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Preparation and microwave dielectric properties of new low-loss $NiZrTa_2O_8$ ceramics

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ABSTRACT

New low-loss $NiZrTa_2O_8$ ceramics were prepared by conventional mixed-oxide method with the crystal structure and microwave dielectric properties being investigated. The phase composition of $NiZrTa_2O_8$ ceramics presented two-phase coexistence, indexed as $NiTiNb_2O_8$ and $Ni_{0.5}Ti_{0.5}NbO_4$. The apparent morphology and grain size were analyzed as a function of sintering temperature. The dielectric constant (ε_r) which changed from 19.02 to 21.38, was mainly dependent on the densities and phase composition. The quality factor ($Q \times f$) was dominated by grain size. In this paper, the optimum performance of $NiZrTa_2O_8$ ceramics were $\varepsilon_r = 20.61$, $Q \times f = 49200$ GHz, sintered at 1325 °C.

1. Introduction

With the fast developing of microwave communication technology, the demands for microwave dielectric ceramics with different properties have continued to increase, prompting large numbers of researchers to devote themselves to investigate and invent more kinds of new microwave dielectric ceramics.

$ZnTiNb_2O_8$ ceramics on microwave application had been firstly reported by D. W. Kim et al. [1]. Since then, the $ZnTiNb_2O_8$ ceramics had attracted increasing interests for their excellent microwave dielectric properties [2,3]. According to previous investigations, it could be known that microwave dielectric properties could be optimized by ionic replacement [4]. In 2012, modification research of $ZnTiNb_2O_8$ ceramics had been carried out by substituting Ti ion as Zr ion by Liao et al. [5]. Hereafter, many compounds, which were obtained by ionic replacement and had the similar formula with $ZnTiNb_2O_8$ system, had been reported on their microwave dielectric properties [6–9]. Ni ion was a good substitution in the type of bivalent ions, especially in minor-substitution, since it could sometimes enormously meliorate microwave dielectric properties of the original system. Therefore, a new compound with composition of $NiZrTa_2O_8$ was designed based on $ZnTiNb_2O_8$ formula with Ni substituting for Zn, Zr substituting for Ti and Ta substituting for Nb.

In this work, the $NiZrTa_2O_8$ ceramics had been successfully prepared for the first time using conventional solid state reaction method, with sintering characteristics, crystal structure and microwave dielectric properties being investigated.

2. Experimental procedure

The $NiZrTa_2O_8$ ceramics were prepared with high purity oxide powders of NiO (99%), ZrO_2 (99%), and Ta_2O_5 (99.9%). These powders were weighed according to the $NiZrTa_2O_8$ formula and the mixtures were milled under distilled water with zirconia ball for 24 h. After the slurries being dried and sieved, the mixtures were calcined in alumina crucible at 1200 °C for 4 h. Then the calcined powders were milled once again under distilled water with zirconia ball for 24 h. Next, the slurries were dried, sieved and pressed into pellets with 10 mm in diameter and 5 mm in thickness. Finally, the pellets were sintered in air at the temperature range of 1275°C–1375 °C.

The crystal structures of these sintered samples were confirmed by X-ray diffraction (XRD) analysis using Rigaku diffractometer (Model D/Max-B, Rigaku Co., Japan) with Cu Kα radiation. Apparent morphology of the sintered samples was observed using a scanning electron microscopy (FE-SEM, SIGMA Zeiss, Germany). The densities of the samples were measured by Archimedes method. The measurement of microwave dielectric properties was carried out by Hakki-Coleman's resonator method using a network analyzer (E5063A, Keysight Co., USA) [10,11].

3. Result and discussion

Fig. 1 showed the XRD patterns of $NiZrTa_2O_8$ ceramics under different sintering temperature. The peak position and peak shape of the XRD patterns were similar under different sintering temperature, suggesting that the phase composition and crystal structure of $NiZrTa_2O_8$ ceramics had no obvious change among the whole sintering range. According to the XRD data, the phase composition of $NiZrTa_2O_8$ ceramics, which presented two-phase coexistence, was indexed as $Ni_{0.5}Ti_{0.5}NbO_4$ structure (ICDD-PDF #52-1875: Tetragonal, *P*4$_2$/*mnm*) and $NiTiNb_2O_8$ structure (ICDD-PDF #52-1874: Orthorhombic, *Pbcn*). For detail, the ratio of relative intensities between $Ni_{0.5}Ti_{0.5}NbO_4$ and $NiTiNb_2O_8$ was calculated and listed in Fig. 1. It was observed that the relative intensities between the two phase compositions presented a slight change with the sintering temperature ranged from 1275 °C to 1325 °C and then tended to gentle, suggesting that the phase compositions of $NiZrTa_2O_8$

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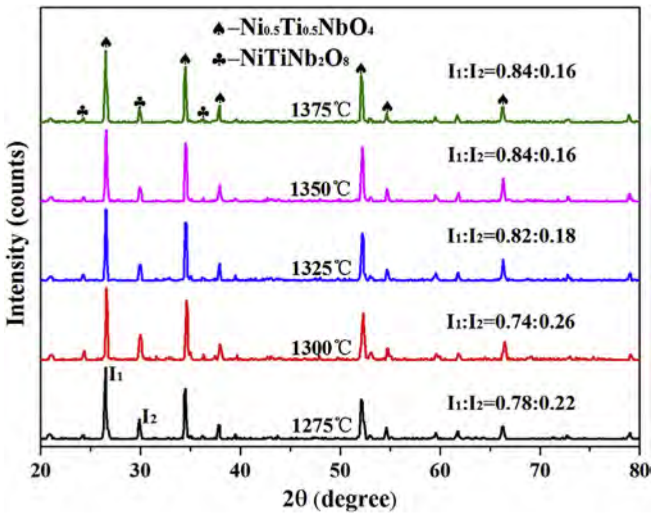

Fig. 1. The XRD patterns of $NiZrTa_2O_8$ ceramics under different sintering temperature.

ceramics were stable under different sintering temperature. As a results, the phase composition and crystal structure of $NiZrTa_2O_8$ had no obvious change under different sintering temperature, which were related to intrinsic microwave dielectric properties.

Generally, the extrinsic microwave dielectric properties were analyzed by apparent morphology. Fig. 2 showed the SEM images of $NiZrTa_2O_8$ ceramics at different sintering temperature. All the samples were well sintered with flat boundary and no pores, suggesting that the dense samples could be obtained under the sintering temperature in this paper. In addition, the grain size (GS) of each sample was measured and listed in Fig. 2 [1]. It was obvious that the grain size of each sample was different and tended to increase as the sintering temperature increased. Although the grain size had an increasing tendency as a function of the sintering temperature, it was not always a good situation for microwave dielectric properties. As shown in Fig. 2, it was important to note that the abnormal grain growth had been observed, which would deteriorate the performance of the ceramics.

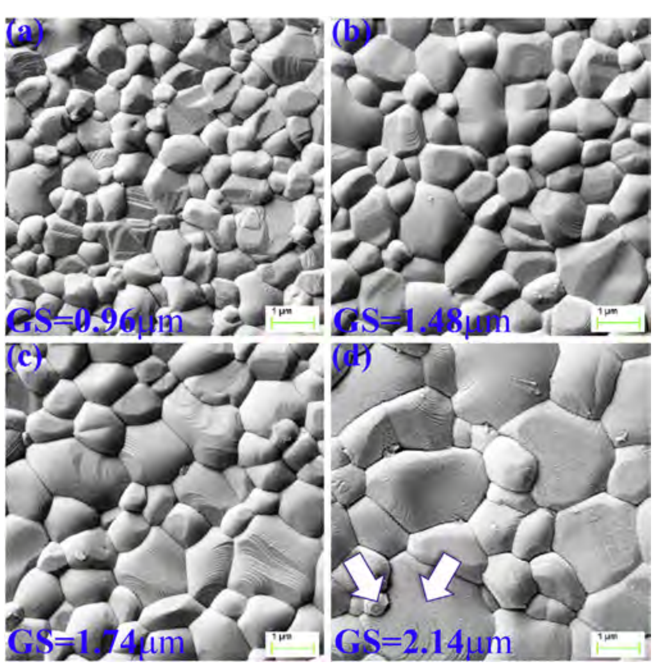

Fig. 2. The apparent morphology of $NiZrTa_2O_8$ ceramics sintered at (a) 1275 °C, (b) 1300 °C, (c) 1325 °C, (d) 1375 °C.

The dielectric constant, which was associated with volume of microwave device, had been measured and illustrated in Fig. 3. It was found that the dielectric constant of $NiZrTa_2O_8$ ceramics increased to maximum ($\varepsilon_r = 21.38$) at 1300 °C and then decreased with the increase of sintering temperature. It was known that the dielectric constant was dependent not only on intrinsic factors such as molecular polarizability but also on extrinsic factors including densities, phase composition, impurities and so on. Based on the same raw materials and according to the XRD analysis, the molecule, phase composition and impurities of $NiZrTa_2O_8$ ceramics sintered at different temperature were similar, suggesting that these influencing factors could be taken no account of in this paper. Therefore, the densities of samples were considered as the main factor influencing the dielectric constant. As shown in Fig. 3, compared the curves of dielectric constant and densities, the variation between the two curves was consistent, which confirmed our above analysis. In addition, it was important to note that the fluctuation of dielectric constant (Δ) within the sintering temperature zone was very small ($\Delta = 2.36$), indicating that there was no significant change in dielectric constant under different sintering temperature.

The $Q \times f$ values, representing the quality factors of dielectric resonator, were tested and drawn under different sintering temperature, as shown in Fig. 4. The $Q \times f$ values of $NiZrTa_2O_8$ ceramics increased to a maximum value of 49200GHz when sintered at 1325 °C and then decreased with the further increasing sintering temperature. It was well known that the influence for $Q \times f$ values at microwave frequency included two parts: (1) intrinsic factors like lattice vibrational modes, (2) extrinsic factors such as phase compositions, grain size and impurities. In this paper, considering the stable phase composition, consistent crystal structure and the same raw materials, the $Q \times f$ values were mainly dependent on grain size. It was noted that the variation of $Q \times f$ values was consistent with that of grain size with sintering temperature ranged from 1275 °C to 1325 °C. But inverse tendency between them had been observed when the sintering temperature was further increased, resulting in abnormal grain growth.

4. Conclusion

New dielectric material with $NiZrTa_2O_8$ formula was designed

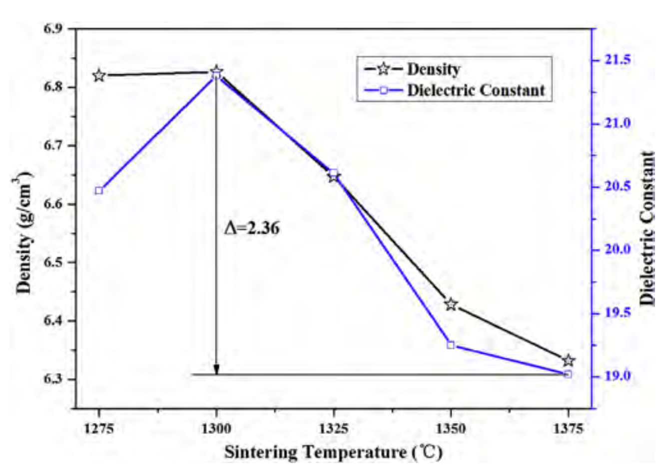

Fig. 3. The dielectric constant and the density of $NiZrTa_2O_8$ ceramics under different sintering temperature.

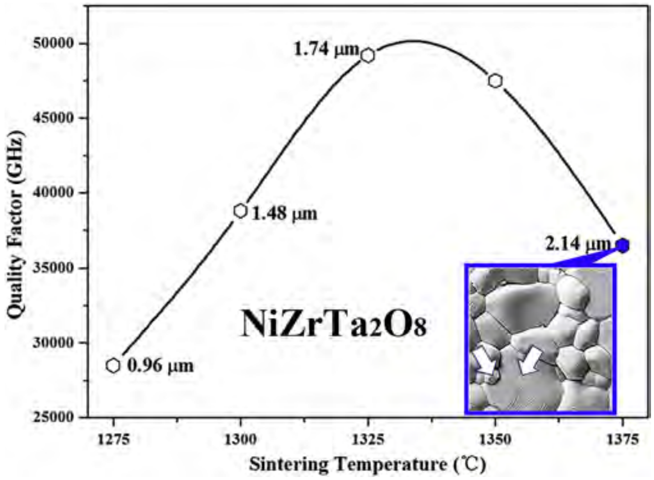

Fig. 4. The Q × ƒ values of $NiZrTa_2O_8$ ceramics under different sintering temperature.

and prepared for the first time. The dense $NiZrTa_2O_8$ ceramics were obtained with the sintering temperature ranged from 1275 °C to 1375 °C. The phase structure of $NiZrTa_2O_8$ ceramics was characterized as coexistence of two phases and there was no obvious change under different sintering temperature. The grain growth was analyzed by apparent morphology. The grain size increased monotonically as a function of the sintering temperature, while abnormal grain growth had been observed when the sintering temperature was too high. For microwave dielectric properties, the dielectric constant of $NiZrTa_2O_8$ ceramics had no obvious change during the whole sintering range, which was related to densities of the samples. The Q × ƒ values, which were mainly dependent on grain size, increased at first and then decreased as a function of sintering temperature. The typical microwave dielectric properties of $NiZrTa_2O_8$ ceramics were $\varepsilon_r = 20.61$, Q × ƒ = 49200 GHz, sintered at 1325 °C.

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