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Letter

Low-loss microwave dielectrics using rock salt oxide Li_2MgTiO_4

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ABSTRACT

Rock-salt-structured Li_2MgTiO_4 ceramic was prepared by the conventional mixed oxide route and its microwave dielectric properties were investigated. The microstructures of the ceramics were characterized by SEM. The dielectric properties of the ceramics exhibited a significant dependence on the sintering condition and crystal structure. A new microwave dielectric material, Li_2MgTiO_4 sintered at 1360 °C has a dielectric constant (ε_r) of ~17.25, a $Q \times f$ of ~97,300 GHz (where f = 9.86 GHz, is the resonant frequency) and a τ_f of ~−27.2 ppm/°C. The microwave dielectric properties of the ceramic are reported for the first time.

1. Introduction

Ceramics with low-dielectric loss and high-dielectric constant are the primary requirement to produce good performance passive components such as dielectric resonators, filters and antennas. Moreover, a rapid growth of the wireless communication market in recent years has further led to extensive research and development in the area of microwave dielectrics. Although some materials with excellent properties have been developed for commercial applications [1–3], active work is still being carried out in search of sophisticated materials to adapt different frequency band applications [4–9].

Previous studies of rock salt type oxides containing lithium showed that these materials are potential ionic conductors and the Li_2MTiO_4 (M = Mn, Fe, Co, and Ni) dielectrics were first investigated as a new series of lithium cathode materials [10,11]. Mg_2TiO_4, however, is extensively studied as dielectric material because of its dielectric constant is stable with temperature and frequency [2]. Therefore, the introduction of lithium in magnesium titanium oxides is of great interest for the generation of new dielectrics. In fact, Li_2MgTiO_4 were prepared via solid-state and sol–gel methods and its low-frequency dielectric properties were also investigated [11]. However, to the best of our knowledge, the microwave dielectric properties of this composition have not been reported to date.

2. Experimental procedure

Mixed oxide powders of Li_2MgTiO_4 were prepared from Li_2CO_3, MgO and TiO_2 with purity higher than 99.9% by conventional solid state method. Because MgO is hygroscopic, it was dried at 600 °C for 5 h to remove moisture retains. The weighed raw materials were mixed by ball milling with agate media in alcohol for 24 h, and the mixtures were dried and calcined at 1000 °C for 2 h. The calcined powders were dried and ball milled for 24 h. The fine powder with 3 wt% of a 10% solution of PVA as a binder (PVA 500, Showa, Japan), granulated by sieving through a 200 mesh, and pressed into pellets with 11 mm in diameter and 5 mm in thickness. All samples were prepared using an automatic uniaxial hydraulic press at 2000 kg/cm^2. These pellets were sintered at temperatures of 1300–1420 °C for 2 h in air. The heating rate and the cooling rate were both set at 10 °C/min.

The crystal phases of the sintered ceramics were identified by XRD using Cu Kα (λ = 0.15406 nm) radiation with a Siemens D5000 diffractometer (Munich, Germany) operated at 40 kV and 40 mA. The microstructures were evaluated for thermal-etched surfaces by scanning electron microscopy (SEM; Philips XL-40FEG, Eindhoven, the Netherlands). The apparent densities of the sintered pellets were

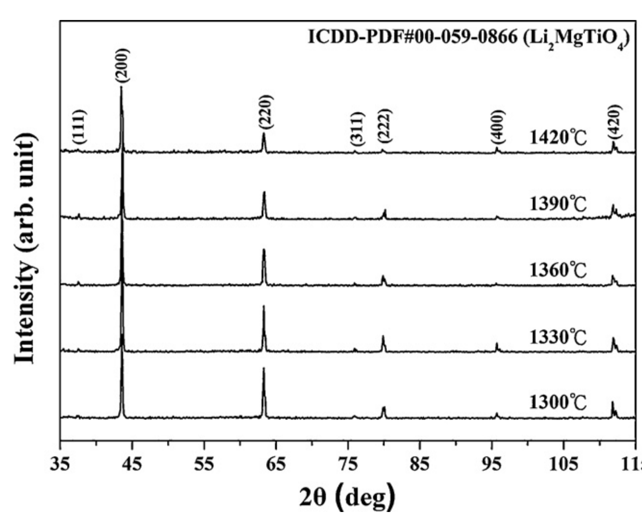

Fig. 1. X-ray diffraction patterns of Li_2MgTiO_4 ceramics sintered at different temperatures for 2 h.

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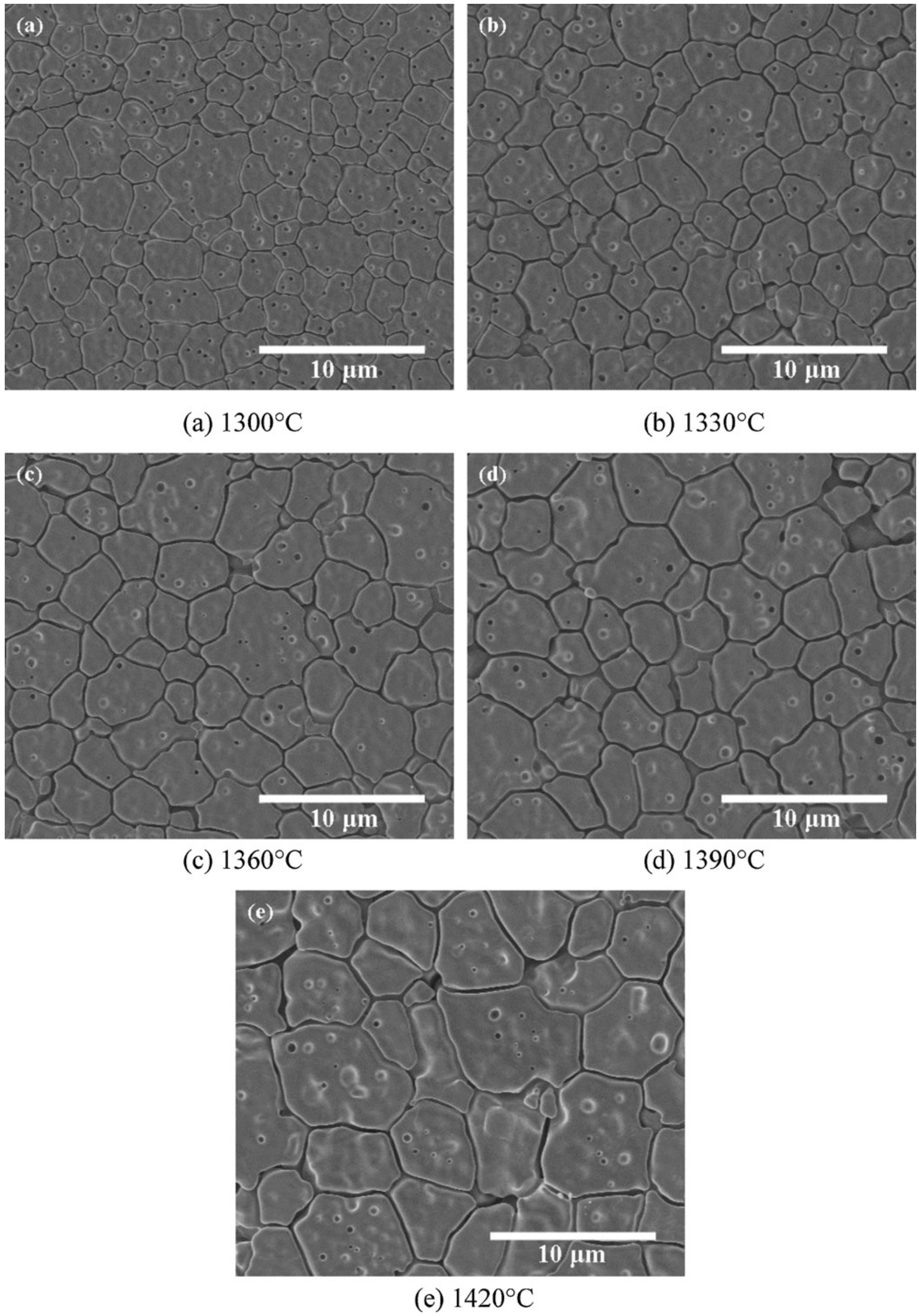

Fig. 2. Secondary electron SEM micrographs of Li_2MgTiO_4 ceramics sintered at (a) 1300, (b) 1330, (c) 1360, (d) 1390, and (f) 1420 °C for 2 h.

measured by commonly the Archimedes method. The dielectric constant (ε_r) and the quality factor values (Q) at microwave frequencies were measured using the Hakki–Coleman dielectric resonator method [12,13]. A system combining an HP8757D network analyzer (HP, Palo Alto, CA) and an HP8350B sweep oscillator (HP, Palo Alto, CA) was employed in the measurement. For temperature coefficient of resonant frequency (τ_f), the technique is the same as that of quality factor measurement. The test cavity was placed over a thermostat in the temperature range of 25–80 °C. The τ_f value (ppm/°C) was calculated by noting the change in resonant frequency (Δf)

$$\tau_f = \frac{f_2 - f_1}{f_1(T_2 - T_1)} \tag{1}$$

where f_1 andf_2 represent the resonant frequencies at T_1 and T_2, respectively.

3. Results and discussion

Fig. 1 illustrates the XRD patterns recorded from the Li_2MgTiO_4 ceramics sintered at different temperatures for 2 h. The diffraction pattern of rock-salt-structured Li_2MgTiO_4 is fully indexed as a single cubic crystal (ICDD-PDF#00-059-0866), belonging to the space group Fm-3m (225).

The thermal-etched section microstructural photographs of Li_2MgTiO_4 ceramics sintered at 1300–1420 °C are illustrated in Fig. 2. The average grain size (AGS) of Li_2MgTiO_4 ceramics obviously increased from 2.1 to 4.3 µm as the sintering temperature increased from 1300 to 1420 °C. The small AGS, in turn, would give rise to

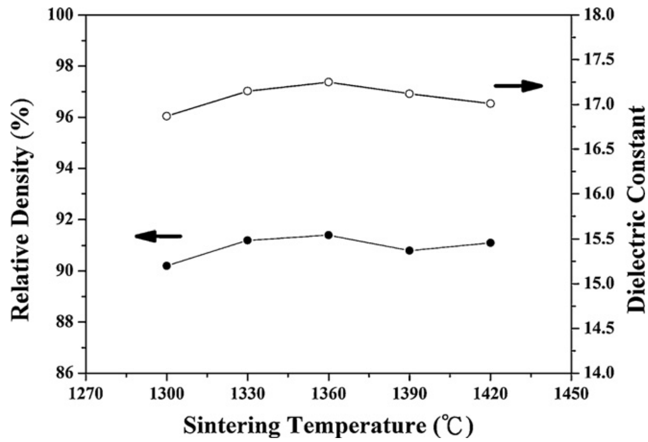

Fig. 3. Relative density and dielectric constant of Li_2MgTiO_4 ceramics sintered at different temperatures for 2 h.

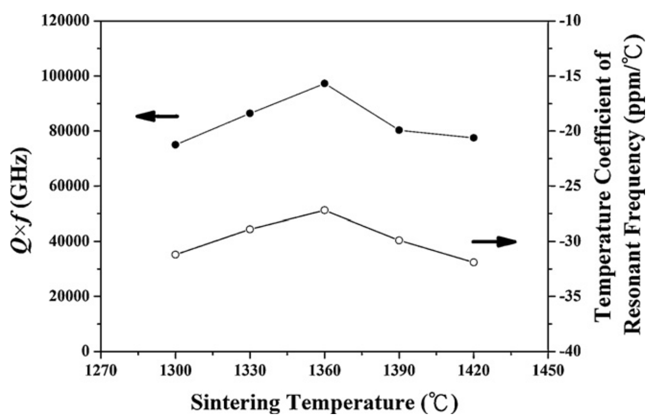

Fig. 4. $Q \times f$ and τ_f values of Li_2MgTiO_4 ceramics sintered at different temperatures for 2 h.

a great amount of grain boundary. As illustrated in Fig. 2, small pores existed in the grains in all specimens, causing the low relative densities in Li_2MgTiO_4 ceramics. However, some pores started to appear at grain boundary for specimen sintered at 1420 °C, which might be a result from an over-sintering.

Fig. 3 demonstrates the relative density and dielectric constant of the Li_2MgTiO_4 ceramics as a function of its sintering temperature for 2 h. The theoretical density of Li_2MgTiO_4 is 3.465 g/cm^3 and the densities apparently increased with increasing sintering temperature. After reaching its maximum at 1360 °C, it slightly decreased. Variation of dielectric constant was consistent with that of density and the highest dielectric constant of ∼17.25 can be achieved at 1360 °C. The dielectric constant primarily depends on the composition and to some extent on the grain size and the density. It seemed that the dielectric constant was mainly controlled by the density of the specimen. In addition, increasing sintering temperature does not necessarily lead to a higher dielectric constant.

The $Q \times f$ and τ_f (temperature coefficients of resonant frequency) values of the Li_2MgTiO_4 ceramics as a function of its sintering temperature conducted for 2 h are demonstrated in Fig. 4. Variation of $Q \times f$ was also consistent with that of density. The quality factor ($Q \times f$) values of Li_2MgTiO_4 samples progressively increased from 75,000 to the highest of 97,300 GHz as the sintering temperature increased from 1300 to 1360 °C. Densification of the ceramics plays an important role in controlling the dielectric loss, and same phenomenon has been shown for other microwave dielectric materials.

Table 1
Microwave dielectric properties of Li_2MgTiO_4 ceramics system sintered at different temperatures for 2 h.

Sintering temperature (°C)	Relative density (%)	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)
1300	90.23	16.87	75,000	−31.2
1330	91.23	17.15	86,400	−28.9
1360	91.45	17.25	97,300	−27.2
1390	90.77	17.12	80,300	−29.9
1420	91.13	17.01	77,500	−31.9

However, it has reported that density has little effect on the $Q \times f$ when relative density is above 90%. Therefore, it may be extrinsic effect on the microwave dielectric loss. Specimen sintered at 1360 °C shows not only less pores but also a relatively uniform morphology which reduces the lattice defects and imperfection leading to a lowering of dielectric loss. Further, the $Q \times f$ data over the entire temperature range (1300–1420 °C) show that specimen using Li_2MgTiO_4 can be considered as a high- and stable-$Q \times f$ material. The τ_f value is well-known to be influenced by the composition, the additive and the second phase of the materials. It was not sensitive to the sintering temperature because there was no alternate composition at different temperatures and the unit cell volume retained constant. Consequently, the τ_f value would remain in the range from −27.2 to −31.9 ppm/°C for specimens sintered at 1300–1420 °C.

Table 1 summarizes the microwave dielectric properties of Li_2MgTiO_4 ceramics system sintered at different temperatures for 2 h. It indicates that 1360 °C is the optimized sintering temperature for specimen to achieve an excellent combination of microwave dielectric properties.

4. Conclusion

The dielectric properties of Li_2MgTiO_4 ceramics have been studied at the microwave frequency regions. The best dielectric properties of the material were obtained at 1360 °C. Specimen using Li_2MgTiO_4 has an ε_r of ∼17.25, a $Q \times f$ of ∼97,300 GHz, and a τ_f of ∼−27.2 ppm/°C. The system has an excellent combination of microwave dielectric properties, which makes it a suitable candidate for applications as microwave passive components.

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