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Microwave dielectric properties of glass-MCT low temperature co-firable ceramics

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Abstract

Interaction between the $(Mg_{0.95}Ca_{0.05})TiO_3$ (MCT) microwave dielectric materials and BaBSiO glass materials was examined. Rigorous interaction occurs when the MCT-glass composite materials were densified at 800–900 °C, which, fortunately, does not seriously degrade the microwave dielectric properties of the materials such that dielectric constant (K) = 8–10 was achieved for MCT-glass composite materials, no matter whether they were in pellet or tape form. The MCT–glass composites with MCT–BaBSiO = 50:50 (vol.%) possess low shrinkage characteristics, when densified at 800–900 °C for 10 min, and were used as constrain layer for an LTCC sandwich. LTCC tapes possessing very small x–y shrinkage ($\leqq 0.05\%$) and good microwave properties ($k = 8.0$) are thus synthesized.

Keywords: Microwave dialectric property; Glass-ceramic composite; Low-temperature co-firable ceramics

1. Introduction

Low temperature co-firable ceramics (LTCC) possessing good microwave dielectric properties have recently been widely investigated, due to the necessity for miniaturization of microwave devices to reduce the size of wireless communication systems.[1–3] However, the microwave dielectric materials, which possess a high quality factor and large dielectric constant, usually need very high sintering temperature and long soaking time to achieve a high enough density. On the other hand, use of the Ag material as conducting material for transmission lines and ground planes is needed in order to minimize the microwave absorption loss. Reduction of the sintering temperature for the microwave material to a level co-firable with Ag-electrode materials is thus called for. Generally, low softening temperature glass materials were mixed with the ceramic materials to reduce the firing temperature needed.[4–9] However, network formers contained in the glass materials may absorb the microwave power profoundly in high frequency regime, degrading the quality factor for the materials.[10] In this paper, the microwave dielectric properties of the glass materials and the effect of processing parameters on the characteristics of the glass-to-ceramic composites were investigated.

2. Experiments

Pure $(Mg_{0.95}Ca_{0.05})TiO_3$, MCT, materials were first prepared by mixed oxide process. MgO, CaO and TiO_2 with a molar ratio of nominal composition $(Mg_{0.95}Ca_{0.05})$-TiO_3 were mixed and then calcined at 1000 °C (2 h). Thus obtained powders were then pulverized down to about 0.5 μm size by a Dynomill. The glass powders of the composition shown in Table 1 was then mixed with $(Mg_{0.95}Ca_{0.05})TiO_3$ in 50-to-50 vol.%, followed by pelletization and then sintering at 800–900 °C for 30 min. The sintered density was measured using Archimedes' method. The crystal structure and the microstructure of the sintered MCT samples were examined by X-ray diffraction analysis (XRD, Rigaku D/max-IIB) and scanning electron microscopy (SEM, Joel JSM-840A), respectively. The microwave dielectric properties were measured using H.P.8722A network analyzer in a resonant cavity or a split-post test fixture.[11–13]

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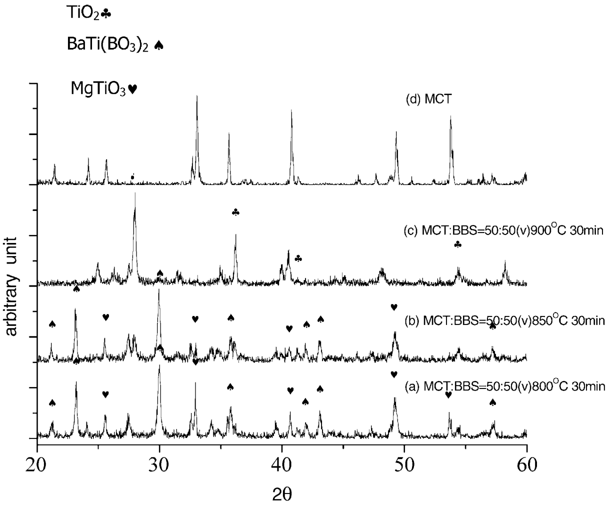
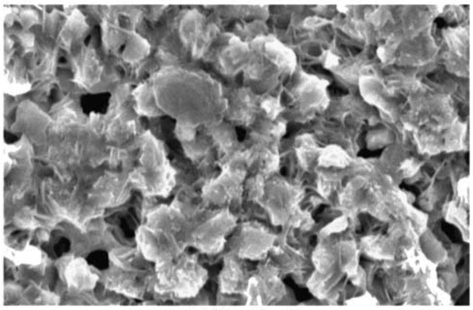
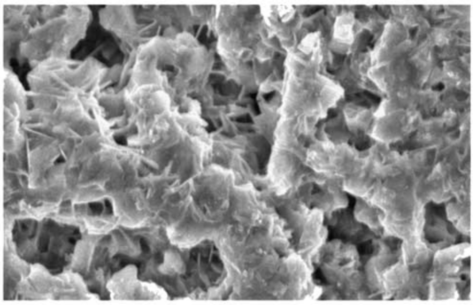
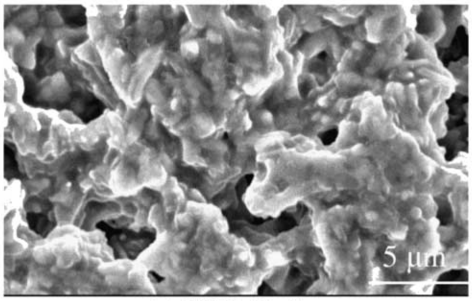


Fig. 1. X-ray diffraction patterns for MCT–BaBSiO glass composites (in pellets form), sintered at (a) 800 °C, (b) 850 °C and (c) 900 °C for 30 min, whereas (d) is the XRD pattern for MCT ceramics. (MCT:BaBSiO = 50:50 (vol.%); MCT = $(Mg_{0.95}Ca_{0.05})TiO_3$).


(a) 800℃ 30 min


(b) 850℃ 30 min



(c) 900℃ 30 min

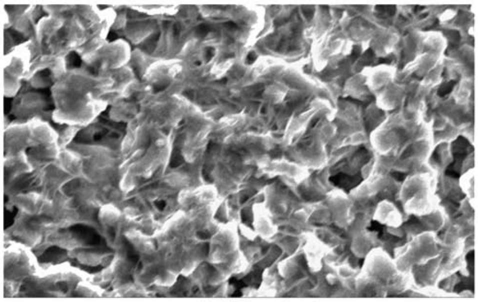
Fig. 2. SEM micrographs for MCT–BaBSiO glass composites (in pellet form), sintered at (a) 800 °C, (b) 850 °C and (c) 900 °C for 30 min.

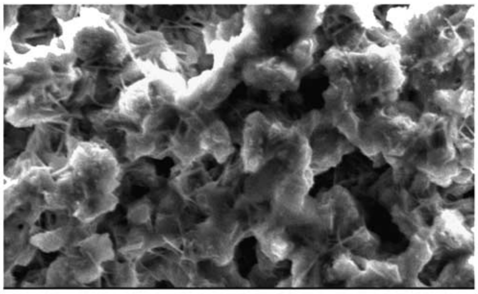
3. Results and discussion

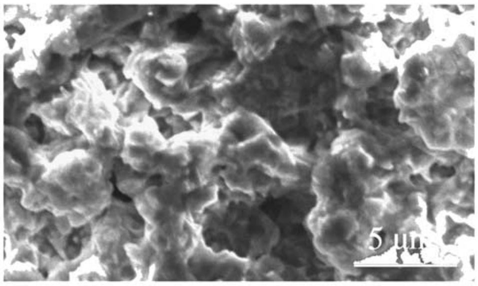
To study the interaction between the MCT ceramics and BaBSiO glass materials, the MCT and glass mixture (50:50 vol.%) were pelletized and then densified at 800–900 °C for 30 min. Fig. 1 shows that the two materials interact rigorously, forming a secondary phase $BaTi(BO_3)_2$. The proportion of MCT phase decreases with increasing processing temperature. SEM micrographs shown in Fig. 2 reveal that the $BaTi(BO_3)_2$ phase is of thin-plate like geometry. Thus form glass-ceramic composites are quite porous, about 75% T. D., possessing

Table 1
The composition of BaBSiO glass

Materials	B_2O_3	SiO_2	BaO
BaBSiO (wt.%)	45	13	42


(a) 800℃ 10 min


(b) 850℃ 10 min



(c) 900℃ 10 min

Fig. 3. SEM micrographs for MCT–BaBSiO glass composites (in tape form), sintered at (a) 800 °C, (b) 850 °C and (c) 900 °C for 10 min. (MCT:BaBSiO = 50:50 vol.%; MCT = $(Mg_{0.95}Ca_{0.05})TiO_3$).

dielectric constant about K = 10.0–10.6, regardless of the processing temperature and soaking time interval.

The MCT and glass mixture (50:50 vol.%) was then tape-castled, debindered and then densified at 800–900 °C for 10 min, forming MCT–glass composite tapes. Fig. 3 indicates that tapes are even more porous (about 70% T. D.), which is not much denser than the green pellets. The MCT-glass composite tapes possess slightly smaller microwave dielectric properties ($K \approx 7.8$–9.0). It should be mentioned that microwave dielectric properties for the MCT-glass pellets were measured using a cavity method, whereas these for MCT–glass tapes were measured using a split-post test fixture. The latter method contain a fixed gap between the two dielectric resonator posts, which may induce microwave losses, resulting in lower apparent K and $Q \times f$ values. Therefore, it can be concluded that the MCT-glass tapes preserve the same microwave dielectric properties of the materials. X-ray diffraction patterns shown in Fig. 4 indicate again the rigorous interaction between the MCT ceramics and BaBSiO glass materials.

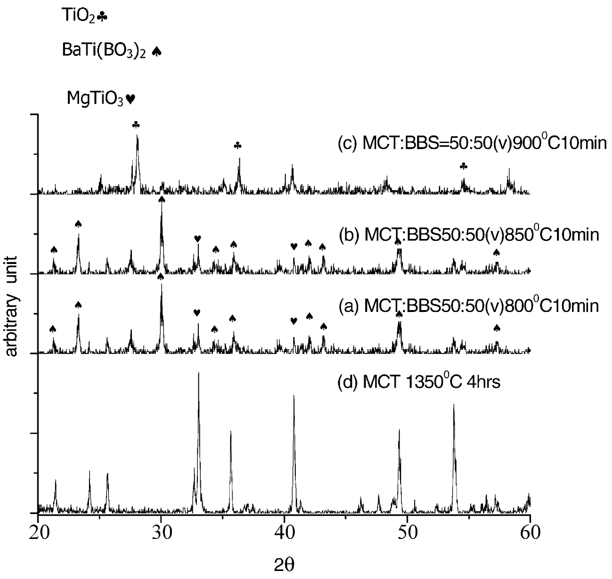


Fig. 4. X-ray diffraction patterns for MCT–BaBSiO glass composites (in tape form), sintered at (a) 800 °C, (b) 850 °C and (c) 900 °C for 10 min. (MCT:BaBSiO = 50:50 vol.%; MCT = $(Mg_{0.95}Ca_{0.05})TiO_3$).

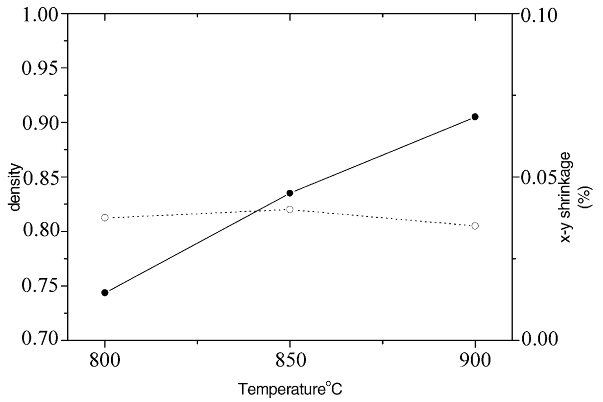
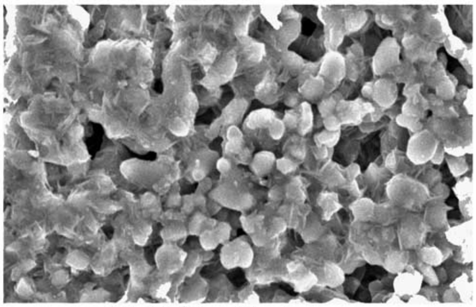
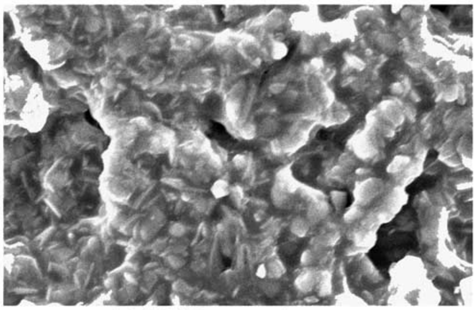


Fig. 5. The variation of sintered density (solid line) and x–y shrinkage (dotted line) for LTCC sandwiched tapes with densification temperature. The LTCC sandwiched tapes contain a glass layer sandwiched in between 2 MCT-glass (50:50 vol.%) composite layers.

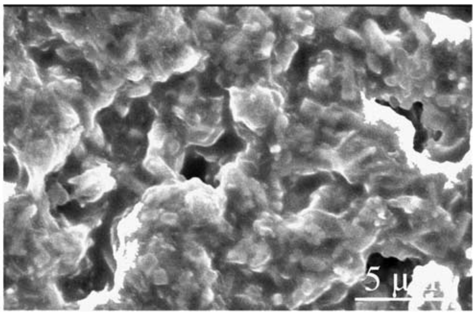
One of the interesting phenomena for the MCT–glass composite tapes is that the MCT–glass (50:50 vol.%) materials possess very low shrinkage characteristics, which can potentially be used as constrain layer to hinder the shrinkage of a LTCC tapes in x- and y-direction. A multilayer LTCC tape, containing a BaBSiO glass sandwiched in between the two MCT–glass (50:50 vol.%) composite materials, was thus made. Fig. 5 shows that the sintered density of such a LTCC sandwich increases with processing temperature from 75% to 90% T. D. (solid line), but the x–y shrinkage was maintained at a very low value (≦0.05%).



(a) 800°C 10 min

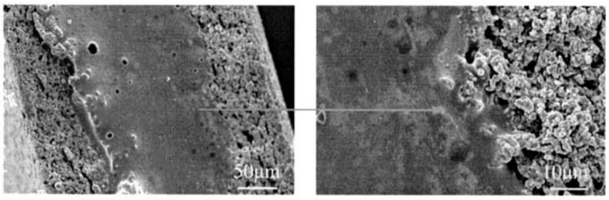


(b) 850°C 10 min

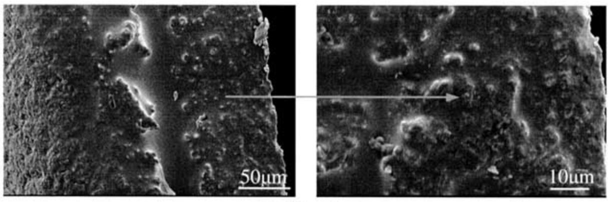


(c) 900°C 10 min

Fig. 6. Surface morphology of LTCC sandwiched tapes densified at (a) 800 °C, (b) 850 °C and (c) 900 °C for 10 min.



(a) 800℃ 10min



(b) 850℃ 10min

Fig. 7. Cross-sectional micrographs for LTCC sandwiched tapes densified at (a) 800 °C and (b) 850 °C for 10 min.

SEM micrographs shown in Fig. 6 reveals that the surface porosity decreases with increased firing temperature, which is even more clearly illustrated by cross-sectional micrographs in Fig. 7(a) and (b) for 800 °C (10 min) and 850 °C (10 min) densified LTCC sandwich, respectively. The K and $Q \times f$ values of the LTCC sandwich are insignificantly different from the MCT-glass composited tapes. Such a phenomenon is understandable, as the higher sintering density tends to compensate for the decrease in MCT proportion in LTCC sandwich. Figs. 6 and 7 infers that the relative thickness of glass and MCT-glass composite layers need to be closely controlled, such that the proportion of glass layer is sufficient to fill in the pores in MCT-glass composite layers without leaving behind unreacted glass.

4. Conclusion

Interaction between the MCT microwave dielectric materials and BaBSiO glass materials was examined. Rigorous interaction occurs when the MCT–glass composite materials were densified at 800–900 °C, which, fortunately, does not seriously degrade the microwave dielectric properties of the materials such that K = 8–10 were achieved for MCT–glass composite materials, no matter whether they are in pellet or tape form. The MCT-glass composites with MCT:BaBSiO = 50:50 (vol.%) possess low shrinkage characteristics, when densified at 800–900 °C for 10 min, and was used as constrain layer for a LTCC sandwich. LTCC tapes possessing very small *x*–*y* shrinkage (≦0.05%) and good microwave properties ($k = 8.0$) are thus synthesized.

Acknowledgements

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