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The sintering and microwave dielectric characteristics of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics

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Abstract

$MgTa_{1.5}Nb_{0.5}O_6$ ceramics were combined from two microwave dielectrics with high $Q \times f$ values and high τ_f values, $MgTa_2O_6$ (sintered at 1500 °C, $\varepsilon_r = 30.5$, $Q \times f = 56{,}900$ GHz, and $\tau_f = 28.3$ ppm/°C) and $MgNb_2O_6$ (sintered at 1300 °C, $\varepsilon_r = 21.7$, $Q \times f = 89{,}900$ GHz, and $\tau_f = -68.5$ ppm/°C) $MgNb_2O_6$, in order to obtain microwave dielectric resonators with τ_f value close to 0 ppm/°C. The sintering and microwave dielectric characteristics of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics were investigated in this study. As the sintering temperature increased from 1300 °C to 1450 °C, the density values, the ε_r values, and the $Q \times f$ values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics increased and saturated at 1450 °C, and τ_f values were shifted to close 0 ppm/°C. The 1450 °C-sintered $MgTa_{1.5}Nb_{0.5}O_6$ ceramics had the microwave dielectric characteristics of $\varepsilon_r = 27.9$, $Q \times f = 33{,}100$ GHz, and $\tau_f = -0.7$ ppm/°C.

Keywords: $MgTa_{1.5}Nb_{0.5}O_6$; Microwave dielectric characteristic

1. Introduction

In general, a dielectric material with a high dielectric constant has a large τ_f value.[1,2] To adjust τ_f value of microwave dielectric resonators close to zero, two or more compounds having negative and positive τ_f values are employed to form a solid solution or mixed phases. Kucheiko reported that zero τ_f value was achieved at $CaTiO_3$–$Ca(Al_{1/2}Ta_{1/2})O_3$ system.[1] Chen et al. reported that small τ_f value was achieved at CaO–Li_2O–Sm_2O_3–TiO_2 (CLST) system,[3] in which the $Li_{1/2}Sm_{1/2}TiO_3$ ($\varepsilon_r = 52$, $Q \times f = 2280$ GHz and $\tau_f = -260$ ppm/°C) and $CaTiO_3$ ($\varepsilon_r = 70$, $Q \times f = 3600$ GHz and $\tau_f = 800$ ppm/°C) were combined.[2]

AB_2O_6 (A = Ca, Mn, Zn, Mg and B = Ta, Nb) compounds have been investigated as microwave dielectric resonator by Kan et al.[4] and Lee et al.[5] The microwave dielectric properties of $MgTa_2O_6$ (sintered at 1400–1550 °C) and $MgNb_2O_6$ ceramics (sintered at 1200–1350 °C) are shown in Table 1. The $Q \times f$ and ε_r values of $MgTa_2O_6$ and $MgNb_2O_6$ ceramics increase with the increase of sintering temperature and saturate at 1500 °C and 1300 °C, respectively, and the τ_f values will also reach a saturation value of 28.5 ppm/°C and −68.5 ppm/°C, respectively. From Table 1, $MgNb_2O_6$ ceramics has high $Q \times f$ value and lower sintering temperature, but it also has large negative τ_f value; $MgTa_2O_6$ ceramics has lower $Q \times f$ value and higher sintering temperature, but it also has large positive τ_f value. Two reasons urge us to develop the microwave dielectric characteristics of $MgTa_2O_6$–$MgNb_2O_6$ ceramics: the first is to fabricate microwave dielectric resonator with τ_f value close to 0 ppm/°C, the second is used $MgNb_2O_6$ to lower the sintering temperature of $MgTa_2O_6$ ceramics. For that $MgTa_{1.5}Nb_{0.5}O_6$ is used as the base composition, and the sintering and microwave dielectric characteristics of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are developed as a function of sintering temperatures.

2. Experimental procedures

Proportional amounts of reagent-grade starting materials of MgO, Nb_2O_5 and Ta_2O_5 were mixed, according to

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Table 1
Microwave dielectric characteristics of $MgTa_2O_6$ and $MgNb_2O_6$ ceramics

Material	Sintering temperature (°C)	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)
$MgTa_2O_6$	1400	25.2	28500	22.4
$MgTa_2O_6$	1450	28.9	44300	27.1
$MgTa_2O_6$	1500	30.5	56900	28.3
$MgTa_2O_6$	1550	30.6	58200	28.5
$MgNb_2O_6$	1300	15.7	34100	−78
$MgNb_2O_6$	1350	20.5	66500	−69.1
$MgNb_2O_6$	1400	21.7	89900	−68.5
$MgNb_2O_6$	1450	21.8	91500	−68.3

the composition $MgTa_2O_6$, $MgNb_2O_6$, and $MgTa_{1.5}Nb_{0.5}O_6$, and ball-milled for 5 h with deionized water. After drying, the powder was ground and calcined at 1000 °C for 2 h. After grinding and drying, the mixed powder was uniaxially pressed into pellets in a steel die. Sintering of these pellets was carried out at temperatures between 1200 °C and 1550 °C under ambient conditions for 4 h.

The crystal phases were analyzed by means of an X-ray powder diffraction method using Cu Kα radiation (Rigaku D-max/IIB). The densities of the sintered specimens, as a function of sintering temperature, were measured by the liquid replacement method using deionized water as the liquid (Archimedes method). To investigate the morphology of the samples, the sintered surfaces of the specimens were observed, using SEM (Hitachi S-2500). Dielectric characteristics at microwave frequency were measured by Hakki and Coleman's dielectric resonator method,[6] which was improved by Courtney.[7] An HP8720ET network analyzer was used for the microwave characteristic measurements. The dielectric constant can be accurately determined by measuring the resonant frequency of the TE_{011} mode and verified by the $TE_{01\delta}$ resonant mode. For convenience, the $Q \times f$-factor was used for evaluating the loss quality, where f is the resonant frequency and Q is the quality factor. The temperature change of the resonant frequency $\Delta f_0/f_0$ and temperature coefficient of resonant frequency τ_f are defined as:

$$\frac{f_0}{f_0} = \frac{f_T - f_0}{f_0} \tag{1}$$

and

$$\tau_f = \frac{f_0}{f_0 \times T}, \tag{2}$$

where f_T and f_0 are the resonant frequency at T °C (20–80 °C) and 20 °C, respectively.

3. Results and discussion

In order to evaluate the effects of the various sintering temperatures on the morphological changes of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics, the surface micrographs of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics sintered at 1300–1400 °C are analyzed by SEM, and the results are shown in Fig. 1. As sintered at 1300 °C, the $MgTa_{1.5}Nb_{0.5}O_6$ ceramics show a porous structure and the isolated grains are easily observed (Fig. 1a). As sintered at 1350 °C, the pores decrease, and the $MgTa_{1.5}Nb_{0.5}O_6$ ceramic reveals a dense structure with almost no pores (Fig. 1b). As sintered at 1400 °C, the $MgTa_{1.5}Nb_{0.5}O_6$ ceramics illustrate homogeneously grain growth (Fig. 1c), and the grain size increases with the increase of sintering temperature. From the micrographs, the grain growth of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics is promoted by the increase of sintering temperature.

Fig. 2 shows the typical X-ray diffraction patterns of $MgTa_{1.5}Nb_{0.5}O_6$, sintered at 1350–1450 °C. $MgTa_2O_6$ is a single phase which belongs to tetragonal structure and has $a = b = 4.7189$ Å and $c = 9.2003$ Å.[8] $MgNb_2O_6$ is a single phase which belongs to orthorhombic structure and has $a = 5.7001$ Å, $b = 14.1875$ Å, and $c = 5.0331$ Å.[8] In this study, the crystal structure of $MgTa_2O_6$ ceramic (sintered at 1500 °C) has $a = b = 4.7173$ Å and $c = 9.2094$ Å, the crystal structure of $MgNb_2O_6$ ceramic (sintered at 1400 °C) has $a = 5.720$ Å, $b = 14.1780$ Å and $c = 5.036$ Å. The $MgTa_{1.5}Nb_{0.5}O_6$ has the crystal structure of orthorhombic,

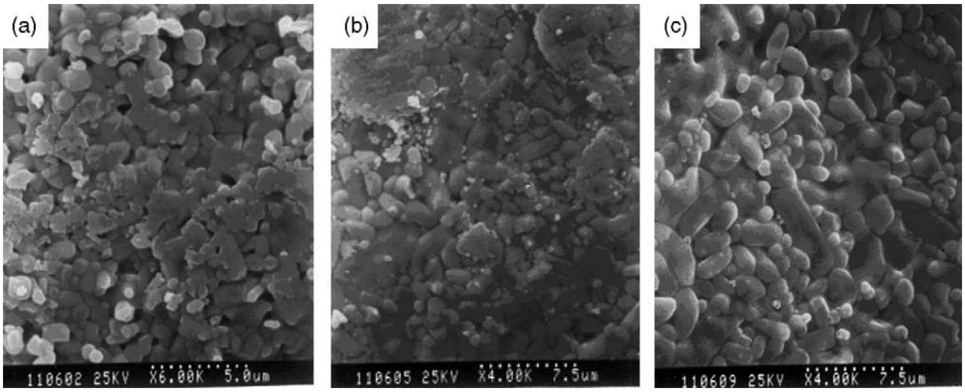

Fig. 1. The microstructures of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics sintered at different temperatures: (a) 1300 °C, (b) 1350 °C, and (c) 1400 °C.

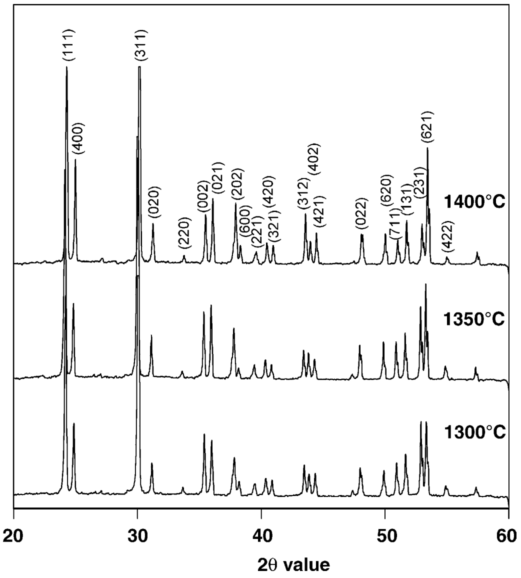

Fig. 2. The X-ray patterns of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics sintered at different temperatures.

which is similar to that of $MgNb_2O_6$ ceramics and exists the lattice constants of $a = 5.484$ Å, $b = 13.724$ Å, and $c = 4.998$ Å. These lattice constants of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are smaller than those of $MgNb_2O_6$, because the Ta^{5+} (0.64 Å) ionic radius is smaller than that of Nb^{5+} (0.69 Å). For $MgTa_{1.5}Nb_{0.5}O_6$ ceramics, even 1450 °C is used as sintering temperature, no impurity phases and raw material phases exist in the ceramic and no decomposition of $MgTa_{1.5}Nb_{0.5}O_6$ can be detected in the XRD patterns.

The density of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics is investigated as a function of sintering temperatures of 1300–1450 °C, and the results are shown in Fig. 3. The theoretical density (TD) of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics calculated from the XRD patterns is 6.071 g/cm^3. As the sintering temperature increases from 1300 °C to 1350 °C, the density of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics critically increases from 84.4% to 93.2%. According to the results in Fig. 1, the decrease of pores will account for this phenomenon. As the sintering temperature is higher than 1350 °C, the density values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are almost saturated, which exhibits a value as high as 96.8% and 96.9% theoretical density for samples sintered at 1400 °C and 1450 °C, respectively. The dielectric constants (ε_r) of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are investigated as a function of sintering temperatures, and the results are also shown in Fig. 3. The ε_r values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramic increase with the increase of sintering temperatures and saturate at about 1400 °C. The relationship between ε_r values and sintering temperatures reveal the same tendency with that between density values and sintering temperatures, because higher sintering temperatures will cause the grain growth and fewer pores, and that will result in a higher ε_r value.

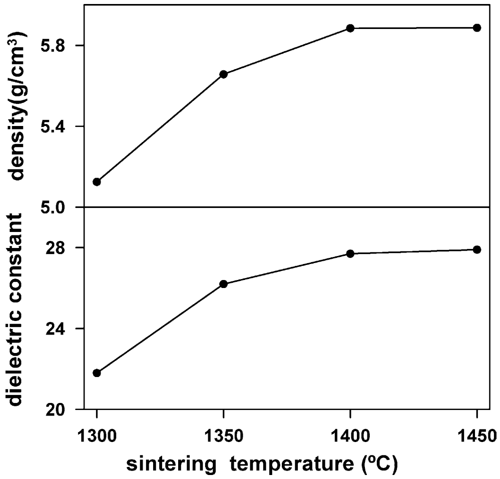

Fig. 3. The density and the dielectric constants of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics sintered at different temperatures.

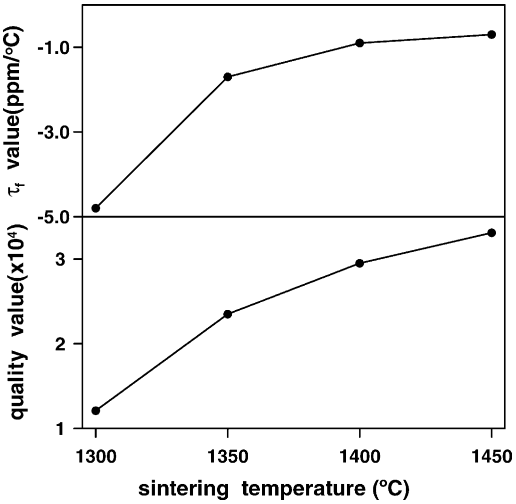

Fig. 4. The quality values ($Q \times f$) and the temperature coefficients of resonant frequency (τ_f) of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics sintered at different temperatures.

Fig. 4 shows the $Q \times f$ values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics as a function of sintering temperatures. As the sintering temperature increases, the $Q \times f$ values increase and reach a maximum value of 33,100 GHz at 1450 °C. The $Q \times f$ values are known to be affected by the morphologies of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics, such as porosity, grain sizes, and the uniformity of grain growth.[4] Even the 1350 °C-sintered ceramics reveal a dense surface, but the grain growth is not uniform and it will lead to a lower $Q \times f$ value. The τ_f values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are also revealed in Fig. 4 as a function of sintering temperatures. As the sintering temperatures increase from 1300 °C to 1400 °C, the τ_f values change steadily from −4.8 ppm/°C to −0.8 ppm/°C. And the τ_f values reach a saturated value of −0.7 ppm/°C at 1450 °C and that is used as the optimized sintering temperature. Although the maximum $Q \times f$ values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are somewhat lower than that of $MgNbO_6$ ceramics, however, the τ_f value is largely improved in this study. It reveals the potential applications of $MgTa_{1.5}Nb_{0.5}O_6$ as microwave devices.

4. Conclusion

For the saturated microwave dielectric characteristics of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics, the ε_r values are smaller than those of $MgTa_2O_6$ ceramics but larger than those of $MgNb_2O_6$ ceramics. The $Q \times f$ values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are smaller than those of $MgTa_2O_6$ and $MgNb_2O_6$ ceramics. However, the τ_f values of $MgTa_{1.5}Nb_{0.5}O_6$ ceramics are more close to 0 ppm/°C than those of $MgTa_2O_6$ and $MgNb_2O_6$ ceramics are. In this study, the $MgTa_{1.5}Nb_{0.5}O_6$ ceramics sintered at 1450 °C will reveal the optimum microwave dielectric characteristics of $\varepsilon_r = 27.9$, $Q \times f = 33{,}100$ GHZ, and $\tau_f = -0.7$ ppm/°C.

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