



Effect of $\text{Li}_2\text{O}-\text{V}_2\text{O}_5$ addition on the sintering behavior and microwave dielectric properties of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics

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Abstract

The effect of Zn^{2+} substitution on the structure and microwave dielectric properties of $\text{Li}_3\text{Mg}_2\text{NbO}_6$ ceramic was investigated. The compositions of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics were prepared by the solid-state reaction method. The phase structure, microstructure and microwave dielectric properties of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics were analyzed via an X-ray diffraction, a scanning electron microscope as well as a network analyzer. All samples sintered at 1080–1240 °C for 4 h remained a single orthorhombic structure, which could be indexed according to the $\text{Li}_3\text{Mg}_2\text{NbO}_6$ phase (JCPDS-PDF #86-0346). The microwave dielectric properties of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics exhibited significant dependence on the sintering condition, microstructure and composition. Excellent microwave dielectric properties ($\epsilon_r \sim 17.2$, $Q \times f \sim 142,331$ GHz, $\tau_f \sim -23.2$ ppm/°C) were obtained in the $x=0.08$ sample sintered at 1120 °C for 4 h in air. In addition, the 0.17 $\text{Li}_2\text{O}-0.83\text{V}_2\text{O}_5$ composition was added as a sintering aid to lower the sintering temperature. The 0.5 wt% 0.17 $\text{Li}_2\text{O}-0.83\text{V}_2\text{O}_5$ added $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic showed fairly good microwave dielectric properties of $\epsilon_r \sim 14$, $Q \times f \sim 83,395$ GHz, $\tau_f \sim -37.2$ ppm/°C when sintered at 925 °C for 2 h. Furthermore, there was no chemical reaction between Ag electrode and the sintering aid added sample, which further demonstrates that the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic modified with 0.17 $\text{Li}_2\text{O}-0.83\text{V}_2\text{O}_5$ would be a promising candidate material for low-temperature cofired ceramic (LTCC) applications.

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1. Introduction

In recent years, the study of microwave dielectric ceramics has been focusing on the low-loss dielectrics because the carrier frequencies of interest in the communication systems are being extended to higher frequency bands, such as the millimeter wave range [1]. A high quality factor (Q) dielectric plays a prominent role in preventing from the drop in Q at high frequency. From the device design point of view, a good combination of high relative permittivity ϵ_r (for miniaturization), high Q (for selectivity), near zero temperature coefficient of resonant frequency τ_f (for stability), low processing temperature (for low cost), low cost of raw materials (for low cost of the device), and low bulk density (for lightness of electronic

module) is required for a microwave dielectric material [2–5]. The complex perovskites such as $\text{Ba}(\text{Zn}_{1/3}\text{Ta}_{2/3})\text{O}_3$ (BZT) and $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$ (BMT) show low dielectric loss with low τ_f [6,7]. They have important applications as resonators and filters in microwave communication systems. However, they generally require a high sintering temperature and a relatively high cost. Moreover, a rapid growth of the cell phone and wireless communication markets has further led to extensive research and development in the area of microwave dielectrics with low loss and low cost [1].

A number of high- Q microwave dielectric materials have been reported in the last few years. George et al. reported that the $\text{Li}_2\text{ATi}_3\text{O}_8$ ($A=\text{Mg}, \text{Zn}$) ceramics possessed excellent microwave dielectric properties ($\text{Li}_2\text{MgTi}_3\text{O}_8$ ceramic showed $\epsilon_r \sim 27.2$, $Q_u \times f \sim 42,000$ GHz, $\tau_f \sim +3.2$ ppm/°C, and $\text{Li}_2\text{ZnTi}_3\text{O}_8$ ceramic had $\epsilon_r \sim 25.6$, $Q_u \times f \sim 72,000$ GHz, $\tau_f \sim -11.2$ ppm/°C when both were sintered at 1075 °C for 4 h [8]). Later

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on, Huang et al. investigated the systems of $\text{Li}_2(\text{Zn}_{1-x}\text{A}_x)\text{Ti}_3\text{O}_8$ ($\text{A}=\text{Mg}^{2+}$, Co^{2+} ; $x=0.02\text{--}0.1$) and $(\text{Mg}_{1-x}\text{M}_x)_2\text{TiO}_4$ ($\text{M}=\text{Zn}^{2+}$, Co^{2+} and Mn^{2+}) [9–12]. The $Q \times f$ value of $\text{Li}_2\text{ZnTi}_3\text{O}_8$ could be improved from 72,000 GHz to 150,000 GHz and 140,000 GHz as 6% Mg^{2+} and 8% Co^{2+} were used, respectively. Analogously, after partial replacement of Mg^{2+} by Zn^{2+} , Co^{2+} , or Mn^{2+} , the $Q \times f$ values of dielectrics ($\text{Mg}_{0.95}\text{Zn}_{0.05})_2\text{TiO}_4$, $(\text{Mg}_{0.95}\text{Co}_{0.05})_2\text{TiO}_4$ and $(\text{Mg}_{0.95}\text{Mn}_{0.05})_2\text{TiO}_4$ could be easily boosted to a higher value than 250,000 GHz and compatible ϵ_r and τ_f values could be retained at the same time. Moreover, the effect of Ni^{2+} substitution for Zn^{2+} on microwave dielectric properties of $(\text{Zn}_{1-x}\text{Ni}_x)_3\text{Nb}_2\text{O}_8$ ($x=0.02\text{--}0.08$) ceramics was also investigated [13]. The same trend could be obtained in the monoclinic-structured $\text{Zn}_3\text{Nb}_2\text{O}_8$ ceramic. Obviously, it could be discovered that with a small amount of the isovalent ionic substitution, the $Q \times f$ values could be improved by twice approximately, which might be due to the crystal lattice distortion caused by a subtle difference between ionic radius.

The $\text{Li}_3\text{Mg}_2\text{NbO}_6$ composition was firstly reported by Yuan et al. [14]. It showed good microwave dielectric properties of $\epsilon_r \sim 16.8$, $Q \times f \sim 79,643$ GHz, $\tau_f \sim -27.2$ ppm/ $^\circ\text{C}$ when sintered at 1250 $^\circ\text{C}$ for 2 h. However, to the best of our knowledge, the research on Mg-site substitution has not been reported up to date. In present study, we managed to replace Mg^{2+} with a small amount of Zn^{2+} . The crystal structure and the microwave dielectric properties of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics were then investigated. A higher $Q \times f$ value was achieved at $x=0.08$ compared with the pure phase when sintered at 1120 $^\circ\text{C}$ for 4 h. In order to reduce the sintering temperature for low-temperature cofired ceramic (LTCC) applications, the $0.17\text{Li}_2\text{O}\text{--}0.83\text{V}_2\text{O}_5$ (abbreviated as LV) composition was added to the $x=0.08$ ceramic sample due to its eutectic melting point of 620 $^\circ\text{C}$ [15], which has been successfully employed in $\text{Li}_{1.0}\text{Nb}_{0.6}\text{Ti}_{0.5}\text{O}_3$ ceramic [16] and other niobate ceramic compositions (e.g., $\text{Mg}_4\text{Nb}_2\text{O}_9$) [17]. The effect of LV additive on the sinterability and microwave dielectric properties of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic was investigated systematically.

2. Experimental procedure

The $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics were prepared by a solid-state reaction method. High-purity (>99.9%) Li_2CO_3 , $(\text{MgCO}_3)_4 \cdot \text{Mg}(\text{OH})_2 \cdot 5\text{H}_2\text{O}$, ZnO , Nb_2O_5 and V_2O_5 were used as the starting materials. Stoichiometric amounts of the chemical powders were weighed and ball-milled in a nylon jar with zirconia balls and ethanol as the media for 4 h. The resultant slurry was then dried and calcined at 925 $^\circ\text{C}$ for 4 h. The $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ powders were re-milled for 6 h with various amounts of LV additive and then mixed together with poly vinyl alcohol (PVA) binder. The granulated powders were subsequently pressed into cylinders with dimensions of 10 mm in diameter and 7–8 mm in height. The $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics were sintered at 1080–1240 $^\circ\text{C}$ for 4 h. The LV modified $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ samples were sintered at 850–950 $^\circ\text{C}$ for 2 h. In order to suppress the lithium

evaporation loss, the pellets were muffled with the powders of the same composition.

The bulk densities of the sintered pellets were measured by the Archimedes method. The crystal structure of the sintered ceramics was examined via an X-ray diffractometer (XRD, D/Max2500 V, Rigaku, Japan) using $\text{CuK}\alpha$ radiation. The grain morphology was analyzed by a scanning electron microscope (SEM, JSM-6490LV, JEOL, Tokyo, Japan). Microwave dielectric properties of the sintered samples were measured by a network analyzer (N5230C, Agilent, Palo Alto, CA) in the frequency range of 8–14 GHz. The dielectric constant was measured by the Hakki–Coleman method modified by Courtney [18], and the unloaded Q values were measured by the cavity method [19]. The τ_f values of the samples were measured in the temperature range from 20 $^\circ\text{C}$ to 80 $^\circ\text{C}$. It can be calculated by the following formula:

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

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

3. Results and discussion

The XRD patterns of the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics sintered at different temperatures for 4 h are shown in Fig. 1(a). All the patterns could be indexed as a single orthorhombic phase (JCPDS-PDF #86-0346), which belongs to the space group Fddd (70), and no secondary phase was observed. Moreover, the cell volume of the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics gradually increased from 894.28 \AA^3 at $x=0$ to 897.81 \AA^3 as $x=0.1$ owing to the substitution of larger Zn^{2+} (0.74 \AA) ions for Mg^{2+} (0.72 \AA) [20] ions, as shown in Fig. 1(b). These results demonstrate that the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ solid solutions were formed in the whole composition range.

Fig. 2 illustrates typical SEM images on the surface of the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic sintered at various temperatures. The grain size increased and the porosity decreased with increasing sintering temperatures from 1080 $^\circ\text{C}$ to 1120 $^\circ\text{C}$. The sample sintered at 1120 $^\circ\text{C}$ showed a dense and uniform microstructure without obvious pores. With further increasing the temperature up to 1240 $^\circ\text{C}$, some grains began to melt and grow abnormally, leading to the indistinct grain boundaries. It suggests that the grain growth was sensitive to the sintering conditions and the Zn-substitution had a significant effect on the grain growth owing to the formation of the liquid phase [21].

Fig. 3 shows the relative density and ϵ_r of the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics as a function of sintering temperatures. It can be seen that the relative densities of all compositions gradually increased with the sintering temperatures and reached a maximum value at their respective optimal sintering temperatures. It can be noted that the specimen with $x=0.08$ had the highest relative density ($\sim 99.0\%$) compared to other compositions. Further increasing the sintering temperature would result in an abnormal grain growth, thus

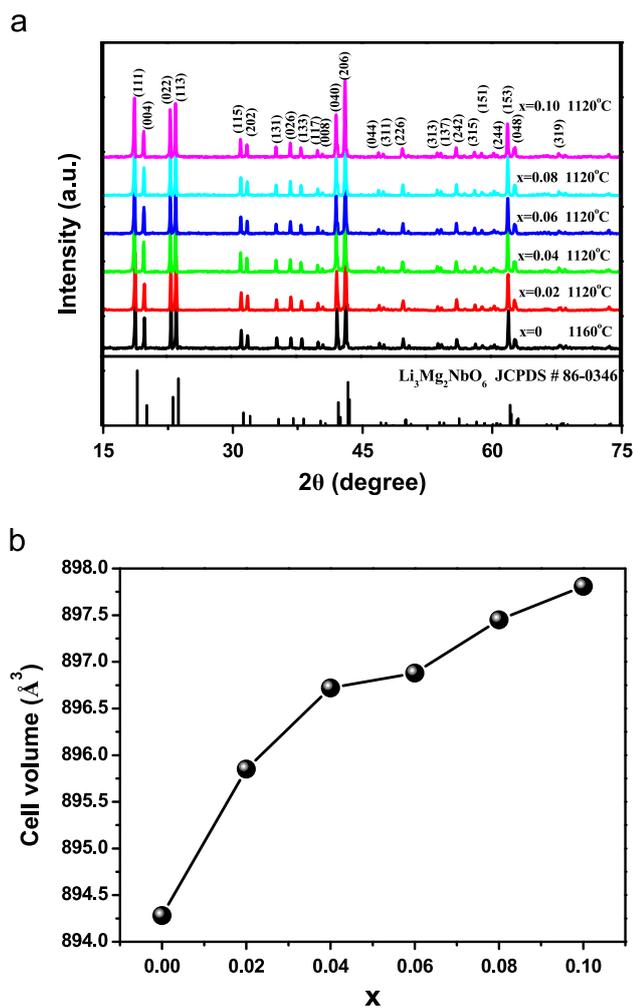


Fig. 1. (a) The XRD patterns of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics sintered at optimal temperatures for 4 h and (b) variation of the cell volume for the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics.

leading to a decrease of the relative density. The ϵ_r value should be mainly determined by the relative density, dielectric polarizabilities and structural characteristics such as the distortion, tilting, and/or rattling spaces of oxygen octahedron in the unit cell [22,23]. In present work, the ϵ_r mainly depended on the relative density of the samples. The variation of ϵ_r was consistent with the relative density. The maximum ϵ_r value of ~ 17.2 was obtained for the specimen of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ sintered at 1120°C for 4 h.

Fig. 4 shows the $Q \times f$ values of the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics sintered at different temperatures. It is well known that the contribution to the quality factor value at microwave frequencies consists of intrinsic and extrinsic losses. The extrinsic factors usually include defect concentration, impurities, grain size, porosity, and secondary phases. The intrinsic factors are generally related with the lattice anharmonicity [24–26]. In this work, the variation of the quality factor followed a similar trend as that of relative density at different temperatures, suggesting the dielectric losses were mainly controlled by the densification. The $Q \times f$ values of the $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ceramics increased as x changed from 0 to 0.08, but would decrease as x was further

increased. The highest value of $Q \times f \sim 142,331$ GHz was obtained in the composition with $x=0.08$. The increment of $Q \times f$ value would be related with the distortion of crystal lattice, which was caused by a small amount of Zn-substitution. Similar behavior had been discovered in other ceramics [9–13]. In general, τ_f is well known to be influenced by the composition, the additive and the second phase of the materials [10]. The inset of Fig. 4 shows the τ_f values of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ as a function of sintering temperatures. The τ_f values were not sensitive to the sintering temperature as expected since there was no change in the composition and no secondary phase was detected at different temperatures. As a result, the τ_f value of the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic would remain in a narrow range from -24.8 ppm/ $^\circ\text{C}$ to -23.0 ppm/ $^\circ\text{C}$ as the samples were sintered at 1080 – 1240°C . The optimum microwave dielectric properties of $\epsilon_r \sim 17.2$, $Q \times f \sim 142,331$ GHz, and $\tau_f \sim -23.2$ ppm/ $^\circ\text{C}$ could be achieved when the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic was sintered at 1120°C for 4 h.

For LTCC applications, LV with a low melting point was used as a liquid phase sintering additive to reduce the sintering temperature of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic. Fig. 5 shows the bulk density of the ceramics sintered at 850 – 950°C as a function of the LV content. It could be found that the density of the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic was dependent on the sintering temperature and the amount of LV. Bulk density of pure $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic increased with increasing the sintering temperature. It was significantly improved after 0.3 wt% LV was added, showing a typical liquid phase sintering characteristic. The bulk density decreased slightly as the LV content was more than 0.5 wt%, probably because of disadvantages provided by too much liquid.

Fig. 6 shows the XRD patterns of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic modified with 0–1.5 wt% LV sintered at 925°C for 2 h. It can be seen that the (111) peak shifted toward higher angles at first, indicating the decrement of the unit cell volume, which could be explained by the fact that V^{5+} (0.54 \AA) substituted for Nb^{5+} (0.64 \AA). It is of particular note that V^{5+} has a very limited solution in Nb-based compounds [27,28]. The substitution of Li^+ (0.76 \AA) for $(\text{Mg}, \text{Zn})^{2+}$ (0.74 \AA and 0.72 \AA , respectively) happened as the LV content was beyond 0.3 wt%, indicating an increment in the unit cell volume, as confirmed by the reflection peak shift toward lower angles. Besides the main phase $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$, three new phases $(\text{Mg}_{0.92}\text{Zn}_{0.08})_4\text{Nb}_2\text{O}_9$, $(\text{Mg}_{0.92}\text{Zn}_{0.08})\text{V}_2\text{O}_6$ and $(\text{Mg}_{0.92}\text{Zn}_{0.08})_{1.5}\text{VO}_4$ were detected when LV was added. The variation of the intensity of secondary phases was not obvious with increasing the LV content. The combination of the partial replacement of $(\text{Mg}_{0.92}\text{Zn}_{0.08})^{2+}$ ions by Li^+ and the substitution of Nb^{5+} by V^{5+} would finally produce the $(\text{Mg}_{0.92}\text{Zn}_{0.08})_4\text{Nb}_2\text{O}_9$ phase. The surplus $(\text{Mg}_{0.92}\text{Zn}_{0.08})^{2+}$ ions reacted with the remaining V_2O_5 to form the V_2O_5 -based compounds ($(\text{Mg}_{0.92}\text{Zn}_{0.08})\text{V}_2\text{O}_6$ and $(\text{Mg}_{0.92}\text{Zn}_{0.08})_{1.5}\text{VO}_4$).

The SEM photographs of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic doped with various LV contents sintered at 925°C for 2 h are illustrated in Fig. 7. The pure $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic could be hardly sintered well at 925°C . The grains were small and many pores could be seen in the sample. As the LV

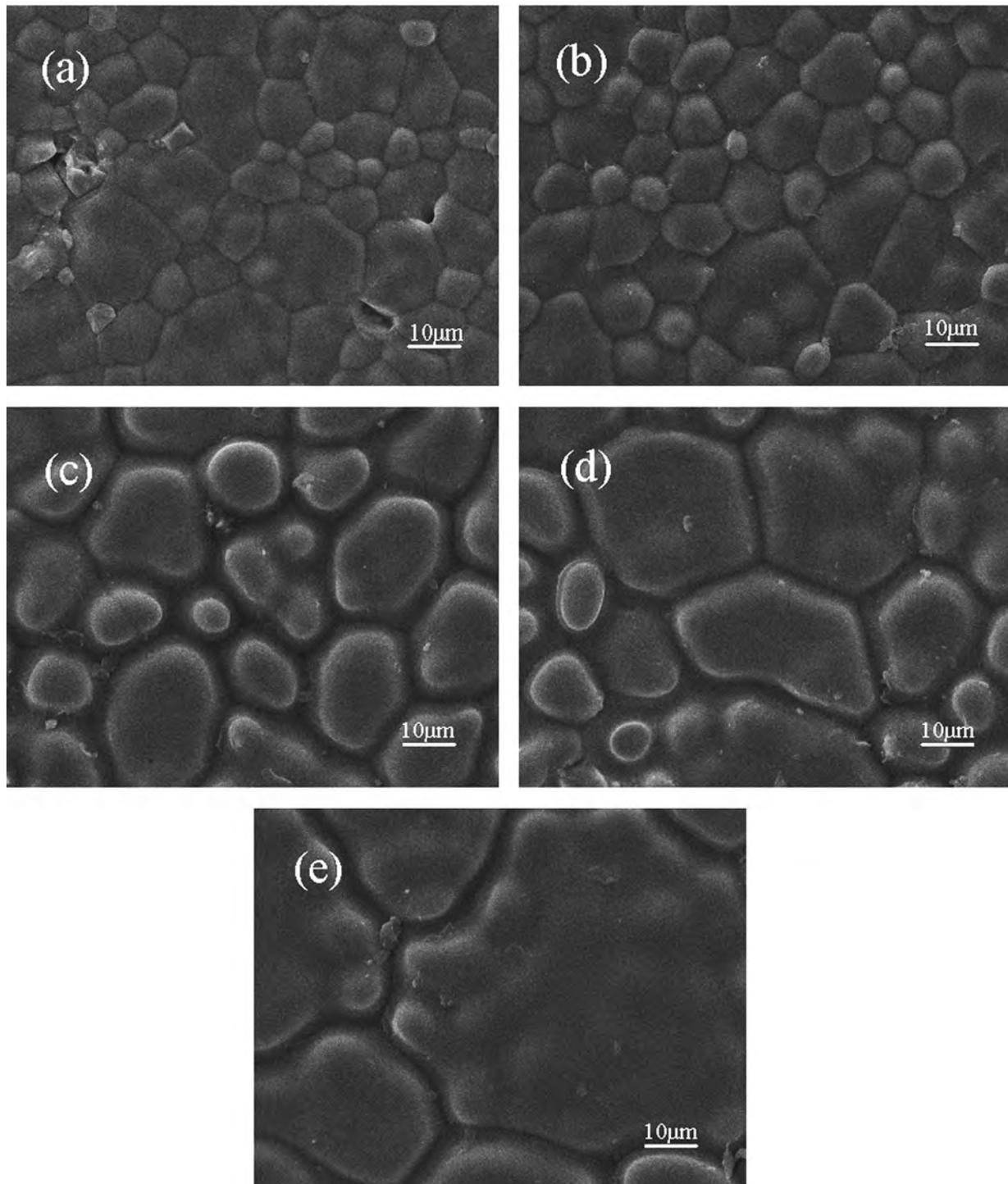


Fig. 2. The SEM images of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramics sintered at (a) 1080 °C, (b) 1120 °C, (c) 1160 °C, (d) 1200 °C, and (e) 1240 °C for 4 h.

content increased, the grains grew bigger, the porosity were reduced and relatively uniform microstructures could be obtained, as confirmed in Fig. 7(b) and (c). Especially, a uniform microstructure with no obvious pores was yielded in the specimen doped with 0.5 wt% LV. Further addition of LV resulted in an inferior microstructure. Some grains exhibited an abnormal growth, which might result in the deterioration of the microwave dielectric properties.

Fig. 8 shows the microwave dielectric properties of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic with various LV contents at different sintering temperatures. The ϵ_r value revealed rather similar tendency with the bulk density. The ϵ_r firstly increased due to the densification improvement benefiting from the LV addition, and then decreased owing to the abnormal grain growth. It is known that the V_2O_5 -based compounds generally have a small ϵ_r . The ϵ_r value of $\text{Mg}_4\text{Nb}_2\text{O}_9$ is ~ 12.9 [28],

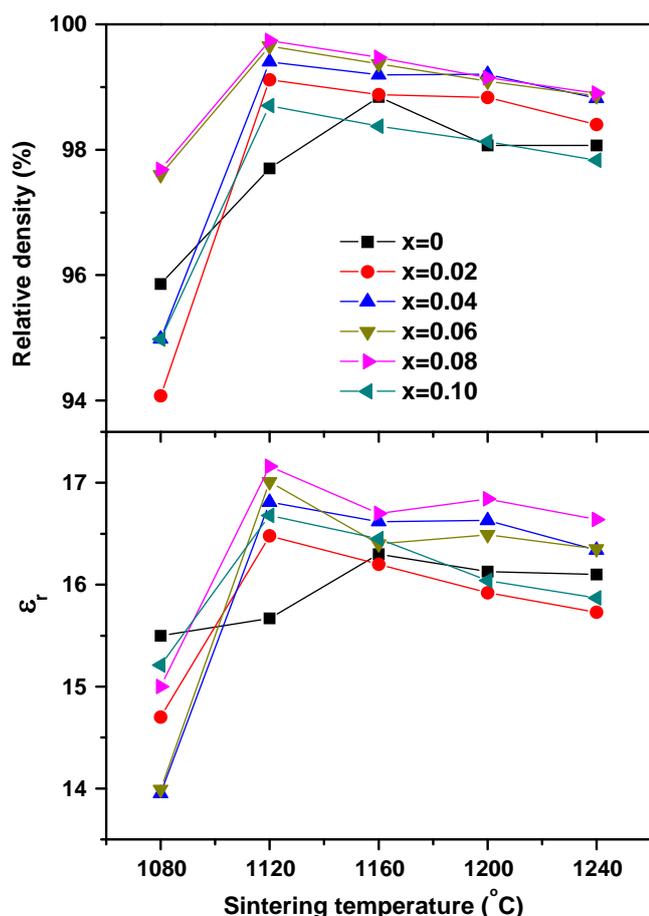


Fig. 3. The variation of the relative density and ϵ_r of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics sintered at different temperatures for 4 h.

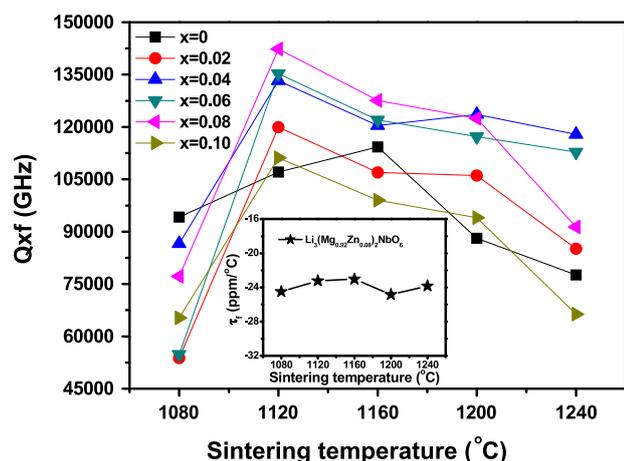


Fig. 4. The variation of $Q \times f$ of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics sintered at different temperatures for 4 h. The inset shows τ_f values of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramics sintered at different temperatures for 4 h.

which is smaller than that of the main phase. The $Q \times f$ value mainly depended on extrinsic contributions. Firstly, the increase of $Q \times f$ value was ascribed to the improvement in density. The maximum $Q \times f$ value was obtained in the sample with 0.5 wt% LV addition sintered at 925 °C. With a further

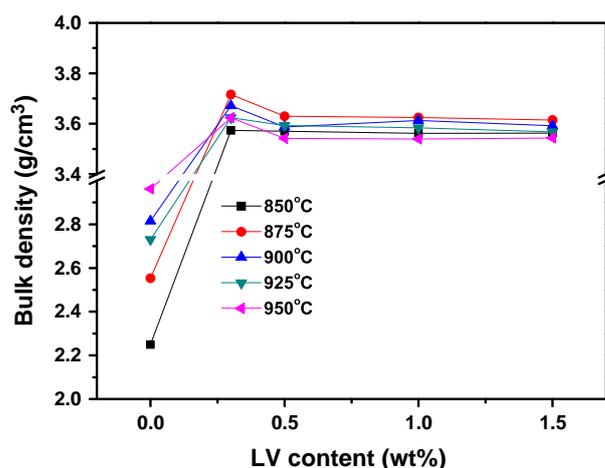


Fig. 5. Bulk densities of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramics with various contents of LV sintered at different temperatures for 2 h.

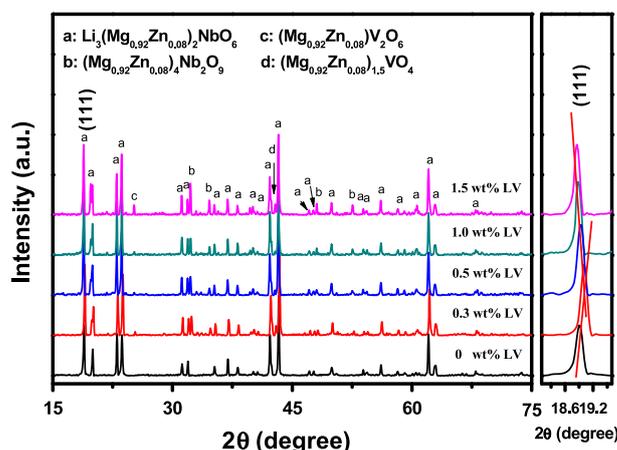


Fig. 6. The XRD patterns of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramics with various contents of LV sintered at 925 °C for 2 h.

addition of LV, the $Q \times f$ value decreased due to the increase in the amount of V_2O_5 -based compounds and pores. Although the $(\text{Mg}_{0.92}\text{Zn}_{0.08})_4\text{Nb}_2\text{O}_9$ phase had a higher $Q \times f$ value, the effect on the properties was not obvious owing to its few amounts. The τ_f value presented a descending tendency as LV content increased, which could be associated with the increasing amount of second phases ($\tau_f \sim -70.4$ ppm/°C for $(\text{Mg}_{0.92}\text{Zn}_{0.08})_4\text{Nb}_2\text{O}_9$) [28]. In a word, the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic doped with 0.5 wt% LV and sintered at 925 °C for 2 h exhibits good microwave dielectric properties of $\epsilon_r \sim 14$, $Q \times f \sim 83,395$ GHz and $\tau_f \sim -37.2$ ppm/°C. For evaluating the chemical compatibility of the 0.5 wt% LV doped $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic with the Ag electrode, the ceramic powder mixed with 20 wt% Ag powder was fired at 925 °C for 2 h. The corresponding XRD pattern is presented in Fig. 9. The result shows that none of any new phases were formed after cofiring, which indicated that the studied composition owns a good chemical stability against Ag electrode under sintering conditions. This further demonstrates that the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic modified

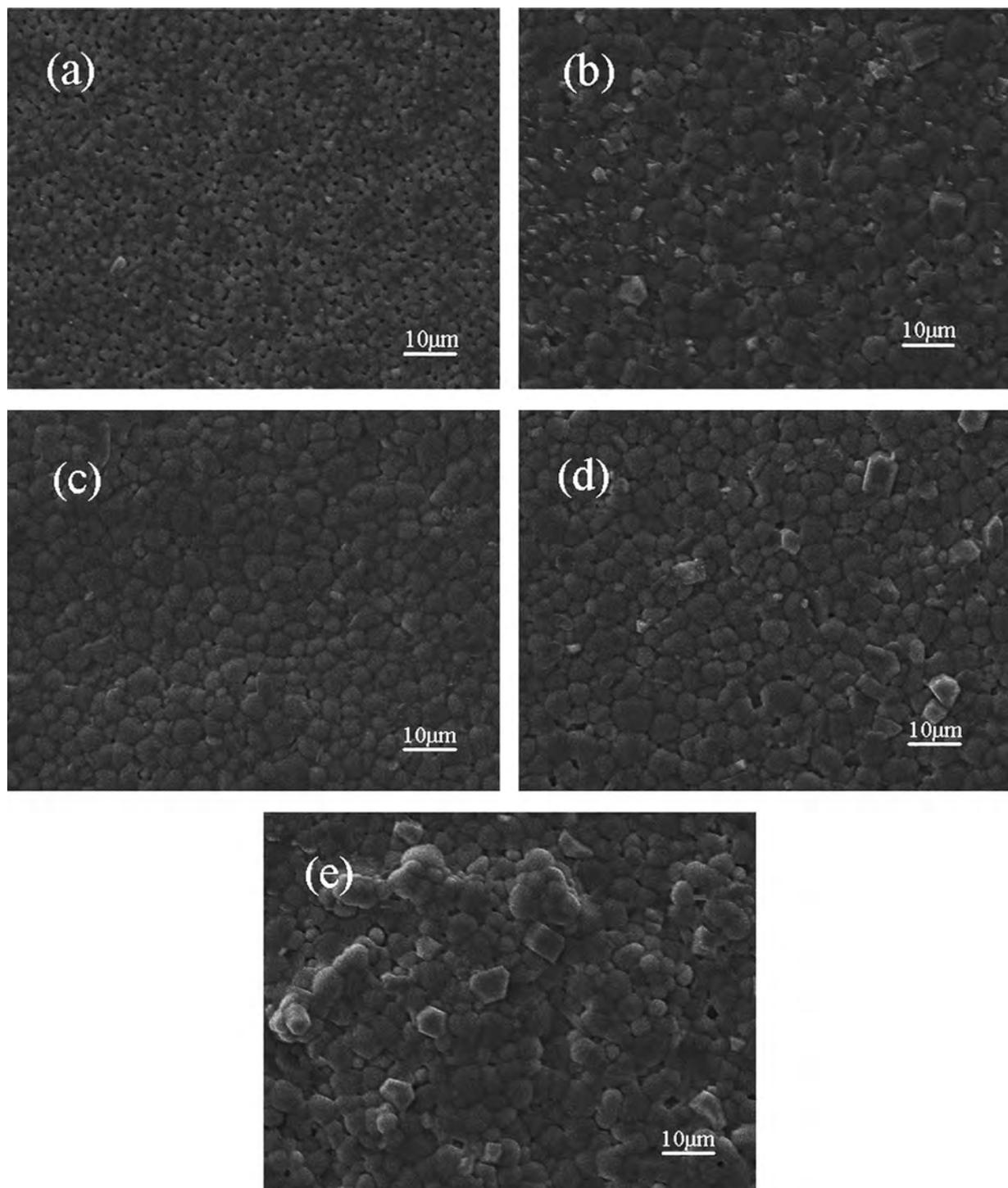


Fig. 7. The SEM images of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramics with (a) 0 wt% LV, (b) 0.3 wt% LV, (c) 0.5 wt% LV, (d) 1.0 wt% LV, and (e) 1.5 wt% LV sintered at 925°C for 2 h.

with 0.5 wt% LV would be a promising candidate material for LTCC applications.

4. Conclusion

The crystal structure and microwave dielectric properties of $\text{Li}_3(\text{Mg}_{1-x}\text{Zn}_x)_2\text{NbO}_6$ ($0 \leq x \leq 0.1$) ceramics were studied in present work. A single orthorhombic phase was formed for all

the compositions. The highest ϵ_r value of 17.2 and the maximum $Q \times f$ value of 142331 GHz were obtained for the $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ sample. The improvement of $Q \times f$ value might be due to the distortion of crystal lattice induced by different ionic substitution. The τ_f values were not sensitive to the sintering temperature, which varied in a narrow range from $-24.8 \text{ ppm}/^\circ\text{C}$ to $-23.0 \text{ ppm}/^\circ\text{C}$. Moreover, a small amount of $0.17\text{Li}_2\text{O}-0.83\text{V}_2\text{O}_5$ could effectively lower the

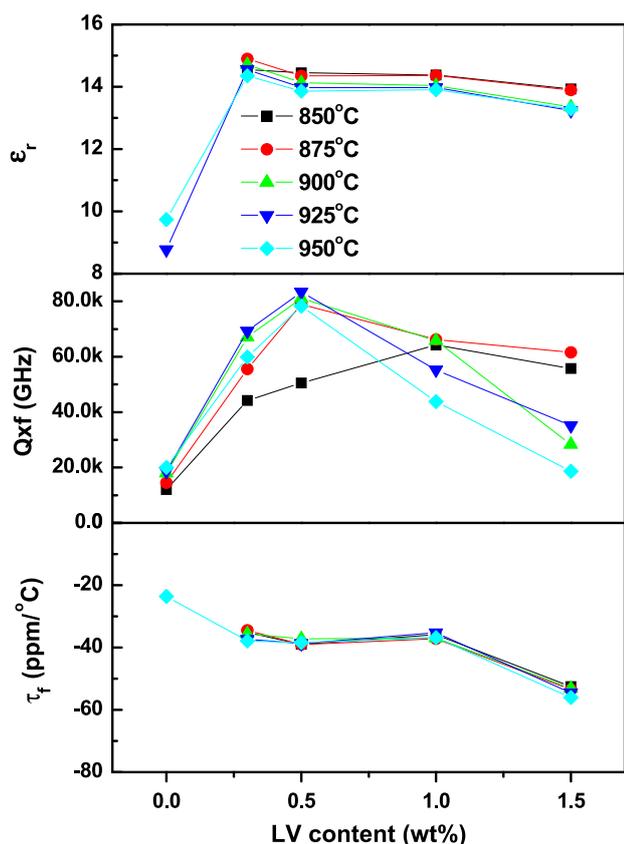


Fig. 8. The microwave dielectric properties of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramics with various contents of LV sintered at different temperatures for 2 h.

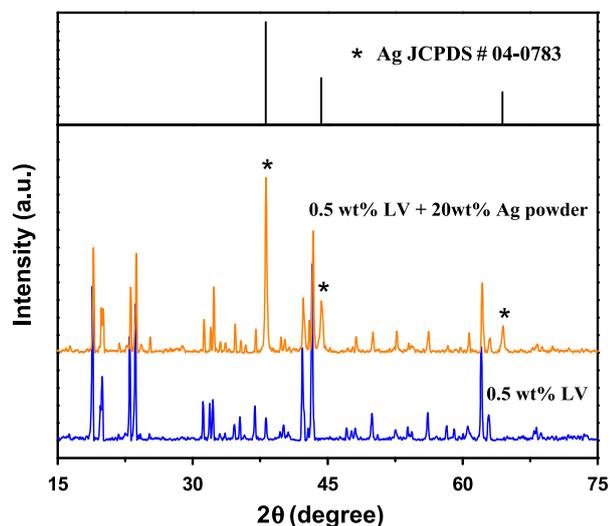


Fig. 9. The XRD pattern of the 0.5 wt% LV doped $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic mixed with 20 wt% Ag powder sintered at 925 °C for 2 h.

sintering temperature of $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic from 1120 °C to 925 °C. However, the formation of the secondary phases deteriorated the microwave dielectric properties. Nevertheless, excellent microwave dielectric properties of $\epsilon_r \sim 14$, $Q \times f \sim 83395$ GHz and $\tau_f \sim -37.2$ ppm/°C were achieved for the 0.5 wt% $0.17\text{Li}_2\text{O}-0.83\text{V}_2\text{O}_5$ doped $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$

ceramic sintered at 925 °C. Moreover, the LV doped $\text{Li}_3(\text{Mg}_{0.92}\text{Zn}_{0.08})_2\text{NbO}_6$ ceramic has a good chemical compatibility with Ag electrode, which makes it be a suitable candidate material for LTCC applications.

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