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# Microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics produced by reaction-sintering method

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**Abstract** $Li_2MgTi_3O_8$ ceramics were prepared by reaction-sintering method (free calcination) for the first time and its microwave dielectric properties were investigated. A single phase of $Li_2MgTi_3O_8$ ceramic was confirmed by XRD pattern. The variation of microstructures was analyzed by SEM. With increasing sintering temperature, the bulk density decreased, the $\varepsilon_r$ and $Q \times f$ increased firstly and then decreased, while the $\tau_f$ changed slightly and remained around 1.5 ppm/ °C. In particular, $Li_2MgTi_3O_8$ ceramics sintered by reaction-sintering method at 1100 °C for 5 h exhibited fine combination microwave dielectric properties of $\varepsilon_r = 23.0$, $Q \times f = 54\,052$ GHz (at 7.29 GHz) and $\tau_f = 1.5$ ppm/ °C.

## 1 Introduction

With the development of wireless communication, low cost microwave dielectric materials with high quality factor ($Q \times f$), high relative dielectric constant ($\varepsilon_r$) and near-zero temperature coefficient of resonant frequency ($\tau_f$) are strongly desired [1, 2]. The microwave dielectric materials are advantageous in terms of compactness, light weight, temperature stability and low cost in the production of high frequency devices. However, many ceramics cannot satisfy these requirements simultaneously, and their properties need to be tuned [3, 4].

In recent years, the dielectric materials in $Li_2O$–MgO–$TiO_2$ system have attracted much attention, since they crystallize in many different structure types and exhibit interesting physical and chemical properties [5, 6]. Among them, the crystal structure of $Li_2MgTi_3O_8$ was first reported by Hiroo [7]. Recently, George and Sebastian first reported its microwave dielectric properties of $\varepsilon_r = 27.2$, $Q \times f =$ 42,000 GHz and $\tau_f = 3.2$ ppm/°C [8]. The comparatively intrinsic low sintering temperature (~1100 °C) and excellent combination dielectric properties make this ceramic promising for application in microwave dielectric resonators, filters and a new low-temperature co-fired ceramics material. Presently active works have been done to decrease its sintering temperature, improve the microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics by doping various additives or suitable ions substitution [9–12]. Reaction-sintering (hereafter referred to as RS) has increased in popularity for its high efficiency by bypassing calcination and subsequent pulverization stages. Several materials, such as $MgTiO_3$, $BaTi_5O_{11}$ and $Ni_4Nb_2O_9$, have been successfully prepared using this method [13–15]. Although the spinel structure $Li_2MgTi_3O_8$ ceramics has been studied by some researchers, however, works of this ceramics by the RS process are not readily available. In the present work, $Li_2MgTi_3O_8$ ceramics was prepared using RS method. The resultant microwave dielectric properties were analyzed based on the densification, phase constituents, and microstructures of the ceramics. Reaction-sintering method was proved to be a simple and efficient method to produce pure phase $Li_2MgTi_3O_8$ ceramics with excellent dielectric properties.

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## 2 Experimental

The starting materials are high-purity oxide powders (>99.9 %): $Li_2CO_3$, MgO and $TiO_2$. Predried raw materials were weighed in stoichiometric $Li_2MgTi_3O_8$ and ball milled for 7 h in a nylon jar with agate balls and ethanol as media. The milled powders were dried. After drying, the powders with 5 wt% PVA as a binder were pressed into pellets 10 mm in diameter and 5 mm in thickness under a pressure of 200 MPa. The green compacts were sintered between 1025 and 1125 °C for 5 h in air.

The bulk densities of the sintered ceramics were measured by Archimedes' method. The crystal structures were analyzed using X-ray diffraction (XRD) with Cu Ka radiation (Rigaku D/MAX2550, Japan). The microstructure of pellets was investigated using a scanning electron microscope (SEM, Fei Quanta 200, Holland). The microwave dielectric properties of sintered samples were measured using a network analysis (ZVB20, Rohde & schwarz, Germany) with the $TE_{01\delta}$ shielded cavity method. The temperature coefficient of resonant frequency ($\tau_f$) was calculated with the following equation:

$$\tau_f = \frac{f_{85} - f_{25}}{f_{25} \times (85 - 25)} \quad (1)$$

## 3 Results and discussions

Figure 1 shows the XRD powder patterns of $Li_2MgTi_3O_8$ prepared by RS method sintered at different temperatures. As can be seen, for all samples, a single phase was detected within the detectable level of XRD. All of the diffraction peaks are consistent with a cubic spinel structure with space group $P4_332$ (PDF 89–1308). The lattice parameters were calculated to be a = b = c = 8.3854 Å, which are a littler larger than that reported earlier [7]. In addition, the XRD patterns of the $Li_2MgTi_3O_8$ ceramics system have no significant change with sintering temperatures in the range 1025–1125 °C.

Typical surface SEM micrographs of $Li_2MgTi_3O_8$ ceramics sintered at various temperatures and times are demonstrated in Fig. 2. A porous structure with small grain size was observed for the specimen sintered at 1025 °C, as shown in Fig. 2a, b revealed that most residual porosity occurred as isolated pores at triple junctions, which indicated that it reached the final stage of sintering. In addition, a noticeable grain growth and a relatively uniform surface morphology were developed at 1100 °C. However, abnormal grain growth and microcracks were observed at temperature higher than 1100 °C or sintering time longer than 5 h (Seen Fig. 2d, f), which might deteriorate the microwave dielectric properties of the ceramics.

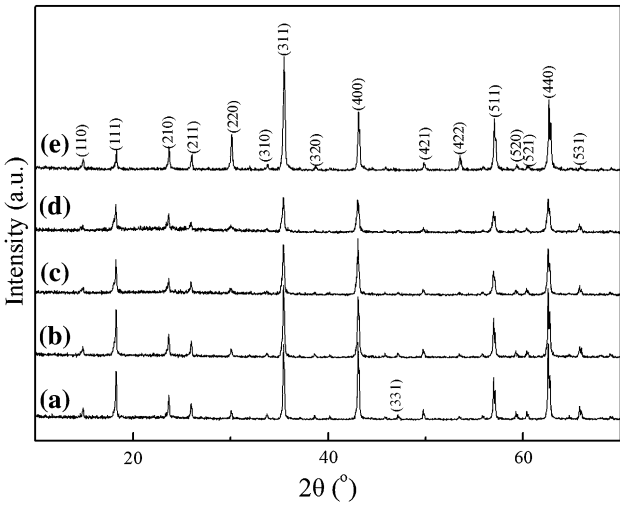


**Fig. 1** XRD powder patterns of $Li_2MgTi_3O_8$ sintered at different temperature: *a* 1025 °C, *b* 1050 °C, *c* 1075 °C *d* 1100 °C, *e* 1125 °C

Figure 3 illustrates the bulk density of $Li_2MgTi_3O_8$ ceramics as a functional of sintering temperature and sintered at 1100 °C for different dwell time. The bulk density of samples decreased with increasing sintering temperature and dwell time, which might be attributed to the evaporation of Li, a similar phenomenon was also reported by Chen et al. [9]. In addition, the reduction in density was also due to the entrapped porosity developed by the abnormal grain growth [16]. The maximum density of 3.1 g/cm$^3$ was obtained for the pellets sintered at 1025 °C for 5 h, which was only reached 88.6 % of theoretical density (3.5 g/cm$^3$). This poor densification maybe connected with the reaction-sintering process, a similar phenomenon was also founded in $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics by RS method [17].

The microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics sintered at various temperatures are exhibited in Fig. 4. From Fig. 4, the $\varepsilon_r$ value initially increased and then decreased after reaching a maximum value at 1050 °C. The increase in $\varepsilon_r$ could be explained the decrease of open pores and grain boundaries, while its degradation was due to the poor densification. Additionally, the $\varepsilon_r$ value of $Li_2MgTi_3O_8$ ceramics in our research was relatively lower than reported value by George et al. [8], which is attributed to the lower relative density in our specimens caused by RS process. With increasing sintering temperature, the Q × f value increased to a maximum value and thereafter slightly decreased. The microwave dielectric loss in bulk ceramics fall into two categories: intrinsic and extrinsic. It is generally accepted that dielectric loss of a material decreases with increase in grain size and increases with increase in porosity and other defects present in the material as the grain boundaries and defects act as scattering centres for the microwave radiation [18, 19]. Based on the above XRD, SEM, density results, the

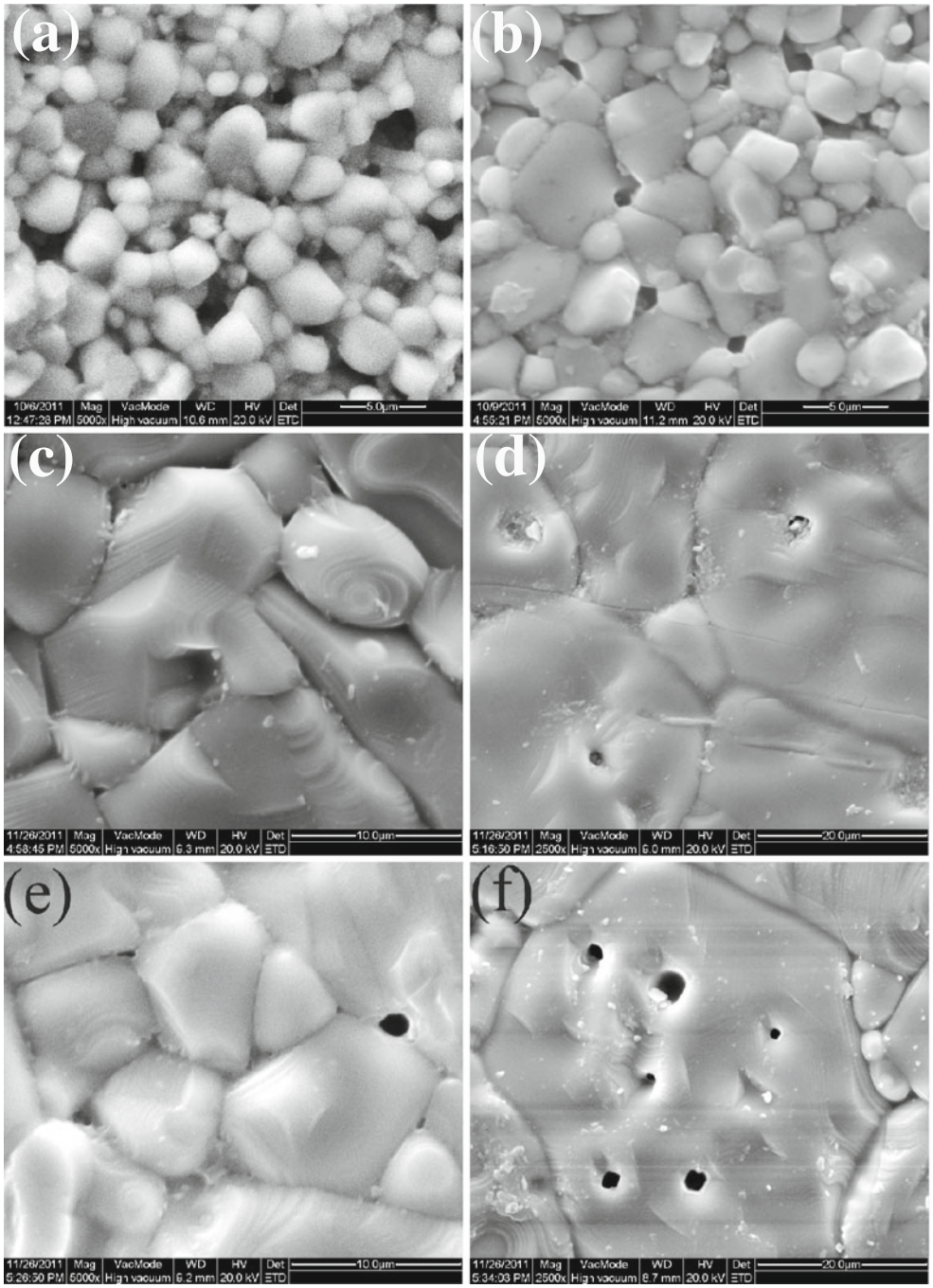


**Fig. 2** Typical surface SEM micrographs of $Li_2MgTi_3O_8$ ceramics prepared at different conditions: **a** 1025 °C/5 h, **b** 1050 °C/5 h, **c** 1100 °C/5 h, **d** 1125 °C/5 h, **e** 1100 °C/3 h, **f** 1100 °C/8 h

increase in Q × f value could be correlated with the effect of grain size, which is due to large grain resulted in less grain boundary, while it decrease was due to the abnormal grain growth and microcracks as shown in Fig. 2d. Moreover, the $\tau_f$ value was almost independent of the sintering temperature (around 1.5 ppm/ °C). This was expected because there was no second phase detected throughout the experiment. Table 1 demonstrates the microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics sintered at 1100 °C for different times. It indicated that the $Li_2MgTi_3O_8$ ceramics sintered at 1100 °C for 5 h has optimum microwave dielectric properties of $\varepsilon_r = 23.03$, Qxf = 54052 GHz, $\tau_f = 1.5$ ppm/ °C.

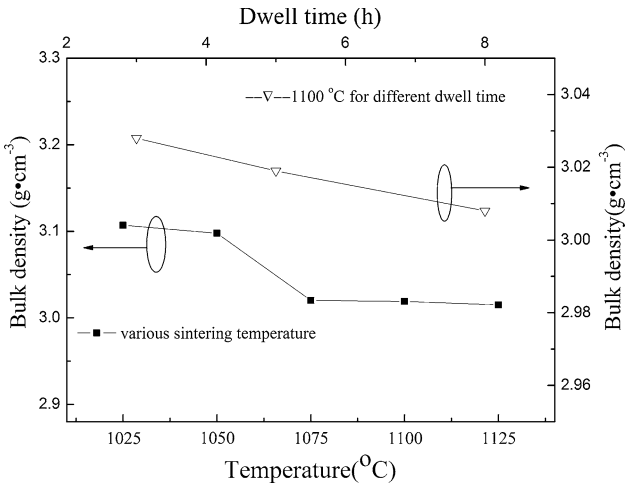


**Fig. 3** Bulk density of $Li_2MgTi_3O_8$ ceramics as a functional of sintering temperature and sintered at 1100 °C for different dwell times

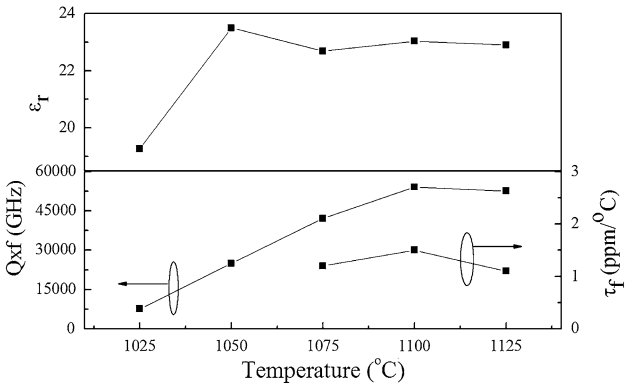


**Fig. 4** Microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics sintered at various temperatures

**Table 1** Microwave dielectric properties of $Li_2MgTi_3O_8$ ceramics sintered at 1100 °C for different dwell times

| dwell time (h) | $\varepsilon_r$ | Qxf (GHz) | $\tau_f$ (ppm/ °C) |
|---|---|---|---|
| 3 | 22.68 | 52534 | 1.98 |
| 5 | 23.03 | 54052 | 1.53 |
| 8 | 23.10 | 52173 | 1.85 |

## 4 Conclusions

The reaction-sintering is a simple and effective method for preparation $Li_2MgTi_3O_8$ ceramics. The $Li_2MgTi_3O_8$ ceramics demonstrated a higher Qf value than those of samples by conventional sintering method, in spite of its higher porosity. With increasing sintering temperature, the bulk density decreased, the $\varepsilon_r$ and $Q \times f$ increased firstly and then decreased, while the $\tau_f$ changed slightly and remained around 1.5 ppm/ °C. Typically, $Li_2MgTi_3O_8$ ceramics sintered by reaction-sintering method at 1100 °C for 5 h exhibited excellent combination microwave dielectric properties of $\varepsilon_r = 23.0$, $Q \times f$=54052 GHz (at 7.29 GHz) and $\tau_f = 1.5$ ppm/ °C.

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