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Journal of the European Ceramic Society 26 (2006) 1909–1912

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High Q dielectric resonator material with low dielectric constant for millimeter-wave applications

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Abstract

Al_2O_3—TiO_2 ceramics has great promise as a dielectric resonator material for millimeter-wave use. The Al_2O_3—TiO_2—MnO system provides low dielectric constant and high quality factor (Q). TiO_2 and Al_2O_3 can compensate each other's temperature coefficient of resonant frequency. It is important to suppress the appearance of Al_2TiO_5 phase for obtaining high Q-value. In this study, dense Al_2O_3—TiO_2 mixed ceramics without Al_2TiO_5 phase was synthesized by lowering the sintering temperature. The addition of MnO decreases the sintering temperature from 1400 to 1300 °C. In this temperature range, Al_2TiO_5 phase is hardly formed. X-ray powder diffraction (XRD) patterns show that the ceramics is composed of Al_2O_3 and TiO_2 phases only. Dense and well dispersed mixture of Al_2O_3 and TiO_2 grains is observed by scanning electron microscopy (SEM).

The $0.9Al_2O_3$—$0.1TiO_2$ ceramics doped with 0.4 mol% of MnO has dielectric constant of 12.4, $Q \times f$ of 274,000 GHz at 76 GHz, and temperature coefficient of resonant frequency of +0.4 ppm/°C. The dielectric constant and the temperature coefficient of resonant frequency can be controlled by TiO_2 content. This ceramics is suitable for millimeter-wave applications because of its low dielectric constant, high Q-value, and small temperature coefficient of resonant frequency.

Keywords: Sintering; Dielectric properties; Al_2O_3—TiO_2; Al_2TiO_5

1. Introduction

As a result of the contribution of dielectric materials, telecommunication technologies such as mobile phones and wireless LAN have developed significantly. Utilized frequency has also increased from microwave to millimeter-wave range because large quantity of information must be transported with high speed. Dielectric resonator materials for millimeter-wave use are required to have high quality factor (Q), relatively low dielectric constant (ε_r) and small temperature coefficient of resonant frequency (τ_f).

Al_2O_3 with low ε_r (10) and a high Q has promise as millimeter-wave application material but cannot be used for practical application because of large negative τ_f (−60 ppm/°C). Addition of TiO_2 can compensate the τ_f with its positive τ_f (+450 ppm/°C). But it is difficult to obtain high Q-value and small τ_f because of the formation of Al_2TiO_5 phase.[1,2] Tzou et al. reported that Al_2O_3—TiO_2 ceramics achieved $\tau_f = 0$ ppm/°Cby lowering the sintering temperature with a low melting point glass.[2] However, the $Q \times f$ values of these ceramics were relatively low (about 10,000 GHz) and dense ceramics was not obtained at a low sintering temperature of 1300 °C. In this study, the addition of MnO was attempted to synthesize the Al_2O_3—TiO_2 mixed ceramics without the formation of Al_2TiO_5 phase.

1.1. Experimental procedure

High purity (>99.9%) Al_2O_3, TiO_2, and $MnCO_3$ powders were weighed and mixed with vinyl acetate emulsion. Well dispersed slurries were dried at 100 °C. The mixed powders were pressed under 350 MPa to form cylindrical pellets of 12 mm in diameter and 6 mm thickness. The pellets were sintered in the range between 1250 and 1400 °C for 4 h in air. The crystal structure of the sintered pellets was investigated by X-ray powder diffraction (XRD). The microstructure was observed by scanning electron microscopy (SEM) with wavelength dispersive X-ray spectrometry (WDX). The microwave dielectric properties (ε_r, Q, and τ_f) were evaluated using a pair of parallel conducting silver plates in the TE_{011} mode with dielectric resonator method.[3] τ_f was estimated by comparing of the resonant frequencies measured at 25 and 55 °C.

$$\tau_f = \frac{f_{0\,55\,^\circ\mathrm{C}} - f_{0\,25\,^\circ\mathrm{C}}}{\Delta T \times f_{0\,25\,^\circ\mathrm{C}}} \times 10^6 \quad (\mathrm{ppm}/^\circ\mathrm{C})$$

$$\text{where} \quad \Delta T = 55 - 25 = 30\,^\circ\mathrm{C}$$

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doi:10.1016/j.jeurceramsoc.2005.09.019

ε_r and Q in millimeter-wave range were also evaluated by dielectric resonator method using NRD guides.[4]

2. Results and discussion

Fig. 1 shows the sintering temperature dependence of dielectric properties for $(1-x)Al_2O_3$–$xTiO_2$ ceramics, where $x = 0.10, 0.15$ and 0.20. When the ceramics is sintered at 1300 °C, ε_r and $Q \times f$ are low, and τ_f cannot be measured because resonant peaks do not appear due to low $Q \times f$. The $Q \times f$ has maximum value on sintering at 1350 °C. The τ_f changes to negative values with the increase in sintering temperature from 1350 to 1400 °C. These results indicate the low reproducibility of $Q \times f$ and τ_f.

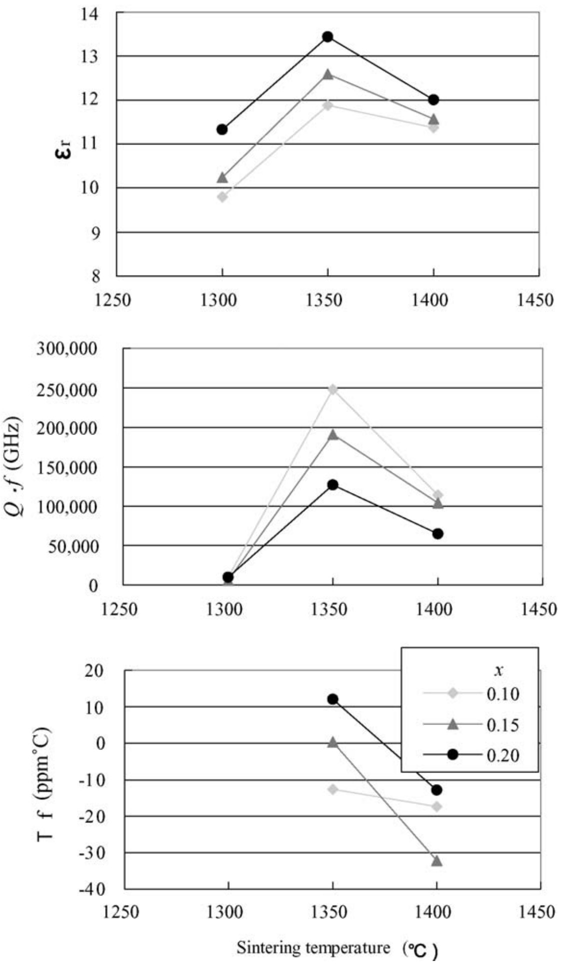

Fig. 1. Sintering temperature dependence of dielectric properties for $(1-x)Al_2O_3$—$xTiO_2$ ceramics.

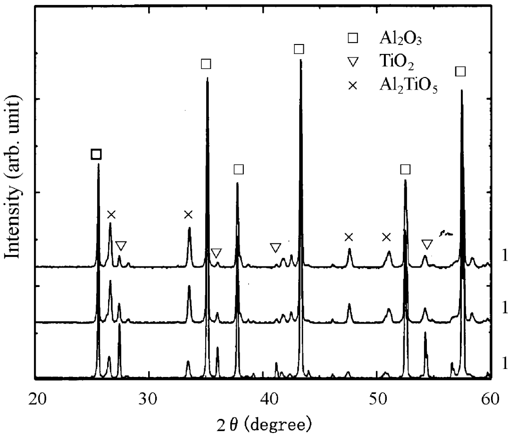

Fig. 2. XRD patterns of $0.9Al_2O_3$—$0.1TiO_2$ ceramics sintered at 1300–1400 °C for 4 h.

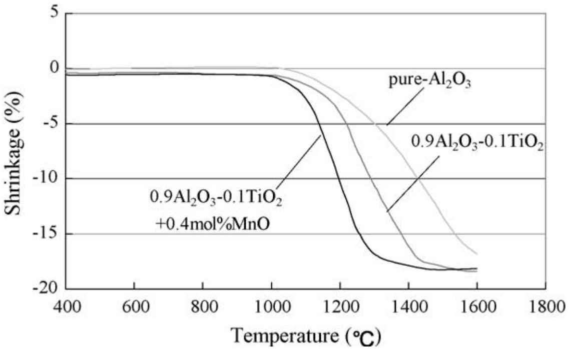

Fig. 3. Shrinkage at sintering for MnO added Al_2O_3—TiO_2 ceramics.

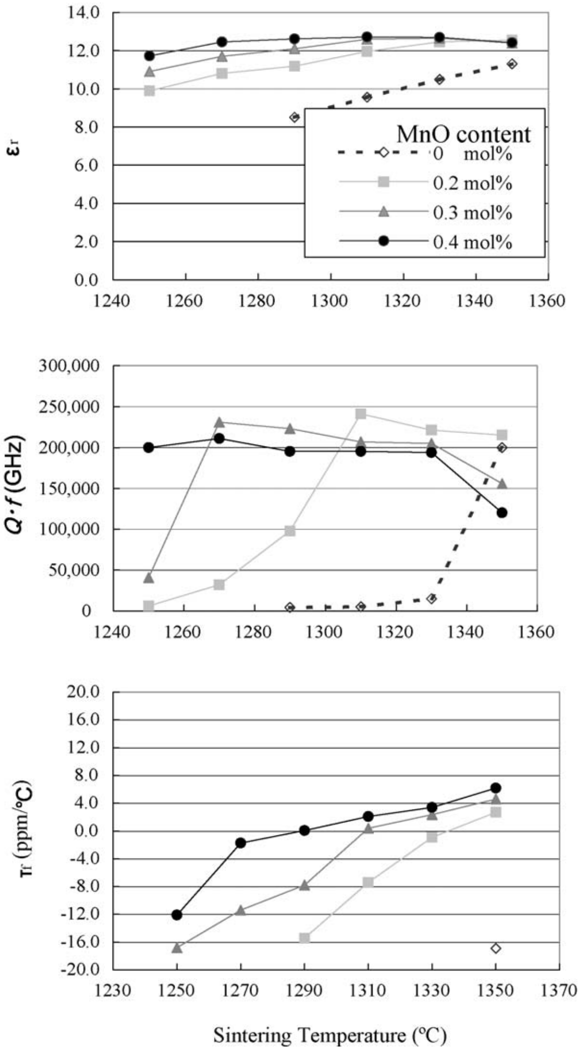

Fig. 4. Sintering temperature dependence of dielectric properties for MnO added $0.9Al_2O_3$—$0.1TiO_2$ ceramics.

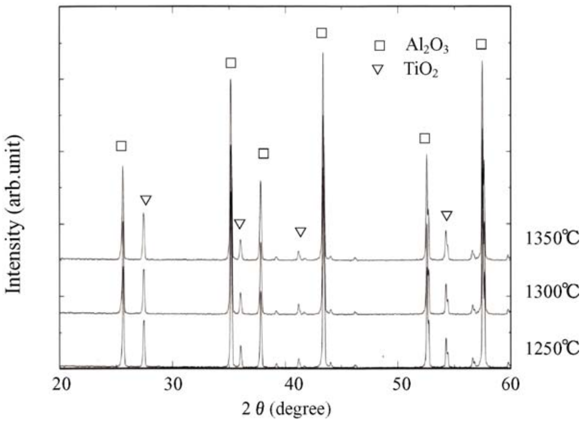

Fig. 5. XRD patterns of 0.4 mol% of MnO added $0.9Al_2O_3$—$0.1TiO_2$ ceramics and sintered at 1250–1350 °C.

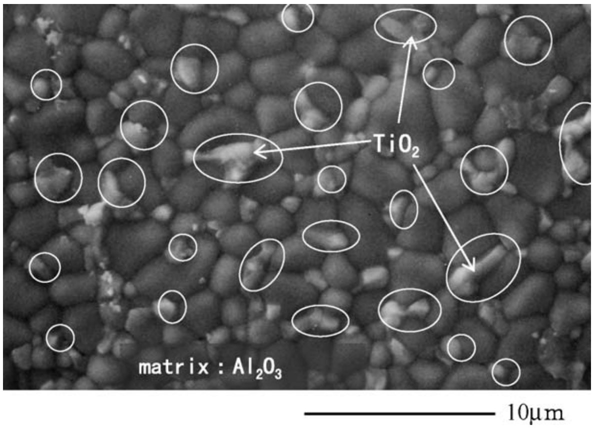

Fig. 6. SEM micrograph of $0.9Al_2O_3$—$0.1TiO_2$ ceramics with 0.4 mol% of MnO sintered at 1300 °C.

The XRD patterns of $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1300–1400 °C for 4 h are shown in Fig. 2. The peak height of Al_2TiO_5 rises with increase of sintering temperature accompanied by a decrease of the peaks of TiO_2. This result indicates that the decrease in $Q \times f$ (shown Fig. 1) is due to the following reaction, which occurred at around 1300 °C.

$$Al_2O_3 + TiO_2 \rightarrow Al_2TiO_5$$

Fig. 3 shows the shrinkage during sintering for $0.9Al_2O_3$–$0.1TiO_2$ ceramics with 0.4 mol% of MnO as measured by thermo-mechanical analysis (TMA). Pure Al_2O_3 ceramics cannot be densified even if it is sintered at 1600 °C. However, addition of TiO_2 lowers the densification temperature at about 1400 °C. MnO makes further reduction of the densification temperature for Al_2O_3–TiO_2 ceramics to about 1300 °C. This improvement is thought to be produced by enhancement of mass transport.

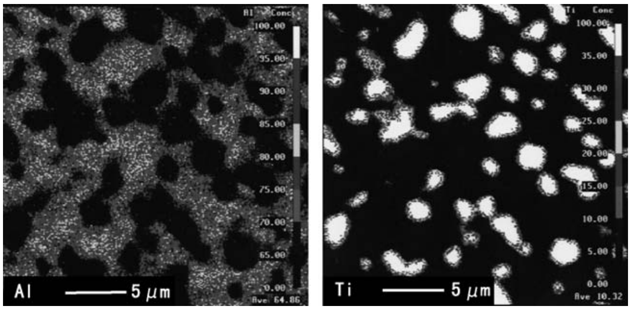

Fig. 7. Al and Ti mapping of $0.9Al_2O_3$—$0.1TiO_2$ ceramics with 0.4 mol% of MnO sintered at 1300 °C.

Table 1
Microwave and millimeterwave dielectric properties of $0.9Al_2O_3$—$0.1TiO_2$ ceramics with 0.4 mol% of MnO

$Q \times f$ (GHz)	f_0 (GHz)	Measurement method
211,000	12.1	Dielectric resonator method
274,000	75.9	Dielectric rod resonator method using NRD guide

The sintering temperature dependence of dielectric properties for MnO added $0.9Al_2O_3$–$0.1TiO_2$ ceramics is shown in Fig. 4. The ceramics with larger amount of MnO shows high $Q \times f$ at lower sintering temperatures. For example, the ceramics with

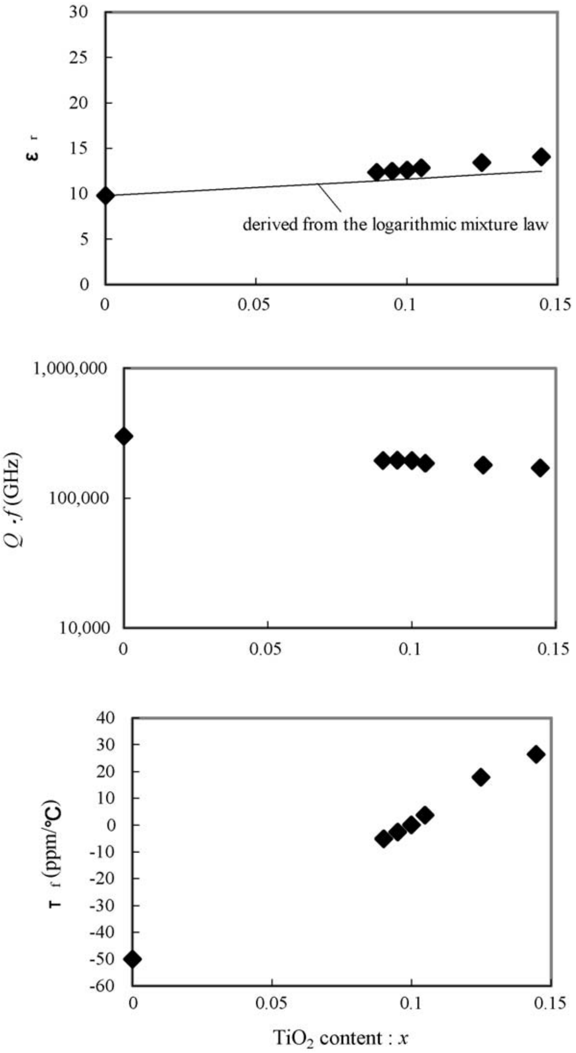

Fig. 8. TiO_2 content dependence of dielectric properties for $(1-x)Al_2O_3$—$xTiO_2$ ceramics with 0.4 mol% of MnO sintered at 1300 °C.

0.4 mol% of MnO showed high $Q \times f$ of 201,000 GHz when sintered at a low temperature of 1250 °C.

XRD patterns of the 0.4 mol% of MnO added $0.9Al_2O_3$–$0.1TiO_2$ ceramics and sintered at different temperatures are shown in Fig. 5. The XRD patterns indicate that these ceramics are composed of Al_2O_3 and TiO_2 phases only. XRD peaks of Al_2TiO_5 phase were not detected even in the ceramics sintered at 1350 °C. Addition of MnO not only lowers the sintering temperature but also suppresses the formation of Al_2TiO_5 phase. The SEM micrograph and WDX element mapping of the 0.4 mol% of MnO added $0.9Al_2O_3$–$0.1TiO_2$ ceramics are shown in Figs. 6 and 7, respectively. The ceramics is composed of light grains and dark grains in SEM micrograph (Fig. 6). WDX mapping reveals that light grains are TiO_2 and dark ones are Al_2O_3. Al_2TiO_5 grains are not observed. These microstructure analyses indicate that the obtained ceramics are composed of well dispersed Al_2O_3 and TiO_2 grains without Al_2TiO_5 grains.

Dielectric properties of the 0.4 mol% of MnO added $(1-x)Al_2O_3$–$xTiO_2$ ceramics as a function of TiO_2 content are shown in Fig. 8. The ε_r, Q, and τ_f vary linearly with TiO_2 content.

It is well known that the ε_r of complex dielectrics follows the logarithmic mixture law.[5] The ε_r derived from the logarithmic mixture law is also shown in Fig. 8. By comparing between experimental result and calculation, it is confirmed that the ε_r changes in accordance with the logarithmic mixture law.

The ε_r and the $Q \times f$ evaluated at 76 GHz are shown in Table 1. The ceramics have high $Q \times f$ value in the millimeter-wave frequency range. Since the $Q \times f$ at 76 GHz is larger than that at 10 GHz, the accuracy is not sufficient enough to discuss the relationship between $Q \times f$ and resonant frequency. Further study with greater accuracy is needed to find the relationship between $Q \times f$ and resonant frequency.

3. Conclusions

Dense Al_2O_3–TiO_2 mixed ceramics were synthesized by lowering the sintering temperature with addition of MnO. This ceramics is suitable for millimeter-wave applications due to its low ε_r of 12.4, high $Q \times f$ of 274,000 GHz at 76 GHz, and small τ_f of +0.4 ppm/°C.

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