

Phase evolution and microwave dielectric properties of $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La , $x \leq 0.3$) ceramics



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ABSTRACT

$(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La) ceramics were prepared by conventional solid state reaction method. Solid solutions crystallizing in a monoclinic structure with a space group $P21/c$ were obtained in the $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ($x \leq 0.3$) ceramics. For the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ ceramics, tetragonal Nd_2MoO_6 phase with a space group $I4_1/acd$ was detected when x value reached 0.2 besides the Bi_2MoO_6 solid solution. Temperature stability and quality factor Q_f value at microwave frequencies were improved by appropriate substitution for Bi^{3+} by Nd^{3+} and/or La^{3+} in Bi_2MoO_6 system. When $x=0.2$, for both $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics, Q_f values reached maximum about 30,200 GHz and 32,900 GHz, respectively. TCF value shifted towards zero with substitution of Bi^{3+} by Nd^{3+} and/or La^{3+} . When $x=0.3$, TCF values of $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics were -70 and -85 ppm/ $^\circ\text{C}$, respectively. Relation between crystal structure, microstructure and microwave dielectric properties were discussed.

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1. Introduction

The development of high-speed and high-frequency digital devices, which are capable of transferring large amounts of information in a short time is one of the main strategic drivers of the consumer electronics market. Meanwhile, demands for miniaturization and performance enhancement of microwave devices have increased with the development of the modern communication system. Thus, there have been increasing investigations on microwave dielectric material with a high-quality factor (Q_f value) to decrease the noise and insertion loss selectivity and a near-zero temperature coefficient of resonant frequency (TCF value) for achieving excellent thermal stable microwave devices [1]. At the same time, different microwave devices need materials with different permittivities. For example, as microwave substrates, the use of low relative permittivity (ϵ_r) materials lower than that of alumina ($\epsilon_r=9.8$) can reduce the signal propagation delay sufficiently to offer immense potential for high-frequency applications

[2,3], while a large dielectric constant (ϵ_r) is also required for the microwave dielectric material to miniaturize the microwave resonators because the size of the device is inversely proportional to $\epsilon_r^{1/2}$ [4].

A great number of excellent microwave dielectric materials were developed, such as $\text{Sr}_{2-x}\text{Ca}_x\text{Al}_2\text{SiO}_7$ [5], $\text{ZnO-Nb}_2\text{O}_5$ [6], $\text{Bi}(\text{Nb}, \text{Ta}, \text{Sb})\text{O}_4$ [7–9], $\text{Li}_2\text{O-Nb}_2\text{O}_5\text{-TiO}_2$ [10], $(\text{Zr}, \text{Sn})\text{TiO}_4$ [11], $\text{BaO-TiO}_2\text{-Nb}_2\text{O}_5$ [12], and the $(\text{A}_1\text{A}_2)(\text{B}_1\text{B}_2)\text{O}_3$ complex perovskite system [13]. While most of them need high sintering temperatures above 1200 $^\circ\text{C}$. Nowadays, investigations on new compounds with intrinsically lower sintering temperatures have attracted much more attention [14–17]. In the $\text{Bi}_2\text{O}_3\text{-MoO}_3$ system, six compounds with pure phase were reported and all of them were glass-free low-firing microwave dielectric materials. [18] The Bi_2MoO_6 ceramic with a monoclinic structure was reported to possess a low sintering temperature (750 $^\circ\text{C}$), a high Q_f value (16,700 GHz), and a negative TCF value (-114 ppm/ $^\circ\text{C}$). TCF value could be tailored to near zero by ceramic composites, such as $\text{Bi}_2\text{MoO}_6\text{-TiO}_2$ composites, and a temperature stable low-firing microwave dielectric material was obtained in our previous work [19]. Substitution is another good way to modify TCF values of microwave dielectrics [20–23]. Rare earth cations, such as La^{3+} and Nd^{3+} , have the same chemical valence and similar ionic radii to Bi^{3+} , so solid solutions

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might be formed in the $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La) system. According to literatures' reports, both Nd_2MoO_6 and La_2MoO_6 can crystallize as tetragonal crystal structure with a $I4_1/acd$ space group [24,25]. The La_2MoO_6 is a kind of optical materials which can be activated with the trivalent rare earth ions [26], and the Nd_2MoO_6 ceramic, which can be densified well at 1350°C , is a microwave dielectric material with a permittivity (ϵ_r) of 13.8, a Qf value of 66,400 GHz, and a TCF value of $-53\text{ ppm}/^\circ\text{C}$ [27]. In the present work, $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La , $x \leq 0.3$) ceramics were prepared by conventional solid state reaction method, and sintering behavior, phase evolution, microstructure and microwave dielectric properties were investigated in detail.

2. Experimental procedure

Proportionate amounts of reagent-grade Bi_2O_3 (> 99%, Shu-Du Powders Co. Ltd., Chengdu, China), La_2O_3 (> 99.99%, Sinopharm Chemical Reagent Co., Ltd, Shanghai, China), Nd_2O_3 (> 99.99%, Sinopharm Chemical Reagent Co., Ltd, Shanghai, China), and MoO_3 (> 99%, Fuchen Chemical Reagents, Tianjin, China) were prepared according to the stoichiometric compound formulation of $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La , $x=0.0, 0.1, 0.2$, and 0.3). Powders were mixed and milled for 4.5 h using a planetary mill (Nanjing Machine Factory, Nanjing, China). The mixtures were dried and calcined at $550\text{--}650^\circ\text{C}$ for 5 h. The calcined powders were ball milled again for 5 h, and then the dried powders were pressed into cylinders (10 mm in diameter and 4–5 mm in height) in a steel die with 5 wt% PVA binder addition under a uniaxial pressure of 20 MPa. Samples were sintered at temperatures from 830 to 1030°C for 2 h.

Densities of the sintered specimens were measured by Archimedes method. To examine phase structure, sintered ceramics were crushed into powders. Room temperature X-ray diffraction (XRD) measurement was performed by using a XRD with $\text{Cu K}\alpha$ radiation (Rigaku D/MAX-2400 X-ray diffractometry, Tokyo, Japan). The ss-fired samples were observed by using scanning electron microscopy (SEM) (JSM-6460, JEOL, Tokyo, Japan) to examine grain morphology. Dielectric properties at microwave frequencies were measured according to the TE_{018} dielectric resonator method with a network analyzer (HP 8720 Network Analyzer, Hewlett-Packard) and a temperature chamber (Delta 9023, Delta Design, Poway, CA). Temperature coefficient of resonant frequency TCF was calculated with the following formula:

$$\text{TCF} = \frac{f_T - f_{T_0}}{f_{T_0} \times (T - T_0)} \times 10^6 \text{ ppm}/^\circ\text{C}, \quad (1)$$

where f_T and f_{T_0} are the TE_{018} resonant frequencies at temperature T and T_0 (room temperature), respectively.

3. Results and discussion

Fig. 1 presents X-ray diffraction patterns of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics. Almost all the diffraction peaks of the samples (except $(\text{Bi}_{0.7}\text{Nd}_{0.3})_2\text{MoO}_6$ ceramics) can be indexed on a monoclinic cell, which was similar with parent Bi_2MoO_6 . It indicates that solid solutions, which crystallized as single monoclinic phase structure with space group $P21/c$, were obtained in $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ($x \leq 0.3$) and $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ ($x \leq 0.2$) ceramics. The diffraction peaks (080) and (002) separated from each other with the substitution of Bi^{3+} by Nd^{3+} and/or La^{3+} , because the cell parameters varied with substitution. In the $(\text{Bi}_{0.7}\text{Nd}_{0.3})_2\text{MoO}_6$ ceramics, a small amount of tetragonal Nd_2MoO_6 phase with space group $I4_1/acd$ was detected besides the Bi_2MoO_6 solid solution phase. As we know, both pure Nd_2MoO_6 and La_2MoO_6 crystallize as tetragonal crystal structure with $I4_1/acd$ space group [24,25], which is quite different from that of Bi_2MoO_6 , so solid solutions couldn't be obtained in the whole range of $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La). According to Shannon's result, ionic radius of La^{3+} (1.160 Å) is much closer to that of Bi^{3+} (1.170 Å), and the ionic radius of Nd^{3+} (1.109 Å) is much smaller [28]. Thus, the solid solubility of $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ is a bit higher than that of $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$, as the case here.

Fig. 2 shows the bulk densities and relative densities of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics as a function of sintering temperature. Bulk densities of $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La) ceramics decreased with the contents of Nd^{3+} and/or La^{3+} due to their smaller atomic weights than that of Bi. For parent Bi_2MoO_6 ceramic, its bulk density reached maximum value and relative density kept above 96% when sintering temperature $\geq 850^\circ\text{C}$. The sintering temperature increased with the substitution of Bi^{3+} by Nd^{3+} and/or La^{3+} . For both $(\text{Bi}_{0.7}\text{Nd}_{0.3})_2\text{MoO}_6$ and $(\text{Bi}_{0.7}\text{La}_{0.3})_2\text{MoO}_6$ ceramics, they could not be densified well unless the sintering temperatures were higher than 970°C . Meanwhile, the bulk densities and relative densities would decrease when sintering temperatures went further, for all the $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La) ceramics, which might result from the Bi volatilization at higher sintering temperatures. So the sintering

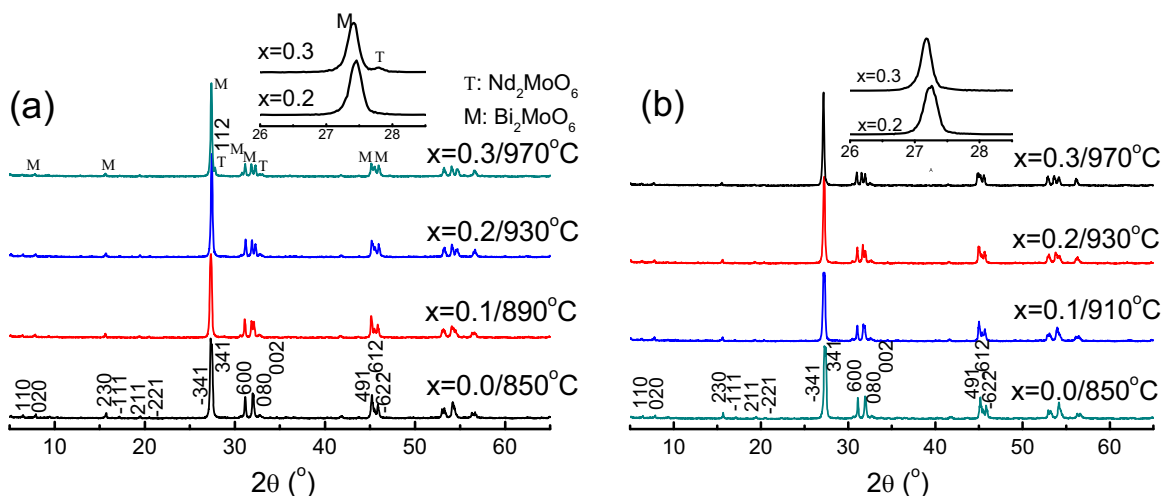


Fig. 1. X-ray diffraction patterns of (a) $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and (b) $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics.

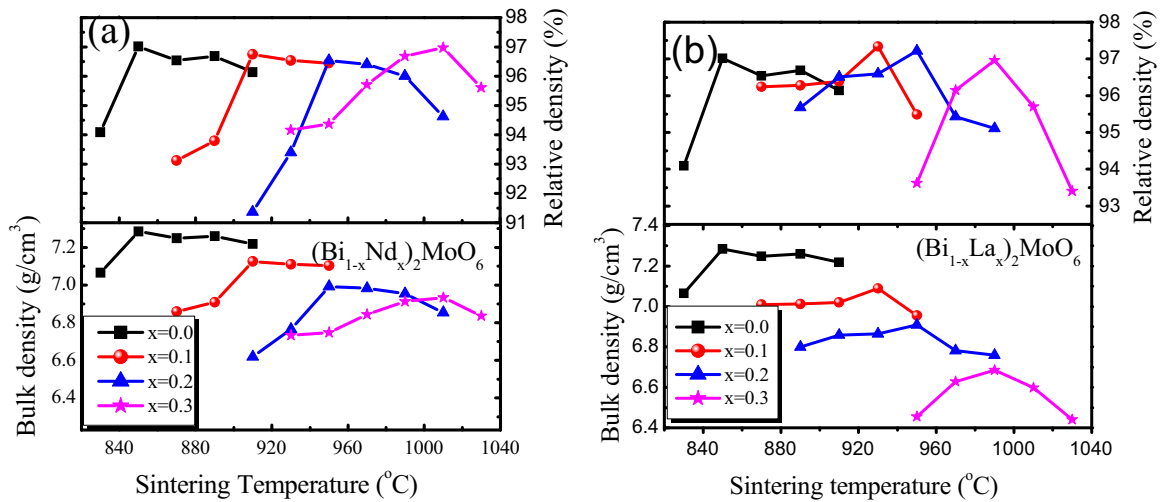


Fig. 2. Bulk densities and relative densities of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ (a) and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ (b) ceramics as a function of sintering temperature.

temperatures could not be too high to densify $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ ($\text{Ln}=\text{Nd}$ and La) ceramics.

Fig. 3 presents SEM images of as-fired surfaces of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics. All the ceramics had been well densified with little pores inside. The grains were uniform and there was no secondary phase grains in all $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ($x \leq 0.3$) ceramics and $(\text{Bi}_{0.9}\text{Nd}_{0.1})_2\text{MoO}_6$ ceramic. In the $(\text{Bi}_{0.8}\text{Nd}_{0.2})_2\text{MoO}_6$ and $(\text{Bi}_{0.7}\text{Nd}_{0.3})_2\text{MoO}_6$ ceramics, a small amount of secondary phase grains with octahedral shape were detected in the grain boundaries, which might be tetragonal Nd_2MoO_6 grains according to the XRD results above. It means that the secondary phase appeared in $(\text{Bi}_{0.8}\text{Nd}_{0.2})_2\text{MoO}_6$ ceramic, but the amount was too small to be detected by XRD experimental. On the other hand, the secondary phase grains gathered to the surface

of the sample, so they can only be observed on the surface from SEM image.

Microwave dielectric properties of $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics were shown in Fig. 4. Permittivity decreased gradually with the substitution for Bi^{3+} by Nd^{3+} and/or La^{3+} because of their smaller dielectric polarizabilities of Nd^{3+} (5.01 \AA^3) and La^{3+} (6.07 \AA^3) than Bi^{3+} (6.12 \AA^3) [29]. Permittivity decreased from 31.5 of pure Bi_2MoO_6 ceramic to 22.6 at $x=0.3$ for $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ ceramics and to 20.7 at $x=0.3$ for $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics, respectively. Permittivity of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ ceramic was a little larger than that of $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramic with the same x value, which might be attributed to the influence of the secondary phase Nd_2MoO_6 .

It is shown in Fig. 4(b) that Qf value first increased remarkably

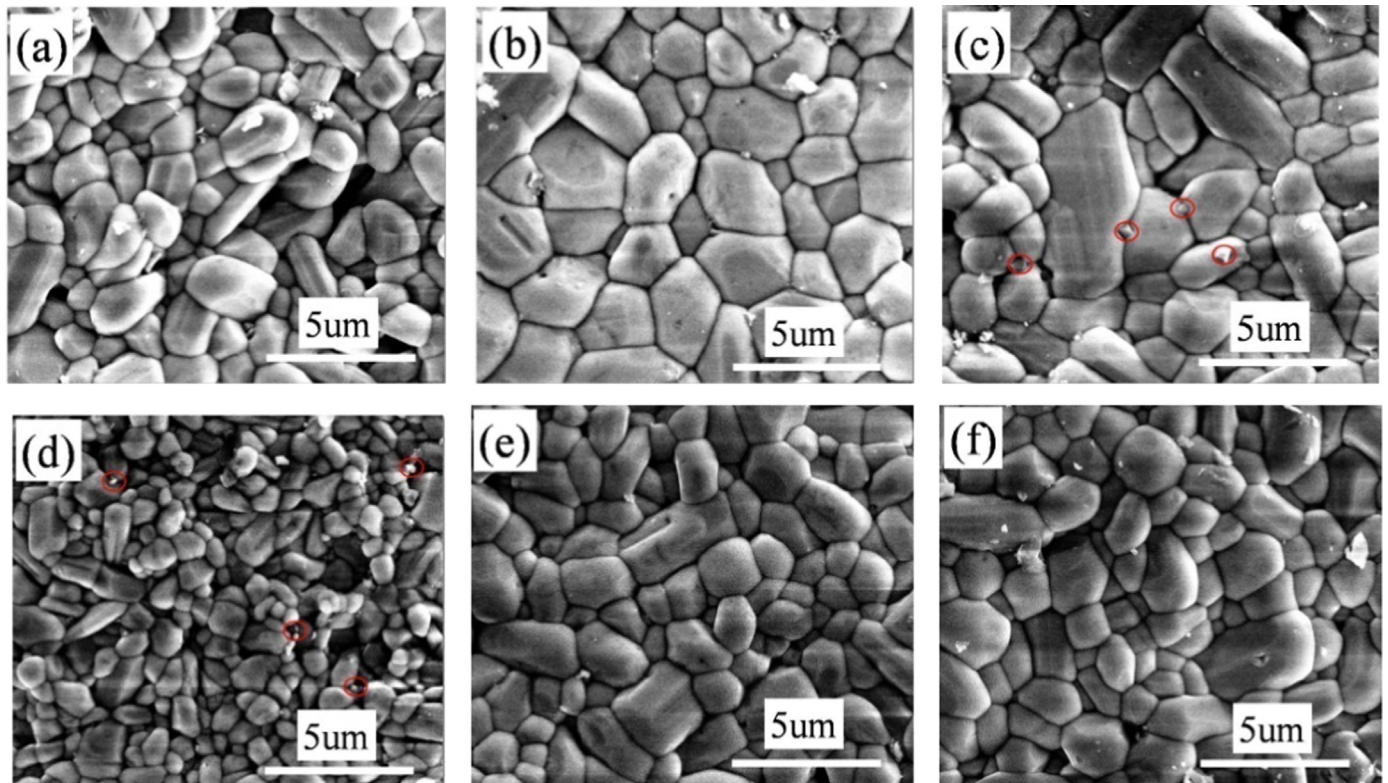


Fig. 3. SEM images of as-fired surface of (a) Bi_2MoO_6 sintered at 850°C , (b) $(\text{Bi}_{0.9}\text{Nd}_{0.1})_2\text{MoO}_6$ sintered at 910°C , (c) $(\text{Bi}_{0.8}\text{Nd}_{0.2})_2\text{MoO}_6$ sintered at 930°C , (d) $(\text{Bi}_{0.7}\text{Nd}_{0.3})_2\text{MoO}_6$ sintered at 970°C , (e) $(\text{Bi}_{0.9}\text{La}_{0.1})_2\text{MoO}_6$ sintered at 890°C , and (f) $(\text{Bi}_{0.8}\text{La}_{0.2})_2\text{MoO}_6$ sintered at 930°C .

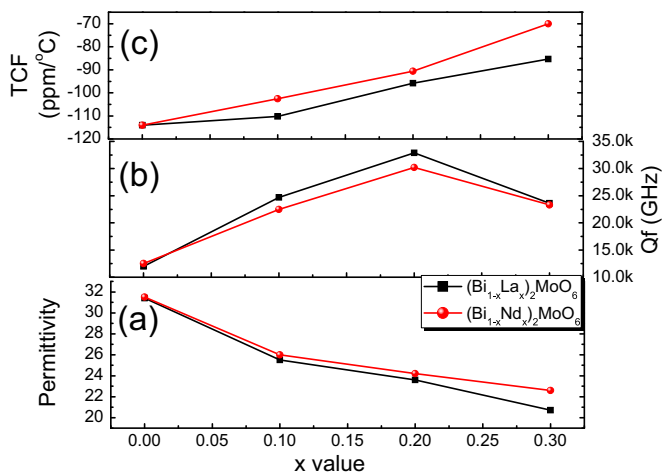


Fig. 4. Microwave dielectric properties of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics: (a) permittivity; (b) Qf value; (c) TCF value.

Table 1

Sintering Temperatures and microwave dielectric properties of several typical microwave dielectrics.

Sample	S.T (°C)	Permittivity	Qf value (GHz)	TCF (ppm/°C)	Reference
SmAlO ₃	1650	20.4	65,000	-74	[31]
LaAlO ₃ +0.25 wt% CuO	1460	20.7	48,000	-80	[33]
BaTe ₂ O ₆	650	21	50,300	-51	[32]
NdAlO ₃	1650	22.3	58,000	-33	[31]
LiNb ₃ O ₈	1075	24	58,000	-96	[30]
$(\text{Bi}_{0.8}\text{La}_{0.2})_2\text{MoO}_6$	930	23.6	32,900	-96	This work
$(\text{Bi}_{0.8}\text{Nd}_{0.2})_2\text{MoO}_6$	930	24.2	30,200	-91	This work
Li ₂ ZnTi ₃ O ₈	1075	25.6	72,000	-11.2	[34,35]
Li ₂ MgTi ₃ O ₈	1075	27.2	42,000	+3.2	[34,35]

S.T.: Sintering Temperature.

and then decreased slightly with the increase of substitution amount. Maximum Qf values were achieved at $x=0.2$, and the maximum Qf values were 30,200 GHz in $(\text{Bi}_{0.8}\text{Nd}_{0.2})_2\text{MoO}_6$ ceramics and 32,900 GHz in $(\text{Bi}_{0.8}\text{La}_{0.2})_2\text{MoO}_6$ ceramics, respectively. Decrease in Qf value in $x=0.3$ sample might be caused by the escape of Bi at high sintering temperature. TCF value of the $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ (Ln=Nd and La) ceramics shifted towards zero with the substitution for Bi^{3+} by Nd^{3+} and/or La^{3+} , as shown in Fig. 4(c). When $x=0.3$, the TCF values of $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics were -70 and -85 ppm/°C, respectively. It means that both temperature stability and Qf value can be improved by appropriate substitution of Bi^{3+} by Nd^{3+} and/or La^{3+} in Bi_2MoO_6 system. Table 1 presents the sintering temperatures and microwave dielectric properties of several typical microwave dielectrics with permittivity 20–24 [30–33]. Comparing with these dielectrics, the $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ (Ln=Nd and La) ceramics were nontoxic and processed a lower sintering temperature, which makes it possible for low temperature co-fired ceramics technology.

4. Conclusions

Microwave dielectric ceramics, $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$, were prepared by conventional solid state reaction method. Solid solutions with monoclinic structure were obtained for $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics when $x \leq 0.3$. While in the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ ceramics, tetragonal Nd_2MoO_6 phase with space group $I4_1/acd$ appeared when $x \geq 0.2$ besides the Bi_2MoO_6 solid

solution phase. With the increase of substitution amount, sintering temperature of the $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics increased from 850 °C to about 970 °C, and permittivity decreased from 31.5 to 22.6 and 20.7, respectively. Both temperature stability and Qf value were improved. Maximum Qf values were 30,200 GHz in $(\text{Bi}_{0.8}\text{Nd}_{0.2})_2\text{MoO}_6$ ceramics and 32,900 GHz in $(\text{Bi}_{0.8}\text{La}_{0.2})_2\text{MoO}_6$ ceramics, respectively. TCF value of $(\text{Bi}_{1-x}\text{Ln}_x)_2\text{MoO}_6$ (Ln=Nd and La) ceramics shifted towards zero with the substitution of Bi^{3+} by Nd^{3+} and/or La^{3+} . When $x=0.3$, TCF values of $(\text{Bi}_{1-x}\text{Nd}_x)_2\text{MoO}_6$ and $(\text{Bi}_{1-x}\text{La}_x)_2\text{MoO}_6$ ceramics were -70 and -85 ppm/°C, respectively.

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References

- [1] M.T. Sebastian, Dielectric Materials for Wireless Communication, Elsevier, Oxford, 2008.
- [2] B.D. Hutton, K. Landskron, W.J. Hunks, M.R. Bennett, D. Shukaris, D.D. Perovic, G.A. Ozin, Materials chemistry for low-k materials, Mater. Today 9 (2006) 22–31.
- [3] M.T. Sebastian, R. Ubic, H. Jantunen, Low-loss dielectric ceramic materials and their properties, Int. Mater. Rev. 60 (2015) 392–412.
- [4] D. Zhou, D. Guo, W.B. Li, L.X. Pang, X. Yao, D.W. Wang, I.M. Reaney, Novel temperature stable high- ϵ_r microwave dielectrics in the Bi_2O_3 - TiO_2 - V_2O_5 system, J. Mater. Chem. C 4 (2016) 5357–5362.
- [5] K.M. Manu, C. Karthik, R. Ubic, M.T. Sebastian, Effect of Ca^{2+} substitution on the structure, microwave dielectric properties of $\text{Sr}_2\text{Al}_2\text{SiO}_7$ ceramics, J. Am. Ceram. Soc. 96 (2013) 342–384.
- [6] O.A. Shlyakhtin, Y.J. Oh, Low temperature sintering of $\text{Zn}_3\text{Nb}_2\text{O}_8$ ceramics from fine powders, J. Am. Ceram. Soc. 89 (2006) 3366–3372.
- [7] N. Wang, M.Y. Zhao, Z.W. Yin, Effects of Ta_2O_5 on microwave dielectric properties of BiNbO_4 ceramics, Mater. Sci. Eng. B 99 (2003) 238–242.
- [8] C.L. Huang, M.H. Weng, The microwave dielectric properties and the microstructures of $\text{Bi}(\text{Nb}, \text{Ta})\text{O}_4$ ceramics, Jpn. J. Appl. Phys. 38 (Part 1) (1999) 5949–5952.
- [9] D. Zhou, H. Wang, X. Yao, L.X. Pang, Dielectric behavior and cofiring with silver of monoclinic BiSbO_4 ceramic, J. Am. Ceram. Soc. 91 (2008) 1380–1383.
- [10] A.Y. Borisevich, P.K. Davies, Crystalline structure and dielectric properties of $\text{Li}_{1+x}\text{Nb}_{1-x-3y}\text{Ti}_{x+4y}\text{O}_3$ M-phase solid solutions, J. Am. Ceram. Soc. 85 (2002) 573–578.
- [11] L.X. Pang, D. Zhou, Y.H. Chen, H. Wang, Structural and microwave dielectric behavior of $(\text{Li}_{1/4}\text{Nb}_{3/4})$ substituted $\text{Zr}_x\text{Sn}_y\text{Ti}_z\text{O}_4$ ($x+y+z=2$) system, Mater. Chem. Phys. 125 (2011) 641–645.
- [12] I.S. Cho, J.R. Kim, D.W. Kim, D.W. Kim, K.S. Hong, Microwave dielectric properties and far-infrared spectroscopic analysis of $\text{Ba}_{5+n}\text{Ti}_n\text{Nb}_4\text{O}_{15+3n}$ ($0.3 < n < 1.2$) ceramics, J. Eur. Ceram. Soc. 27 (2007) 3081–3086.
- [13] X.M. Chen, D. Liu, R.Z. Hou, X. Hu, X.Q. Liu, Microstructures and microwave dielectric characteristics of $\text{Ca}(\text{Zn}_{1/3}\text{Nb}_{2/3})\text{O}_3$ complex perovskite ceramics, J. Am. Ceram. Soc. 87 (2004) 2208–2212.
- [14] M. Udovic, M. Valant, D. Suvorov, Phase formation and dielectric characterization of the Bi_2O_3 - TeO_2 system prepared in an oxygen atmosphere, J. Am. Ceram. Soc. 87 (2004) 591–597.
- [15] A. Feteira, D.C. Sinclair, Microwave dielectric properties of low firing temperature $\text{Bi}_2\text{W}_2\text{O}_9$ ceramics, J. Am. Ceram. Soc. 91 (2008) 1338–1341.
- [16] D. Zhou, L.X. Pang, H.D. Xie, J. Guo, B. He, Z.M. Qi, T. Shao, X. Yao, C.A. Randall, Crystal structure and microwave dielectric properties of a novel ultra-low temperature fired $(\text{AgBi})_0.5\text{WO}_4$ ceramic, Eur. J. Inorg. Chem. 2 (2014) 296–301.
- [17] D. Zhou, W.B. Li, L.X. Pang, J. Guo, Z.M. Qi, T. Shao, Z.X. Yue, X. Yao, Sintering behavior and dielectric properties of ultra-low temperature fired silver molybdate ceramics, J. Am. Ceram. Soc. 97 (2014) 3597–3601.
- [18] D. Zhou, H. Wang, L.X. Pang, C.A. Randall, X. Yao, Bi_2O_3 - MoO_3 binary system: an alternative ultralow sintering temperature microwave dielectric, J. Am. Ceram. Soc. 92 (2009) 2242–2246.
- [19] L.X. Pang, H. Wang, D. Zhou, X. Yao, A new temperature stable microwave dielectric with low-firing temperature in Bi_2MoO_6 - TiO_2 system, J. Alloy. Compd. 493 (2010) 626–929.
- [20] D. Zhou, L.X. Pang, J. Guo, Z.M. Qi, T. Shao, Q.P. Wang, H.D. Xie, X. Yao, C.A. Randall, Influence of Ce substitution for Bi in BiVO_4 and the impact on the phase evolution and microwave dielectric properties, Inorg. Chem. 53 (2014) 1048–1055.

- [21] P. Mohanty, S. Keshri, M.K. Sinha, V.R. Gupta, Study on microwave dielectric properties of corundum type $(\text{Mg}_{1-x}\text{Co}_x)_4\text{Ta}_2\text{O}_9$ ($x=0-0.6$) ceramics for designing a microwave low pass filter, *Ceram. Int.* 42 (2016) 5911–5920.
- [22] L.X. Pang, W.G. Liu, D. Zhou, Temperature stable high K microwave dielectric ceramics of Bi_3NbO_7 doped by V_2O_5 , *Ceram. Int.* 41 (2015) 5182–5185.
- [23] D. Zhou, J. Guo, X. Yao, L.X. Pang, Z.M. Qi, T. Shao, Phase evolution and microwave dielectric properties of $(\text{Li}_{0.5}\text{Bi}_{0.5})(\text{W}_{1-x}\text{Mo}_x)\text{O}_4$ ($0.00 \leq x \leq 1.0$) ceramics with ultra-low sintering temperatures, *Funct. Mater. Lett.* 4 (2012) 1250042.
- [24] V.A. Efremov, A.V. Tyulin, V.K. Trunov, Actual structure of tetragonal $\text{Ln}_2\text{O}_2\text{MoO}_4$ and factors, determining the forming structure of the coordination polyhedral, *Koord. Khim.* 13 (1987) 1276–1282.
- [25] J.S. Xue, R.M. Antonio, L. Soderholm, Polymorphs of Ln_2MoO_6 : a neutron diffraction investigation of the crystal structures of La_2MoO_6 and Tb_2MoO_6 , *Chem. Mater.* 7 (1995) 333–340.
- [26] F. Meng, X. Zhang, H. Li, J.S. Hyo, Synthesis and spectral characteristics of La_2MoO_6 : Ln^{3+} ($\text{Ln}=\text{Eu}, \text{Sm}, \text{Dy}, \text{Pr}, \text{Tb}$) polycrystals, *J. Rare Earths* 30 (2012) 866–870.
- [27] Y.C. Chen, M.Z. Weng, Improving quality factor of Nd_2MoO_6 ceramics by removing moisture content, *J. Mater. Sci.: Mater. Electron.* 26 (2015) 3502–3505.
- [28] R.D. Shannon, Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides, *Acta Cryst.* A32 (1976) 751–767.
- [29] R.D. Shannon, Dielectric polarizabilities of ions in oxides and fluorides, *J. Appl. Phys.* 73 (1993) 348–366.
- [30] S.O. Yoon, J.H. Yoon, S.H. Shim, Y.K. Pyon, Microwave dielectric properties of LiNb_3O_8 ceramics with TiO_2 additions, *J. Eur. Ceram. Soc.* 26 (2006) 2031–2034.
- [31] S.Y. Cho, I.T. Kim, K.S. Hong, Microwave dielectric properties and applications of rare earth aluminates, *J. Mater. Res.* 14 (1999) 114–119.
- [32] D.W. Kwon, M.T. Lanagan, T.R. Shrout, Microwave dielectric properties of BaO-TeO_2 binary compounds, *Mater. Lett.* 61 (2007) 1827–1831.
- [33] C.S. Hsu, C.L. Huang, Effect of CuO additive on sintering and microwave dielectric behavior of LaAlO_3 ceramics, *Mater. Res. Bull.* 36 (2001) 1939–1947.
- [34] S. George, M.T. Sebastian, Low-temperature sintering and microwave dielectric properties of $\text{Li}_2\text{ATi}_3\text{O}_8$ ($\text{A}=\text{Mg}, \text{Zn}$) ceramics, *Int. J. Appl. Ceram. Technol.* 8 (2011) 1400–1407.
- [35] S. George, M.T. Sebastian, Synthesis and microwave dielectric properties of novel temperature stable high Q, $\text{Li}_2\text{ATi}_3\text{O}_8$ ($\text{A}=\text{Mg}, \text{Zn}$) ceramics, *J. Am. Ceram. Soc.* 93 (2010) 2164–2166.