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Dielectric properties of high-Q $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics at microwave frequency

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Abstract

$(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) ceramics were prepared via the conventional solid-state method and their dielectric properties were investigated in the microwave frequency region. Formation of solid solutions was confirmed by the X-ray diffraction analysis, the EDX analysis, and the measured lattice parameters. A small amount of Zn substitution for Mg produced a large increase in $Q \times f$ due to a high packing fraction as well as a high relative density of the ceramics. By increasing x from 0.00 to 0.06, the $Q \times f$ of the specimen could be tremendously improved from 141,000 to a maximum of 210,700 GHz (at 10.52 GHz), demonstrating an unique potential for low-loss microwave applications. The τ_f values were found to retain in the range from −54.2 to −62.3 ppm/°C for all compositions because the resultant change of unit cell volume was small. Also, a remarkable lowering of the sintering temperature down to ∼300 °C was observed when Mg was totally substituted by Zn, thereby making it possible for low firing applications.

Keywords: Microwave dielectric properties; High-Q

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1. Introduction

The continuing development of microwave dielectric materials for applications including substrates, resonators, filter, and antennas used in wireless communication systems demands new low-cost dielectrics with high performance. In particularly, a low dielectric constant material with a high $Q \times f$ has become increasingly viable for reducing the signal propagation delay and crosstalk in ultra high frequency applications.[1] The search for low-loss dielectric ceramics therefore, has emerged to become a primary issue in the last few years.[1–7]

Ilmenite-structured $MgTiO_3$ ($\varepsilon_r \sim 16$, $Q \times f \sim 160{,}000$ GHz, $\tau_f \sim -50$ ppm/°C)[8] and spinel-structured Mg_2TiO_4 ($\varepsilon_r \sim 14$, $Q \times f \sim 150{,}000$ GHz, $\tau_f \sim -50$ ppm/°C)[9,10] are both recognized as useful dielectrics for microwave applications because of their relatively low-cost and extremely low dielectric loss. The $MgTi_2O_5$ phase, which is known for its much lower $Q \times f$ ($\varepsilon_r \sim 17.4$, $Q \times f \sim 47{,}000$ GHz, $\tau_f \sim -66$ ppm/°C),[11,12] has always formed as an intermediate phase of $MgTiO_3$ and therefore becomes difficult to completely eliminate itself from the $MgTiO_3$ sample as prepared by mixed oxide route. In addition, the phenomenon of Mg_2TiO_4 being thermally decomposed into $Mg_{2+2\delta}Ti_{1-\delta}O_4$ and $MgTiO_3$ phases was reported earlier by Petrova et al., where δ is a function of temperature.[13] Consequently, the tetragonal-structured $Mg_{1.8}Ti_{1.1}O_4$ ($\varepsilon_r \sim 15.74$, $Q \times f \sim 141{,}000$ GHz, $\tau_f \sim -52.4$ ppm/°C)[14] is likely to appear as a second phase during the synthesis of Mg_2TiO_4. Both $MgTiO_3$ and $Mg_{1.8}Ti_{1.1}O_4$ are not harmful to the dielectric properties of Mg_2TiO_4 because they possess similar properties. With partial substitution of Mg by Zn or Co, the $Q \times f$ of the dielectrics $(Mg_{0.95}Zn_{0.05})_2TiO_4$ ($\varepsilon_r \sim 15.48$, $Q \times f \sim 275{,}300$ GHz, $\tau_f \sim -34$ ppm/°C)[15], $(Mg_{0.95}Co_{0.05})_{1.8}Ti_{1.1}O_4$ ($\varepsilon_r \sim 16.11$, $Q \times f \sim 207{,}500$ GHz, $\tau_f \sim 52.6$ ppm/°C)[16], and $(Mg_{0.95}Co_{0.05})_2TiO_4$ ($\varepsilon_r \sim 15.7$, $Q \times f \sim 286{,}000$ GHz, $\tau_f \sim 52.5$ ppm/°C)[17] can be easily boosted to a value higher than 200,000 GHz while retaining compatible values of ε_r andτ_f. Since the dielectric properties of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ and its end member $Zn_{1.8}Ti_{1.1}O_4$ are still remained "unknown",

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it should be of interest to characterize the dielectric response of these ceramics and its accompanying solid solutions $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ at microwave frequency.

In this study, $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solutions were synthesized to investigate their microwave dielectric properties because the ionic radius of Zn^{2+} (0.74 Å, $CN = 6$)[18] are similar to that of Mg^{2+} (0.72 Å, $CN = 6$).[18] Consequently, the compound under study not only showed a tremendous lowering in the dielectric loss but also retained comparably decent values of ε_r and τ_f. The resultant microwave dielectric properties were analyzed based on the densification, X-ray diffraction (XRD) patterns, and the microstructures of the ceramics. The correlation between the microstructure and the $Q \times f$ value was also investigated.

2. Experimental procedure

$(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ samples were synthesized by a conventional solid-state method from individual high-purity oxide powders (>99.9%) made up of MgO, ZnO, and TiO_2. Because MgO is hygroscopic, it was fired at 600 °C for 5 h to remove moisture retained therein. A stoichiometric mix of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ compound was ground in distilled water for 24 h in a ball mill using agate balls. The mixtures were dried and calcined at 800–1200 °C for 4 h. The calcined powders were dried, ball milled for 24 h. The fine powders with 5 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 1030–1420 °C for 4 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 CuKα ($\lambda = 0.15406$ nm) radiation with a Siemens D5000 diffractometer (Munich, Germany) operated at 40 kV and 40 mA. The surface microstructures were evaluated by scanning electron microscopy (SEM; Philips XL-40FEG, Eindhoven, the Netherlands). The apparent densities of the sintered pellets were measured by the common Archimedes method. The dielectric constant (ε_r) and the quality factor values (Q) at microwave frequencies were measured (at 10.52 GHz) using the Hakki–Coleman dielectric resonator method.[19,20] 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. An identical technique was applied for the measurement of the temperature coefficient of resonant frequency (τ_f). The test cavity was placed over a thermostat in the temperature range of 25–80 °C. The τ_f (ppm/°C) can be calculated by considering the change in the resonant frequency (Δf). The temperature coefficient of resonant frequency (τ_f) can be defined as

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

where f_1 and f_2 represent the resonant frequencies at T_1 and T_2, respectively.

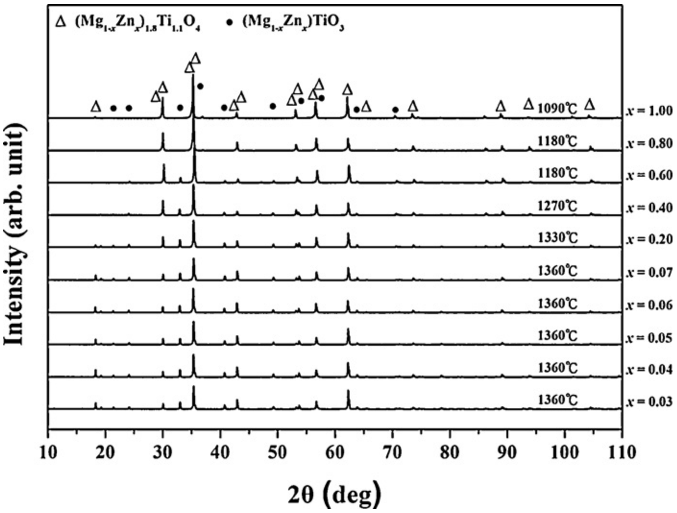


Fig. 1. X-ray diffraction patterns of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) ceramics sintered at different sintering temperatures for 4 h.

3. Results and discussion

The room temperature X-ray diffraction (XRD) patterns recorded from the $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) ceramics sintered at different temperatures for 4 h are illustrated in Fig. 1. Strong XRD peaks of a spinal tetragonal phase of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ type (indexed as $Mg_{1.8}Ti_{1.1}O_4$, ICDD-PDF#00-054-1226, the space group P4122 (91)) was classified as the main phase and its position and relative intensity only varied slightly, which implied the compounds tend to form a continuous solid solution. In addition, a very small amount of $(Mg_{1-x}Zn_x)TiO_3$ (can be indexed as $MgTiO_3$; $\varepsilon_r \sim 17.1$, $Q \times f$ ~264,000 GHz at 7 GHz, $\tau_f \sim -40.3$ ppm/°C)[21] second phase was also detected in the whole composition range and almost eliminated at $x \geq 0.8$. The formation of $(Mg_{1-x}Zn_x)TiO_3$ second phase is most likely due to the thermal decomposition mechanism proposed by Petrova et al.[13] Lattice parameters of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics sintered at 1360 °C were measured and presented in Table 1, which further confirmed the formation of solid solutions. Except for $x = 0.06$ and 0.07, the cell volume is increased in a linear trend as Zn content is increased, which is also expected because the ionic radius of Zn^{2+} (0.74 Å, $CN = 6$) is larger than that of Mg^{2+} (0.72 Å, $CN = 6$).

Fig. 2 shows the change of shrinkage of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics with various amounts of x. The results illustrate that the onset temperature of shrinkage is lowered with the small partial substitution of Mg by Zn and is a function of Zn content. Notice that the densification of $Mg_{1.8}Ti_{1.1}O_4$ ceramics with ≥60 mol% Zn substitution begins at a temperature below 1000 °C. The shrinkage curves provide a more clear inspection of the contribution of Zn substitution to the densification of $Mg_{1.8}Ti_{1.1}O_4$ ceramics.

Table 1
Lattice parameters and cell volume of the $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics.

	Lattice parameters (Å) of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics		Cell volume (Å^3)	Reference
x value	*a* = *b*	*c*		
0.03	5.9682 (±0.0014 Å)	8.4353 (±0.0020 Å)	300.46	This work
0.04	5.9676 (±0.0015 Å)	8.4374 (±0.0022 Å)	300.47	This work
0.05	5.9678 (±0.0016 Å)	8.4390 (±0.0024 Å)	300.55	This work
0.06	5.9663 (±0.0016 Å)	8.4382 (±0.0023 Å)	300.37	This work
0.07	5.9667 (±0.0015 Å)	8.4377 (±0.0022 Å)	300.39	This work
0.20	5.9694 (±0.0013 Å)	8.4430 (±0.0019 Å)	300.86	This work
0.40	5.9747 (±0.0009 Å)	8.4345 (±0.0013 Å)	301.09	This work
0.60	5.9750 (±0.0006 Å)	8.4358 (±0.0016 Å)	301.16	This work
0.80	5.9759 (±0.0007 Å)	8.4366 (±0.0018 Å)	301.28	This work
1.00	5.9768 (±0.0006 Å)	8.4372 (±0.0014 Å)	301.39	This work

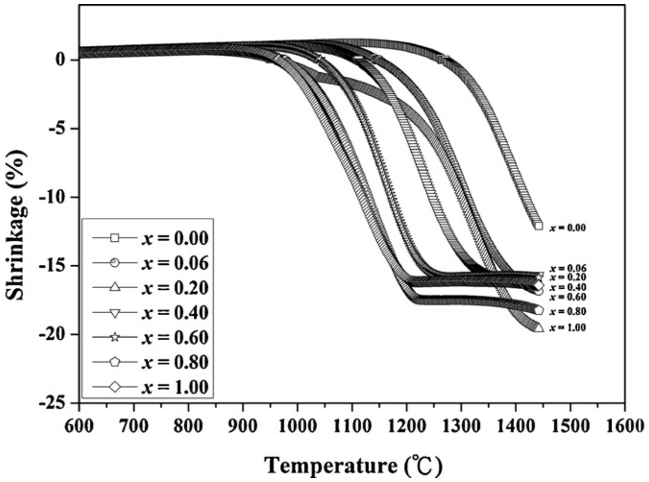


Fig. 2. Shrinkage of the $Mg_{1.8}Ti_{1.1}O_4$ samples with 0–100 mol% Zn substitution.

The SEM photographs of thermal-etched $(Mg_{0.94}Zn_{0.06})_{1.8}Ti_{1.1}O_4$ ceramics sintered at different temperatures for 4 h are illustrated in Fig. 3. Porous microstructure can be seen for specimen sintered at 1300 °C and the grain size increases as the sintering temperature increases due to a grain growth. Well-developed microstructure can be achieved at temperatures 1330–1360 °C and further increase in the sintering temperature leads to an over-sintering which would give rise to a rapid grain growth, and grains of relatively large size up to >20 μm can be seen at 1420 °C. The Energy dispersive X-ray (EDX) analysis of corresponding spots A–D from Fig. 3(c) is shown in Table 2 (EDX with a resolution of 183 eV). Accordingly, both $(Mg_{0.94}Zn_{0.06})_{1.8}Ti_{1.1}O_4$ and $(Mg_{0.94}Zn_{0.06})TiO_3$ phases can be identified and it is in agreement with the XRD results.

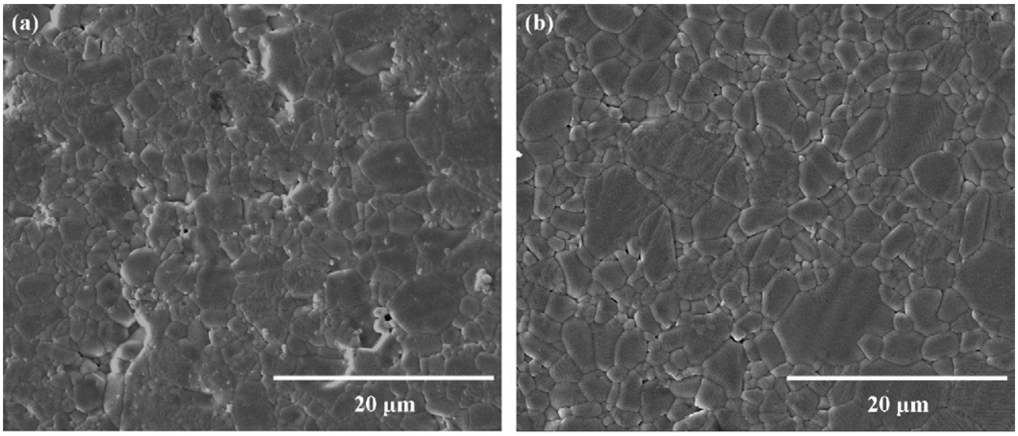
Table 2
The EDX data of the $(Mg_{0.95}Zn_{0.05})_{1.8}Ti_{1.1}O_4$ ceramics for spot A–D.

Spots	Atom (%)			
	Mg (K)	Zn (L)	Ti (K)	O K)
A	36.23	4.02	24.11	35.65
B	36.30	3.00	23.26	37.44
C	30.77	3.35	28.41	37.48
D	31.31	3.78	28.52	36.40

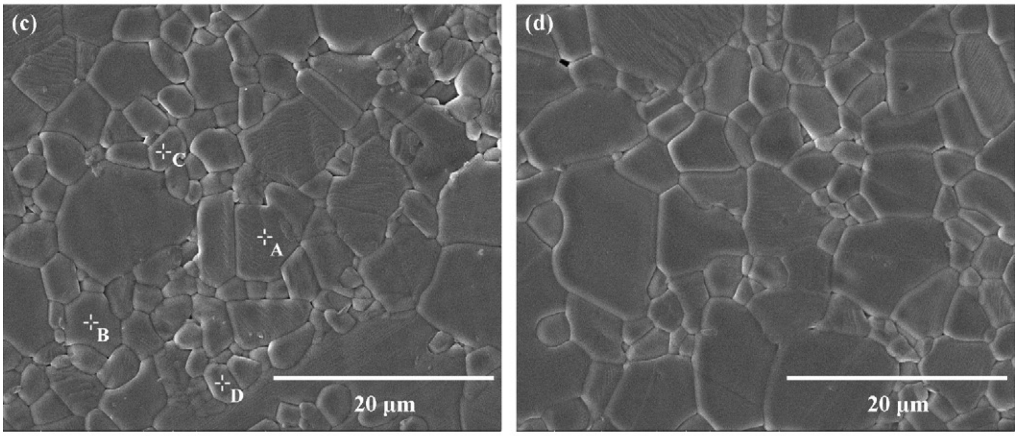
The relative density of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.00$–1.00) solid solutions is demonstrated in Fig. 4. With increasing sintering temperature, the relative density of specimens initially increases to a maximum of 97.81% at $x = 0.06$ and thereafter it decreases all the way down to 91% at $x = 1.0$. Notice that specimens with $x = 0.03$–0.04 have the highest relative density at 1390 °C, $x = 0.05$–0.07 at 1360 °C, and the breaking temperature continuingly decreases with the increase of *x* down to a value of 1090 °C at $x = 1.0$, which suggests the Zn substitution is capable of significantly lowering the sintering temperature of the specimens. The results are in good agreement with the variations of shrinkage curves shown in Fig. 2.

The dielectric constant of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solutions sintered at different temperatures for 4 h is illustrated in Fig. 5. Variation of dielectric constant is consistent with that of density. By increasing the *x* from 0.03 to 1.0, not only the sintering temperature is lowered, the dielectric constant is also increased from 16.28 to 20.01, suggesting the Zn substitution has a significant effect on the dielectric constant of the specimens. In addition, the dielectric constants of $(Mg_{0.95}Zn_{0.05})_{1.8}Ti_{1.1}O_4$ (~16.33) and $(Mg_{0.95}Zn_{0.05})TiO_3$ (~17.1)[21] are similar to each other, which implies the presence of the second phase is not expected to bring much deviation to the dielectric constant of the specimen.

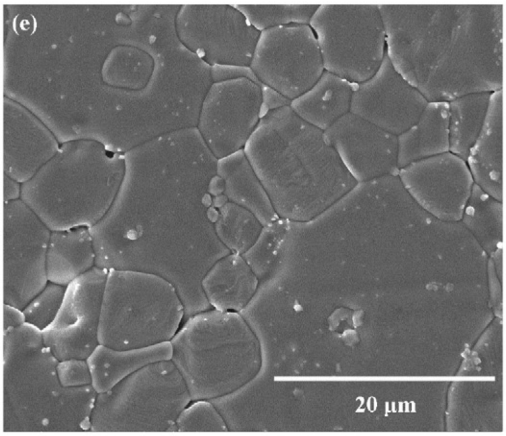
The $Q \times f$ values of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solutions sintered at different temperatures for 4 h are demonstrated in Fig. 6. The microwave dielectric loss is mainly caused not only by the intrinsic loss such as lattice vibration modes, but also by the extrinsic losses such as density, second phases, impurities, surface morphology, and lattice defect.[22] Since the variation of $Q \times f$ is consistent with that of relative density, it suggests the dielectric loss of the specimen is controlled by the relative density. According to Kim et al.,[23] the $Q \times f$ value also could be largely dependent on the packing fraction defined



(a) 1300°C (b) 1330°C



(c) 1360°C (d) 1390°C



(e) 1420°C

Fig. 3. SEM micrographs of $(Mg_{0.94}Zn_{0.06})_{1.8}Ti_{1.1}O_4$ ceramics sintered at (a) 1300 °C, (b) 1330 °C, (c) 1360 °C, (d) 1390 °C, and (e) 1420 °C for 4 h.

by the summing the volume of packed ions over the volume of a primitive unit cell and it can be expressed as[23,24]

$$\text{Packing fraction}\,(\%) = \frac{\text{volume of packed ions}}{\text{volume of primitive unit cell}} = \frac{\text{volume of packed ions}}{\text{volume of unit cell}} \times Z \qquad (2)$$

where Z is the number of formula units per unit cell. The results are demonstrated in Fig. 7. Accordingly, the dependence of the $Q \times f$ of the $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics on the packing fraction for small amount of Zn substitution for Mg ($x = 0.03$–0.2) is confirmed. However, the $Q \times f$ value is inversely proportional to the packing fraction for $x > 0.2$. Because the relative density shows a huge drop from 96.48% at $x = 0.2$ to

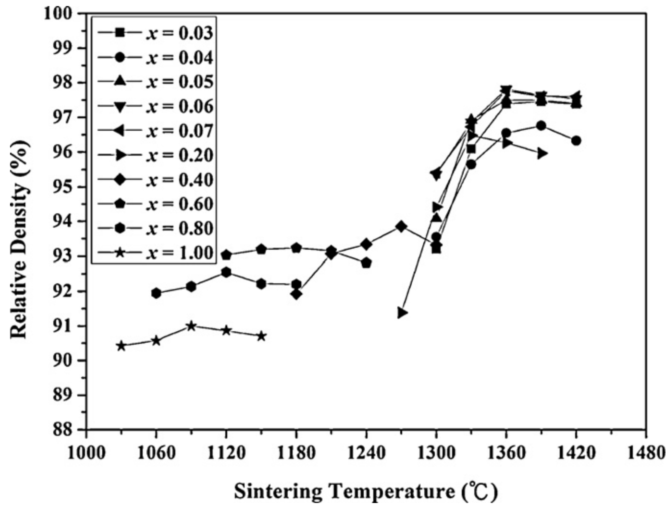


Fig. 4. Relative density of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solution sintered at different temperatures for 4 h.

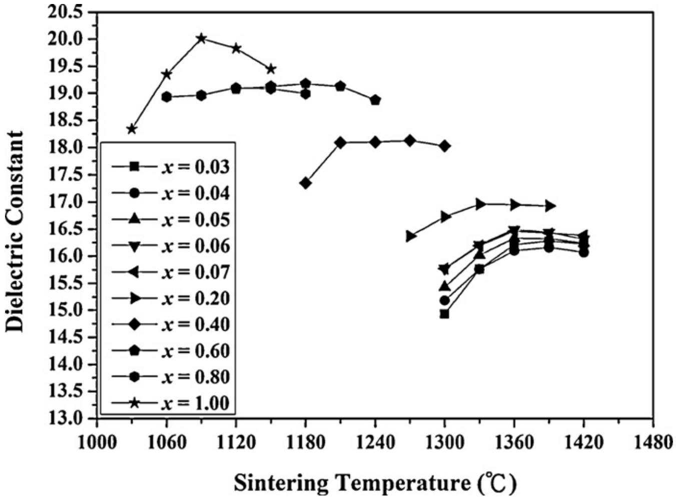


Fig. 5. Dielectric constant of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solution sintered at different temperatures for 4 h.

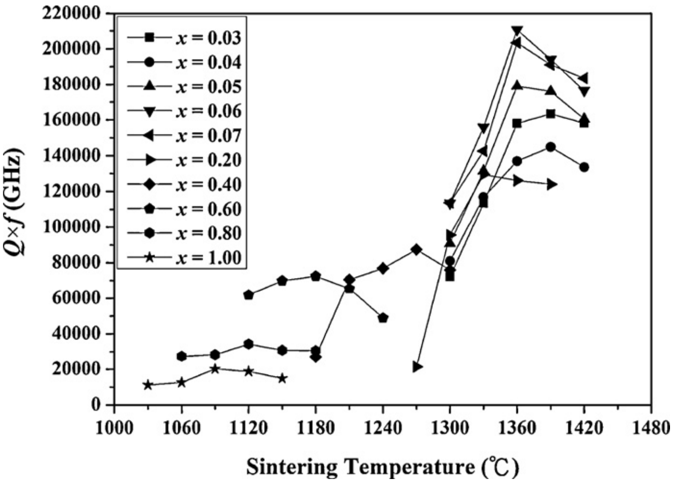


Fig. 6. $Q \times f$ values of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solution sintered at different temperatures for 4 h.

91.0% at $x = 1.0$, and the second phase has a similar $Q \times f$ value, the effect of packing fraction on $Q \times f$ for $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ may be neglected. In other words, the decreased relative density is considered to be the most possible extrinsic reason for this deviation for $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics with $x > 0.2$. A maximum $Q \times f$ value of 210,700 GHz (at 10.52 GHz) could be achieved for specimen using $(Mg_{0.94}Zn_{0.06})_{1.8}Ti_{1.1}O_4$ sintered at 1360 °C for 4 h.

Fig. 8 illustrates the temperature coefficient of resonant frequency (τ_f) of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ (0.00–1.00) solid solutions sintered at different temperatures for 4 h. It is generally known that the τ_f can be correlated to τ_ε by the following equation:

$$\tau_f = -\frac{1}{2}\tau_\varepsilon - \alpha \qquad (3)$$

where α is the linear thermal expansion coefficient of the dielectric (α is typically in the range of 3–15 ppm/°C for most of dielectric ceramics[25]). Yoon et al. found that τ_ε was proportional to the relative magnitude of unit cell volume.[26] Notice that the τ_f remains virtually unchanged with

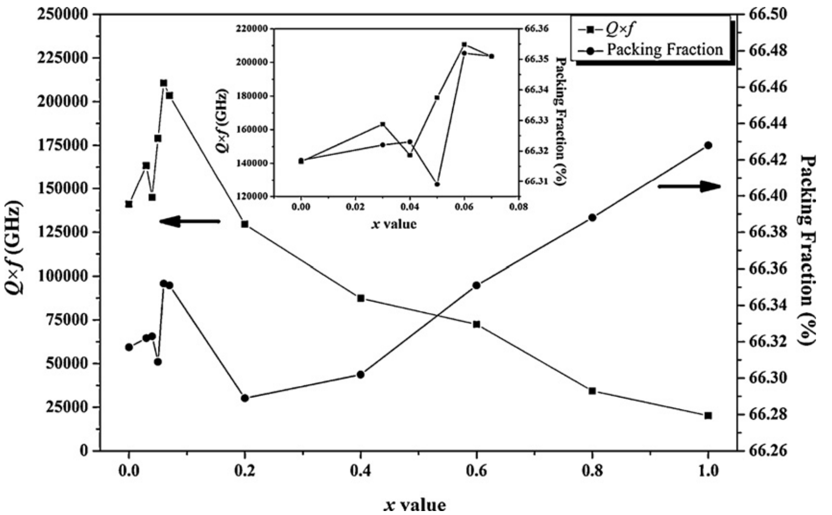


Fig. 7. The packing fraction and $Q \times f$ value of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solutions.

Table 3
Microwave dielectric properties of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramics system sintered at different temperatures for 4 h.

x value	Relative density (%)	ε_r	$Q \times f$ (GHz)	τ_f (ppm/°C)	Sintering temperature (°C)
0.00[a]	96.53	15.74	141,000	−52.4	1450 °C
0.03	97.45	16.28	163,300	−54.2	1390 °C
0.04	96.76	16.16	144,900	−54.5	1390 °C
0.05	97.50	16.33	179,000	−59.7	1360 °C
0.06	97.81	16.49	210,700	−62.3	1360 °C
0.07	97.76	16.46	203,500	−58.4	1360 °C
0.20	96.48	16.96	129,500	−56.1	1330 °C
0.40	93.85	18.13	87,400	−57.4	1270 °C
0.60	93.23	19.18	72,500	−57.8	1180 °C
0.80	92.55	19.10	34,300	−54.4	1120 °C
1.00	91.00	20.01	20,200	−57.9	1090 °C

[a] ref. [12].

different unit cell volumes but having the same structure of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$, because the variation of unit cell volume is relatively small (<0.4%) and the change of τ_ε can therefore be neglected. Moreover, the τ_f of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ and $(Mg_{1-x}Zn_x)TiO_3$ are similar and it is also not sensitive to the sintering temperature. Consequently, the τ_f values more or less vary in the range from −54.2 to −62.3 ppm/°C for specimens with $x = 0.03$–1.00 at 1090–1390 °C.

Table 3 summarizes the microwave dielectric properties of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ceramic system. These dielectric properties and densities vary substantially with sintering temperature and composition. It indicates that Zn content has an effect on the sintering temperature of the specimen and a reduction of 300 °C in the sintering temperature can be achieved for $x = 1.0$. With the increase of x in the compound, the relative density increases up to a maximum of 97.81% at $x = 0.6$ and decreases thereafter. It also possesses the highest $Q \times f$ (∼210,700 GHz), indicating an approximate 50% reduction in the dielectric loss compared to that of pure $Mg_{1.8}Ti_{1.1}O_4$ (∼141,000 GHz). For pure $Zn_{1.8}Ti_{1.1}O_4$ ceramic, it can be sintered with a lower temperature at 1090 °C and possesses the following microwave dielectric properties; $\varepsilon_r \sim 20.01$, $Q \times f \sim 20{,}200$ GHz, $\tau_f \sim -57.9$ ppm/°C.

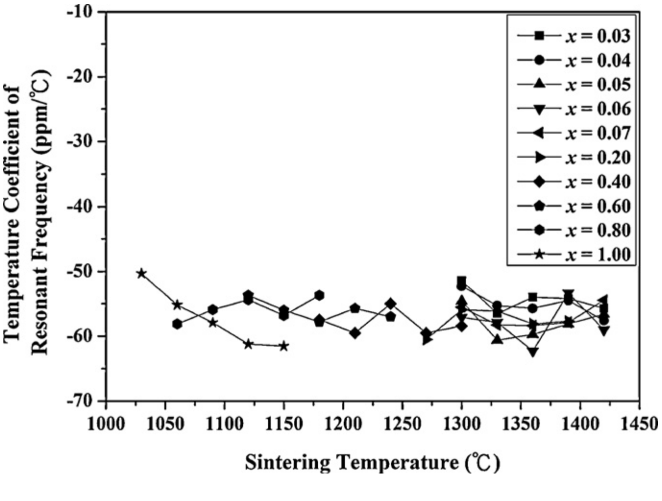


Fig. 8. τ_f values of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solution sintered at different temperatures for 4 h.

4. Conclusion

The microwave dielectric properties of $(Mg_{1-x}Zn_x)_{1.8}Ti_{1.1}O_4$ ($x = 0.03$–1.00) solid solution have been studied in present study. XRD patterns exhibit a single spinal tetragonal as the main phase in the whole composition range, except for small amount of ilmenite phase being detected as a second phase. Both extrinsic (relative density) and intrinsic (packing fraction) factors have effects on $Q \times f$ for specimens with small amount of Zn substitution ($x = 0.03$–0.2), but the extrinsic one becomes a dominant factor when $x > 0.2$. A maximum $Q \times f$ value of 210,700 GHz (at 10.52 GHz) can be achieved for specimen using $(Mg_{0.94}Zn_{0.06})_{1.8}Ti_{1.1}O_4$, which indicates an approximate 50% reduction in the dielectric loss compared to that of pure $Mg_{1.8}Ti_{1.1}O_4$. The τ_f remains virtually unchanged in the entire compositional range since the variation of unit cell volume is small (<0.4%). In addition, the Zn substitution can significantly lower the sintering temperature of the specimens.

Acknowledgement

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