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Preparation and microwave properties of $ZnTiNb_2O_8/SmTiNbO_6$ composite dielectric ceramics

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Abstract. In order to get a microwave dielectric ceramics having near zero τ_f, different ratio of $ZnTiNb_2O_8$ and $SmTiNbO_6$ powders are sintered together at different temperature. XRD and SEM of these composite ceramics are tested to analyze the composition. The microwave properties and density of the composite ceramics are measured in order to find the best ratio of $ZnTiNb_2O_8$ and $SmTiNbO_6$. For (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ ceramics, the ceramics have very good microwave properties(ε_r = 24.63, Q*f=11846GHz, τ_f = 0.16ppm/℃) when x is 0.35.

1 Introduction

Limitless use of wireless data transfer becomes a prime motivator in developing advanced communication device [1, 2]. This leads to integration of antenna in miniature devices such as in a mobile phone [3]. Despite environmental friendly, portable, and lightweight, a low loss and thermally stable antenna is an important feature for this innovation [4]. Meanwhile, in antenna itself, only selected materials with specific parameters can be fit in. High-permittivity ceramics make it possible to reduce the scale of passive microwave devices. These ceramics must fulfill the requirements of high permittivity(ε_r), high Quality factor (Q*f) as inversely very low dielectric losses(tan δ) and an extremely low temperature coefficient of resonant frequency (τ_f).

2 Experimental procedure

Dried reagent-grade ZnO (99.99% purity), TiO_2 (99.9% purity), Nb_2O_5 (99.9% purity) Sm_2O_3 (99.99% purity) were weighed out, according to the composition of $ZnTiNb_2O_8$ and $SmTiNbO_6$. The powders were ball-milled for 16h in isopropan-2-ol, followed by calcinations at a certain temperature for 4h respectively. Pellets, 10mm in diameter and about 5mm thick, were prepared by uniaxial pressing of powders at an applied load of ～

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0.7 tons, and subsequently sintered for 4h at different temperatures. The bulk densities of the sintered pellets were measured by the Archimedes method.

X-ray diffraction (XRD) was performed at a step size of 0.02° with a scan rate of 1°/min using a Phillips X-ray diffractometer equipped with a CuKα source (λ = 1.5405Å) operating at 40kV and 30mA. Scanning electron microscopy (SEM) was performed on the as-polished surfaces using a JEOL JSM-6400.

Microwave measurements were carried out using the transmission resonant cavity technique in an Au-coated brass cavity and by using an ADVANTEST R3767CH vector network analyzer. A Peltier device was used to heat up the cavity in order to measure the resonant frequency, f_0, between 25℃ and 80℃. A linear fit to the data was used to calculate τ_f.

3 Results and discussion

3.1 Phase composition analysis of $ZnTiNb_2O_8$/$SmTiNbO_6$ composite dielectric ceramics

When $ZnTiNb_2O_8$ powder and $SmTiNbO_6$ powder are mixed and sintered, as the amount of $SmTiNbO_6$ increases, $ZnTiNb_2O_8$ will react with $SmTiNbO_6$ by Eq.1 , and the products are respectively $(Zn_{0.15}Nb_{0.3}Ti_{0.55})O_2$ and $SmNbO_4$. When $SmTiNbO_6$ is excessive, there will be surplus $SmTiNbO_6$. Because the temperature coefficient of the resonance frequency of $(Zn_{0.15}Nb_{0.3}Ti0_{.55})O_2$ is very large in the positive direction, we should add as little $SmTiNbO_6$ powder as possible or reduce the sintering temperature to make the reaction incomplete. Only in this way can the resonance frequency temperature of $ZnTiNb_2O_8$/$SmTiNbO_6$ microwave composite ceramic material be tending to zero.

$$0.4SmTiNbO_6+0.15\ ZnTiNb_2O_8 \longrightarrow (Zn_{0.15}Nb_{0.3}Ti_{0.55})\ O_2+ 0.4SmNbO_4 \quad (1)$$

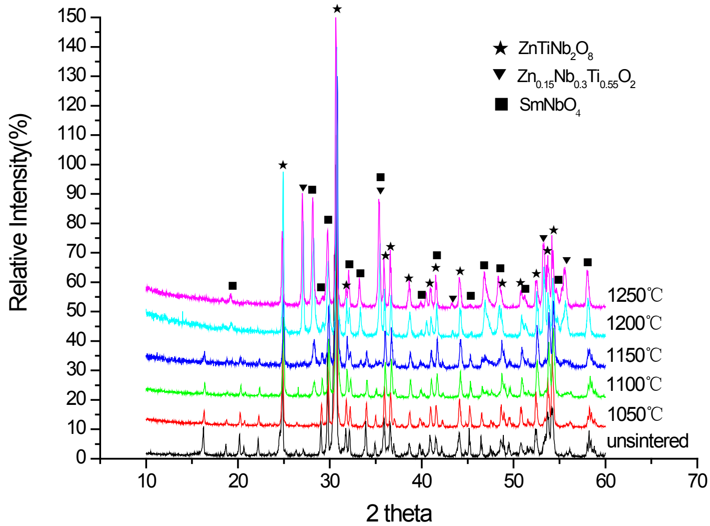

Fig. 1. XRD patterns of $0.65ZnTiNb_2O_8$-$0.35SmTiNbO_6$ composite ceramics sintered at different temperature.

In order to obtain a microwave dielectric composite ceramic with a zero temperature coefficient of resonance frequency, we choose an excess of $ZnTiNb_2O_8$ and $SmTiNbO_6$ powder to composite, and reduce the sintering temperature as much as possible to reduce the reaction between the two matrix materials. Taking x=0.35 as an example, after mixing the two base materials and sintering at different temperatures, the XRD pattern is shown in Figure 1. As the temperature rises, the XRD diffraction peaks continue to change. It can be seen that when the sintering temperature is 1100°C, the two base materials begin to react until 1250°C, and the reaction is complete. The analysis shows that there are three components in the ceramic: $(Zn_{0.15}Nb_{0.3}Ti_{0.55})_{O2}$, $SmNbO_4$ and $ZnTiNb_2O_8$. In order to ensure the compactness of the ceramics and minimize the reaction between the two matrix materials, and to facilitate the comparison of the microwave performance of microwave dielectric ceramics with different $SmTiNbO_6$ contents, we choose the sintering temperature of all the components of the composite ceramics to be 1150°C.

3.2 Microwave properties and microstructure of (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ composite dielectric ceramics

The (1-x) $ZnTiNb_2O_8$-x $SmTiNbO_6$ composite ceramic with the same content of $SmTiNbO_6$ is sintered at 1150℃, and the permittivity of the ceramic obtained is shown in Figure 2. When x is relatively small, $(Zn_{0.15}Nb_{0.3}Ti_{0.55})O_2$ and $SmNbO_4$ are formed which have low permittivity(ε_r), so the permittivity of the composite ceramic shows a downward trend[5,6]. When x=0.35, the permittivity reaches to the minimum value of 24.63. However, as x continues to increase, there are more and more remaining $SmTiNbO_6$ in the composite ceramic, and permittivity of $SmTiNbO_6$ is 49.55, so the permittivity of (1-x) $ZnTiNb_2O_8$-x $SmTiNbO_6$ composite ceramics will change again. As x increases, it increases.

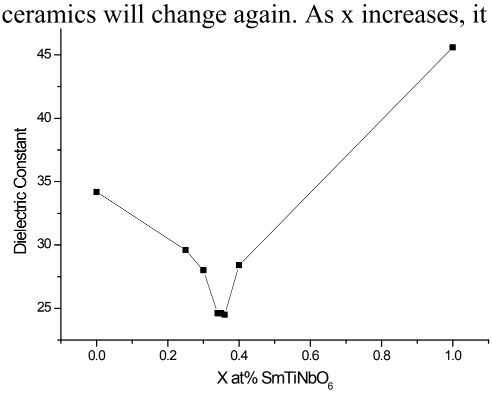

Fig. 2. The permittivity of (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ composite ceramics sintered at 1150℃.

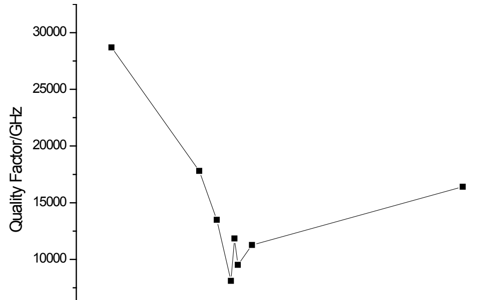

Fig. 3. The quality factor of (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ composite ceramcis sintered at 1150℃.

It can be seen from Figure 3 that with the increase of x, the quality factor of (1-x) $ZnTiNb_2O_8$-x $SmTiNbO_6$ composite ceramics firstly decreases and then increases. Due to the continuous increase in the amount of multiphase production and the relatively low density of the composite ceramics, the quality factor of the composite ceramics decreases with the increase of x. When x = 0.35, the Q**f* value reaches a minimum of 11846, and When x>0.35, the value of Q**f* increases with the increase of x. This is because at this time, the two matrix materials no longer react. As the content of $SmTiNbO_6$ continues to increase, the density of the ceramic also increases. The continuous increase is shown in Figure 4, and $SmTiNbO_6$ ceramics have a higher quality factor. Therefore, the quality factor Q**f* of (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ composite ceramics increases with the increase of x.

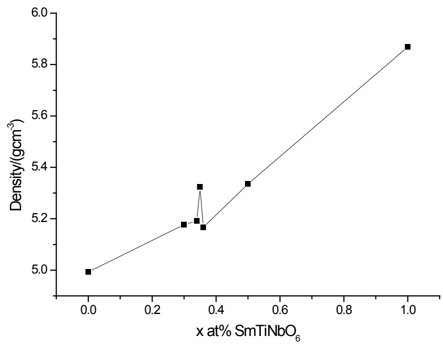

Fig. 4. The density of (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ composite ceramics sintered at 1150℃.

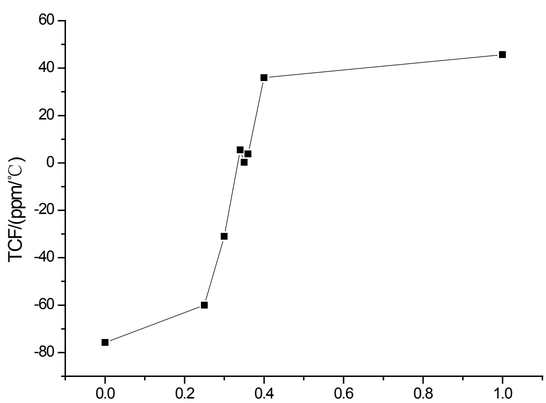

Fig. 5. The temperature coefficient of resonant frequency of (1-x)$ZnTiNb_2O_8$-x$SmTiNbO_6$ composite ceramics sintered at 1150℃.

The relationship between the temperature coefficient of the resonance frequency of the (1-x) $ZnTiNb_2O_8$-x $SmTiNbO_6$ composite ceramic and x is shown in Figure 5. The temperature coefficient of the resonance frequency of $(Zn_{0.15}Nb_{0.3}Ti_{0.55})O_2$ and $SmTiNbO_6$ both are positive. According to the changes in the composition of the composite ceramic and the Lichnetecker mixing rule, we can know that any one of the two components increasing will cause the temperature coefficient of the resonant frequency of the composite ceramic to change in the positive direction [7,8,9]. When x = 0.35, the temperature coefficient of the resonant frequency of the composite ceramic is 0.1594ppm/℃.

3.3 The density and the microstructure of the composite ceramic

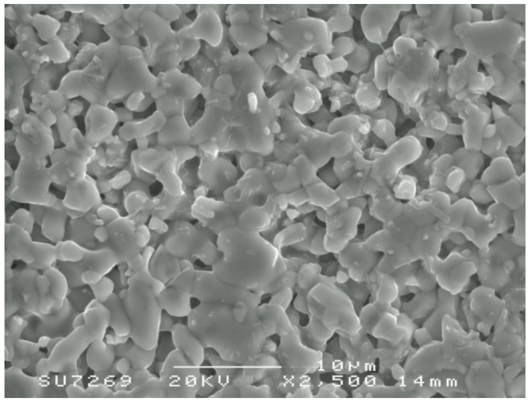

Fig. 6. SEM image of 0.65 $ZnTiNb_2O_8$-0.35$SmTiNbO_6$ composite ceramics sintered at 1150℃.

Since the density of $SmTiNbO_6$ ceramics (5.7929g/cm^3) is greater than the density of $ZnTiNb_2O_8$ ceramics (5.0935g/cm^3) [9,10], as x increases, the density of the composite ceramic increases, as shown in Figure 4. But this doesn't mean that the porosity of ceramics has decreased, the reason is the increase in the content of $SmTiNbO_6$ ceramics which has higher density. In order to reduce the reaction of the two matrix materials, the sintering temperature is too low, which makes the porosity of the multiphase ceramics too high. As shown in Figure 6, the composite ceramics basically do not form porcelain, and the ceramic particles are only closely packed together which seriously affects the microwave dielectric properties of multiphase ceramics.

4 Conclusions

By studying the preparation and microwave properties of (1-x) $ZnTiNb_2O_8$-x $SmTiNbO_6$ composite ceramics, when x = 0.35 and the sintering temperature is 1150℃, the permittivity ε_r is 24.63, the quality factor $Q*f$ is 11846GHz and the temperature coefficient of resonance frequency τ_f is 0.16ppm/℃. It is a microwave dielectric ceramic material with excellent performance. However, the density of ceramics is not large enough and the sintering temperature is too high to be co-fired with Ag (961°C) and Cu (1042°C) electrodes [4,11]. Therefore, in the future it is necessary to add a sintering aid to reduce the sintering temperature of the ceramic and increase its density. And then improve the microwave dielectric properties of the material, especially the permittivity and quality factor.

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