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$BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ composite microwave ceramics with high *Q*-factor and low sintering temperature

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Abstract

Revised thermodynamic equilibrium in the BaO–MgO–Nb_2O_5 pseudo-ternary system has lead to development of a novel composite dielectric material with dielectric constant, $\varepsilon' = 25.5$, efficacy factor, $Q \times f = 160$ THz, and temperature coefficient of the resonant frequency, $\tau_f = +0.5$ ppm/K. The material shows one of the highest *Q*-factors among the Ta-free microwave dielectric resonators. It also does not contain volatile Zn and Co elements. Other important property of the title compound is low sintering temperature of 1320 °C which significantly reduces the processing cost.

Keywords: A. Dielectric resonators; C. Phase equilibria; D. Wireless; D. Microwave

1. Introduction

Solid-state microwave resonators based on dielectric ceramics with appropriate dielectric constant, low dielectric loss, and temperature-stable resonance frequency are the core elements of the wireless base station filters, microwave oscillators and input multiplexers for communication satellites. Although the field of dielectric resonators (DRs) for microwaves has matured several decades ago, the new product requirements together with increasing price of raw materials have been major driving forces behind the development of novel low-loss DRs.

Because ceramics with zero temperature coefficient of the resonance frequency ($\tau_f \approx 0$ ppm/K) are rare, several alternative methods have been adopted to achieve temperature-invariant DRs. The first method involves preparation of the single phase ceramics by alloying two materials with similar crystal structure and opposite signs of τ_f.[1] Temperature-invariant dielectric resonators based on solid solutions of $LaAlO_3$–$Ca(Sr)TiO_3$, $BaZn_{1/3}Nb_{2/3}O_3$–$BaCo_{1/3}Nb_{2/3}O_3$, $BaZn_{1/3}Nb_{2/3}O_3$–$SrZn_{1/3}Nb_{2/3}O_3$ have been successfully realized using this approach.[1–5] The second method involves preparation of a composite ceramics consisting of two different chemical/structural phases with opposite τ_f. This approach was utilized in $Ag(Nb_{0.65}Ta_{0.35})O_3$–$Ag(Nb_{0.35}Ta_{0.65})O_3$, Al_2O_3–TiO_2, $MgTiO_3$–$CaTiO_3$ and Mg_2TiO_4–$CaTiO_3$ composites.[6–11]

In many cases preparation of dense composite ceramics suffers from chemical (thermodynamic) incompatibility of the comprising phases that brings about undesirable secondary phases. As a result, ceramic processing becomes rather complex as one has to carefully consider both equilibrium thermodynamics and reaction kinetics to minimize the chemical reaction and inter-diffusion between the main ceramic constituents.[6] An alternative solution is to find two ceramic counterparts having opposite signs of the τ_f that can co-exist in thermodynamic equilibrium. The simplest example of such a composite system consists of the two compounds *A* and *B* with zero gradient of the chemical potential, $\mu_i^A - \mu_i^B = 0$, where μ_i^A and μ_i^B are chemical potentials of constituent *i* in phases *A* and *B*, respectively. Such conditions are usually satisfied when the two phases are connected by a tie line in the binary or ternary phase diagram.[12] Here the author reports on a practical example that highlights the remarkable potential of the aforementioned approach.

$BaMg_{1/3}Nb_{2/3}O_3$ was selected as one of the starting compounds owing to its low dielectric loss, $\tan\delta \leq 5.0 \times 10^{-5}$ at 10 GHz, and suitable dielectric constant, $\varepsilon' \approx 32$.[13,14] The crucial drawback of the $BaMg_{1/3}Nb_{2/3}O_3$-based DR is a large $\tau_f \approx +25$ ppm/K which is beyond commercially viable range of $-3 \leq \tau_f \leq +5$ ppm/K. Recently Kolodiazhnyi et al.[15] reported on the revised sub-solidus thermodynamic equilibria in the BaO–MgO–Ta_2O_5 pseudo-ternary system where they found

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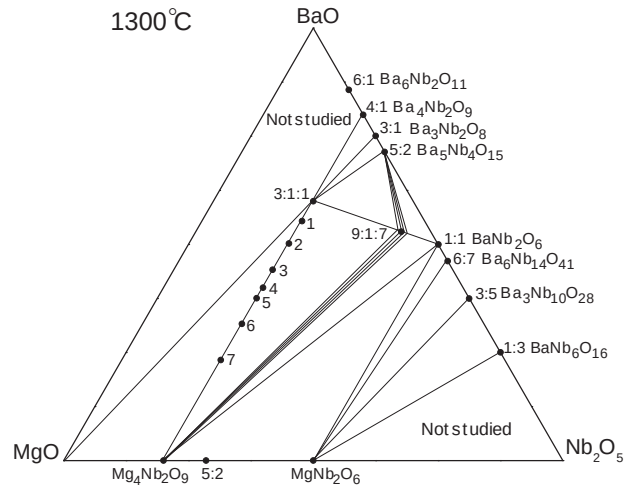

Fig. 1. Subsolidus phase diagram of BaO–MgO–Nb_2O_5 adopted from Ref. 16. Only two ternary compounds have been identified, perovskite-type 3:1:1 = $Ba_3MgNb_2O_9$ and tetragonal tungsten bronze-type 9:1:7 = $Ba_9MgNb_{14}O_{45}$. Black dots numbered 1–7 are the samples along the $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ tie line reported in this study. Their detailed chemical compositions are given in Table 1.

that the $BaMg_{1/3}Ta_{2/3}O_3$ perovskite phase forms a tie line with corundum-type $Mg_4Ta_2O_9$. In a follow-up study of the BaO–MgO–Nb_2O_5 pseudo-ternary system, the author confirmed that a similar tie line exists between the $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$ phases.[16] The relevance of the latter studies is recognized by the fact that both $Mg_4Ta_2O_9$ and $Mg_4Nb_2O_9$ show very low dielectric loss (i.e., tan $\delta \leq 3.0 \times 10^{-5}$ at 10 GHz), and, most importantly, a negative τ_f of −70 ppm/K.[17,18] It is obvious, therefore, that the $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ composite ceramic offers a unique opportunity of achieving the temperature-invariant low-loss DR while completely avoiding the chemical incompatibility issues.

2. Experimental

A series of the chemical compositions along the $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ tie line can be represented by the hypothetical chemical formulae $Ba_{4-5x}Mg_{5x}Nb_2O_9$, where $0.2 \leq x \leq 0.8$. The location of the selected compositions on the BaO–MgO–Nb_2O_5 pseudo-ternary diagram is shown by black dots numbered 1, 2, 3, . . ., 7 in Fig. 1. Samples were prepared from 99.9% pure $BaCO_3$, MgO and Nb_2O_5 purchased from Tokai Chemy, Japan. The mixtures with $0.24 \leq x \leq 0.57$ were calcined at 1050 °C for up to 10 h. The calcined product was wet-milled for 20 h. 2 wt% of polyvinyl alcohol binder was added to the powder and the powder was molded by press into compacts of 7 mm diameter and 4 mm height. The compacts were heat treated at 500 °C in air to remove the binder and then sintered at 1320 °C for up to 10 h. The end-members, $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$, were sintered at 1490 and 1400 °C, respectively. Phase composition was studied by powder X-ray diffraction (Rigaku Ultima III X-ray diffractometer with Cu K_α X-ray source). Lattice parameters were obtained from Rietveld refinement of the X-ray data using RIETAN 2000.[19] Quantitative phase analysis of the composite ceramics was performed by electron probe microanalysis (EPMA) using wavelength-dispersive (WDS) X-ray spectroscopy (model JXA-8500F, JEOL). The as sintered ceramic DRs had diameter of *ca.* 5.9 mm. The height of the DRs was adjusted using the SiC abrasive powder to 2.6 mm to make certain that the first resonance mode is of the $TE_{01\delta}$-type. The DR was placed on the low-loss quartz support of 5 mm diameter and 4.3 mm height in the center of the commercial cylindrical resonance cavity (QWED, Poland) having inner diameter of 24 mm and height of 16 mm. The resonances of the microwave power in transmission mode (s^{21} parameter) were observed using HP 8719C Vector Network Analyzer. The dielectric constant, ε', and unloaded Q-factor were calculated using the QWED software.[20] Temperature coefficient of the resonance frequency, τ_f, was measured in the temperature interval of +20 to +90 °C. A more detailed description of the sample preparation and dielectric measurements are given elsewhere.[21]

3. Results and discussion

Before reporting on the dielectric properties of the $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ composite ceramics, the author would like to briefly address the BaO–MgO–Nb_2O_5 subsolidus phase equilibria shown in Fig. 1. At 1300 °C only two ternary compounds were identified, i.e., perovskite 3:1:1 = $Ba_3MgNb_2O_9$ and tetragonal tungsten bronze (TTB) 9:1:7 = $Ba_9MgNb_{14}O_{45}$. Similar to the BaO–MgO–Ta_2O_5 phase equilibria,[15] the 3:1:1 phase forms tie lines with $Mg_4Nb_2O_9$, MgO, $Ba_4Nb_2O_9$ and 9:1:7 phases. The solidus temperature along the $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ pseudo-binary was found to be 1360 ± 5 °C according to the differential scanning calorimetry data.[16] In spite of the similar ionic radii of Nb^{5+} and Ta^{5+}, the Nb-based analogue of the 4:1:5 = $Ba_4MgTa_{10}O_{30}$ phase was not found in the BaO–MgO–Nb_2O_5 pseudo-ternary diagram. Wang et al.[22] have reported that the substitution limit of Nb for Ta in $Ba_4MgTa_{10-z}Nb_zO_{30}$ does not exceed $z = 2$.

It is known that the minute amounts of secondary phases with high dielectric loss adversely affect the Q-factor of ceramics. For example, formation of the 9:1:7 $Ba_9MgTa_{14}O_{45}$ secondary phase in the $BaMg_{1/3}Ta_{2/3}O_3$ ceramics sintered above 1600 °C causes drastic decrease in the Q-factor.[23] To avoid traces of the relaxor-type 9:1:7 $Ba_9MgNb_{14}O_{45}$ phase with high dielectric loss,[24] samples prepared along the $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ tie line were formulated according to the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ with a small Nb deficiency. Their detailed compositions are given in Table 1.

The powder X-ray diffraction data shown in Fig. 2 for sample 4 revealed that the final product is comprised of $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$ phases in agreement with the phase diagram reported in Fig. 1. The intensity of the diffraction peaks correlates with the relative volume of the $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$ phases of the target compositions. Rietveld refinement of the lattice parameters and weight fraction confirmed that the sample 4 consists of 76 wt% of $BaMg_{1/3}Nb_{2/3}O_3$ (space group $P\bar{3}m1$, lattice constants $a = 5.776(1)$ Å and $c = 7.088(8)$ Å) and 24 wt% of $Mg_4Nb_2O_9$ (space group $P\bar{3}c1$, lattice constants $a = 5.161(9)$ Å

Table 1
Chemical composition, sintering temperature, and properties of the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ ceramics.

Sample	x	y	T_{sint} (°C)	ε'	τ_f (ppm/K)	$Q \times f$ (THz)
$BaMg_{1/3}Nb_{2/3}O_3$	0.2	0	1490	32	+25	180
1	0.245	0.002	1320	30	+17.2	171
2	0.296	0.002	1320	29	+13.4	163
3	0.362	0.002	1320	27	+7.4	152
4	0.400	0.002	1320	26	+3.7	150
5	0.425	0.002	1320	25	+0.5	160
6	0.485	0.002	1320	23	−9.1	170
7	0.569	0.002	1320	21	−18.8	184
$Mg_4Nb_2O_9$	0.8	0	1400	13	−69.5	190

and $c = 14.021(7)$ Å). The X-ray diffraction pattern did not show any MgO reflections. This is not unusual because the sensitivity of the laboratory X-rays to the phases containing light elements does not exceed 3–5% and can be attributed to the low concentration and weak X-ray electron scattering of the minute amount of the MgO phase.

In agreement with the X-ray diffraction data, the microstructural analysis shown in Fig. 3 confirmed that the title compound consists of two major phases, $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$. In addition, small amount of the MgO phase was detected by EPMA in line with the accurate sample composition formulated with a small Nb deficiency. As mentioned above, the Nb deficiency was introduced to suppress the formation of the relaxor-type 9:1:7 phase, $Ba_9MgNb_{14}O_{45}$. In contrast to the 9:1:7 phase, MgO demonstrates very low dielectric loss of $Q \times f \geq 300$ THz.[25] Therefore, the presence of a small amount of MgO in the composites is not expected to detrimentally affect the overall Q-factor.

Microwave dielectric properties of the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ composite are summarized in Fig. 4, panels (a)–(c) as well as Table 1. Panel (a) in Fig. 4 shows linear dependence of the dielectric constant which decreases from 32 to 12 as composition changes from $x = 0.2$ to 0.8. In contrast to the composites with large difference in the dielectric constant of constituent phases, the linear dependence of ε' is expected in the compounds with not so different values of ε' of their end members. In the latter case, the effective dielectric constant, ε_{eff} is given by $\varepsilon_{eff} \approx \sum_{j=1}^{n} \gamma_j \varepsilon_j$, where γ_j is a volume fraction of the phase with dielectric constant ε_j. In the former case of significantly different ε_j, the effective dielectric constant requires calculations of the electric filling factors of constituent phases for a given electromagnetic mode.[10] The validity of the simple sum rule for the effective dielectric constant in our case is further supported by the nearly liner composition dependence of the τ_f shown in panel (b) of Fig. 4. A gradual change of τ_f from +10 to −10 ppm/K in the $0.326 \leq x \leq 0.500$ range makes this composite material technologically attractive for microwave communication industry. At $x = 0.425$ the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ composite shows $\varepsilon' = 25$, $Q \times f = 160$ THz and $\tau_f = +0.5$ ppm/K.

Dielectric loss, tan δ, in the microwave frequency band is conventionally reported as Q-factor ($Q = 1/\tan \delta$). In most cases of the microwave ceramics the Q-factor decreases (almost) linearly with frequency so that the product of $Q \times f$ is adopted as a figure of merit when comparing different microwave materials. Panel (c) in Fig. 4 shows the $Q \times f$ dependence of the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ as a function of x measured at $f = 10$ GHz.

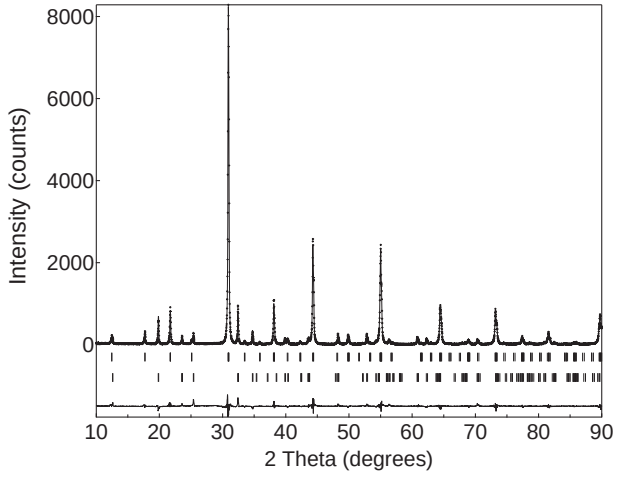

Fig. 2. X-ray diffraction pattern of sample 4 (+). Calculated diffraction pattern from Rietveld refinement (solid line). The firs and second rows of vertical bars indicate the positions of expected Bragg peaks for $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$ phases, respectively. The difference between observed and calculated data is shown as a solid line at the bottom of the plot.

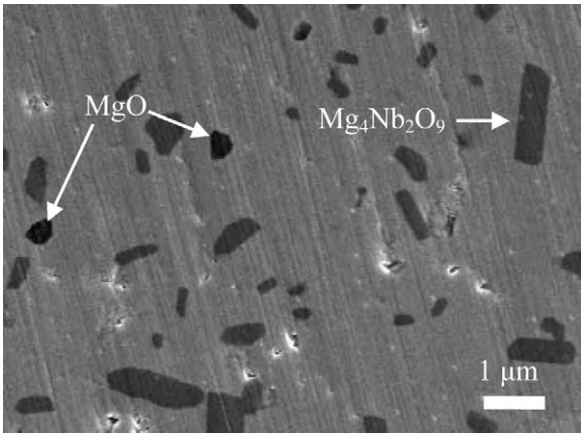

Fig. 3. Backscattered electron microscopy of the polished surface of the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ ceramics with $x = 0.296$ and $y = 0.002$ sintered at 1320 °C. Due to small Nb deficiency, in addition to the two major phases, $BaMg_{1/3}Nb_{2/3}O_3$ (light gray) and $Mg_4Nb_2O_9$ (dark gray), a minute amount of the MgO second phase (seen as a black inclusions due to the small electron scattering cross-section) was detected.

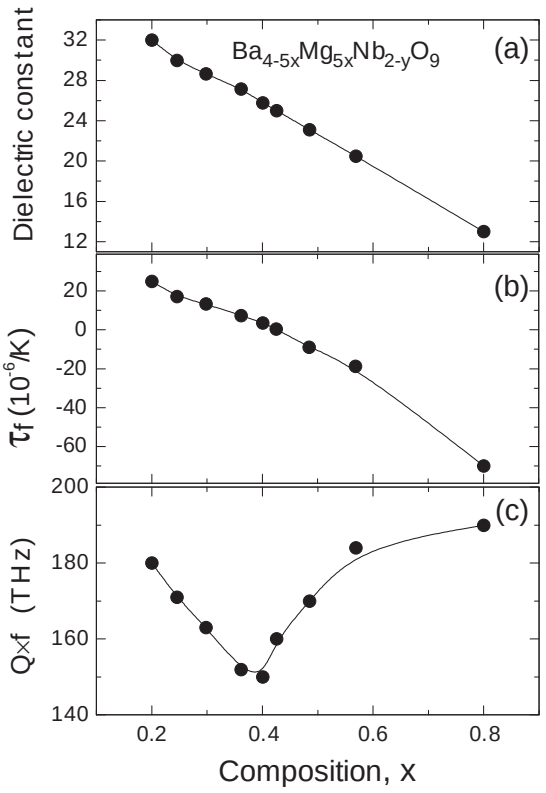

Fig. 4. Composition dependence of the dielectric constant (a), temperature coefficient of the resonance frequency, τ_f (b) and product of the Q-factor and resonance frequency, $Q \times f$ (c) with x in the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$. The values of y correspond to those listed in Table 1.

The end members of the title compound show $Q \times f = 180$ and 190 THz for $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$, respectively. The $Q \times f$ graph shows a broad minimum with $Q \times f = 150$ THz at $x = 0.400$. The origin of this broad minimum is not clear at the moment. One of the reasons could be the effect of the residual lattice strain on the optical phonon damping constant. Because the $Q \times f$ minimum corresponds to the approximately equal volumes of the $BaMg_{1/3}Nb_{2/3}O_3$ and $Mg_4Nb_2O_9$ phases, the lattice strain due to their different thermal expansion coefficients will effectively decrease the phonon lifetimes which will ultimately result in the increase in tan δ.[27] As a volume fraction of the second phase decreases towards the one of the end members of the composite, so does the lattice strain effect. As a result, the $Q \times f$ value shows improvement for $x = 0.2$ and 0.8. While previous experience with microwave ceramics suggests that the $Q \times f$ values are usually improved after optimization of the processing conditions,[13] the author remains sceptical that the optimized $Q \times f$ of the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ composite will exceed 185 THz. Although this value is significantly below the $Q \times f = 350$ THz of Ta-based microwave dielectric resonators,[26] it appears to be the highest among the Ta-free temperature stabilized DRs. For example, related dielectric ceramics based on $BaZn_{1/3}Nb_{2/3}O_3$–$BaCo_{1/3}Nb_{2/3}O_3$ solid solution shows a maximum of $Q \times f = 80$ THz.[28] Another advantage of the $Ba_{4-5x}Mg_{5x}Nb_{2-y}O_9$ composite is its low sintering temperature. In contrast to $BaZn_{1/3}Nb_{2/3}O_3$–$BaCo_{1/3}Nb_{2/3}O_3$ with sintering temperature of 1400–1430 °C, the title compound is sintered at as low as 1320 °C owing to its relatively low solidus temperature of 1360 °C.

In conclusion, the author reported on one example of the composite ceramics for microwave dielectric resonators utilizing the 'thermodynamic equilibrium' approach that avoids chemical incompatibility issues. The $BaMg_{1/3}Nb_{2/3}O_3$–$Mg_4Nb_2O_9$ composite dielectric ceramics obtained according to this approach shows the highest $Q \times f$ value among the Ta-free dielectric resonators. It also offers a low-cost processing route at reduced sintering temperature. A detailed knowledge of the phase equilibria diagrams is required to expand this approach to other composite microwave dielectric ceramics.

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