Effect of B_2O_3 On the Microstructure and Microwave Dielectric Properties of $Ba(Mg_{1/3}Nb_{2/3})O_3$ Ceramics

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Abstract

The influences of different doping contents of additive B_2O_3 on the sintering behavior, microstructure and microwave properties of $Ba(Mg_{1/3}Nb_{2/3})O_3$ (BMN) microwave ceramics were studied. The main phase in the sintered ceramics was crystallized as $Ba(Mg_{1/3}Nb_{2/3})O_3$ (BMN). $Ba_2B_2O_5$ as secondary phase was observed with $x \geq 10$. Scanning electron microscopy (SEM) analysis revealed that the photograph of optimally nonstoichiometric ceramics sintered at 900 °C for 5 h displayed a dense microstructure. The dielectric properties and microstructures of BMN ceramics were very sensitive to the nonstoichiometry. The addition of B_2O_3 had a positive influence on the $Q \times f$ value, for example the sample with $x = 7$ showed the highest $Q \times f$ value of 89800 GHz. Finally, excellent microwave dielectric properties ($\epsilon_r = 31.6$, $Q \times f = 89,800$ GHz and $\tau_f = 3.6$ ppm/°C) were achieved for the sample with $x = 7$ sintered at 900 °C for 5 h.

1. Introduction

In the production of miniaturized devices, low densification sintering temperature is very important for ceramic multilayer structures, thus they can be co-fired with low-loss conductors such as silver or copper. Low temperature co-fired ceramic (LTCC) technology has remarkable advantages over the previous packaging technologies in packaging applications with high density, high speed, good heat dissipation and sealing [1-2]. In recent years, some ceramic-glass compositions [3-5] have been used in microwave devices, such as low dielectric constant materials using alumina and suitable glass combinations, and high dielectric constant materials with low dielectric loss in the range of 20-100. As microwave filters, complex perovskite ceramics $Ba(A_{1/3}B_{2/3})O_3$ ($A = Mg, Zn$; $B = Ta, Nb$) have displayed notable properties for the microwave ceramics. Particularly, $Ba(Mg_{1/3}Nb_{2/3})O_3$ (BMN) ceramic have broad application prospects in electronic systems with preferable dielectric properties of $\epsilon_r = 32$, $Q \times f = 56000$ GHz, $\tau_f = 33$ ppm/°C [6-8]. As we know, the main problem of the BMN sintering is that the densification sintering temperature is approximately 1550 °C, and it takes a long time for annealing to obtain ordered BMN phase [6]. In addition, although BMN ceramic possesses superior $Q \times f$ value (56000 GHz [6]), its τ_f value (33 ppm/°C [6]) is unacceptably high, which needs to be adjusted according to practical application. In fact, the higher the sintering temperature is, the higher the cost is. So, it is necessary to reduce the densification sintering temperature to save the manufacturing cost. In order to reduce the sintering temperature and improve the microwave dielectric properties, a lot of researches have been carried out. Tian [9] added 3 mol.% $BaWO_4$ to BMN ceramics and lowered the sintering temperature to 1500°C, obtaining $\epsilon_r = 31$, $Q \times f = 82,300$ GHz and $\tau_f = 32$ ppm/°C. Shan [10] used 0.25 wt% V_2O_5 to reduce the sintering temperature of the ceramics system to 1350 °C, achieving superior microwave dielectric performance ($\epsilon_r = 31.7$, $Q \times f = 42,100$ GHz and $\tau_f = 22.7$ ppm/°C). Sun [11] reported that $Ba([Mg_{1-x}Co_x]_{1/3}Nb_{2/3})O$ ($x = 0.8$) ceramics could be well sintered at 1400 °C, and good microwave dielectric performance of $\epsilon_r = 31.7$, $Q \times f = 76,900$ GHz and $\tau_f = + 3.3$ ppm/°C were achieved. Zhang [12] added $BaSnO_3$ to decline the sintering temperature of the ceramics system to 1450°C, with excellent microwave performance of $\epsilon_r = 29.02$, $Q \times f = 90,000$ GHz and $\tau_f = 6.3$ ppm/°C. Wang [13] reported that Ba_{1-x}

$x\text{Ca}_x(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ ($x = 0.005$) ceramics could be sintered at 1500 °C, achieving good microwave performance ($\epsilon_r = 31.64$, $Q \times f = 74,421$ GHz and $\tau_f = 14.59$ ppm/°C).

In this article, we tried to sinter BMN ceramics with B_2O_3 addition. By the addition of B_2O_3 , we expected the ceramics system to lower the sintering temperature and improve the microwave dielectric performance. More significantly, the effect of B_2O_3 on phase development, microstructure and microwave dielectric performance of BMN ceramics were investigated in detail.

2. Experimental Procedures

2.1 Specimen preparation

The samples were synthesized via the conventional solid-state reaction technique. For the synthesis of $\text{Ba}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ powders, BaCO_3 , MgO , Nb_2O_5 (purity $\geq 99\%$, ChengDu Kelong Chemical Co., Ltd, Chengdu, China) were mixed and ball-milled in a nylon jar with zirconia balls and deionized water for 16 h, and after drying, these powder were calcined at 1200°C for 5 h. Then the calcined powders were re-milled for 18 h with x ($x = 1 \sim 15$) wt% B_2O_3 of $\geq 99\%$ purity. Later, with 6wt% PVA as binder, the fine powders was pressed into cylindrical samples of 16 mm $\times$ 8 mm and then sintered at 900 °C for 5 h.

2.2 Characterization

The XRD analyses were carried out using a Philips X'Pert diffractometer within the 2θ range from 15° to 85° with $\text{CuK}\alpha$ radiation. The lattice parameters were calculated by the Rietveld refinement method using Jade. The apparent densities of sintered samples were measured by the Archimedes method. The relative density was defined as relative density (%) = $D_2/D_1 \times 100\%$, where D_1 and D_2 are the X-ray theoretical density and the measured density, respectively. Microstructures of the samples were observed using scanning electron microscopy (SEM, FEI Inspect F) The cation ordering degree (S) of the specimens was

obtained by the equation [14]:
$$S = \sqrt{\frac{(I_{100}/I_{102})_{\text{obs}}}{(I_{100}/I_{102})_{\text{calc}}}}$$
, in which $(I_{100}/I_{102})_{\text{obs}}$ and $(I_{100}/I_{102})_{\text{calc}}$ are the intensity ratio of the (100) superlattice reflection to that of (102) main reflection from the observed and the calculated values of complete ordering, respectively. The dielectric characteristics at microwave frequencies were measured by the Hakki-Coleman dielectric resonator method in the TE_{011} mode using a network analyzer (HP83752A) [15]. The sintered samples used for this study had the ratio: diameter/length = 1.8-2.2. The τ_f values were determined using the equation: $\tau_f = (f_{t2} - f_{t1}) / (f_{t1} \times (t_2 - t_1))$, where f_{t1} and f_{t2} are the resonant frequencies at $t_1 = 25$ °C and $t_2 = 85$ °C, respectively.

3. Results And Discussion

Generally speaking, the densification of the ceramics is mainly dependent on the sintering temperature. The relative densities of the samples are showed as a function of sintering temperature and B_2O_3 content in Fig. 1. The samples were sintered in the range from 850 °C to 950 °C for 5 h. For all samples, the

highest relative density was always obtained at 900 °C and subsequently decreased with sintering temperature concentration. The increase of sinterability was due to the decrease of porosity and grain growth. At the same time, the reduction of sinterability was ascribed to the increase of porosity and the formation of secondary phase. Additionally, the relative density first increased to the maximum value and then dropped with increasing x . Similar results were observed in $\text{Ba}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ ceramics with BaWO_4 addition [16], where the change of grain and porosity led to the variation of relative density. All in all, it was proved that the perfect sintering temperature of the samples remained to be 900 °C with the increase of x . Therefore, all B_2O_3 -doped samples were sintered at 900 °C in this study.

BMN ceramics with various B_2O_3 contents were sintered at 900 °C for 5 h, and figure 2 displayed the XRD patterns of these samples. From the XRD patterns, we could find that the main phase of the samples was BMN phase with a complex perovskite crystal structure. For the samples with $x \leq 7$, BMN peaks without secondary phases were observed, indicating the existence of a pure BMN phase. However, When $x \geq 10$, peaks for the $\text{Ba}_2\text{B}_2\text{O}_5$ phase appeared and the diffraction peak intensity of secondary phase was increased with B_2O_3 concentration. When increasing B_2O_3 content, no secondary phase except $\text{Ba}_2\text{B}_2\text{O}_5$ phase was found in the samples. When B_2O_3 was added to $\text{Ba}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ ceramic, B_2O_3 reacted with BaO , forming the $\text{Ba}_2\text{B}_2\text{O}_5$ phase. A similar result was observed in the $\text{PbO}-\text{B}_2\text{O}_3$ -added BaTiO_3 and B_2O_3 -added $\text{Ba}_5\text{Nb}_4\text{O}_{15}$ ceramics [17-18]. However, the BaB_2O_4 phase was not observed in B_2O_3 -added BMN. As we know, the densification sintering temperature of BMN was very high (about 1550 °C), so it was difficult to sinter. However, BMN ceramics could be well sintered at 900 °C with B_2O_3 addition. Therefore, it was reasonably believed that the secondary phase containing BaO and B_2O_3 formed with B_2O_3 addition, which was beneficial to the improvement of sintering property of BMN ceramics.

The lattice parameters and unit cell volume of the BMN ceramics with x ($x = 1 - 15$) wt% B_2O_3 were calculated and the results are displayed in Fig. 3. With increasing the doping amount of B_2O_3 from 1 wt% to 15 wt%, the lattice parameters and the unit cell volume were decreased. Meanwhile, the c/a ratio first increased to the peak value and then decreased. And the samples possessed the peak value in c/a ratio at $x = 7$. The most important indicator of the hexagonal distortion caused by the cation ordering was the c/a ratio in the BMN ceramics[16, 19]. According to previous work, the increase of c/a ratio was beneficial to improve the $Q \times f$ value of the ceramic system [20]. This study revealed that the perfect c/a ratio could be achieved for the samples with appropriate B_2O_3 addition.

Figure 4 displays the SEM photographs of the BMN ceramics with varying amounts of B_2O_3 sintered at 900 °C for 5 h. We can see that BMN ceramic with $x \leq 7$ sintered at 900 °C showed many pores and incompact microstructure, which could be explained that there were not enough liquid phases to fill the residual pores. When increasing B_2O_3 content, the microstructures of the samples were improved greatly, which was caused by the formation of liquid phases. Obviously, the abundant liquid phases arising from more B_2O_3 addition could accelerate the mass transportation in the sintering process, which contributed to the densification of the systems. When $x = 7$, the sample showed the best microstructure of all

samples, suggesting that appropriate B_2O_3 addition in BMN ceramics could obtain a dense microstructure and excellent properties. However, from the SEM images of the samples with $x > 7$, a considerable number of pores were obvious. And the formation of pores could be caused by the grain growth with surplus B_2O_3 content addition.

Figure 5 presents the apparent density and dielectric constant (ϵ_r) of BMN ceramics with x ($x = 1 - 15$) wt% B_2O_3 sintered at 900 °C for 5 h. Let's first analyze the apparent density. When $x \leq 7$, the apparent density of the samples was very low firstly and increased with increasing B_2O_3 content. As shown in Figs. 4(a)–4(d), the porous microstructure formed in the samples sintered at 900 °C changed to a dense structure with B_2O_3 concentration. Therefore, the increase of apparent density with B_2O_3 concentration was explained by the variation in the microstructure. When $x \geq 10$, the apparent density began to decrease when large amounts of B_2O_3 were added. Associating with Fig. 4(e) and Fig. 4(f), the dense microstructure was developed with moderate B_2O_3 addition, but the loose microstructure with a few residual pores formed when the B_2O_3 content increased. Therefore, the variation in apparent density with B_2O_3 was very closely related to the microstructure of the samples. The dielectric properties of all samples were tested at 1 MHz. Figure 5 illustrates the dielectric constant (ϵ_r) of the samples as a function of B_2O_3 addition. As we know, the dielectric constant were mainly related to the apparent density, phase composition and microstructure [21-22]. As shown in Fig. 5, the dielectric constant (ϵ_r) increased gradually when $x = 1 \sim 7$, and then decreased when $x > 7$, showing the similar variation tendency as the apparent densities. Therefore, it could be inferred that the dielectric constant of the samples were closely related to the apparent density and located in the range of 29.3-31.6. Because of the dense microstructure, the sample with $x = 7$ sintered at 900 °C exhibited the highest dielectric constant (31.6). Meanwhile, the apparent density of the samples decreased with the surplus B_2O_3 addition, thus, the decrease of the dielectric constant could be attributed to the decrease in the density of the samples.

The correlations between the cation ordering degree (S) and the dielectric loss are shown in Fig. 6, which indicate that the cation ordering degree increase with B_2O_3 concentration, while the dielectric loss decrease, accordingly. That is, an inverse relationship existed between the cation ordering degree (S) and the dielectric loss from $x = 1$ to $x = 15$. According to the above analysis, the S value of the samples reached the maximum value at $x = 7$, and then decreased sharply with B_2O_3 concentration, which was the same with the changing tendency of c/a ratio. As reported by Tang [23], the dielectric loss was related to the cation ordering degree, the phase composition and the crystal defects. Especially, the cation ordering degree was very closely related to the dielectric loss of materials. Additional, many defects in the crystal lattice, such as lattice distortion, pores and lattice defect were also the key influence on the dielectric loss of ceramics. For the sample with $x = 7$, the dielectric loss decreased to 1.6×10^{-4} , which was attributed to the reduction of pores and the increase of cation ordering degree. Thus, the dielectric loss decreased gradually at $x \leq 7$ with B_2O_3 concentration. Moreover, combining with Fig. 2, the increase of dielectric loss was due to the formation of secondary phase $Ba_2B_2O_5$.

The $Q \times f$ values and τ_f values of BMN ceramics with x ($x = 1 - 15$) wt% B_2O_3 sintered at 900 °C for 5 h are presented in Fig. 7. When increasing B_2O_3 content, the $Q \times f$ values first increased to the maximum value of 89800 GHz with $x = 7$, and dropped thereafter. The $Q \times f$ values of the samples initially increased with B_2O_3 concentration, which was attributed to the increase in apparent density. When surplus B_2O_3 was added to the BMN ceramics, an inhomogeneous phase with secondary phase $Ba_2B_2O_5$ was developed. Therefore, the decrease of $Q \times f$ values with increasing B_2O_3 content could be ascribed to the appearance of the secondary phase $Ba_2B_2O_5$. Moreover, since the apparent density decreased with B_2O_3 concentration, the decline in $Q \times f$ values was also explained by the decrease of apparent density. As shown in Fig. 7, the variation of the temperature coefficient of resonant frequency (τ_f) was in accord with the proportion of B_2O_3 . It was obvious that the τ_f values decreased first to 3.6 ppm/°C ($x = 7$), and then increased to 15.8 ppm/°C ($x = 15$) with the increase of B_2O_3 content. The τ_f values were determined by the additive content, the secondary phase and the degree of densification [24-25]. Associating Fig. 5 with Fig. 7, the τ_f value in Fig. 7 displayed the opposite changing tendency as that of apparent density in Fig. 5. Thus, it was reasonably believed that the increase of apparent density could be the key influence on the decrease in the τ_f value when $x \leq 7$. However, the τ_f value began to increase to 15.8 ppm/°C with x further increased to 15. The variation of τ_f value showed a ring trend due to the decrease of density, which was ascribed to the surplus B_2O_3 addition.

Table 1. The microwave dielectric properties of some typical $Ba(Mg_{1/3}Nb_{2/3})O_3$ -based ceramics

Year	Ceramics composition	Sintering temperature (°C)	ϵ_r	$Q \cdot f$ (GHz)	τ_f (ppm/°C)	Reference
1983	Ba(Mg _{1/3} Nb _{2/3})O ₃	1550	32	56000	33	[6]
1998	Ba(Mg _{1/3-x} Nb _{2/3})O ₃ ($x = 0.02$)	1450	32	96000	30	[26]
2004	Ba(Mg _{1/3} Nb _{2/3})O ₃ + 3 mol.% BaWO ₄	1500	31	82300	32	[9]
2007	Ba(Mg _{1/3} Nb _{2/3})O ₃ + 0.25 wt% V ₂ O ₅	1350	31.7	42100	22.7	[10]
2013	Ba([Mg _{1-x} Co _x] _{1/3} Nb _{2/3})O ₃ ($x = 0.8$)	1400	31.7	76900	3.3	[11]
2015	(1-x)Ba(Mg _{1/3} Nb _{2/3})O ₃ -xBaSnO ₃ ($x = 0.2$)	1450	29.02	90000	6.3	[12]
2019	Ba _{1-x} Ca _x (Mg _{1/3} Nb _{2/3})O ₃ ($x = 0.005$)	1500	31.64	74421	14.59	[13]
2021	Ba(Mg _{1/3} Nb _{2/3})O ₃ + 7 wt% B ₂ O ₃	900	31.6	89800	3.6	This work

As is known to all, BMN ceramics have attracted much attention due to their excellent dielectric properties. However, the main problem of the BMN sintering is that the densification sintering temperature is approximately 1550 °C, which is a great difficulty for the application of substrate materials. In this study, the sample with $x = 7$ could be well sintered at 900 °C for 5 h and showed good microwave dielectric properties: $\epsilon_r = 31.6$, $Q \times f = 89,800$ GHz, $\tau_f = 3.6$ ppm/°C. Additionally, Table 1 presents the microwave dielectric properties of some typical Ba(Mg_{1/3}Nb_{2/3})O₃-based ceramics. As shown in Table 1, with the change of time, the evident progress has been made in improving microwave dielectric properties and sintering properties. The above results showed that B₂O₃ addition not only improved the microwave dielectric properties, but lowered the densification sintering temperature.

4 Conclusions

The BMN ceramics with x ($x = 1 \sim 15$) wt.% B₂O₃ were synthesized via the conventional solid-state ceramic route for studying the effect of B₂O₃ addition on the sintering behavior and microwave dielectric properties. The phase composition, microstructure and the microwave dielectric properties of BMN ceramics were studied as a function of B₂O₃ content. XRD test proved the solid solution BMN was the main phase and Ba₂B₂O₅ as the secondary phase. B₂O₃ worked as a sintering aid and improved the liquid phase sintering mechanism, which promoted noticeably the crystal growth and lowered the densification sintering temperature by approximately 650 °C. As increasing x from 1 to 7, the grain

growth was promoted, moreover the τ_f values and dielectric losses dropped from 28.9 ppm/°C to 3.6 ppm/°C and from 9.2×10^{-4} to 1.6×10^{-4} respectively, and the $Q \times f$ values and cation ordering degree increased from 36,200 GHz to 89,800 GHz and from 0.768 to 0.886 respectively. However, when the B_2O_3 addition was extremely excessive, with the increase of x up to $x \geq 10$, the density, $Q \times f$ value and cation ordering degree decreased greatly, while the τ_f values and dielectric loss increased remarkably. At last, BMN ceramics with 7 wt% B_2O_3 sintered at 900 °C for 5 h displayed the excellent microwave dielectric properties of $\epsilon_r = 31.6$, $Q \times f = 89,800$ GHz, $\tau_f = 3.6$ ppm/°C, which were expected to be used in microwave oscillator, mixer, detector and other devices.

Declarations

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Figures

Figure 1

Figure 2

XRD patterns of BMN ceramics with x (x = 1 - 15) wt% B2O3 sintered at 900 °C for 5 h

Figure 3

Lattice parameters and unit cell volume of the BMN ceramics with x (x=1 - 15) wt% B₂O₃ sintered at 900°C for 5 h

Figure 4

SEM photographs of BMN ceramics doped by x wt% B₂O₃ sintered at 900 °C for 5 h with (a) x = 1, (b) x = 3, (c) x = 5, (d) x = 7, (e) x = 10, (f) x = 15

Figure 5

Apparent density and dielectric constant of BMN ceramics with x (x = 1 - 15) wt% B₂O₃ sintered at 900 °C for 5 h

Figure 6

The relationships between the cation ordering degree (S) and the dielectric loss with B2O3 concentration

Figure 7

$Q \times f$ values and τ_f values of BMN ceramics with x (x = 1 - 15) wt% B₂O₃ sintered at 900 °C for 5 h