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Processing of low-fired glass-free $Li_2MgTi_3O_8$ microwave dielectric ceramics

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ABSTRACT

An approach in which the raw materials of Li_2CO_3, MgO and TiO_2 were ball milled before mixing (abbreviated as LMT2) was reported. For the traditional ceramic technique (abbreviated as LMT1), the raw materials of Li_2CO_3, MgO and TiO_2 were directly used without further processing. The powders from LMT2 tended to be smaller and uniform and the ceramics exhibited higher densities than that of LMT1, which significantly lowered the sintering temperature from 1075 °C to 925 °C. LMT2 ceramics sintered at 925 °C exhibited good microwave dielectric performance with $\varepsilon_r = 27.0$, $Q \times f = 58{,}480$ GHz (at ~ 5.8 GHz), $\tau_f = 0.45$ ppm/°C. Moreover, LMT2 ceramic was found to be chemically compatible with Ag. The results indicate that the pretreatment of raw materials is a relatively simple and particularly effective method to obtain glass-free $Li_2MgTi_3O_8$ LTCC ceramics.

1. Introduction

In recent years, low temperature co-fired ceramics (LTCC) multilayer devices have been reported to reduce the size of component, meeting the increasing requirements of communication systems on the miniaturization of device size [1–4]. Due to the high conductivity and comparatively low cost, Ag has been widely used in LTCC technology as a metallic electrode. Owing to the low melting point of silver (~961 °C), microwave dielectrics used in LTCC devices must be sintered at temperatures below 950 °C [5–8]. So it is very important to develop microwave dielectric materials with having low sintering temperatures and chemical compatibility with Ag electrode.

The use of sintering aids, such as V_2O_5, $BaCu(B_2O_5)$, B_2O_3 and CuO etc., is a common method to lower the sintering temperature of dielectric ceramics [9–15]. However, the microwave dielectric properties of materials will be degraded because the addition of glass or oxides forms a large amount of amorphous phase in the final ceramics. Therefore, a number of studies are focused on search for new materials with having relatively lower temperatures (<950 °C) and good microwave dielectric properties (low ε_r, high Q and near-zero τ_f), such as $Li_2M_2^{2+}Mo_3O_{12}$ [16], $Li_3M^{3+}Mo_3O_{12}$ [16], $Li(Mg_{1-x}Co_x)PO_4$ [17], $(Li_{0.5}Sm_{0.5})WO_4$ [18], $(K_{0.5}Ln_{0.5})MoO_4$ (Ln = Nd and Sm) [19], $BaLn_2(MoO_4)_4$ (Ln = Nd and Sm) [20], $(1-x)BiVO_4-xYVO_4$ [21], $(1-x)BiVO_4-xTiO_2$ [22].

$Li_2MgTi_3O_8$ spinel ceramic has attracted much scientific and commercial attention because of its excellent microwave dielectric properties. West and Hernandez et al. [23,24] first investigated the structure of $Li_2MgTi_3O_8$. Recently, George et al. [25] and Zhou et al. [26] reported that $Li_2MgTi_3O_8$ ceramic sintered at 1075 °C for 4 h exhibited good microwave dielectric performance with a moderate ε_r of ~27.2, high $Q \times f$ of ~42,000 GHz and near-zero τ_f of ~3.2 ppm/°C. However, higher sintering temperature restricted its further application in LTCC devices. As we know, chemical processing for starting powders with smaller particle sizes benefits for developing new material systems with low sintering temperatures (normally below 1100 °C) [27]. Moreover, the powders with uniform and small size are in favor of the formation of final phase and the reduction of secondary phase, which increases the microwave performance of materials [28]. In this paper, we obtained starting powders with smaller particle sizes by physical grinding method and investigated their effects on the sinterability and microwave dielectric performances of $Li_2MgTi_3O_8$ ceramic. Furthermore, the chemical compatibility between the dielectric and silver was also reported.

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2. Experimental procedures

Specimens of $Li_2MgTi_3O_8$ ceramic were prepared by LMT1 and LMT2 methods. For the LMT1, the analytical reagent raw materials (≥99%, Guo-Yao Co. Ltd., Shanghai, China) of Li_2CO_3, MgO and TiO_2 were directly mixed without further processing. But for the LMT2, the raw materials of Li_2CO_3, MgO and TiO_2 were ball milled firstly in a polyamide pot with ZrO_2 balls for 4 h using ethanol as dispersants before mixing, respectively. Then, the LMT1 and LMT2 ceramics were synthesized by the same procedures as follows: stoichiometric proportion of the above raw materials was mixed in the high-purity alcohol (≥99.5%) medium using zirconia balls for 4 h. Then the mixtures were dried and calcined at 900 °C for 4 h. Subsequently, the mixtures were milled in the same way as the raw powders. After drying, the powders were mixed with 5 wt% polyvinyl alcohol (PVA) and pressed into the cylinders with 12 mm in diameter and 6–7 mm in thickness by uniaxial pressing under a pressure of 200 MPa. Finally, the samples were sintered at 900–1100 °C for 4 h in air at a heating rate of 5 °C/min and then cooled in furnace to room temperature. In order to investigate whether the LMT2 ceramics react with Ag electrode or not, 20 wt% Ag powders was mixed with the calcined LMT2 powders and co-fired at 925 °C for 4 h.

The phase analysis of these ceramics was carried out by an X-ray diffractometer (XRD) (CuKα1, 1.54059 Å, Model X'Pert PRO, PANalytical, Almelo, Holland) operated at 40 kV and 40 mA. The microstructural observation of the samples was performed using a scanning electron microscope (SEM) (Model JSM6380-LV SEM, JEOL, Tokyo, Japan). The apparent densities of the sintered samples were measured by the Archimedes method using distilled water as a medium. Dielectric behaviors at microwave frequencies were measured by the $TE_{01\delta}$ shielded cavity method [29] in the frequency range of 4–9 GHz using a Network Analyzer (ModelN5230A, Agilent Co., CA, 10 MHz–40 GHz) and a temperature chamber (DELTA9039, Delta Design, USA) with an open invar cavity. The temperature coefficients of resonant frequency (τ_f) were calculated using Equation (1):

$$\tau_f = \frac{f_T - f_0}{f_0(T - T_0)}, \qquad (1)$$

where f_T, f_0 were the $TE_{01\delta}$ resonant frequencies at the measuring temperature T (85 °C) and T_0 (25 °C), respectively.

3. Results

3.1. Initial microstructure of LMT samples

Fig. 1 shows the SEM images of the mixtures with Li_2CO_3, MgO and TiO_2 prepared by LMT1 and LMT2 routes. Obviously, for the LMT1 route in which the raw materials were directly used without further processing, the degree of crumbling between the Li_2CO_3, MgO and TiO_2 is not same, which can be explained by the different plasticity of raw materials. As a result, there are some huge particles in the mixtures for the LMT1 powders, as shown in Fig. 1 (a). In contrast, smaller size and more uniform powder particles (~0.5 μm) are gained by the LMT2 [Fig. 1 (b)]. Smaller particles indicate larger contact area between Li_2CO_3, MgO and TiO_2, which promotes the reaction of powders in the calcined procedure.

3.2. Phase composition and structure of LMT ceramics

Fig. 2 demonstrates the room-temperature XRD patterns of LMT2 and LMT1 ceramics sintered at 925 °C and 1075 °C, respectively. A single-phase $Li_2MgTi_3O_8$ with cubic structure (PDF code: 01-089-1308) was formed and no secondary phase could be observed in diffraction patterns. The diffraction peaks of LMT2 ceramics were much sharper than that of LMT1 ceramics, indicating a better crystallization for LMT2 ceramics. It was attributed to smaller starting powders of LMT2, which enlarges the contact area between Li_2CO_3, MgO and TiO_2 particles and promotes the reaction during calcination. It turned out that these different morphologies of the mixtures directly influenced the microstructure and sintering characteristics of $Li_2MgTi_3O_8$ ceramics.

The SEM micrographs of the surface for $Li_2MgTi_3O_8$ ceramics sintered at 925 °C and 1075 °C are illustrated in Fig. 3. With

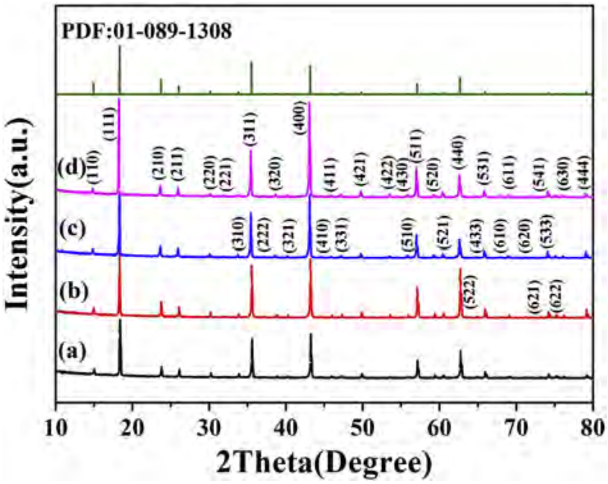

Fig. 2. XRD patterns of LMT1 and LMT2 ceramics sintered at 925 °C and 1075 °C for 4 h: (a) LMT1, 925 °C; (b) LMT2, 925 °C; (c) LMT1, 1075 °C; (d) LMT2, 1075 °C.

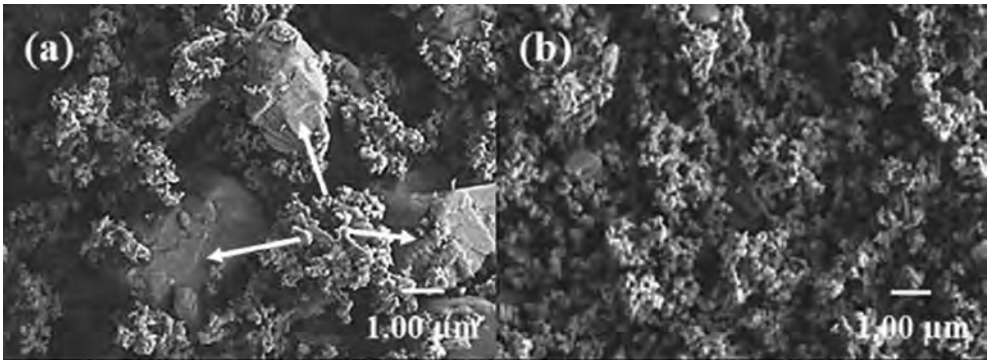

Fig. 1. The morphologies of the mixtures with Li_2CO_3, MgO and TiO_2 by LMT1 (a) and LMT2 (b).

increasing the sintering temperature, the grains grew and the microstructure of $Li_2MgTi_3O_8$ ceramics exhibited fewer pores and became denser. At the same sintering temperature, it can be observed obviously that the ceramics have a clearly different grain morphology and density. For LMT1, there are many visible pores, but LMT2 ceramics displayed denser microstructure and bigger grain size than that of LMT1. Moreover, it was easier to observe the re-crystallization of LMT2 ceramics than that of LMT1 ceramics when the sintering temperatures were 925 °C and 1075 °C. Due to the fact that small size particles have high interface energy, more contact points and shorter diffusion distances among particles, the size of raw powders has a great influence on the grain growth of LMT ceramics.

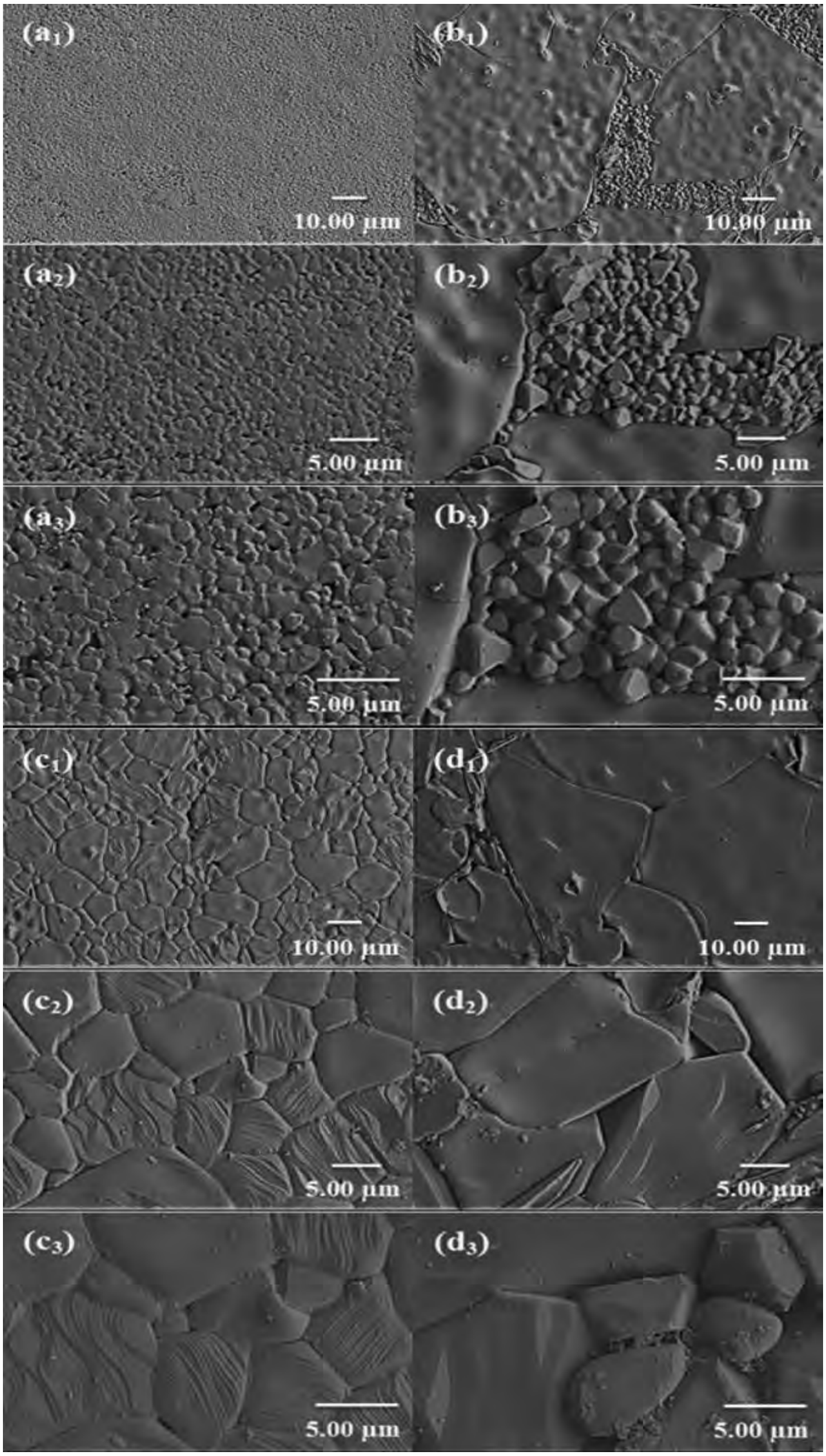

Fig. 3. SEM micrographs of LMT1 and LMT2 ceramics sintered at their optimized temperatures: (a_1)–(a_3) LMT1, 925 °C; (b_1)–(b_3) LMT2, 925 °C; (c_1)–(c_3) LMT1, 1075 °C; (d_1)–(d_3) LMT2, 1075 °C.

3.3. Bulk density and microwave dielectric properties of LMT ceramics

Fig. 4 shows the bulk density, relative permittivity, quality factor and temperature coefficients of resonant frequency (τ_f) of LMT ceramics as a function of the sintering temperature. As shown in Fig. 4(a), when the sintering temperature was increased from 900 °C to 1100 °C, the density of $Li_2MgTi_3O_8$ ceramics increased firstly, reached a maximum value (LMT1: ~3.369 g/cm^3; LMT2: ~3.381 g/cm^3) when the sintering temperatures was increased to 1075 °C (LMT1) and 925 °C (LMT2), and then decreased with further increasing the sintering temperature. The densities of LMT2 ceramics are slightly higher than that of LMT1, especially at low sintering temperatures. Generally, the density is related to porosity. When the sintering temperature was 925 °C, few pores can be found in LMT2 ceramic, but there are a large number of pores in LMT1 ceramic. For this reason, the density of LMT2 is greater than LMT1 at low sintering temperature. In addition, compared with the LMT1 ceramics, the optimal temperature of LMT2 ceramics decreased from 1075 °C (LMT1) to 925 °C (LMT2). These results showed that smaller and more uniform powder particles could promote the densification of $Li_2MgTi_3O_8$ ceramics.

Fig. 4(b) illustrates the relative permittivity (ε_r) of $Li_2MgTi_3O_8$ ceramics as a function of the sintering temperature. As the sintering temperatures was increased from 900 °C to 1100 °C, ε_r presents a trend similar to that of density since the density and ε_r of ceramic are associated with the elimination of the pores [30,31]. The permittivity of LMT1 and LMT2 specimens increased at first, got the optimum value about 26.8 at 1075 °C (LMT1) and 27.0 at 925 °C (LMT2), then decreased with further increasing the sintering temperature. Obviously, LMT2 ceramics obtained larger optimum permittivity than LMT1 owing to the higher relative densities, especially at low temperature.

Fig. 4(c) demonstrates the quality factor of $Li_2MgTi_3O_8$ ceramics as a function of the sintering temperature. The variation of $Q \times f$ values mainly resulted from densification changes and the average grain size [32]. Due to the lower densities, the microwave quality factors of LMT1 ceramics were lower than LMT2 at low sintering temperatures. With increasing the sintering temperatures, the densities of LMT2 ceramics rapidly approached to that of LMT1 ceramics, and quality factors of LMT2 ceramics were higher than that of LMT1 ceramics. That was because quality factors were mainly affected by the densification changes.

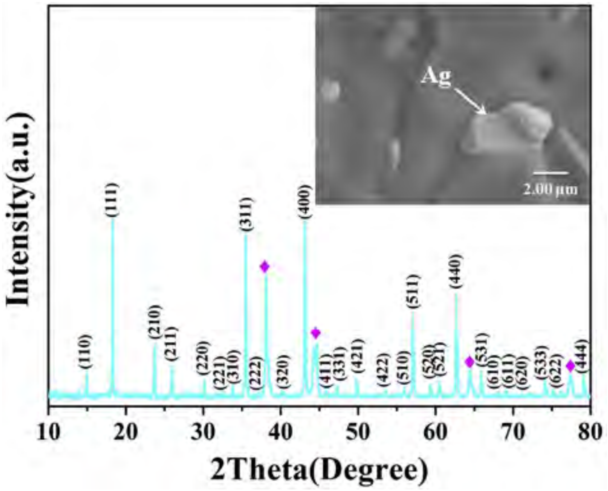

Fig. 5. XRD patterns of LMT2 ceramic mixed with 20 wt% Ag sintered at 925 °C for 2 h. Inset shows a backscattered electron image of LMT2 sample cofired with Ag.

As the sintering temperature was increased from 900 °C to 1100 °C, the τ_f values of LMT2 ceramics decreased at first, and reached a minimum of 0.45 ppm/°C at 925 °C, as shown in Fig. 4(d). Due to the same material systems, the τ_f values of LMT1 ceramics presented the similar trend to that of LMT2 ceramics with increasing the sintering temperature, and got a minimum of 3.0 ppm/°C as the sintering temperature was 1075 °C. Those results

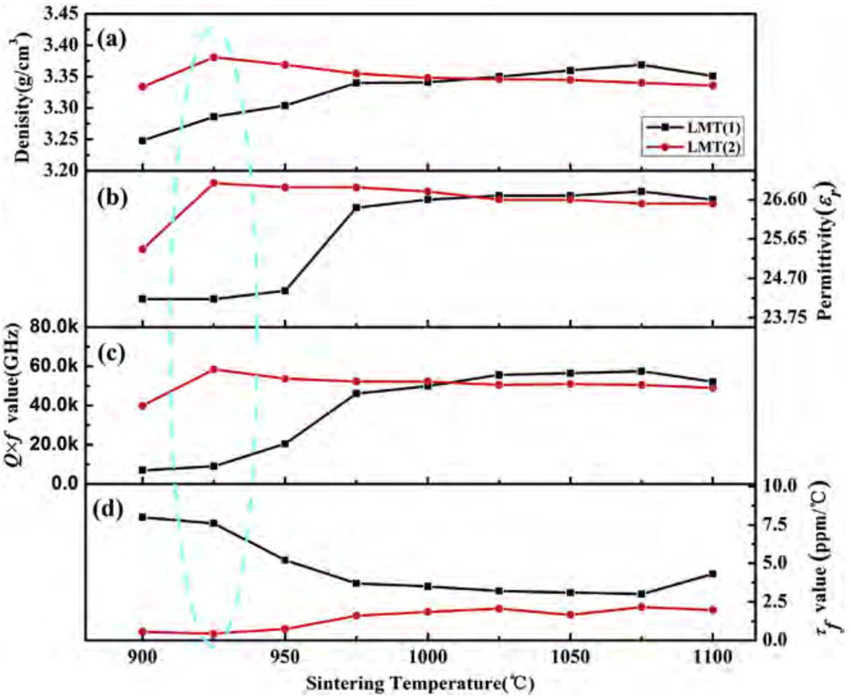

Fig. 4. The bulk density (ρ) (a), relative permittivity (ε_r) (b), $Q \times f$ (c) and τ_f value (d) of LMT1 and LMT2 ceramics as a function of the sintering temperatures.

Table 1
The comparison of properties between $Li_2MgTi_3O_8$ (LMT1, LMT2) ceramics and the previous systems.

Ceramic composition	Sintering temperature (°C)	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)	Ref.
$ZnNb_2O_6$	~1200	23.2	84,500	−76.0	[33]
$Ba(Mg_{1/3}Ta_{2/3})O_3$	~1550	25	176,400	4.4	[34]
$Ba(Zn_{1/3}Ta_{2/3})O_3$	~1550	30	168,000	0.5	[35]
$0.5LaCa_{0.5}Zr_{0.5}O_3$-$0.5CaTiO_3$	~1575	26.0	15,000	−67.0	[36]
$0.9MgTiO_3$-$Ca_{0.6}La_{0.8/3}TiO_3$	~1300	25.45	82,500	0.5	[37]
$Nd(Mg_{1/2}Ti_{1/2})O_3$	~1650	23.0	36,900	−49.0	[38]
LMT1	~900	24.2	6950	8.0	Our work
LMT2	~900	25.4	39,910	0.55	
LMT1	~925	24.2	8990	7.6	
LMT2	~925	27.0	58,450	0.45	
LMT1	~975	26.4	46,140	3.7	
LMT2	~975	26.9	52,350	1.6	
LMT1	~1075	26.8	57,560	3.0	
LMT2	~1075	26.5	50,600	2.2	
LMT1	~1100	26.6	52,140	4.3	
LMT2	~1100	26.5	49,010	2.0	

indicated that LMT2 ceramics had lower sintering temperatures than LMT1 based on the similar sintering ability, and LMT2 has obtained a near-zero τ_f value.

3.4. Chemical compatibility between the calcined $Li_2MgTi_3O_8$ powders and Ag

To evaluate the chemical compatibility between LMT2 ceramic and silver electrode, the calcined LMT2 powders was cofired with 20 wt% Ag powders at 925 °C for 2 h and analyzed the interactions between the low-fired powders and electrodes. Fig. 5 presents the XRD patterns and backscattered electron image (BEI) of the co-fired sample. The XRD pattern showed no formation of any other phase except for $Li_2MgTi_3O_8$ and Ag. In the BEI micrograph of Fig. 5, white Ag grains could be observed apart from LMT2 grains, indicating that the LMT2 ceramic did not react with Ag powders and no other phase was formed. The results show that the LMT2 material exhibits a good chemical compatibility with silver powders.

The comparison of properties between $Li_2MgTi_3O_8$ (LMT1, LMT2) ceramics and various material systems is listed in Table 1. Some niobate, tantalate and Ti-based compounds demonstrated good microwave dielectric properties with appropriate permittivity (ε_r) and high quality factor ($Q \times f$) [33–38]. But higher sintering temperatures (≥1200 °C) and larger τ_f values restricted their further applications in LTCC devices. In contrast, $Li_2MgTi_3O_8$ ceramics exhibited high quality factor ($Q \times f$) and near-zero τ_f at a low temperature (900 °C–1100 °C). Moreover, it was observed that the microwave dielectric properties of LMT2 ceramic were better than that of LMT1 ceramic, especially at 925 °C. In addition, LMT2 ceramic presented good chemical compatibility with silver powders. These results indicate that equivalent particle size of the raw powders can promote the sintering ability and improve the microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics.

4. Discussion

The powders from LMT2 tended to be smaller (~0.5 μm) and more uniform, indicating high interface energy, more contact points and shorter diffusion distances among particles. As a result, the sinteringability of LMT ceramics was improved, which significantly enhances the microwave dielectric properties of LMT ceramic. As we know, chemical processing for starting powders with smaller particle sizes (include nanosized powders) benefited for developing new material systems with low sintering temperatures. The preparation of nanosized powders needs high-cost raw materials and complicated technique. In the present work, we tried to develop a method with low cost and easy procedure to obtain smaller starting powders, which was easy to apply in the commercial production. The starting materials with particle size of ~0.5 μm were obtained by planet ball-milling method. The particle size is ~0.5 μm when the ball milling time is 4 h. And more ball milling time will not affect the particle size of powders. Different milling time on the effects of particle size will not be studied in this paper. So we think that the grain size of ~0.5 μm leads to lower sintering temperature.

In other words, LMT2 method is a useful strategy to optimize the microwave dielectric properties and lower the sintering temperature of creamics, which promotes further applications of materials in LTCC devices, such as filter, antenna, etc.

5. Conclusion

The pretreatment of raw materials has a great effect on the microstructure, sintering behavior and microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics. Compared with the traditional ceramic processing, the LMT2 method greatly lowered the sintering temperature and optimized the microwave dielectric performance of LMT ceramics. LMT2 ceramic sintered at 925 °C for 4 h exhibited favorable microwave dielectric properties of $\varepsilon r = 27.0$, $Q \times f = 58{,}480$ GHz (at ~5.8 GHz), and $\tau f = 0.45$ ppm/°C. The backscattered electron image and XRD analysis showed no reaction between the dielectric ceramic and silver powders. The results indicate that LMT2 might be a candidate for LTCC devices.

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