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# Microwave dielectric characteristics of $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics

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**Abstract**

The sintering characteristics, phase composition, and microwave dielectric properties of $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1300–1500 °C have been investigated. Results show that $Nb^{5+}$ and $Al^{3+}$ can co-substitute for $Ti^{4+}$ and form $Ti_{0.8}Al_{0.1}Nb_{0.1}O_2$, which can lower effectively the sintering temperature, and improve the quality factor of $0.9Al_2O_3$–$0.1TiO_2$ ceramics.


*Keywords:* C. Dielectric properties; Ceramics; $Nb_2O_5$ addition; $Al_2O_3$–$TiO_2$

## 1. Introduction

Dielectric materials for microwave applications have been used in mobile phones, wireless local area networks (LAN), and intelligent transport systems (ITS). Recently, they are planned to be applied in ultrahigh speed wireless LAN in millimeterwave, because they can reduce the resource of electromagnetic wave. Dielectric ceramics for millimeterwave applications are also planned for use in ITS, including a car anti-collision system [1].

In order to meet this application, high performance microwave dielectric materials with a high quality factor ($Q$), and a low dielectric constant ($\varepsilon_r$) are required. Moreover, near-zero temperature coefficient of resonant frequency ($\tau_f$) is also needed to provide stability at various service temperatures. $Q$ value is generally evaluated as $Q{\cdot}f$ ($f$ is resonant frequency) because $Q{\cdot}f$ is almost independent on resonant frequency [2].

Alumina ($Al_2O_3$) is used as a ceramic substrate material because of its high thermal conductivity, suitable dielectric constant, and low dielectric loss. However, it has a large negative $\tau_f$ value (−60 ppm/°C) [3,4]. In order to shift the $\tau_f$ value of $Al_2O_3$ ceramics, $TiO_2$ with positive $\tau_f$ value (+450 ppm/°C) [5] can be introduced to form a composition of $Al_2O_3$–$TiO_2$. It shows that the $\tau_f$ value of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1550 °C can reach +1.5 ppm/°C. To lower the sintering temperature of the $0.9Al_2O_3$–$0.1TiO_2$ composition, suitable oxides, low-melting-point glasses, and smaller particle size of the starting materials should been used. In this study, the effects of $Nb_2O_5$ content on microwave dielectric properties of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics are reported. Relationships among the sintering temperature, phase compositions, and microwave dielectric properties of the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics are also discussed.

## 2. Experimental procedures

$Al_2O_3$ and $TiO_2$ with purity higher than 99.5% were used as starting materials, and mixed according to the composition of $0.9Al_2O_3$–$0.1TiO_2$ by ball-milling for 3 h in deionized water. After drying and grinding, the powder was calcined at 1100 °C for 3 h. The calcined $0.9Al_2O_3$–$0.1TiO_2$ powder was mixed with $Nb_2O_5$ by ball-milling in deionized water for 1 h. After drying and mixing with polyvinylalcohol as binder, the mixed powder was pressed into pellets in a steel die. After debindering, these pellets were sintered at 1300–1500 °C for 3 h.

The crystal phases of the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics were analyzed by X-ray powder diffraction (XRD) using Cu Kα radiation and electron-probe microanalysis (EPMA). The $\varepsilon_r$ and $Q{\cdot}f$ value were measured in the $TE_{011}$ mode by the Hakki and Coleman's dielectric resonator method which was improved by Courtney [6,7], using an Advantest

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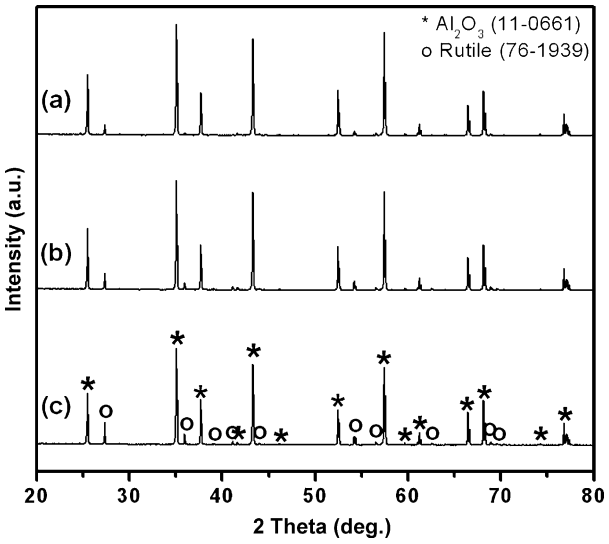


Fig. 1. X-ray diffraction patterns of the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1400 °C: (a) 0.3 wt.%; (b) 0.5 wt.%; and (c) 0.8 wt.%.

Table 1
Element content in the white area in the $0.9Al_2O_3$–$0.1TiO_2$ ceramics with 0.5 wt.% $Nb_2O_5$ sintered at 1400 °C.

| Element | Al | Ti | Nb | O |
|---|---|---|---|---|
| Content (at.%) | 2.82 | 27.42 | 3.06 | 66.71 |

R3767C network analyzer. The $\tau_f$ value was defined by formula:

$$\tau_f = \frac{1}{f(20)} \times \frac{f(80) - f(20)}{80 - 20} \quad (1)$$

where $f(20)$ and $f(80)$ are the resonant frequency at 20 and 80 °C, respectively.

## 3. Results and discussion

Fig. 1 shows XRD patterns of the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1400 °C for 3 h. $Al_2O_3$ and $TiO_2$ are two main crystalline phases in the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ system. With the increase of $Nb_2O_5$, the X-ray diffraction intensity of $Al_2O_3$ phase decreases while the intensity of $TiO_2$ phase increases.

Fig. 2 shows backscattered electron image and mapping of the elements by EPMA for the $0.9Al_2O_3$–$0.1TiO_2$ ceramics with 0.5 wt.% $Nb_2O_5$ sintered at 1400 °C. It can be observed clearly from the backscattered electron image that two phases exist in the system. The white area represents rutile phase while the gray area is $Al_2O_3$ phase, which agrees well with the above XRD results, as shown in Fig. 1.

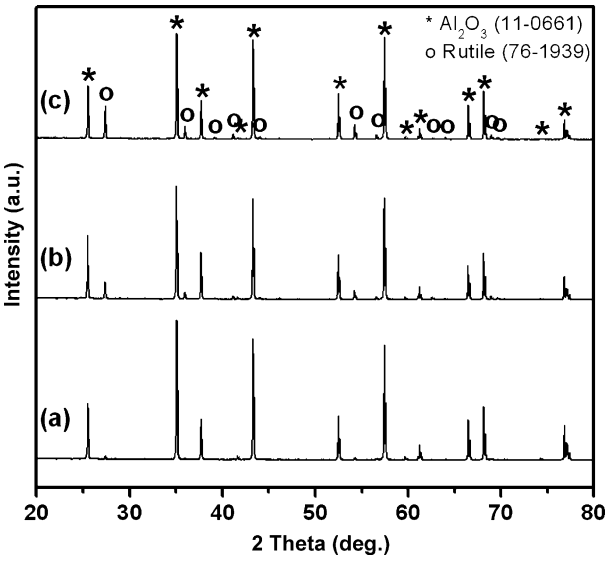


Fig. 3. X-ray diffraction patterns of the 0.5 wt.% $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1300–1500 °C, respectively. (a) 1300 °C; (b) 1400 °C; and (c) 1500 °C.

Table 1 shows element content in the white area in the $0.9Al_2O_3$–$0.1TiO_2$ ceramics with 0.5 wt.% $Nb_2O_5$ sintered at 1400 °C. A small amount of $Al^{3+}$ and $Nb^{5+}$ cations is included in the rutile phase, because the equal mole $Al^{3+}$ and $Nb^{5+}$ cations can co-substitute for $Ti^{4+}$ cation and form $Ti_{0.8}Al_{0.1}Nb_{0.1}O_2$ solid solution. Therefore, with the increase of $Nb_2O_5$ content, the amount of $Ti_{0.8}Al_{0.1}Nb_{0.1}O_2$ solid solution phase increases while that of $Al_2O_3$ phase reduces in the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ system, which results into the enhancement in the X-ray diffraction intensities of rutile phase and the weakening in those of $Al_2O_3$ phase, as shown in Fig. 1.

Fig. 3 shows XRD patterns for the 0.5 wt.% $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1300–1500 °C for 3 h. With the increase of the sintering temperature, the X-ray diffraction intensities of $Al_2O_3$ phase reduce gradually while those of rutile phase enhance obviously, which indicates that the increase in sintering temperature can promote the formation of $Ti_{0.8}Al_{0.1}Nb_{0.1}O_2$ solid solution.

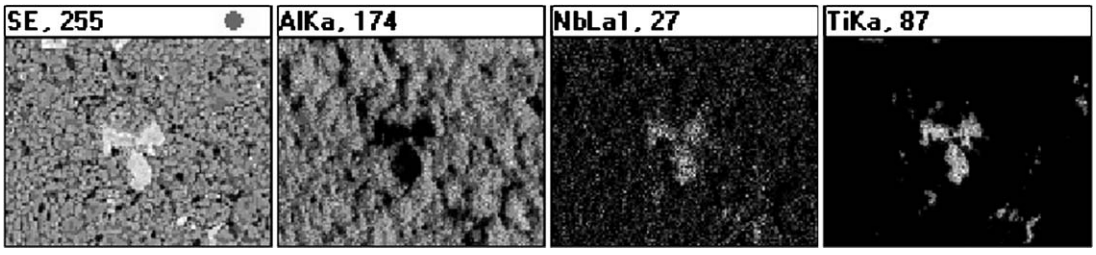


Fig. 2. Backscattered electron image and mapping of the elements by EPMA for the $0.9Al_2O_3$–$0.1TiO_2$ ceramics with 0.5 wt.% $Nb_2O_5$ sintered at 1400 °C.

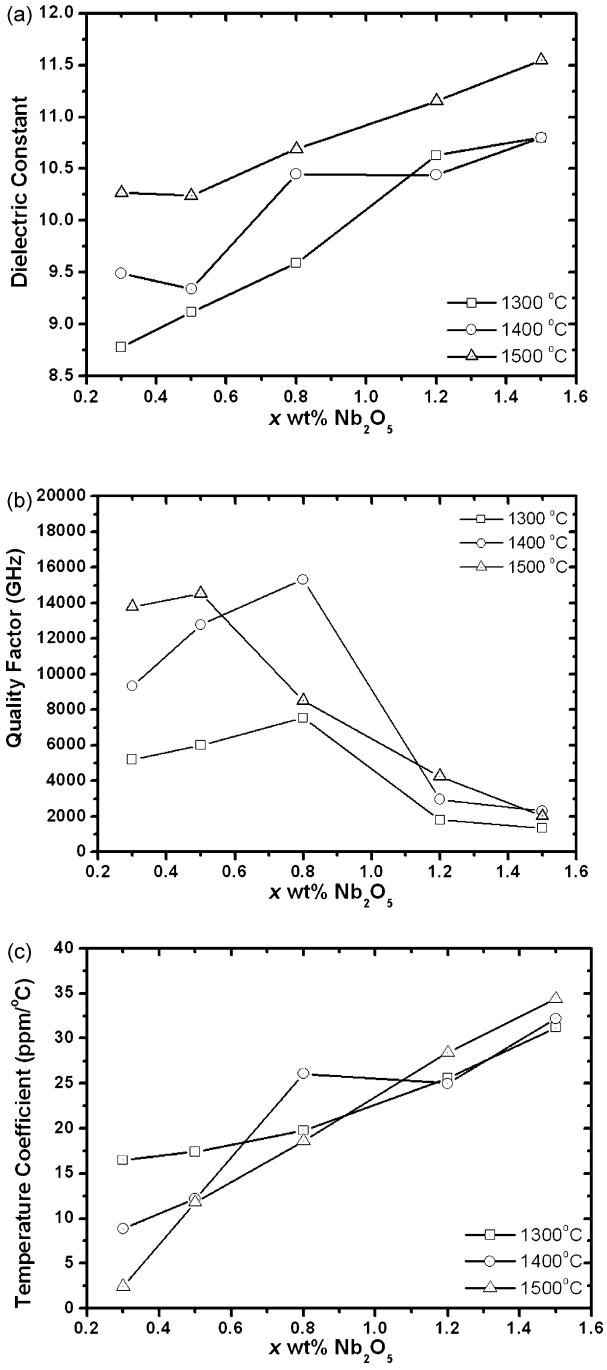


Fig. 4. Microwave dielectric properties of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics as a function of sintering temperature and $Nb_2O_5$ content.

Microwave dielectric properties of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics as a function of sintering temperature and $Nb_2O_5$ content are shown in Fig. 4. With the increase of $Nb_2O_5$ content or sintering temperature, the $\varepsilon_r$ value takes on an increasing tendency, and the $Q{\cdot}f$ value increases initially and then reduces. Furthermore, the $\tau_f$ value increases gradually with $Nb_2O_5$ content while it is nearly independent on sintering temperature.

An increase in $Nb_2O_5$ content or sintering temperature can result in an increase in the amount of $Ti_{0.8}Al_{0.1}Nb_{0.1}O_2$ solid solution in the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ system, as shown in Figs. 1 and 3 respectively. According to Shannon [8], the polarizability of $Nb^{5+}$ cation is 3.98 Å, which is bigger than that of $Ti^{4+}$ ($\alpha$ = 2.94 Å). As a result, the $\varepsilon_r$ value increases with the augment of $Nb_2O_5$ content and sintering temperature (see Fig. 4(a)).

The $Q{\cdot}f$ value is generally affected not only by the lattice vibrational modes, but also by the density, the second phases, the impurity, the lattice defect, crystallizability, and inner stress. The increase in sintering temperature is beneficial to the densification and crystallizability until the $Q{\cdot}f$ value reaches the maximum. The further increase in sintering temperature will result in the appearance of abnormal grains and pores and consequently lead to the reduction of the $Q{\cdot}f$ values [9]. It can be observed from Fig. 4(b) that the $Q{\cdot}f$ value can reach the maximum 15,320 GHz when the sintering temperature is equal to 1400 °C, lower than the densification temperature of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics by about 150 °C, which indicates that $Nb_2O_5$ addition can enhance the sinterability of the $0.9Al_2O_3$–$0.1TiO_2$ system. Therefore, the increase in $Nb_2O_5$ content can improve the density and crystallizability, which results in the increase of the $Q{\cdot}f$ value. However, at the same time, the amount of the impurity and defect increases with $Nb_2O_5$ content, which deteriorates the $Q{\cdot}f$ value. Therefore, as the $Q{\cdot}f$ value increases initially and then reduces with the increase of $Nb_2O_5$ content.

$Nb_2O_5$ addition can heighten the $\tau_f$ value of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics, as shown in Fig. 4(c). When $x$ is equal to 0.3, the $\tau_f$ can be controlled to near zero value. It is not obvious that the effect of sintering temperature on the $\tau_f$ value. To understand clearly the relationship among $Nb_2O_5$ content, sintering temperature and $\tau_f$ value, further investigation must be performed.

Compared with $0.9Al_2O_3$–$0.1TiO_2$ ceramics sintered at 1550 °C with an $\varepsilon_r$ value of 11.8, a $Q{\cdot}f$ value of 8000 GHz and a $\tau_f$ value of +1.5 ppm/°C, the $Nb_2O_5$-added $0.9Al_2O_3$–$0.1TiO_2$ ceramics have more excellent sinterability and $Q{\cdot}f$ value.

## 4. Conclusion

$Nb_2O_5$ addition can lower effectively the sintering temperature of the $0.9Al_2O_3$–$0.1TiO_2$ ceramics. The equal mole $Nb^{5+}$ and $Al^{3+}$ can co-substitute for $Ti^{4+}$ and form $Ti_{0.8}Al_{0.1}Nb_{0.1}O_2$ solid solution. With the increase of $Nb_2O_5$ content or sintering temperature, the $\varepsilon_r$ value takes on an increasing tendency, and the $Q{\cdot}f$ value increases initially and then reduces. Furthermore, the $\tau_f$ value increases gradually with $Nb_2O_5$ content while it is nearly independent on sintering temperature.

## References

[1] W. Werising, in: B.C.H. Steele (Ed.), Electronic Ceramics, Elsevier, London, 1991, p. 67.
[2] G. Wolfram, H.E. Gobel, Mater. Res. Bull. 16 (1981) 1455.
[3] N.M. Alford, S.J. Penn, J. Appl. Phys. 80 (1996) 5895.
[4] S.J. Penn, N.M. Alford, A. Templeton, X. Wang, M. Xu, M. Reece, K. Schrapel, J. Am. Ceram. Soc. 80 (1997) 1885.

[5] A. Templeton, X. Wang, S.J. Penn, N.M. Alford, J. Am. Ceram. Soc. 83 (2000) 95.
[6] B.W. Hakki, P.D. Coleman, IEEE Trans. Microwave Theory Tech. 16 (1960) 402.
[7] W.E. Courtney, IEEE Trans. Microwave Theory Tech. 18 (1970) 476.
[8] D. Shannon, J. Appl. Phys. 73 (1993) 6991.
[9] W. Lei, W.-Z. Lu, J.-H. Zhu, X.-H. Wang, Mater. Lett. 61 (2007) 4066.