Title: Temperature Stable $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ Microwave Dielectric Ceramics with Ultra-Low Sintering Temperatures

Authors: Xian Xue, Xiaofeng Yuan, Rong Ma, Qibin Yuan, Hong Wang

PII: S0955-2219(19)30349-8
DOI: https://doi.org/10.1016/j.jeurceramsoc.2019.05.029
Reference: JECS 12516

To appear in: *Journal of the European Ceramic Society*

Received date: 13 October 2018
Revised date: 14 May 2019
Accepted date: 18 May 2019

Please cite this article as: Xue X, Yuan X, Ma R, Yuan Q, Wang H, Temperature Stable $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ Microwave Dielectric Ceramics with Ultra-Low Sintering Temperatures, *Journal of the European Ceramic Society* (2019), https://doi.org/10.1016/j.jeurceramsoc.2019.05.029

Temperature Stable $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ Microwave Dielectric Ceramics with Ultra-Low Sintering Temperatures

Xian Xue[1], Xiaofeng Yuan[1], Rong Ma[1], Qibin Yuan[1], Hong Wang[1, 2, *]

[1]*School of Electronic and Information Engineering & State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China*

[2]*Department of Materials Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, China*

**Corresponding author. E-mail: wangh6@sustc.edu.cn (H. Wang)*

Abstract

In this study, the novel temperature-stable (1-*x*)Ag_2MoO_4-*x*$Ag_{0.5}Bi_{0.5}MoO_4$ microwave dielectric ceramics were prepared by a modified solid-state reaction method. The phase composition, microstructures and microwave dielectric properties of the (1-*x*)Ag_2MoO_4-*x*$Ag_{0.5}Bi_{0.5}MoO_4$ ceramics were investigated. All the compounds can be sintered well at ultra-low temperatures (< 540 °C). The XRD and SEM analysis indicate that the Ag_2MoO_4 and the $Ag_{0.5}Bi_{0.5}MoO_4$ can coexist with each other. When x = 0.65, the ceramics exhibit the best microwave dielectric properties with a relative permittivity of 23.9, a Q × f value of 16200 GHz (at 7.3 GHz) and a near-zero TCF value of -2.4 ppm/°C at 520 °C. The results indicate that temperature-stable (1-*x*)Ag_2MoO_4-*x*$Ag_{0.5}Bi_{0.5}MoO_4$ ceramics are promising candidates for low temperature co-fired ceramics (LTCC) applications.

Keywords：Dielectric properties; ultra-low sintering; near-zero TCF

1. Introduction

In recent years, with the rapid development of wireless communication, low-temperature co-fired ceramics (LTCC) technology with excellent electrical, mechanical, thermal and technological characteristics, has become the main-stream technology for passive integration. This technique greatly improves the reliability and promotes miniaturization of the microwave devices. To meet the requirement of LTCC technology, the microwave dielectric ceramics with a suitable relative permittivity, a high Q × f value, a near zero temperature coefficient of the resonant frequency (TCF value) and an ultra-low sintering temperature are needed [1-3]. Considering the industrial cost and the stability of the device to temperature, lowering sintering temperature and adjusting temperature coefficient of the ceramics to near zero are always the research focuses in the related fields. Currently, many ULTCC materials with sintering temperature lower than 700°C were reported [4]. However, most

of them have negative TCF values, which limit their applications. Among all the reported low-loss microwave dielectric ceramic materials, there are only three types of temperature-stable ceramics with sintering temperature lower than 600 °C. The detail information of them is listed in Table 2. Thus, the temperature-stable ceramics with ultra-low sintering temperature are still in urgent need for different applications.

Generally, there are two effective approaches to adjust the TCF value, which are solid solution method and compositing two materials with opposite TCF values. The first one requires that the radius and the electronegativity of the substituted ions are close. Therefore, it is difficult to find a suitable ion to regulate the TCF value. The second method, which is not limited by the materials' structures, is widely applied to adjust the TCF value [5-10]. Recently, the Ag_2O-MoO_3 binary system with ultra-low sintering temperature and good microwave dielectric properties was reported. The Ag_2MoO_4 ceramics with a relative permittivity of 8.1, a Q × f value of 17000 GHz and a temperature coefficient of resonance frequency about -133 ppm/°C could be densified well at 450 °C [11]. However, the negative TCF value limits its LTCC application. On the contrary, the $Ag_{0.5}Bi_{0.5}MoO_4$ ceramics sintered at 690 °C have positive TCF value of approximately +57 ppm/°C, a relative permittivity of 30.4 and a Q × f = 12600 GHz [12]. By the second method, it is possible to compound these two phases to get a near zero TCF value. To improve the density and avoid the side effect of the residual PVA, a modified solid-state reaction method was also applied, which was proposed in our previous study [13]. Compared with the traditional solid-state reaction method, this method with isostatic pressing technology and the substituting of PVA with ethanol has been proved to be effective in improving the density and dielectric properties of ceramics [13-14]. All the ceramic composites in this paper were prepared by this method.

In this paper, these two materials with close sintering temperatures were selected and the novel temperature-stable $(1-x)Ag_2MoO_4$-$xAg_{0.5}Bi_{0.5}MoO_4$ ceramics sintered at ultra-low temperatures have been obtained. Their phase compositions, microstructures, and microwave dielectric properties were also systematically studied. By tuning the chemical constituents and sintering temperature, the TCF value of this two-phase composite could be successfully adjusted to near zero.

2. Experimental procedure

The $(1-x)Ag_2MoO_4$-$xAg_{0.5}Bi_{0.5}MoO_4$ ($0.00 \leq x \leq 1.00$) composite ceramics with ultra-low sintering temperature were synthesized by a modified solid-state reaction method. The reagent-grade raw materials Ag_2O (>99.7 %; Tianjin Guangfu Fine Chemical Research Institution, Tianjin, China),

MoO_3 (>99.5 %; Yutong Chemical Reagents, Tianjin, China) and Bi_2O_3 (>99 %; Sinopharm Chemical Reagent Co. Ltd., Shanghai, China) were weighted and mixed according to the corresponding stoichiometric ratios. Then the mixtures were milled for 4 h in ethanol medium by a planetary mill (Nanjing Machine Factory, Nanjing, China) at 150 rpm with ZrO_2 balls. The dried powders were then calcined at 420 °C for 6 h and cooled down in the furnace. After being re-milled for 4h, the dried powders were sprayed with ethanol to be moist. Then the powders were pressed into cylindrical disks (about 10 mm in diameter and 5 mm in thickness) at a uniaxial press of 150 MPa. Then, all the cylinders were pressed again at 200 MPa for 3 min via a cold isostatic press (CIP AIP3-12-60C, American Institute of Physics, MD, America). All the green presses were heated at 120 °C for 2 h to remove the ethanol then sintered at 440-540 °C for 6h with a heating rate of 2 °C/min.

The crystalline structures of samples were characterized at room temperature by a X-ray diffractometer (SHIMADZU XRD-7000, Tokyo, Japan) with Cu-Ka radiation of 1.54 Å. To observe the microstructures, the scanning electron microscope (SEM) (FEI Quanta FEG 250, FEI, Hillsboro, Oregon, USA) was used. The sintered ceramics were polished and thermally etched at different temperatures for 25 minutes (x = 0.2, etched at 430°C; x = 0.4, 0.6, etched at 440°C; x = 0.65, 0.7, 0.8, etched at 450°C). The bulk densities of sintered samples were measured by Archimedes method. The network analyzer (8720ES, Agilent, Palo Alto, CA) and a temperature chamber were used to measure the dielectric behaviors of the sintered ceramics according to the TE01δ shielded cavity method. And the TCF (τ_f) values of those ceramics were calculated by the following formula:

$$\tau_f = (f_{85} - f_{25})/(60 \times f_{25}) \times 10^6 \text{ (ppm/°C)} \quad (1)$$

where, f_{25} and f_{85} are the $TE_{01\delta}$ resonant frequencies at 25 °C and 85 °C, respectively.

3. Results and discussion

This series of (1-x)Ag_2MoO_4-$xAg_{0.5}Bi_{0.5}MoO_4$ ceramics (x = 0.2, 0.4, 0.6, 0.65, 0.7, 0.8) is called AMAB for short. Fig. 1 presents the X-ray diffraction spectrum of the AMAB ceramics calcined at 420 °C. It is clear that both Ag_2MoO_4 phase and $Ag_{0.5}Bi_{0.5}MoO_4$ phase have been formed and no extra phases were detected as x value changed, which indicates that the tetragonal $Ag_{0.5}Bi_{0.5}MoO_4$ ceramics and cubic Ag_2MoO_4 ceramics can coexist well with each other.

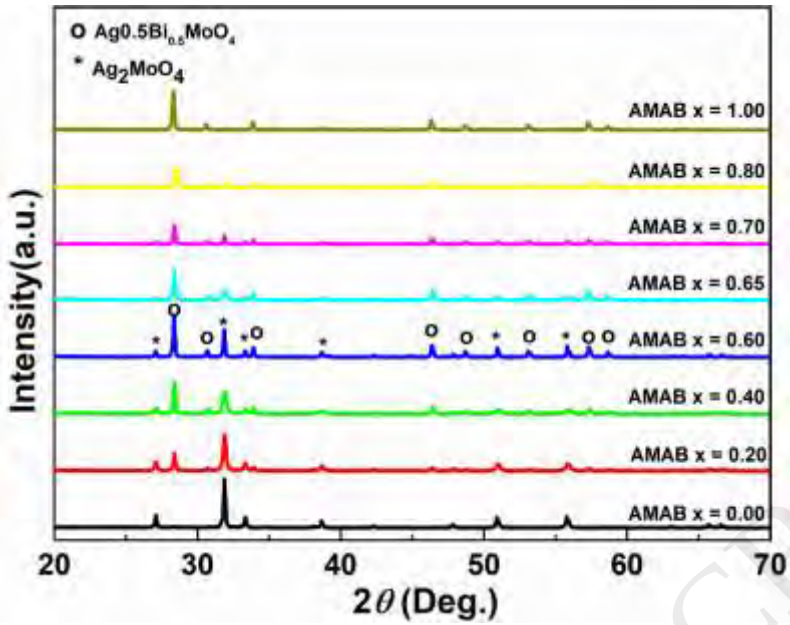

Fig. 1. XRD patterns of (1-*x*)Ag_2MoO_4-*x*$Ag_{0.5}Bi_{0.5}MoO_4$ ceramics calcined at 420 °C.

Fig. 2 shows the relative densities of the AMAB ceramics sintered at their optimal sintering temperatures as a function against the *x* value. Due to the water-soluble properties, the samples were all kept at 120 °C for 4 h in the air oven and then cooled to room temperature. Subsequently, Archimedes method with ethanol as solvent was used to measure the densities of the samples. The density of the composite ceramics can be predicted as follows:

$$\rho_{theo} = v_1\rho_1 + v_1\rho_1 \quad (2)$$

where the subscripts of '1' and '2' represent the material 1 and material 2, respectively. ρ_{theo} is the theoretical density of the composite, ρ_1 and ρ_2 are the theoretical densities of Ag_2MoO_4 and $Ag_{0.5}Bi_{0.5}MoO_4$, respectively. v_1 and v_2 indicate the volume fractions of corresponding materials. Here, the theoretical density of Ag_2MoO_4 is 6.285 g/cm^3 according to PDF card No.# 01-1002 and that of $Ag_{0.5}Bi_{0.5}MoO_4$ calculated from lattice constants is 6.524 g/cm^3. As shown in Fig. 2, all the relative densities of the AMAB ceramics are above 95%. This result indicates that the series of AMAB ceramics can be easily densified.

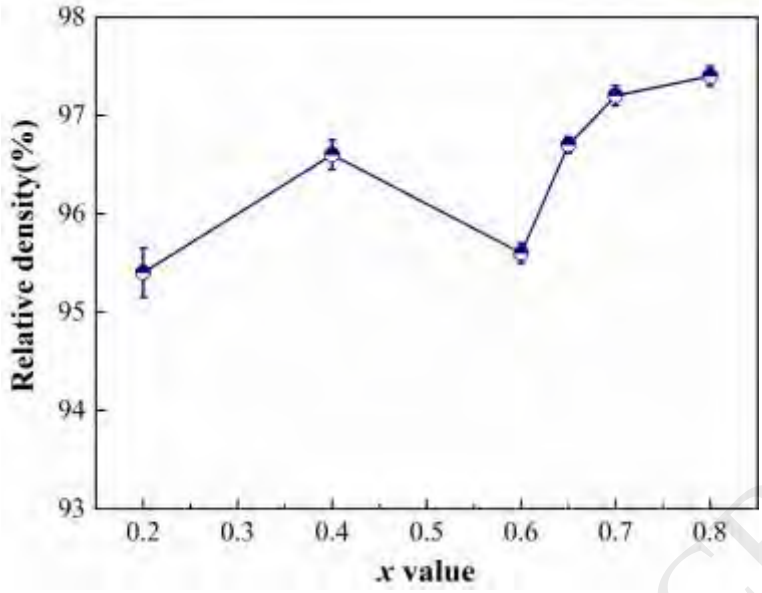

Fig. 2. Relative density of the AMAB ceramics sintered at their optimal sintering temperatures.

Scanning electron micrographs for the thermally etched surfaces of the AMAB ceramics sintered at their optimal sintering temperatures (S.T.) are shown in Fig.3. All the AMAB ceramics show compact microstructures. It can be observed that two types of grains can coexist, which is in accordance with the XRD results. The grains with a larger size and a smaller size belong to Ag_2MoO_4 phase and $Ag_{0.5}Bi_{0.5}MoO_4$ phase, respectively. Since the sintering temperature of AMAB composites is lower than that of pure $Ag_{0.5}Bi_{0.5}MoO_4$ ceramics, the grains of $Ag_{0.5}Bi_{0.5}MoO_4$ phase have not fully grown in the sintering process. As the x value increases, the number of $Ag_{0.5}Bi_{0.5}MoO_4$ grains increases significantly and the densification temperature of AMAB ceramics rises from 460 °C to 530 °C. When the sintering temperature is higher than 520 °C, some irregular grains are observed as shown in Fig. 3(e) and Fig. 3(f), which may be attributed to the excessive sintering temperature for pure Ag_2MoO_4 phase. Too high sintering temperature results in the abnormal growth of the grains. In addition, according to the previous work, there might be some pores deeply inside the abnormal grains[15].

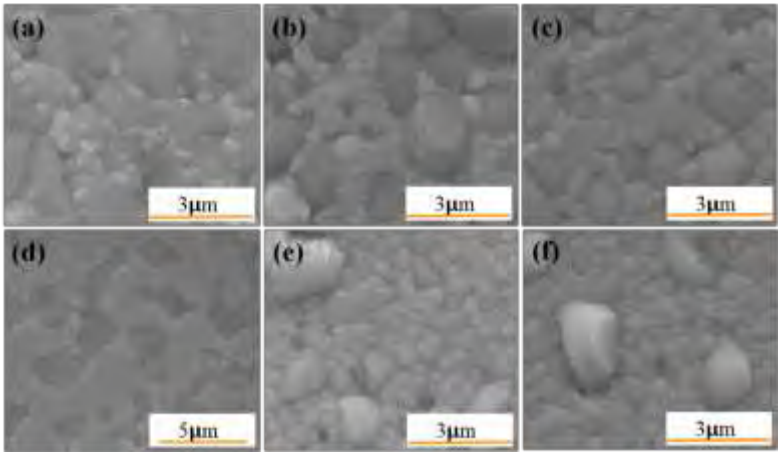

Fig. 3. SEM images of thermally etched AMAB ceramics: (a) x = 0.2, S.T. = 460°C, (b) x = 0.4, S.T. = 470°C, (c) x = 0.6, S.T. = 480°C, (d) x = 0.65, S.T. = 520°C, (e) x = 0.7, S.T. = 530°C, (f) x = 0.8, S.T. = 530°C.

The microwave dielectric properties of the AMAB ceramics sintered in air at their optimal temperatures are shown in Fig. 4. The corresponding values have been obtained from the related samples sintered at their optimal sintering temperatures. When the composition of multiphase materials distributes randomly, there are several models to calculate the effective dielectric constant. Among them, Lichtenecker empirical logarithmic rule is applied to this work due to its relative simplicity and effectiveness [16].

$$\ln \varepsilon = v_1 \ln \varepsilon_1 + v_2 \ln \varepsilon_2 \quad (3)$$

Where, ε is the relative permittivity. To ensure the reliability of the theoretical calculation, the Ag_2MoO_4 and $Ag_{0.5}Bi_{0.5}MoO_4$ ceramics were also prepared by the modified solid-state reaction method. The microwave dielectric properties of them are listed in Table 1. With the increase of *x* value, the measured and calculated relative permittivities increase gradually. The interfacial effects in the composite material may increase the dielectric constant of the ceramics, resulting in measured values higher than the calculated values.

According to the Lichtenecker empirical logarithmic rule, the semi-empirical linear formula for calculating TCF value of multiphase ceramics can be deducted as,

$$\tau_f = v_1 \tau_{f1} + v_2 \tau_{f2} \quad (4)$$

where, τ_{f1} and τ_{f2} are the temperature coefficients of the resonant frequency (TCF value) of material 1 and material 2, respectively. The TCF value of the Ag_2MoO_4 ceramic is -124 ppm/°C and it is +60 ppm/°C for $Ag_{0.5}Bi_{0.5}MoO_4$ ceramic. Thus, for the purpose of temperature stability, the theoretical x value is 0.71. Mathematically, the origin TCF value can be calculated as follows:

$$\tau_f = -\alpha_L - \tau_\varepsilon/2 \quad (5)$$

It can be seen that the TCF value is closely related to the temperature coefficient of the relative relative permittivity (τ_ε) and the linear expansion coefficient (α_L). However, according to the previous work, the thermal expansion coefficient can be influenced by many factors, such as porosity [17]. It can be seen from the Fig. 4 that the measured TCF value increases linearly with *x* value. As the *x* value increases, the TCF value of the AMAB ceramics has been gradually adjusted from a negative value (-12 ppm/°C) to a positive value (+28 ppm/°C). When x = 0.65, the TCF value is -2.4 ppm/°C, which is near zero. The difference between the measured value and the theoretical value may be ascribed to the porosity in the samples. However, the difference is not significant, indicating that the semi-empirical linear formula is applicable to predict the TCF value of the composite mixtures.

For multiphase ceramics, the Q × f value can be predicted according to the well-known general empirical model:

$$1/\mathrm{Q} \times \mathrm{f} = v_1/(\mathrm{Q}_1 \times \mathrm{f}_1) + v_2/(\mathrm{Q}_2 \times \mathrm{f}_2) \quad (6)$$

where, $\mathrm{Q} \times \mathrm{f}$ is the quality factor of the mixture, $\mathrm{Q}_1 \times \mathrm{f}_1$ and $\mathrm{Q}_2 \times \mathrm{f}_2$ are the quality factors of two components. The Q × f values of Ag_2MoO_4 and $Ag_{0.5}Bi_{0.5}MoO_4$ are 28000 GHz and 15000 GHz, respectively. When the mole fraction of $Ag_{0.5}Bi_{0.5}MoO_4$ rises, the Q × f value of the AMAB ceramics decreases due to the lower Q × f value of pure $Ag_{0.5}Bi_{0.5}MoO_4$. As reported [18-19], the Q × f value is influenced by many factors, such as grain boundary, grains size, porosity, and liquid phase. Thus the measured Q × f values are lower than the calculated values.

Table 1 The microwave dielectric properties of the Ag_2MoO_4 ceramic and the $Ag_{0.5}Bi_{0.5}MoO_4$ ceramic prepared by the modified solid-state reaction method and traditional solid-state reaction method

Sample	S. T. (°C)	$\boldsymbol{\varepsilon_r}$	Q × f (GHz)	TCF (ppm/°C)	Density(g/cm^3)	Ref.
Ag_2MoO_4	440	8.98	28000	-124	5.7	This work
$Ag_{0.5}Bi_{0.5}MoO_4$	580	36.8	15000	60	6.38	This work
Ag_2MoO_4	450	8.08	17000	-133	6.02	[11]
$Ag_{0.5}Bi_{0.5}MoO_4$	690	30.4	12600	57	--	[12]

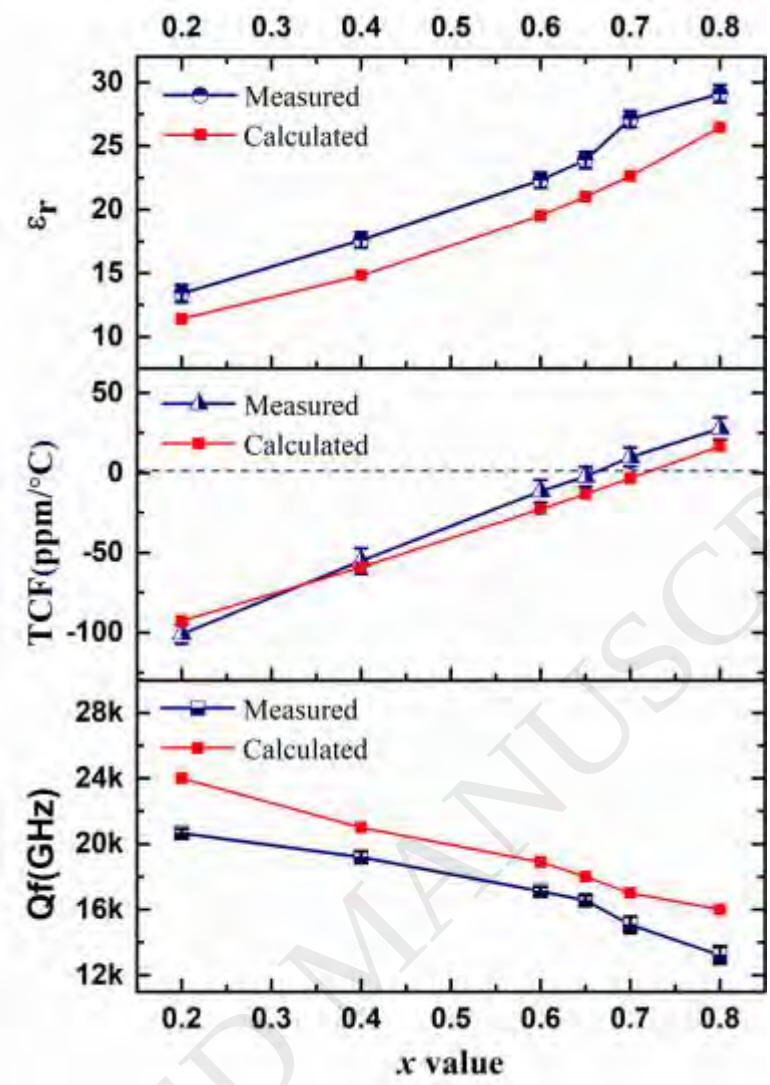

Fig. 4. Microwave dielectric properties of the AMAB ceramics sintered at their optimal temperatures

The microwave dielectric properties of the AMAB (x=0.65) ceramics sintered at temperatures ranging from 440 °C to 540 °C are shown in Fig. 5. When sintered at 520 °C, the $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ ceramics exhibit the best properties with a relative permittivity of 23.9, a Q × f value of 16200 GHz (at 7.3 GHz), and a near zero TCF value of -2.4 ppm/°C. The relative permittivity improves with the increasing of the sintering temperature and reaches the optimal value of 23.9 at around 520 °C and then decreases. This trend is attributed to the porosity of ceramic samples [20-21]. The porosity in the samples decreases with the sintering temperature at first then increases slightly because of the increasing closed pores in the abnormal grains. The TCF value fluctuates from -5.6 ppm/℃ to -2.4 ppm/℃ in the whole sintering temperature range. When sintered at 520 °C, the grain boundary and porosity of $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ ceramic drastically decrease due to its

homogeneous and compact microstructures, and its Q × f value reaches a maximum of 16200 GHz. Subsequently, the abnormal growth of grain makes the increase of grain boundary and porosity, resulting in the decease of the Q × f value. Table 2 shows some selected ultra-low temperature co-fired ceramics with temperature stability. Compared with the reported temperature-stable ceramics with low-sintering temperatures, the $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ ceramic does not have the highest Q × f value. However, considering the highly toxic of tellurium oxide, the $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ ceramic in our work has the lowest sintering temperature and excellent microwave dielectric properties comprehensively [22-24].

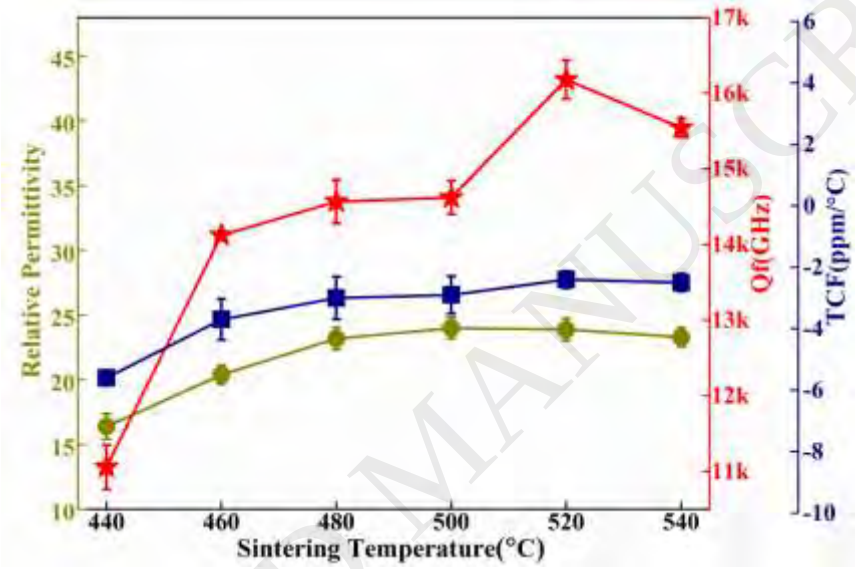

Fig. 5 Microwave dielectric properties of the $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ ceramics as a function of sintering temperature.

Table 2 The sintering temperature, microwave dielectric properties, and densities of some selected ultra-low temperature co-fired ceramics with temperature stability

Sample	S. T. (°C)	ε_r	Q × f (GHz)	TCF (ppm/°C)	Ref.
$BaTe_4O_9$ + 40wt%$TiTe_3O_8$	575	25.0	19300	-3	[22]
0.47$BaTe_4O_9$-0.53$TiTe_3O_8$	560	28.0	12000	4	[23]
0.25$Na_2Mo_2O_7$-0.75$Na_{0.5}Bi_{0.5}MoO_4$	580	24.0	13000	3	[24]
0.35Ag_2MoO_4-0.65$Ag_{0.5}Bi_{0.5}MoO_4$	520	23.9 ± 0.1	16200 ± 700	-2.4 ± 0.2	This work

4. Conclusion

In this work, the temperature-stable $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ microwave dielectric ceramics sintered at low temperatures were prepared by a modified solid-state reaction method. Two types of grains distribute randomly and coexist with each other according to the XRD and SEM results.

The samples sintered at 520 °C exhibit a high relative density of 97 % and good microwave dielectric properties with a relative permittivity of 23.9, a Q × f value of 16200 GHz (at 7.3 GHz) and a near-zero TCF value of -2.4 ppm/°C. Compared with other temperature-stable ceramics sintered at ultra-low sintering temperatures, the $0.35Ag_2MoO_4$-$0.65Ag_{0.5}Bi_{0.5}MoO_4$ ceramics in this work possess superior comprehensive advantages, such as better temperature stability, lower sintering temperature and environment-friendly feature, providing more possibilities for its application in LTCC microwave devices.

Acknowledgements

The work was supported by the National Key Research Program (Grant No. 2017YFB0406303) and the National Natural Science Foundation of China (No: 61631166004). The SEM work was done at International Center for Dielectric research, Xi'an Jiaotong University, Xi'an, China.

References

[1] M.T. Sebastian, H. Jantunen, Low loss dielectric materials for LTCC applications: a review, Int. Mater. Rev. 53 (2008) 57-90.

[2] M.T. Sebastian, R. Ubic, H. Jantunen, Low-loss dielectric ceramic materials and their properties, Int. Mater. Rev. 60 (2015) 392-412.

[3] H. Yu, J. Liu, W. Zhang, S. Zhang, Ultra-low sintering temperature ceramics for LTCC applications: a review, J. Mater. Sci. - Mater. Electron. 26 (2015) 9414–9423.

[4] M.T. Sebastian, H. Wang, H. Jantunen, Low temperature co-fired ceramics with ultra-low sintering temperature: a review, Curr. Opin. Solid State Mater. Sci. 20 (2016) 151–170.

[5] K.P. Surendran, N. Santha, P. Mohanan, M.T. Sebastian, Temperature stable low loss ceramic dielectrics in $(1-x)ZnAl_2O_4$-$xTiO_2$ system for microwave substrate applications, Eur. Phys. J. B. 41(2004) 301-306.

[6] Y.C. Liou, Y.T. Chen, W.C. Tsai, Synthesis of temperature-stable microwave dielectric ceramics $NiNb_2O_6$ with TiO_2 addition, J. Alloys Compd. 477(2009) 537-542.

[7] H.W. Lee, J.H. Park, S. Nahm, D.W. Kim, J.G. Park, Low-temperature sintering of temperature-stable $LaNbO_4$ microwave dielectric ceramics, Mater. Res. Bull. 45.1(2010) 21-24.

[8] M. Guo, S.P. Gong, G. Dou, D. Zhou, A new temperature stable microwave dielectric ceramics: $ZnTiNb_2O_8$ sintered at low temperatures, J. Alloys. Compd. 509.20(2011) 5988-5995.

[9] Y. Wu, D. Zhou, J. Guo, L.X. Wang, H. Wang, X. Yao, Temperature stable microwave dielectric ceramic $0.3Li_2TiO_3$-$0.7Li(Zn_{0.5}Ti_{1.5})O_4$ with ultra-low dielectric loss, Mater. Lett. 65(2011) 2680-2682.

[10] J. Guo, D. Zhou, H. Wang, Y.H. Chen, Y. Zeng, F. Xiang, Y. Wu, X. Yao. Microwave and infrared dielectric response of temperature stable (1-*x*) $BaMoO_4$-*x*TiO_2 composite ceramics, J. Am. Ceram. Soc. 95(2012) 232-237.

[11] D. Zhou, W.B. Li, L.X. Pang, J. Guo, Z.M. Qi, T. Shao, Sintering behavior and dielectric properties of ultra-low temperature fired silver molybdate ceramics, J. Am. Ceram. Soc. 97(2014) 3597-3601.

[12] D. Zhou, C.A. Randall, L.X. Pang, H. Wang, J. Guo, G.Q. Zhang, Y. Wu, K.T. Guo, L. Shui, X. Yao, Microwave dielectric properties of $(AB)_{1/2}$ MoO_4 (A = Li, Na, K, Rb, Ag) type ceramics with ultra-low firing temperatures, Mater. Chem. Phys. 129(2011) 688–692.

[13] X.F. Yuan, G.Q. Zhang, H. Wang, A novel low temperature sintering (K1-xNax)2Mo2O7 ceramics via modified solid-state reaction method, J. Eur. Ceram. Soc. 38(2018) 4967-4971.

[14] X.F. Yuan, X.Xue, H. Wang, Preparation of ultra‐low temperature sintering ceramics with ultralow dielectric loss in Na2O-WO3 binary system, J. Am. Ceram. Soc. 102(2019) 4014-4020.

[15] G.Q. Zhang, J. Guo, H. Wang, Ultra-low temperature sintering microwave dielectric ceramics based on Ag_2O-MoO_3 binary system, J. Am. Ceram. Soc. 00(2017)1–8.

[16] Y. Imanaka, Multilayered Low Temperature Cofired Ceramics (LTCC) Technology, first ed., Springer-Verlag, New York, 2005.

[17] Z.Z. Weng, Z.H. Han, F. Xiao, D.L. Peng, Low temperature sintering and microwave dielectric properties of $Zn_{1.8}SiO_{3.8}$, ceramics with $BaCu(B_2O_5)$ additive for LTCC applications, Ceram. Int. 44(2018)14145-14150.

[18] S. J. Penn, N. Alford, A. Templeton, X. Wang, M. Xu, M. Reece, K. Schrapel, Effect of Porosity and Grain Size on the Microwave Dielectric Properties of Sintered Alumina, J. Am. Ceram. Soc. 80 (1997) 1885–1888.

[19] N.M. Alford, J. Breeze, X. Wang, S. J. Penn, S. Dalla, S. J. Webb, N. Ljepojevic, X. Aupi, Dielectric Loss of Oxide Single Crystals and Poly-crystalline Analogues from 10 to 320 K, J. Eur. Ceram. Soc. 21 (2001) 2605–2611.

[20] M.N. Rahaman, Ceramic Processing and Sintering, ???: John Wiley & Sons, Inc. (2006)540-615.

[21] W.D. Kingery, H.K. Bowen, D.R. Uhlmann, Introduction to Ceramics, New York City, New York: Wiley, 1976, 448-507.

[22] S.F. Wang, Y.R. Wang, Y.F. Hsu, J.S. Tsai, Densification, microstructure and microwave dielectric properties of ultra-low fire $BaTe_4O_9$-$TiTe_3O_8$ ceramic composites, J. Eur. Ceram. Soc. 30 (2010) 1737–1741.

[23] X. Jiao, C. Zhong, S. Zhang, X. Liu, B. Li, Microwave dielectric properties of $BaO–TiO_2–TeO_2$ ternary system, J. Mater. Sci. 45 (2010) 3331–3335.

[24] G.Q. Zhang, J. Guo, X.F. Yuan, H. Wang, Ultra-low temperature sintering and microwave dielectric properties of a novel temperature stable $Na_2Mo_2O_7$-$Na_{0.5}Bi_{0.5}MoO_4$ ceramic, J Eur Ceram Soc. 38(2018) 813–816.