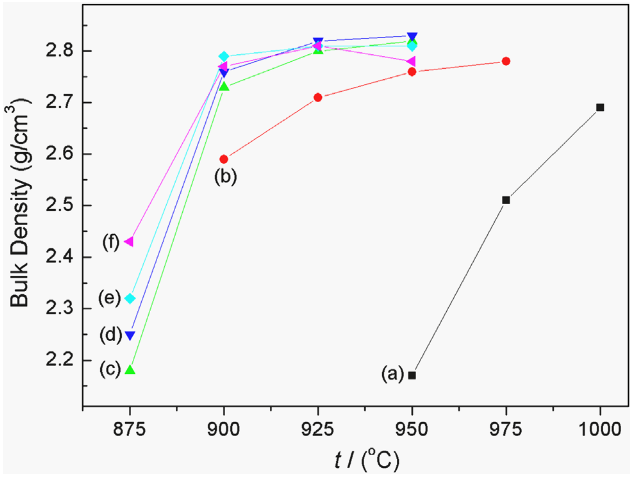


Fig. 7. Bulk densities of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 0.2 wt% CuO and (a) 0.2 wt%, (b) 0.4 wt%, (c) 0.8 wt%, (d) 1.0 wt%, (e) 1.5 wt% and (f) 2.0 wt% Li_2CO_3 additions.

The SEM photographs of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 0.2 wt% CuO and different amounts of Li_2CO_3 sintered at 900 °C for 2 h are illustrated in Fig. 8. When the Li_2CO_3 addition is increased from 0.8 wt% to 1.5 wt%, the grains grow and the specimens become densified, as shown in Fig. 8(a)–(c). However, the surface of the ceramics seems to be porous as the Li_2CO_3 addition reaches 2.0 wt%. One possible explanation is that the volatilization of the excess Li_2CO_3 happens and results in porosity. During the sintering process, the liquid phase from Li_2CO_3 may penetrate the grains completely and then react with $CaSiO_3$ to form $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ second phases, in which case the $CaSiO_3$ grains will be separated from one another by these second phases. Energy dispersive spectrum (EDS) is taken for the composition analysis. From Fig. 9(a) and (b), the coarse grains are found to be $CaSiO_3$, and Al_2O_3 grains are observed around the $CaSiO_3$ grains, which is consistent with our previous work

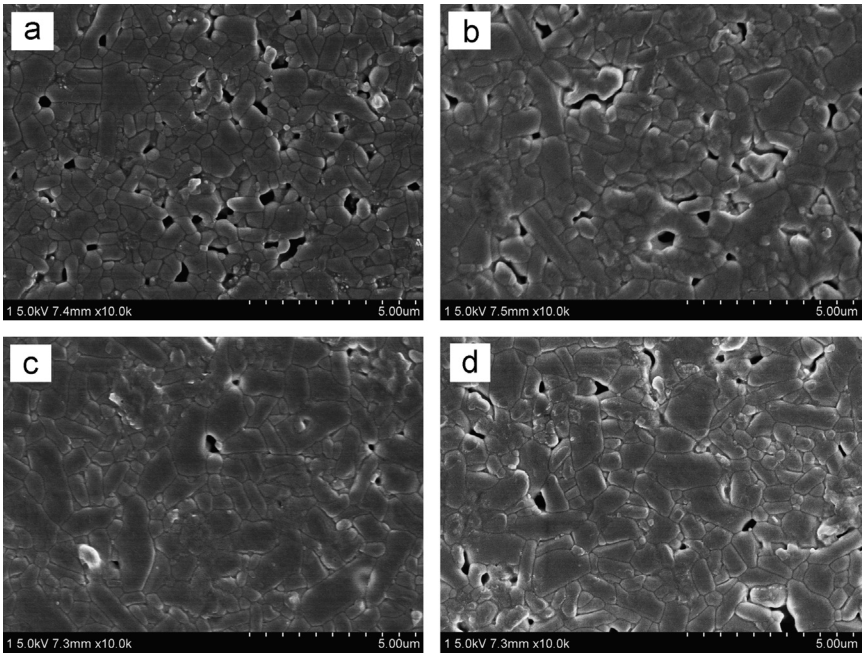


Fig. 8. SEM photographs of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C with 0.2 wt% CuO and (a) 0.8 wt%, (b) 1.0 wt%, (c) 1.5 wt% and (d) 2.0 wt% Li_2CO_3 additions.

[13]. Fig. 9(c) shows Ca element is less than Si element, combined with the XRD analysis, this may mean that there exist $Li_2Ca_3Si_6O_{16}$ phase which can restrict the growth of $CaSiO_3$ grains by surrounding their boundaries. Compared with CuO addition, Li_2CO_3 addition can not only promote the sintering process, but also result in the presence of $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ second phases, which could restrict the $CaSiO_3$ grains growth.

The dielectric constant (ε_r) and quality factor value ($Q \times f$) of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 0.2 wt% CuO and different amounts of Li_2CO_3 sintered at different temperatures are presented in Figs. 10 and 11, respectively. The dielectric constant of specimens is approximately proportional to the sintered bulk density of specimens. The dielectric constant of $CaSiO_3$–1 wt% Al_2O_3 ceramics with 0.2 wt%, 0.4 wt%, 0.8 wt%, 1.0 wt%, 1.5 wt% and 2.0 wt% Li_2CO_3 addition sintered at 900 °C is 6.39, 6.89, 7.03, 7.15 and 7.18, indicating the dielectric constant increases slightly with the increase of the Li_2CO_3 amount. Combined with the empirical rule of $\ln\varepsilon_r = \Sigma_i V_i \ln\varepsilon_{ri}$, where V_i and ε_{ri} are the volume fraction and permittivity of each phase, respectively [12,18], it is considered that the dielectric constant of the $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ compounds is larger than that of $CaSiO_3$–1 wt% Al_2O_3. Fig. 11 shows that the $Q \times f$ value of the $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C gradually increases with the increase of the Li_2CO_3 addition from 0.2 wt% to 1.5 wt%, and a maximum $Q \times f$ value of 21,950 GHz is obtained when the ceramics are incorporated with 0.2 wt% CuO and 1.5 wt% Li_2CO_3. Further increase of the Li_2CO_3 addition deteriorates the microwave dielectric properties, and the $Q \times f$ value decreases to 20,371 GHz as the Li_2CO_3 addition increases to 2.0 wt%. We conclude that high amounts of Li_2CO_3 and CuO additions produce second phases spreading in $CaSiO_3$–1 wt% Al_2O_3 ceramics and degrade the $Q \times f$ value. In addition, volatilization of the excess Li_2CO_3 additions results in porosity, which will deteriorate the microwave dielectric properties.

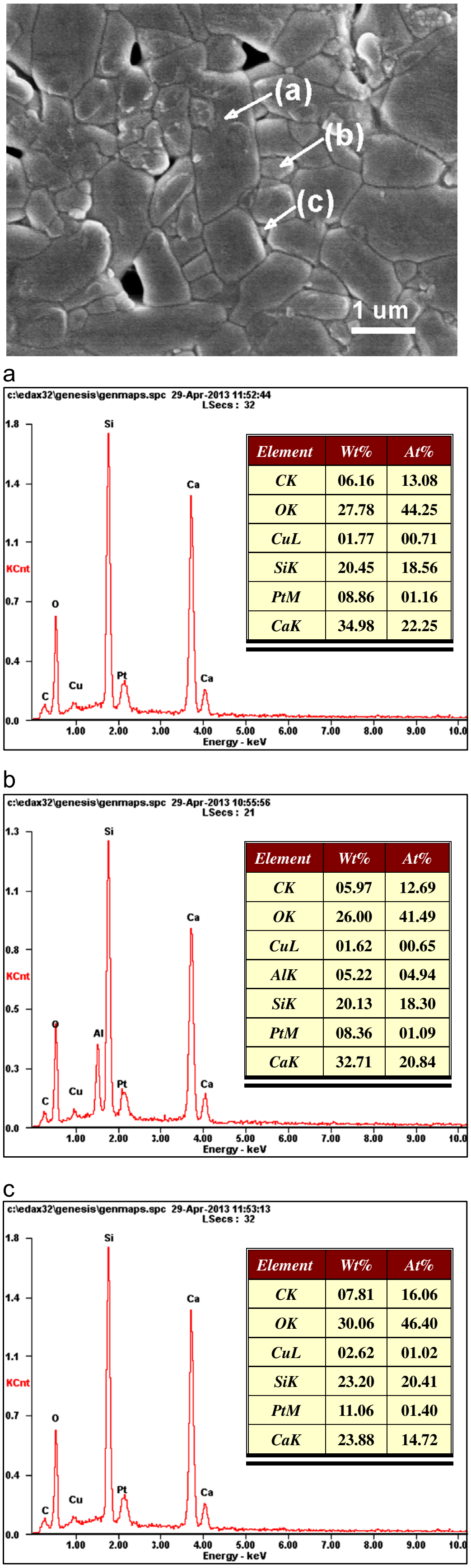
The variation of τ_f as a function of different amounts of CuO and Li_2CO_3 additions is also investigated. The τ_f changes little with different amount of CuO and Li_2CO_3 additions. With the doping concentration of 0.2 wt% CuO and 1.5 wt% Li_2CO_3, the τ_f value of $CaSiO_3$–1 wt% Al_2O_3 ceramic sintered at 900 °C is -46 ppm/°C.

3.3. Effects of the $CaTiO_3$ addition on $CaSiO_3$–1 wt% Al_2O_3 ceramics

In order to obtain a zero temperature coefficient, $CaTiO_3$ was selected as compensation [19]. Fig. 12 shows the microwave dielectric properties (ε_r, $Q \times f$ and τ_f) of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C with 0.2 wt% CuO, 1.5 wt% Li_2CO_3 and various $CaTiO_3$ contents. It is evident that the temperature coefficient (τ_f) decreases with the increase of $CaTiO_3$ content and the τ_f value is -1.22 ppm/°C as the $CaTiO_3$ content reaches 10 wt%. Due to the high dielectric constant ($\varepsilon_r = 170$) and low quality factor value ($Q \times f = 3600$ GHz) [20], $CaTiO_3$ addition leads to the increase of dielectric constant and decrease of $Q \times f$ value. When the $CaTiO_3$ content is 10 wt%, the dielectric constant and $Q \times f$ value of the ceramic is 8.92 and 19,763 GHz, respectively.

3.4. Co-fired with Ag electrode

In order to investigate ceramics slurry for tape casting, the $CaSiO_3$–1 wt% Al_2O_3 powders doped with 0.2 wt% CuO, 1.5 wt% Li_2CO_3 and 10 wt% $CaTiO_3$ were first mixed with solvent and dispersant in a ball mill for 12 h. Then the binder



a

Element	*Wt%*	*At%*
CK	06.16	13.08
OK	27.78	44.25
CuL	01.77	00.71
SiK	20.45	18.56
PtM	08.86	01.16
CaK	34.98	22.25

b

Element	*Wt%*	*At%*
CK	05.97	12.69
OK	26.00	41.49
CuL	01.62	00.65
AlK	05.22	04.94
SiK	20.13	18.30
PtM	08.36	01.09
CaK	32.71	20.84

c

Element	*Wt%*	*At%*
CK	07.81	16.06
OK	30.06	46.40
CuL	02.62	01.02
SiK	23.20	20.41
PtM	11.06	01.40
CaK	23.88	14.72

Fig. 9. SEM and EDS images of $CaSiO_3$–1 wt% Al_2O_3 ceramic sintered at 900 °C with 0.2 wt% CuO and 2.0 wt% Li_2CO_3 additions.

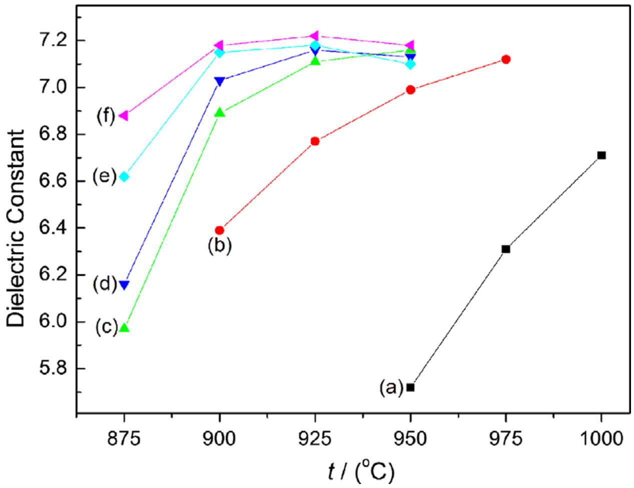


Fig. 10. Dielectric constants of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 0.2 wt% CuO and (a) 0.2 wt%, (b) 0.4 wt%, (c) 0.8 wt%, (d) 1.0 wt%, (e) 1.5 wt% and (f) 2.0 wt% Li_2CO_3 additions.

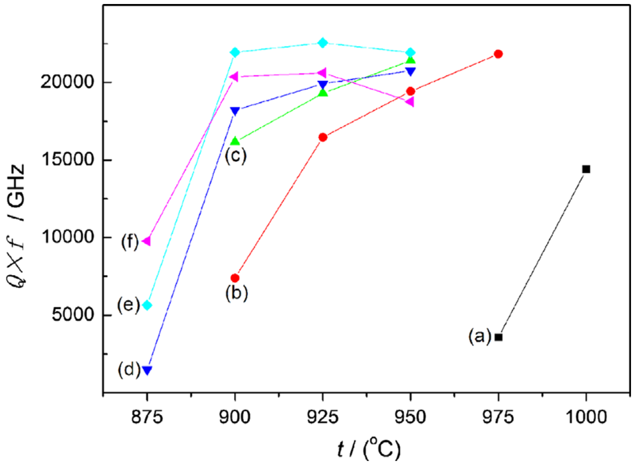


Fig. 11. $Q\times f$ values of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at different temperatures with 0.2 wt% CuO and (a) 0.2 wt%, (b) 0.4 wt%, (c) 0.8 wt%, (d) 1.0 wt%, (e) 1.5 wt% and (f) 2.0 wt% Li_2CO_3 additions.

and plasticizer were added, and mixed for another 12 h to obtain the slurry. The solvent was the mixture of toluol and ethanol, and the dispersant was menhaden fish oil. The binder itself was polyvinyl butyral (PVB), and the compatible plasticizer was butyl benzyl phthalate (S160). The viscosity of the ceramics slurry was 1865 mPa s. Fig. 13 shows the microstructures of the green tapes. In the green state, the average particle size of LTCC ceramics is around 1 μm, and the microstructures of the green tapes are uniform and there is no agglomeration. On the other hand, the surface of the green tape is glabrous. The green tape has very high density and tensile strength.

For compatibility tests, ceramics sheet with Ag electrodes were co-fired and analyzed to detect interactions between the low-fired samples and electrodes. SEM analysis reveals that there is no interaction forming new phases after firing, as shown in Fig. 14. It is obvious that the LTCC materials do not react with Ag electrodes. Therefore, $CaSiO_3$–Al_2O_3 ceramics incorporated with CuO–Li_2CO_3–$CaTiO_3$ additions could be regarded as suitable candidates for LTCC materials due to their

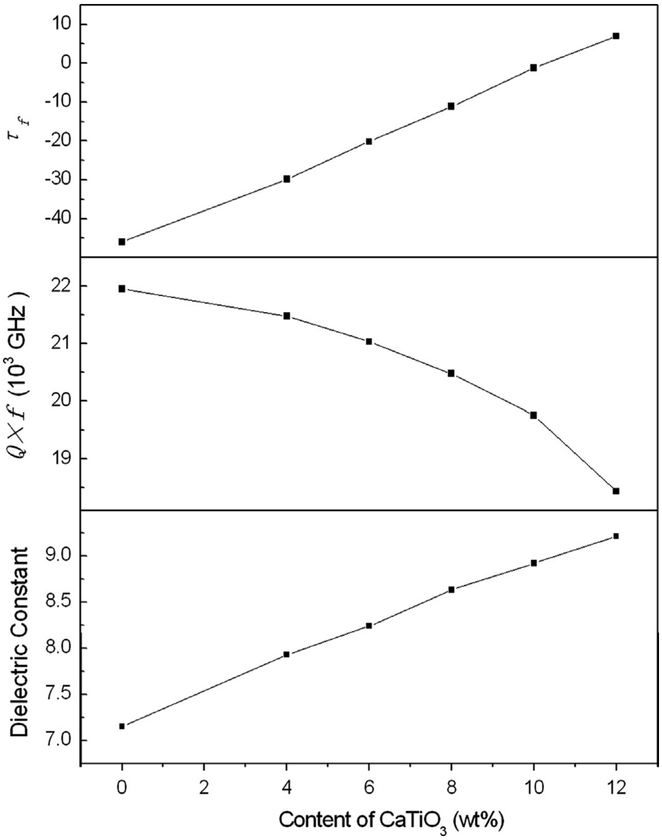


Fig. 12. Microwave dielectric properties of $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C incorporated with 0.2 wt% CuO, 1.5 wt% Li_2CO_3 and different amounts of $CaTiO_3$.

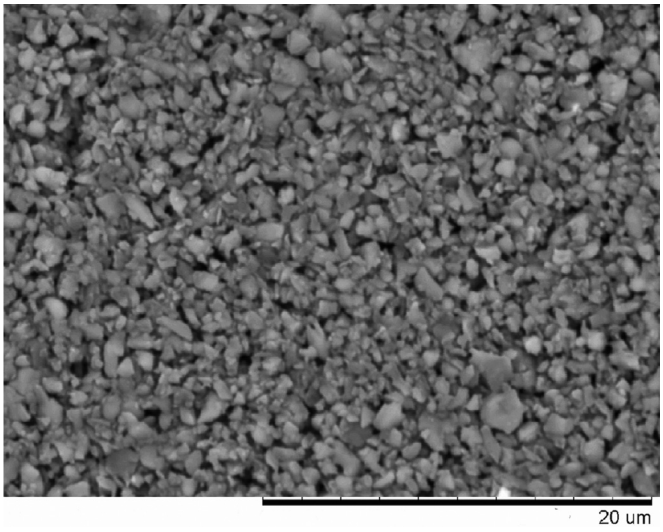


Fig. 13. SEM micrograph of the green tape.

low sintering temperature, good microwave dielectric properties and compatibility with Ag electrodes.

4. Conclusions

A small amount of CuO and Li_2CO_3 addition into the $CaSiO_3$–1 wt% Al_2O_3 ceramic could lower the densification temperature from 1250 °C to 900 °C. With a single addition of 1 wt% Li_2CO_3, the $CaSiO_3$–1 wt% Al_2O_3 ceramic required at least 975 °C to be dense enough, whereas a small amount of CuO addition significantly promoted the sintering process and lowered the densification temperature to 900 °C. However, CuO addition deteriorated the microwave dielectric properties of $CaSiO_3$–1 wt% Al_2O_3 ceramics. Fixing the CuO content at 0.2 wt%, the Li_2CO_3 addition not only promoted the sintering process, but also resulted in the presence of $Li_2Ca_3Si_6O_{16}$ and $Li_2Ca_4Si_4O_{13}$ second phases, which could restrict the $CaSiO_3$ grains growth. Based on 10 wt% $CaTiO_3$ compensation in temperature coefficient, good microwave dielectric properties of $\varepsilon_r = 8.92$, $Q \times f = 19{,}763$ GHz and $\tau_f = -1.22$ ppm/°C were obtained for the 0.2 wt% CuO and 1.5 wt% Li_2CO_3 doped $CaSiO_3$–1 wt% Al_2O_3 ceramics sintered at 900 °C. The as-prepared low-temperature sintering ceramics powders were suitable for the tape casting process, and these ceramics were good candidates for LTCC applications with silver electrode.

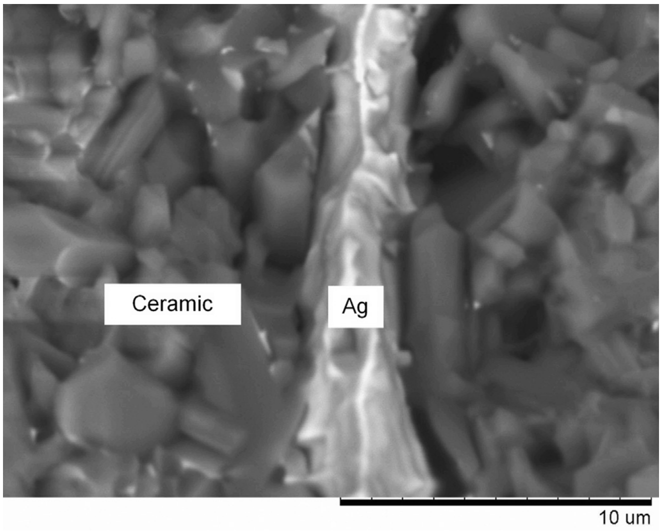


Fig. 14. SEM micrograph of $CaSiO_3$–1 wt% Al_2O_3 ceramics incorporated with 0.2 wt% CuO, 1.5 wt% Li_2CO_3 and 10 wt% $CaTiO_3$ co-fired with Ag in air at 900 °C for 2 h.

Acknowledgment

This work was financially supported by the Science and Technology Innovative Research Team of Zhejiang Province (2010R50016) and the Nature Science Foundation of Zhejiang Province (Y1110648).

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