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To be cited as: *Eur. J. Inorg. Chem.* 10.1002/ejic.201800016

Link to VoR: http://dx.doi.org/10.1002/ejic.201800016

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Relationship of crystal structure and microwave dielectric properties in $Ni_{0.5}Ti_{0.5}NbO_4$ ceramics with Ta substitution

Xin Huang, * [a] Huaiwu Zhang, * [a] Yuanming Lai,[a] Gang Wang,[a] Mingming Li,[a] Caiyun Hong,[a] Jie Li[a]

Abstract: In this work, we prepared $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ microwave dielectric ceramics using traditional solid-state reaction method. The influence of Ta^{5+} substituted at Nb^{5+} site on the crystal structures and dielectric properties of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics was discussed. Structure refinement indicates that oxygen octahedron distortion is affected by Ta substitution. The dielectric constant is dependent on the polarization of NbO_6 octahedral. The Q×f value is improved as Ta content increasing because of the increase of packing fractions. The τ_f value is mostly dominated by distortions of Nb-O bonds (δ_{Nb-O}). And the δ_{Nb-O} reduced with Ta ion doped in Nb site, which makes the τ_f value decreased with increased Ta content. Optimal microwave dielectric properties (ε_r = 48.5, Q×f =17500 GHz and τ_f = 88.6 ppm/°C at the resonant frequency of 6.05GHz) be obtained for $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics at x = 0.2.

Introduction

Recent years, the rapid developments of wireless communication have made enormous demands on materials. Microwave dielectric material is one of those basic materials which are receiving many attentions by researchers. Those materials could be used to fabricate resonators, antennas and filters[1–5]. Low dielectric loss (*Q*×*f* values > 5000) and great dielectric constant (ε_r >10) are benifit to the application of dielectric materials in communication systems. High dielectric constant would promote the device miniaturization. And, the high quality factor helps in maintaining the signal transmission without distortion. Currently, dielectric materials with permittivity of 5~20 are used in high bandwidth antenna substrates, the permittivity of 20~50 for base station resonators and antenna substrates, the permittivity of 60~70 for base station resonators, and the permittivity beyond 120 often used in ultra-small GPS antenna substrates[6]. So that, exploring the new excellent microwave dielectric materials is a hot research area in nowadays.

Generically, most of microwave dielectric materials are oxide ceramics. Their dielectric properties derive from the large ionic polarizabilities of the O^{2-} ion, such as the polarization of NbO_6-octahedra[7], TiO_6-octahedra[8], and MoO_4-tetrahedral[9]. Thus, lots of niobate dielectric ceramics are excellent microwave dielectric materials. There are some representative serials of niobate dielectric ceramics as follows: $A^{2+}Nb_2O_6$[10,11], $A^{2+}TiNb_2O_8$[12], and $A^{2+}ZrNb_2O_8$[13,14] (A^{2+}=Mg, Co, Cu, Ni, and Zn). All of the above ceramics show excellent dielectric properties, especially the $ZnNb_2O_6$[10] (ε_r = 23.2, *Q*×*f* =84500 GHz, τ_f = -75.8 ppm/°C), $ZnTiNb_2O_8$[12] (ε_r = 34.4, *Q*×*f* =56900 GHz, τ_f = -47.94 ppm/°C) and $ZnZrNb_2O_8$[15] ceramics (ε_r = 28.37, *Q*×*f* =63300 GHz, τ_f = -19.9 ppm/°C), τ_f is stand for Temperature stability of resonant frequency. To make niobate dielectric ceramics better application in device fabrication, many efforts have been made by researchers. These efforts can be separated into the following fields[1]. (1) Regulating the τ_f values, a small τ_f value is a benefit to device stability at different temperatures. (2) Improving the *Q*×*f* values, it is a general rule that large dielectric constant shows small *Q*×*f* value, thus, optimize *Q*×*f* values is selectivity to a specific frequency. (3)Reducing the sintering temperatures, low temperature co-fired ceramics (LTCC) technology is an effective way to fabricate device, and those kinds of ceramics need low sintering temperatures below 960 °C.

Lots of reports have indicated that ion substitution is an effective way to regulate dielectric properties of microwave dielectric ceramics[16,17]. For instance, Jinxin Bi et al. [18] reported the Mn substituted for Zn in $ZnZrNb_2O_8$ ceramic have changed its τ_f value from -52 ppm/°C to -5.61 ppm/°C. Yonggui Zhao et al. [19] studied the influence of Ta substitution for Nb on microwave dielectric properties of $Zn_3Nb_2O_8$ ceramic, the *Q*×*f* value be increased from 83300 GHz to 111800 GHz. In previous report[20] that $Ni_{0.5}Ti_{0.5}NbO_4$ ceramics exhibited good dielectric properties (ε_r = 59.95, *Q*×*f* =15094 GHz, τ_f = 111.15 ppm/°C) in the middle dielectric constant ranges (20< ε_r < 60). Thus, in order to increase the *Q*×*f* values of $Ni_{0.5}Ti_{0.5}NbO_4$ ceramics, we prepared $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics through the conventional solid-stated route. Meanwhile, the dielectric properties and crystal structure been studied in this work.

Results and Discussion

The phase analyses of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ powders were conducted through X-ray powder diffraction method. Fig.1 shows

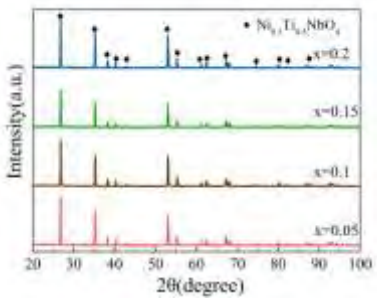

Figure 1. XRD patterns of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at 1140 °C.

the XRD patterns of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics. The reflections from all the samples are identical to a tetragonal rutile-related structure (JCPDS #52-1875) of $Ni_{0.5}Ti_{0.5}NbO_4$ phase. The plane diffraction peaks were indexed to group P4_2/mnm. All the compounds showed a pure phase with Ta^{5+} doping. Rietveld refinements were executed by GSAS software to analyze the crystal structure of the

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$Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics[21]. The Rietveld refined XRD pattern of $Ni_{0.5}Ti_{0.5}Nb_{0.8}Ta_{0.2}O_4$ ceramic is shown in Fig.2. And the crystal structure of $Ni_{0.5}Ti_{0.5}Nb_{0.8}Ta_{0.2}O_4$ ceramic is presented in the insets of Fig. 2, which shows a rutile structure. Meanwhile, the NbO_6 octahedrons can be observed in the insets of Fig. 2. The calculated parameters of crystal structure of all the

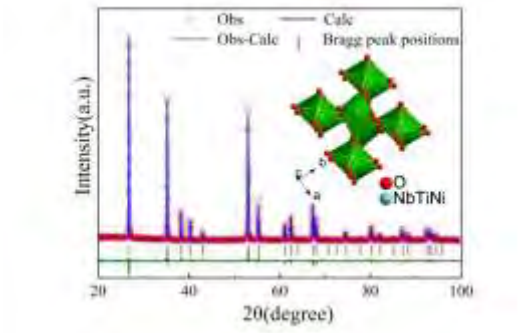

Figure 2. Rietveld refined XRD pattern and crystal structure of $Ni_{0.5}Ti_{0.5}Nb_{0.8}Ta_{0.2}O_4$ ceramics.

samples are listed in Table 1. As it shows that the Rietveld discrepancy factors Rp and Rwp were less than 10% and the x^2 is

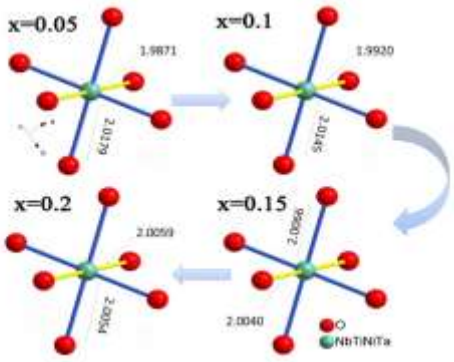

Figure 3. Schematic distortion in NbO_6 for $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics.

less than 2, which indicate the refinements were credible with the investigated parameters. Although the Ta^{5+} ion and Nb^{5+} ion have an identical ionic radius (0.64 Å, CN=6), the volume of unit cell showed a reduction with the increase of Ta content. This trend was in accordance with previous reports[19,22]. We also calculated the bond lengths which listed in Table 2. The result shows that one of Nb-O bond length decreased and the other two increased as Ta content increasing. The Schematic distortions in NbO_6 octahedral for $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics are presented in Fig. 3.

Fig. 4 shows the scanning electron microscopy (SEM) micrographs of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at 1140 °C. As shown in Fig. 4, the fine grains were observed. All the samples showed high densification (Relative density larger than 94%), and the pores were hardly seen. It proved that the $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics have good sintering properties at 1140 °C. With the Ta content increasing, all the grain sizes was decreased. The average grain size of $Ni_{0.5}Ti_{0.5}Nb_{0.95}Ta_{0.05}O_4$(x=0.05) was 14.99 μm and that of $Ni_{0.5}Ti_{0.5}Nb_{0.8}Ta_{0.2}O_4$(x=0.2) was 9.43 μm.

The dielectric constants of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics as a function of sintered temperatures are illustrated in Fig. 5. In general, the dielectric constant is mainly influenced by relative density, the second phase and dielectric polarizabilities[23,24]. As shown in Fig. 4, all the samples possessed high densification, and no second phase was observed from XRD patterns. So that, in this experiment, the dielectric polarizabilities were mainly contribute to the changes of dielectric constant. The structure of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics was a rutile-related tetragonal, all the positive ions occupy the same site to form an oxygen octahedron. So that, the ionic polarization in oxygen octahedron was the key factor to dielectric constant[24]. Correlations between the polarization and

Table 1. Rietveld refinement results for $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics.

Sample	Rwp(%)	Rp(%)	x^2	a(Å)	b(Å)	c(Å)	Volume(Å^3)
X=0.05	8.5	6.67	1.432	4.6977	4.6977	3.0269	66.8
X=0.1	8.76	7.18	1.538	4.6975	4.6975	3.0274	66.805
X=0.15	10.09	8.55	1.232	4.6974	4.6974	3.0274	66.802
X=0.2	8.45	7.05	1.569	4.6978	4.6978	3.0277	66.792

Table 2. Bond length (Å) and distortions of Nb-O bonds for $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics.

Bond length (Å)	Nb-O(1)	Nb-O(2)	Nb-O(3)	δ_{Nb-O}(%)
x=0.05	1.9871(24)	2.0179(12)	2.0179(12)	1.534
X=0.1	1.9920(4)	2.0145(14)	2.0145(14)	1.121
X=0.15	2.0040(5)	2.0066(4)	2.0066(4)	0.129
X=0.2	2.00598(3)	2.00536(3)	2.00536(3)	0.025

dielectric constant are described as Clausius-Mostti equations[25].

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{N\alpha}{3\varepsilon_0} \quad (1)$$

$$\alpha = \frac{4\pi\varepsilon_0 a^3}{n - 1} \quad (2)$$

Where α is polarization and a is the distance between cations and anions, n is the repulsive energy index of electron cloud. According to the calculated length of Nb-O bonds in Table 2, all the Nb-O bonds were classified into two categories. One contains four identical bonds (blue bond in Fig. 3), and another contains two identical bonds (yellow bond in Fig. 3). The yellow bonds were longer than blue bonds with small Ta addition. According to Clausius-Mostti equations, the longer bond lengths decide the intensity of polarization in oxygen octahedron, because the long bond lengths offer the long polarization

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distance between cations and anions. Four blue bonds formed a plane which was crucial to ionic polarization in oxygen octahedron. As the Ta content increased gradually, four yellow bonds were shortened, which means polarization in oxygen octahedron was restricted. This tendency would lead to the decrease of dielectric constant. Wonderfully, the results presented in Fig. 2 indicated that the dielectric constant reduced as Ta content increasing when the samples sintered at the temperature of 1120 ºC to 1160 ºC. Meanwhile, the dielectric constant changed slightly with increased sintering temperatures, this tendency could be explained by the samples already highly densified at the lower sintering temperature.

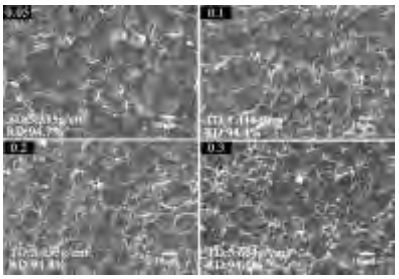

Figure 4. SEM micrographs, theoretical density (TD) and relative density (RD) of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at 1140 ºC.

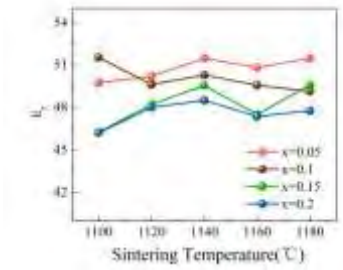

Figure 5. The dielectric constant of the $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at different temperatures.

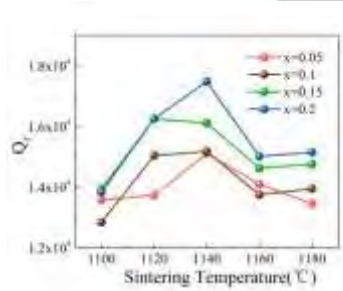

Figure 6. The Q×f values of the $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at different temperatures.

The *Q×f* value is characterization of dielectric loss in microwave frequency. High *Q×f* value means low dielectric loss. Generally, factors which affect the *Q×f* value can be divided into two factors: intrinsic and extrinsic factors. The extrinsic factors contain: grain boundaries, the second phase, porosity and packing fraction[8]. And the intrinsic factors are dominated by lattice vibration modes. Many researchers have proofed that the packing fraction is closely related to *Q×f* value. In their views, high packing fraction leads to high *Q×f* value. The packing fraction is the volume of all the packed ions divide by the volume of primitive unit cell. The packing fraction is given as follows:

$$\text{packing fraction}(\%) = \frac{\text{volume of packed ions}}{\text{volume of primitive unit cell}} \times \text{Z} \qquad (3)$$

According to the structure in JCPDS #52-1875, Z is equal to 1 for $Ni_{0.5}Ti_{0.5}NbO_4$ phase. The volume of packed ions was not changed, because the substituted Ta^{5+} ions have the same ionic radius to Nb^{5+}. So that, the packing fractions is mainly decided by the volume of primitive unit cell. As presented in Table 1, the volume of primitive unit cell increased with the increase of Ta content. Therefore, the packing fraction was increased. As a result, the *Q×f* value was increased simultaneously. This tendency is presented in Fig. 6.

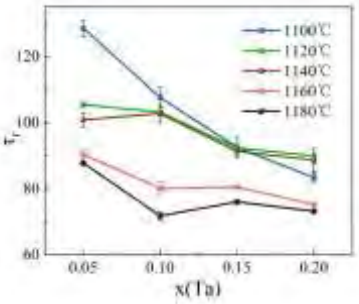

Figure 7. The temperature coefficient of the resonant frequency of the $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at different temperatures.

Fig. 7 illustrates the τ_f values of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics sintered at different temperatures. As Fig. 7 shows that the τ_f values decreased with increased Ta content, which means the dielectric properties become more temperature stabled. As studied by other researchers, the τ_f values were influenced by structural characteristics usually, especially the tilting of oxygen octahedral in microwave materials[26]. Jinxin Bi et al. [18] have studied that the distortions of Nb-O bonds mostly contributed to τ_f values in materials which contains NbO_6 octahedral structures. The distortion is calculated using the following equation:

$$\delta_{Nb-O} = \frac{d_{max} - d_{min}}{d_{ave}} \times 100\% \qquad (4)$$

Where d is the bond distance of individual bond. The calculated values of $\delta_{Nb\text{-}O}$ are listed in table 2. The $\delta_{Nb\text{-}O}$ decreased with increased Ta content, combine with the schematic diagram in Fig. 3, the longer bond lengths became shorter and the shorter bond lengths became longer in $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics as Ta content increasing. So that, temperature stability was strengthen. And τ_f values were decreased with Ta additions.

Finally, the optimal microwave dielectric properties of ε_r = 48.5, Q×f =17500 GHz and τ_f = 88.6 ppm/ºC at the resonant frequency of 6.05GHz were obtained for $Ni_{0.5}Ti_{0.5}Nb_{0.8}Ta_{0.2}O_4$ sintered at 1140 ºC.

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Conclusions

The $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics have been synthesized via the traditional solid-state sintering method. Crystal structure and microwave dielectric properties were studied in this article. The dielectric constant of $Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ ceramics decreased with the increase of Ta content. This tendency is caused by the polarization distance reduced when more Ta substituted in Nb site. Thus, the dielectric constant decreased because the polarization is restricted. The packing fractions increased with Ta substitution. The *Q×f* values have the same variation tendency with packing fraction. The τ_f values are affected mostly by $\delta_{Nb\text{-}O}$. That Ta doped in Nb site has changed the bond length which leaded to the reducing the $\delta_{Nb\text{-}O}$ of NbO_6 octahedral. Therefore, the τ_f value is changed to lower value. The optimal microwave dielectric property of ε_r = 48.5, Q×f =17500 GHz and τ_f = 88.6 ppm/ºC at the resonant frequency of 6.05GHz makes $Ni_{0.5}Ti_{0.5}Nb_{0.8}Ta_{0.2}O_4$ ceramic a better candidate material to fabricate the microwave device.

Experimental Section

$Ni_{0.5}Ti_{0.5}Nb_{1-x}Ta_xO_4$ (x=0.05, 0.1, 0.15, 0.2) ceramics powders were prepared using solid-state reaction method with high purity oxides (NiO (98 %), Ta_2O_5 (99.5 %), TiO_2 (99.9 %) and Nb_2O_5 (99.5 %)). The starting materials were mixed in a ball mill using deionized water as medium in a nylon container with ZrO_2 balls for 10 h. Then, the slurries were dried and pre-sintered at 1040 ºC for 10 h. After that the calcined mixture was re-milled for further 10 h to obtain fine powders, and was pressed into pellet disks with 5%PVA. The disks were been sintered at 1100-1180 ºC for 6h, and furnace-cooled to room temperature. The phase formation was examined by an X-ray diffract-meter (XRD, DX-2700, Haoyuan co.) with Cu Kα radiation. The microstructures and EDX were measured by a scanning electron microscope (JSM-6490, JEOL, Japan). The bulk density was measured by the Archimedes method. The ε_r and *Q×f* values were measured at the frequncey ranges from 300 KHz to 20 GHz by the Hakki-Coleman dielectric resonator method with network analyzer(Agilent N5230A, USA). The τ_f value was calculated by the following equation:

$$\tau_{f=}\frac{f_{85}-f_{25}}{f_{25}\times 60}\times 10^6(\mathrm{ppm/^{\circ}C})$$

where f_{25} and f_{85} are the resonant frequencies at 25 ºC and 85 ºC, respectively.

Acknowledgements

The authors acknowledge the financial support by the National Key Research and Development Plan (No. 2016YFA0300801), and by the development programmer of China (863 Program) (Grant No. 2015AA034102) and by the National Natural Science Foundation of China (No. 51602036, 6167118 and 51672036).

Keywords: crystal structure • microwave dielectric properties • niobate • ionic substitutions

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10.1002/ejic.201800016

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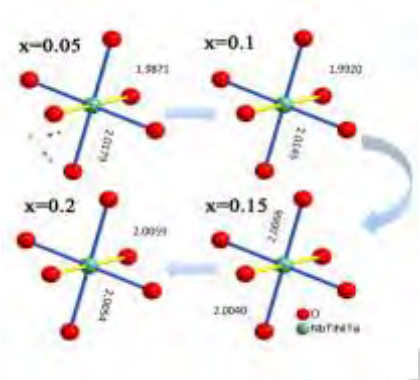
The oxygen octahedron distortion is affected by Ta substitution. The Nb-O bonds can be classified into two categories. The yellow bonds are longer than blue bonds with small Ta addition, As the Ta content increased gradually, four yellow bonds were shortened, which means polarization in oxygen octahedron are restricted. This variation influenced the dielectric properties obviously.



Key Topic*

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