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Ceramics International 39 (2013) 5185–5190

CERAMICS INTERNATIONAL

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Microwave dielectric characteristics of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) ceramics

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Received 16 September 2012; received in revised form 26 November 2012; accepted 5 December 2012
Available online 13 December 2012

Abstract

$Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) ceramics were synthesized by the conventional solid-state reaction route. The effect of M (Zn, Co, and Mn) substitution on the structure, microstructure and microwave dielectric properties of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ has been investigated. The XRD patterns of sintered samples revealed the single-phase formation with spinel structure. With the increase in ionic radius of M, the *Qf* value decrease is attributed to the decrease of packing fraction and grain size. The $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ ceramic sintered at 1075 °C for 4 h showed the best microwave dielectric properties with a dielectric constant of 27.1, a *Qf* value of 44 800 GHz, and a temperature coefficient of resonant frequency of (+)1.9 ppm/°C.

Keywords: Ceramics; Microwave dielectric properties; LMT; X-ray diffraction

1. Introduction

In recent years, microwave dielectric materials play an important part in wireless industry with a wide range of applications from satellite communication and microwave telecommunication to intelligent transport systems [1]. They are widely applied in many kinds of indispensable components such as filters, dielectric resonators and antennas in microwave communication systems [2]. Moreover, to meet the requirements for the rapid growth of cell phone and wireless communication markets, the development of moderate dielectric constant, and low dielectric loss ceramics have received more attention than ever, and further research is still underway [1–5].

Many commercial microwave dielectric ceramics, such as $Ba(B'_{1/3}B''_{2/3})O_3$ (B′ = Mg, Zn; B″ = Ta, Nb), $(Zr_{1-x}Sn_x)TiO_4$, $Ba_2Ti_9O_{20}$, and $MgTiO_3$–$CaTiO_3$ show good microwave dielectric properties with high *Qf* value. However, either they contain expensive chemical elements such as tantalate or niobate, or need high temperature sintering (> 1400 °C) with long annealing time [6–8]. In addition, Hernandez et al. reported the structure and polymorphism of ternary spinal-like phases $Li_{1.33x}Zn_{2-2x}Ti_{1+0.67x}O_4$ ($0.50 < x < 0.90$) [9], and West and colleagues reported on the crystalline structure of ternary complex lithium spinels $Li_2MM'_3O_8$ (M = Mg, Co, Ni, Zn; M′ = Ti, Ge) [10]. However, the microwave dielectric properties have not been investigated in their studies. More recently, Sebastian et al. first reported microwave dielectric properties of $Li_2MgTi_3O_8$ and $Li_2ZnTi_3O_8$ ceramics with ε_r about 25.6–27.5, *Qf* up to 72 000 GHz and τ_f in the range of (−)15–(+)3.2 ppm/°C [11,12]. The excellent microwave dielectric properties, low sintering temperature (< 1100 °C), and low bulk density as well as inexpensive raw materials make the new materials very attractive, and much research work has been done in recent years. For example, the $Li_2CoTi_3O_8$ materials reported by Zhou et al. also showed good combination properties of $\varepsilon_r = 28.9$, *Qf* = 52 600 GHz, τ_f = (+)7.4 ppm/°C [13]. With partial replacement of Zn by Mg and Co, the value of *Qf* could be dramatically promoted to 150 000 GHz for the $Li_2(Zn_{1-x}A_x)Ti_3O_8$ (A = Mg, Co; $x = 0.02$–0.1) solid solutions, associated with an ε_r of ~26.1, and a small τ_f of ~(−)13.4 ppm/°C at A = Mg, $x = 0.05$ [14]. Such phenomenon was also proved to be valid for other ceramics

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http://dx.doi.org/10.1016/j.ceramint.2012.12.015

[5,15,16]. Since the ionic radii of Zn^{2+} (0.74 Å, CN = 6), Co^{2+} (0.745 Å, CN = 6), and Mn^{2+} (0.83 Å, CN = 6) are close to that of Mg^{2+} (0.72 Å, CN = 6) [17], it should be of interest to characterize the dielectric response of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) solid solution systems at microwave frequencies.

In this paper, $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) ceramics were prepared by the solid-state reaction route. Their crystal structure, ceramic microstructure, and microwave dielectric properties were analyzed using a combination of techniques, including X-ray diffraction (XRD), scanning electron microscopy (SEM), and microwave dielectric resonance measurements.

2. Experimental procedure

$Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) powders were prepared by the conventional solid-state reaction route. The starting materials were high-purity grade (99.99%) Li_2CO_3, $(MgCO_3)_4 \cdot Mg(OH)_2 \cdot 5H_2O$, ZnO, Co_2O_3, MnO_2 and TiO_2. The powders were weighed according to the stoichiometry and intimately mixed by ball milling in distilled water for 6 h, using yttria-stabilized zirconia balls. After drying at 100 °C, the powder was calcined at 900 °C for 4 h, and remilled for 12 h. It was then mixed with 7 wt% PVA and again dried and ground well. Cylindrical disks of about 12 mm diameter and 5 mm height were produced under a pressure of 350 MPa. The green compacts were subsequently sintered for 4 h between 1000 °C and 1125 °C in air, using a controlled heating–cooling rate of 5 °C/min.

Phase purity and crystallinity of sintered specimens were studied by the X-ray diffraction (XRD) method using Cu $K\alpha$ radiation (D/max 2500v/pc, Rigaku, Japan). Structure analyses were carried out by the Rietveld refinement. The ceramic microstructures of as-fired surfaces were examined using a SEM (Phillips, XL30, Antilles, Netherlands) operated at 15 kV. The sintered density of the specimen was measured by the Archimedes method. The packing fraction was obtained by the summation of the volume of packed ions over the volume of a primitive unit cell, which could be calculated from Eq. (1) [18]:

$$\textit{Packing fraction}(\%) = \frac{\textit{Volume of the atoms in the cell}}{\textit{Volume of primitive unit cell}} = \frac{\textit{Volume of the atoms in the cell}}{\textit{Volume of unit cell}} \times Z \quad (1)$$

where Z is the number of formula units per unit cell.

The microwave dielectric properties were measured by a Vector Network Analyzer (Model no: N5230C, Agilent Technologies, Santa Clara, CA). The relative permittivity of samples was measured by the Hakki–Coleman method [19], and unloaded quality factor was measured by the cavity method [20]. The temperature coefficient of resonant frequency (τ_f) was measured in the temperature range 25–85 °C. The resonant frequency was noted at regular intervals in the course of heating and the τ_f was calculated using the following equation:

$$\tau_f = \frac{f_2 - f_1}{f_1(T_2 - T_1)} \quad (2)$$

where f_1 is the resonant frequency at temperature T_1 and f_2 at temperature T_2.

3. Results and discussion

The room temperature XRD patterns of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) ceramics sintered at 1075 °C for 4 h are shown in Fig. 1. All the observed (*hkl*) reflections could be indexed based on JCPDS file number 48-0263 for $Li_2MgTi_3O_8$ with cubic crystal symmetry, belonging to the space group $P4_332$ (212), and no additional peaks were observed. In addition, the reflection peaks of the cubic phases slightly shifted toward the higher angle with the substitution of Mg by M (M = Zn, Co, and Mn), which suggested a variation in the unit-cell dimension. In this study, Co and Mn ions were thermodynamically most stable in the valence state of 2 in the sintering temperature range, and following reactions could be used to describe the valence state changes of Co and Mn ions:

$$2Mn^{4+} + 2O^{2-} \rightarrow 2Mn^{2+} + O_2 \quad (3)$$

$$4Co^{3+} + 2O^{2-} \rightarrow 4Co^{2+} + O_2 \quad (4)$$

In order to perform an extensive investigation on the structure and formation of solid solution, lattice parameters, cell volume and theoretical densities of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M = Zn, Co, and Mn) ceramics sintered at 1075 °C for 4 h were measured and presented in Table 1. Because the ionic radii of M^{2+} (0.74 Å for Zn^{2+}, 0.745 Å for Co^{2+}, and 0.83 Å for Mn^{2+}) are similar to that of Mg^{2+} (0.72 Å) [17], Mg^{2+} ions are most likely to be substituted by M^{2+} ions. Both lattice parameters and cell volume showed a small monotonic increase with increasing ionic radii of M^{2+}, for Mg^{2+} ion had a smaller ionic radius than M^{2+} ion. In the case of substituting Mg^{2+} by

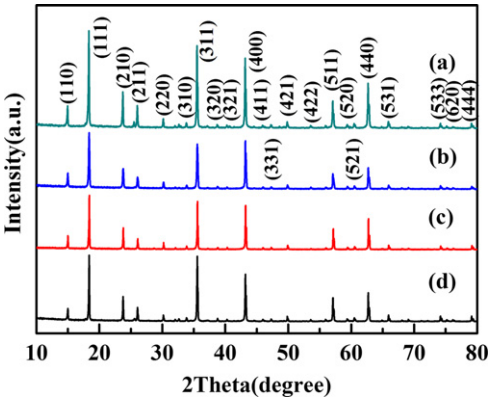

Fig. 1. X-ray diffraction patterns of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C for 4 h with (a) $Li_2MgTi_3O_8$, (b) M = Zn, (c) M = Co, and (d) M = Mn.

Table 1
Lattice parameters, cell volume and theoretical densities of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C for 4 h.

Composition	Lattice parameter $a=b=c$ (Å)	Cell volume (Å^3)	Theoretical density (g/cm^3)	Ref.
$Li_2MgTi_3O_8$	8.3810	588.60	3.5011	PDF #01-48-0263
$Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$	8.3826 ($\pm$0.0005 Å)	588.98	3.5067	This work
$Li_2(Mg_{0.94}Co_{0.06})Ti_3O_8$	8.3830 ($\pm$0.0007 Å)	589.12	3.5123	This work
$Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$	8.3839 ($\pm$0.0006 Å)	589.31	3.5055	This work

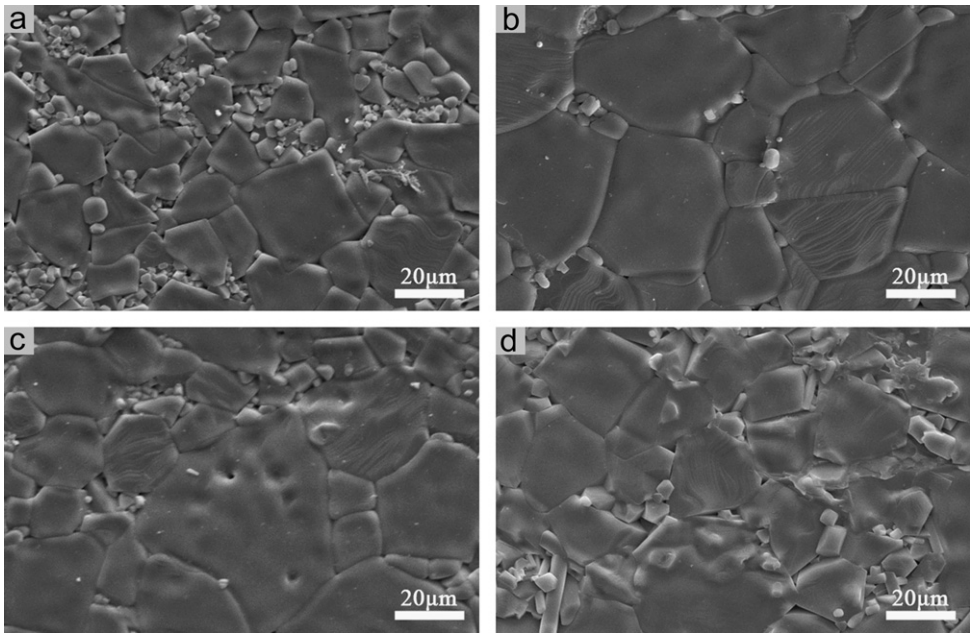

Fig. 2. Scanning electron microscopic micrographs of $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ ceramics sintered at (a) 1050 °C, (b) 1075 °C, (c) 1100 °C for 4 h, and (d) $Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C for 4 h.

Mn^{2+}, the difference between different ionic radii increased to 15.3% and led to an increase in the lattice parameters from $a=b=c=8.381$ Å in $Li_2MgTi_3O_8$ to $a=b=c=8.3839$ Å in $Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$. However, it still retained a spinel structure, implying the formation of a solid solution.

Fig. 2 shows the SEM micrographs of $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ ceramics at different sintering temperatures and $Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$ at 1075 °C for 4 h. For $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ sintered at 1050 °C, distributions of larger grains (~20 μm) embedded in smaller grains (~5 μm) and a small amount of pores were observed. The increase of sintering temperature helped to promote the grain growth, and a significant increase in the grain size was achieved when sintered at 1075 °C, which showed a dense microstructure with grain sizes ranging from 30 to 40 μm. However, as the sintering temperature reached 1100 °C, a porous microstructure started to appear due to the abnormal grain growth and size of some grains were large up to 45 μm. In the case of substituting Mg by Mn^{2+} (0.83 Å, CN=6), the ionic radius difference increased to 15.3% and small amount of Mn^{2+} tended to segregate along the grain boundary, which inhibited the movement of grain boundary, thereby leading to a decrease in grain size as shown in Fig. 2(d). However, a small amount of porosity could still be observed, and it would certainly damage the microwave dielectric properties of the ceramics.

The variation in relative density and relative permittivity (ε_r) of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M=Zn, Co, Mn) ceramics with sintering temperature for 4 h is shown in Fig. 3. The relative density first increased with the increase in sintering temperature up to a maximum at 1075 °C for specimens with M=Zn, Co, and Mn, and then decreased slightly with the increase in sintering temperature. The increased relative density could be contributed to the elimination of pores in the ceramics, and the decrease in relative density above 1075 °C could be ascribed to trapped porosity caused by the evaporation of lithium and abnormal grain growth due to over-sintering [21]. The variation in ε_r with sintering temperature followed the same trend due to the fact that ε_r increased with densification, and a high ε_r of 27.1 could be achieved for $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ when sintered at 1075 °C for 4 h. In addition, the ε_r only showed a small variance with M=Zn, Co, and Mn implying that the influence of composition on ε_r was almost negligible.

On the other hand, the relative permittivity of microwave dielectric ceramics is known to be affected by the molecular volume and ionic polarizability [22]. The observed ionic polarizabilities (α_{obs}) of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ were estimated in order to clarify the effects of Zn^{2+}, Co^{2+}, and Mn^{2+} substitution for Mg^{2+} on the relative permittivity by using the Clausius–Mosotti equation [23] following Eq. (5):

$$\alpha_{obs} = \frac{1[Vm(\varepsilon_r - 1)]}{b(\varepsilon_r + 2)} \quad (5)$$

where Vm and ε_r represent the molar volume and measured relative permittivity, respectively. The molar volumes are determined from the unit cell volumes and formula number Z (in the case of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$, $Z=4$), and the constant value b is $4\pi/3$. The obtained α_{obs} values shown in Table 2 exhibited similar variations in the relative permittivity as shown in Fig. 3, suggesting that in addition to the density, the variance in the relative permittivity could be attributed to the change in ionic polarizability rather than the molar volume in $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics.

Fig. 4 shows the Qf values of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M=Zn, Co, and Mn) ceramics as a function of sintering temperatures. The microwave dielectric loss in ceramics may arise from a combination of extrinsic and intrinsic factors. Extrinsic losses associated with the microstructural imperfections, such as porosity, second phases, grain boundaries, and density, and intrinsic ones related with the lattice vibration modes that can be expected for a particular composition and crystal structure of materials [24,25]. As to the extrinsic factors, densification played an important role in controlling the dielectric, and the variation of Qf was also consistent with that of the density. The Qf values of specimens sintered at 1000 °C were relatively low due to the low densities, as shown in Fig. 3. By increasing the sintering temperature, the Qf gradually increased to a maximum value of 44 800 GHz for M=Zn, 43 500 GHz for M=Co, and 39 700 GHz for M=Mn at 1075 °C, and it decreased thereafter. It has been reported that the impurities, even in trace amounts, cause an increase in the dielectric loss of ceramics [26]. The sintering process sweeps the impurities to the grain boundaries so that the impurities and grain boundaries are linked inextricably [27]. For small grains, there is a possibility of accumulation of impurities at the grain boundaries, which is less for larger grains and would affect the quality factor [28]. It is generally accepted that the ceramics having larger grains show relatively better microwave dielectric properties without considering the inherent properties. Therefore, a lower Qf value of $Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$ compared with that of $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ was attributed to Mn segregating on the grain boundary that resulted in an increase in the dielectric loss of ceramics. Kim et al. [18] reported that the Qf was closely related to the packing fraction of the structure. With the increase of the packing fraction, the lattice vibrations decreased, and then the Qf value would increase. As a result, the intrinsic

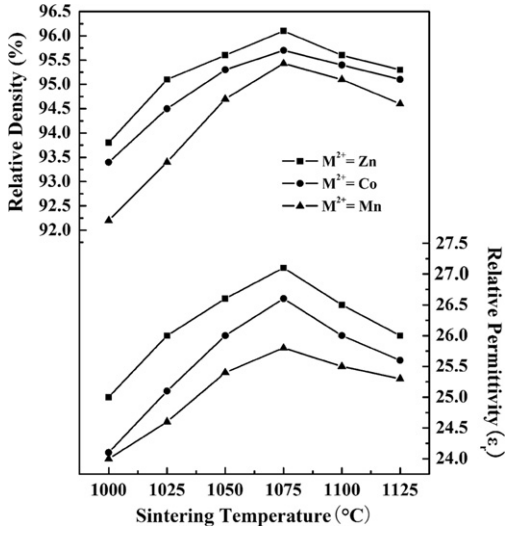

Fig. 3. Relative density and relative permittivity (ε_r) of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at different temperatures for 4 h.

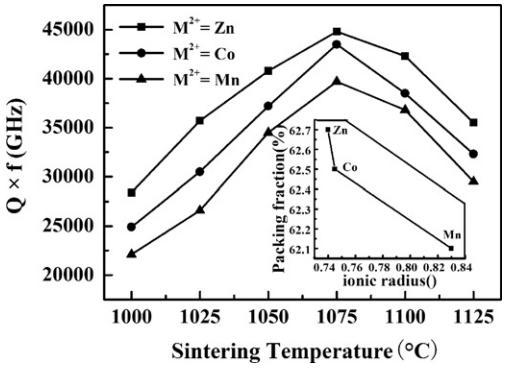

Fig. 4. Qf value of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at different temperatures for 4 h. The inset showed the packing fraction(%) of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C.

Table 2
Molar volume and observed ionic polarizability of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C for 4 h.

Composition	Cell volume (Å^3)	Molar volume (Å^3/mol)	ε_r	α_{obs} (Å^3)
$Li_2MgTi_3O_8$	588.60	147.15	27.2	31.55
$Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$	588.98	147.24	27.1	31.53
$Li_2(Mg_{0.94}Co_{0.06})Ti_3O_8$	589.12	147.28	26.6	31.48
$Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$	589.31	147.33	25.8	31.39

Table 3
Relative densities and microwave dielectric properties of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C for 4 h.

Composition	Sintering temperature (°C)	Relative density (%)	ε_r	Qf (GHz)	τ_f (ppm/°C)	Ref.
$Li_2MgTi_3O_8$	1075	95.5	27.2	42 000	(+) 3.2	[11]
$Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$	1075	96.1	27.1	44 800	(+) 1.9	This work
$Li_2(Mg_{0.94}Co_{0.06})Ti_3O_8$	1075	95.7	26.6	43 500	(+) 4.3	This work
$Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$	1075	95.4	25.8	39 400	(−) 12.5	This work

loss decreases and Qf value increases [29]. In this work, the packing fraction depended on the composition of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M=Zn, Co, and Mn) solid solutions. As shown in the inset of Fig. 4, with the increase in ionic radius of M^{2+} (0.74 Å for Zn^{2+}, 0.745 Å for Co^{2+}, and 0.83 Å for Mn^{2+}), the packing fraction of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M=Zn, Co, and Mn) ceramics would decrease. For this reason, the Qf value decreased with the increase in ionic radius of M.

The relative density and microwave dielectric properties of $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M=Zn, Co, and Mn) solid solution systems under optimum sintering conditions (1075 °C) are summarized in Table 3. It is well known that the τ_f depends on the composition, additives, and second phase of materials, and almost independent of the sintering temperature. It was seen that with substitution of a slightly larger Mn^{2+} (0.83 Å) for a smaller Mg^{2+} (0.72 Å), the τ_f appeared to possess a more negative value of (−)12.5 ppm/°C. This could be a result from a large ionic radius difference (15.3%) between Mg and Mn, which would lead to the change in cell volume and the increase of local lattice distortion. With the increase in ionic radius of M^{2+} (0.74 Å for Zn^{2+}, 0.745 Å for Co^{2+}, 0.83 Å for Mn^{2+}) in $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ ceramics, the relative permittivity (ε_r) slightly decreased from 27.1 to 25.8, the Qf value decreased from 44 800 to 39 400 GHz and the τ_f was in the range from (−)12.5 to (+)4.3 ppm/°C. It was noted that the specimen $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ exhibited a near-zero τ_f of (+)1.9 ppm/°C in combination with a high Qf value of 44 800 GHz and a moderate ε_r of 27.1.

4. Conclusions

The spinel-structured $Li_2(Mg_{0.94}M_{0.06})Ti_3O_8$ (M=Zn, Co, and Mn) solid solution systems were prepared by the conventional solid-state ceramic route. And the microwave dielectric properties of these ceramics were strongly related to the variation of composition. Samples with 0.06 mol% replacement of Mg by Zn and Co were both effective in improving the Qf. In addition, a relatively low Qf value of 39 700 GHz was obtained for $Li_2(Mg_{0.94}Mn_{0.06})Ti_3O_8$ ceramics due to the decrease of packing fraction, density, and grain size. The good microstructure and microwave dielectric properties could be achieved for $Li_2(Mg_{0.94}Zn_{0.06})Ti_3O_8$ ceramics sintered at 1075 °C for 4 h with ε_r=27.1, Qf=44 800 GHz and τ_f=(+)1.9 ppm/°C.

References

[1] N.I. Santha, M.T. Sebastian, Microwave dielectric properties of $A_6B_5O_{18}$-type perovskites, Journal of the American Ceramic Society 90 (2007) 496–501.

[2] C.L. Huang, C.Y. Tai, C.Y. Huang, Y.H. Chien, Low-loss microwave dielectrics in the spinel-structured $(Mg_{1-x}Ni_x)Al_2O_4$ solid solutions, Journal of the American Ceramic Society 93 (2010) 1999–2003.

[3] C.W. Zheng, S.Y. Wu, X.M. Chen, K.X. Song, Modification of $MgAl_2O_4$ microwave dielectric ceramics by Zn substitution, Journal of the American Ceramic Society 90 (2007) 1483–1486.

[4] M.E. Song, J.S. Kim, M.R. Joung, S. Nahm, Y.S. Kim, J.H. Paik, B.H. Choi, Synthesis and microwave dielectric properties of $MgSiO_3$ ceramics, Journal of the American Ceramic Society 91 (2008) 2747–2750.

[5] C.L. Huang, J.J. Wang, Y.P. Chang, Dielectric properties of low loss $(1-x)(Mg_{0.95}Zn_{0.05})TiO_3$–$xSrTiO_3$ ceramic system at microwave frequency, Journal of the American Ceramic Society 90 (2007) 858–862.

[6] M.R. Varma, M.T. Sebastian, Effect of dopants on microwave dielectric properties of $Ba(Zn_{1/3}Nb_{2/3})O_3$ ceramics, Journal of the European Ceramic Society 27 (2007) 2827–2833.

[7] R. Laishram, O.P. Thakur, D.K. Bhattacharya, Improvement in quality factor of $(Zr_{0.8}Sn_{0.2})TiO_4$ microwave ceramics by Mn^{4+} doping, Materials Letters 65 (2011) 1678–1680.

[8] J.K. Plourde, D.F. Linn, H.M. O'Bryan, J.J. Thompson, $Ba_2Ti_9O_{20}$ as a microwave dielectric resonator, Journal of the American Ceramic Society 58 (1975) 418–420.

[9] V.S. Hernandez, L.M.T. Martinez, G.C. Mather, A.R. West, Stoichiometry, structures and polymorphism of spinel-like phases, $Li_{1.33x}Zn_{2-2x}Ti_{1+0.67x}O_4$, Journal of Materials Chemistry 6 (1996) 1533–1536.

[10] H. Kawai, M. Tabuchi, M. Nagata, H. Tukamoto, A.R. West, Crystal chemistry and physical properties of complex lithium spinels $Li_2MM'_3O_8$ (M=Mg, Co, Ni, Zn; M'=Ti, Ge), Journal of Materials Chemistry 8 (1998) 1273–1280.

[11] S. George, M.T. Sebastian, Synthesis and microwave dielectric properties of novel temperature stable high Q, $Li_2ATi_3O_8$ (A=Mg, Zn) ceramics, Journal of the American Ceramic Society 93 (2010) 2164–2166.

[12] S. George, M.T. Sebastian, Low-temperature sintering and microwave dielectric properties of $Li_2ATi_3O_8$ (A=Mg, Zn) ceramics, Journal of Applied Ceramic Technology 8 (2011) 1400–1407.

[13] L. Fang, D. Chu, H. Zhou, X. Chen, Z. Yang, Microwave dielectric properties and low temperature sintering behavior of $Li_2CoTi_3O_8$ ceramic, Journal of Alloys and Compounds 509 (2011) 1880–1884.

[14] C.L. Huang, C.H. Su, C.M. Chang, E. Leite, High Q microwave dielectric ceramics in the $Li_2(Zn_{1-x}A_x)Ti_3O_8$ (A=Mg, Co; x=0.02–0.10) system, Journal of the American Ceramic Society 94 (2011) 4146–4149.

[15] C.L. Huang, C.H. Shen, Phase evolution and dielectric properties of $(Mg_{0.95}M^{2+}_{0.05})Ti_2O_5$ (M^{2+}=Co, Ni, and Zn) ceramics at microwave frequencies, Journal of the American Ceramic Society 92 (2009) 384–388.

[16] C.L. Huang, J.Y. Chen, Microwave dielectric characteristics of $(Mg_{0.95}M_{0.05})Ta_2O_6$ (M=Ni, Zn, Mn) ceramic series, Materials Letters 76 (2012) 28–31.
[17] R.D. Shannon, Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides, Acta Crystallographica A 32 (1976) 751–767.
[18] E.S. Kim, B.S. Chun, R. Freer, R.J. Cernik, Effects of packing fraction and bond valence on microwave dielectric properties of $A^{2+}B^{6+}O_4$ (A^{2+}: Ca, Pb, Ba; B^{6+}: Mo, W) ceramics, Journal of the European Ceramic Society 30 (2010) 1731–1736.
[19] B.W. Hakki, P.D. Coleman, A dielectric resonator method of measuring inductive capacities in the millimeter range, IEEE Transactions on Microwave Theory and Techniques 8 (1960) 402–410.
[20] D. Kajfez, S. Chebolu, M.R. Abdul-Gaffoor, A.A. Kishk, Uncertainty analysis of the transmission-type measurement of Q-factor, IEEE Transactions on Microwave Theory and Techniques 47 (1999) 367–371.
[21] S. George, M.T. Sebastian, Microwave dielectric properties of novel temperature stable high Q $Li_2Mg_{1-x}Zn_xTi_3O_8$ and $Li_2A_{1-x}Ca_xTi_3O_8$ (A=Mg, Zn) ceramics, Journal of the European Ceramic Society 30 (2010) 2585–2592.
[22] R.D. Shannon, Dielectric polarizabilities of ions in oxides and fluorides, Journal of Applied Physics 73 (1993) 348–366.
[23] H. Ogawa, A. Kan, S. Ishihara, Y. Higashida, Crystal structure of corundum type $Mg_4(Nb_{2-x}Ta_x)O_9$ microwave dielectric ceramics with low dielectric loss, Journal of the European Ceramic Society 23 (2003) 2485–2488.
[24] S.J. Penn, N.M. Alford, A. Templeton, X. Wang, M.S. Xu, M. Reece, K. Schrapel, Effect of porosity and grain size on the microwave dielectric properties of sintered alumina, Journal of the American Ceramic Society 80 (1997) 1885–1888.
[25] H. Tamura, Microwave dielectric losses caused by lattice defects, Journal of the European Ceramic Society 26 (2006) 1775–1780.
[26] N.M. Alford, S.J. Penn, Sintered alumina with low dielectric loss, Journal of Applied Physics 80 (1996) 5895–5898.
[27] J.D. Breeze, J.M. Perkins, D.W. McComb, N.M. Alford, Do grain boundaries affect microwave dielectric loss in oxides, Journal of the American Ceramic Society 92 (2009) 671–674.
[28] V.L. Gurevich, A.K. Tagantsev, Intrinsic dielectric loss in crystals, Advances in Physics 40 (1991) 719–767.
[29] Q. Liao, L. Li, X. Ren, X. Ding, E.S. Kim, New low-loss microwave dielectric material $ZnTiNbTaO_8$, Journal of the American Ceramic Society 94 (2011) 3237–3240.