Accepted Manuscript

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PII:	S0254-0584(17)30592-8
DOI:	10.1016/j.matchemphys.2017.07.073
Reference:	MAC 19880
To appear in:	*Materials Chemistry and Physics*
Received Date:	24 April 2017
Revised Date:	14 July 2017
Accepted Date:	21 July 2017

Please cite this article as: Fu Zhifen, Ma Jianli, Liu Peng, Liu Yuan, Novel temperature stable $Li_2Mg_3TiO_6$-$SrTiO_3$ composite ceramics with high *Q* for LTCC applications, *Materials Chemistry and Physics* (2017), doi: 10.1016/j.matchemphys.2017.07.073

Novel temperature stable $Li_2Mg_3TiO_6$-$SrTiO_3$ composite ceramics with high *Q* for LTCC applications

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Abstract:

New temperature stable 4wt% LiF-doped (1-*x*)$Li_2Mg_3TiO_6$-*x*$SrTiO_3$ (LMT-ST, *x* = 0.05, 0.08, 0.10, 0.15) composite ceramics were fabricated by solid-state reaction for low-temperature co-fired ceramics (LTCC) applications. Well-densified LMT-ST composite ceramics were obtained with a relative density of 97.2% at 900 °C. A near zero of τ_f value was obtained by tuning ST composition. 0.9LMT-0.1ST-4wt% LiF ceramics displayed optimum microwave dielectric properties: ε_r = 19.5, $Q\times f$ = 64290 GHz, τ_f = 6.5 ppm/°C at 900 °C. Such samples were compatible with Ag electrodes, which suggests suitability of the developed material for LTCC applications in wireless communication systems.

Keywords: Dielectric Properties; Microstructure; $Li_2Mg_3TiO_6$; Composite Ceramics

1. Introduction

In recent years, the low-temperature co-fired ceramic (LTCC) applications have aroused considerable attention due to their alternating compact of dielectric ceramic

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substrates and internal metallic electrode layers. The microwave dielectric materials used in LTCC field have appropriate dielectric constant ($\varepsilon_r \leq 25$), higher $Q \times f$ value ($Q \times f \geqslant 5000$ GHz), a near-zero temperature coefficient of resonant frequency (τ_f), and co-fired with metal electrodes, such as Ag, Cu. Several microwave dielectric materials, such as Al_2O_3, Mg_2SiO_4, $Mg_4Nb_2O_9$, et al., have been extensively investigated as candidates for high-frequency applications [1-3]. However, their sintering temperatures are too high to reach an ecological and economic aim. Therefore, search for new low-cost, temperature stability and excellent performance LTCC materials is always a primary issue in the last few years.

Many dielectric material systems composed of Bi_2O_3, P_2O_5, Li_2O, V_2O_5, TiO_2, SiO_2, WO_3 and MoO_3 have been developed for LTCC applications [4-11]. Among them, Li-containing compounds such as Li_2WO_4, Li_3AO_4 (A = Nb, Ta, Sb), Li_2TiO_3, Li_2SnO_3 and Li_2MnO_3 have gained considerable attention due to their excellent microwave dielectric properties and lower sintering temperatures[12-17]. In our previous study, $Li_2Mg_3TiO_6$ (LMT) ceramics were fabricated at 1280 °C and displayed a cubic rock salt structure [18]. E.S. Kim et al. reported Li_2O-MgO-TiO_2 ceramics at 1360 °C due to its excellent microwave dielectric properties [19]. However, its higher sintering temperature and larger τ_f value hindered their practical applications in LTCC. LiF-doping enhanced sinterability of LMT ceramics and effectively lowered sintering temperatures to 950 °C without degrading its dielectric properties [20]. A near zero τ_f value was obtained for LMT-based ceramics by substitution of Zn or Ca for Mg, respectively [21-22]. It is known that $SrTiO_3$ (ST)

has large positive τ_f value (~ +1650 ppm/°C) and good microwave dielectric properties of ε_r = 290, $Q \times f$ = 4800 GHz [23]. Additionally, it has been reported as an independent phase in several diphasic systems [24-25]. So, one can expected that a diphasic composite material with a near-zero τ_f value and high Q may be obtained by combining ST with LMT.

In our previous study, low-fired LiF-doped LMT ceramics were obtained at 950 °C with τ_f ~ -44 ppm/°C [20]. In this paper, ST was employed as an τ_f compensator for LiF-doped LMT ceramics. The effects of ST and LiF on the microstructures, crystal compositions and microwave dielectric properties of Low-fired (1-x)$Li_2Mg_3TiO_6$-$xSrTiO_3$-4wt% LiF (LMT-ST) composite ceramics were investigated systematically.

2. Experimental procedure

Li_2CO_3 (98%), MgO (99.99%), TiO_2 (99.99%), $SrCO_3$ (99.9%) and LiF (99.99%) powders were used as starting materials. By calcining mixtures of Li_2CO_3-MgO-TiO_2 and $SrCO_3$-TiO_2 of stoichiometric compositions, LMT and ST powders were synthesized through the conventional solid-state route at 1000 °C for 4 h. Then, (1-x)LMT-xST-4wt% LiF (x = 0.05, 0.08, 0.10, 0.15) mixtures were prepared by mixing the LMT, ST and LiF powders at several weight ratios. The mixtures were ball milled for 8 h using ZrO_2 balls in an alcohol medium. The milled powders were dried, mixed with 5wt% PVA as a binder, and then screened with a 120 mesh. Subsequently, the powders were pressed into cylindrical disks (11.5 mm in diameter and about 6 mm in height) under a pressure of 200 MPa. Before sintering at 850~950 °C on alumina

plates in air for 6 h, all the pellets were pretreated at 550 °C in air for 2 h to expel the binder. Finally, 0.9LMT-0.1ST-4 wt% LiF mixtured with 10 wt% silver (Ag) powders were remilled for 2 h, pressed into cylindrical disks under a pressure of 200 MPa and were sintered at 900 °C in air for 2 h. In order to prevent the evaporation of Li and F in the sintering process, all the pellets were covered with the sacrificial powders, which were composed by LMT-ST-LiF mixing powders.

Sintered behaviors of the mixtured powders were investigated by using thermal gravimetric and differential thermal analysis (TG/DTA, SDT Q600 V8.0 Build 95, American) with heating rate of 10 °C/min in air from room temperature to 1000 °C. The bulk densities of sintered ceramics were measured by the liquid Archimedes method using distilled water. The scanning electron microscope (SEM, Quanta 200, FEI Company, Eindhoven, Holland) and energy dispersive X-ray spectroscopy (EDS) were carried out to examine the microstructure for polished surfaces of specimen. The crystal structures were analyzed using X-ray diffraction (XRD, Rigaku D/MAX2550, Tokyo, Japan) with CuK_α radiation (at 40 kV and 100 mA) and a graphite monochromator in the 2θ range of 20º ~ 70º. The microwave dielectric properties of the specimens were measured using a network analyzer (ZVB20, Rohde & Schwarz, Munich, Germany) with the $TE_{01\delta}$ shielded cavity method. The temperature coefficient resonant frequency (τ_f) was calculated with the following formula:

$$\tau_f = \frac{(f_2 - f_1)\times 10^6}{f_1(T_2 - T_1)} \quad (1)$$

Where, f_1 and f_2 represent the resonant frequency at T_1 and T_2, respectively.

3. Results and discussions

Fig. 1

Fig.1 shows TG/DTA curves of 0.9LMT-0.1ST-4 wt% LiF mixtures. The TG curve exhibited the first weight loss of approximately 3.4 % due to the evaporation of water, which is accompanied by three exothermic peaks in the range of 50~250 °C. The second weight loss of 4.0 % was related to the combustion of residual organic binder (PVC) [26], which is supported by two exothermic peaks in the range of 250 °C ~ 600 °C. No weight loss was observed in the range of 600~950 °C, and a weak endothermic peak around 717 °C is attribute to LiF liquid phase.

Fig. 2

Table 1

In LiF-doped LMT ceramics [20], densification temperatures of LMT ceramics were reduced from 1280 °C to 950 °C due to the elevated boundary diffusion coefficient caused by LiF liquid-phase sintering. Since lithium is volatile and evaporates at elevated sintering temperatures (≥ 1000 °C) [27], lower sintering temperature (950 °C) reduced Li volatilization from 6.0% to 1.1% and then improved density of LMT-base ceramics from 3.35 g/cm^3 to 3.44 g/cm^3. The bulk density and relative density of 4wt% LiF-doped (1-x)LMT-xST ($0.05 \leq x \leq 0.15$) ceramics are presented in Fig. 2. The bulk density, theoretical density and relative density of samples sintered at optimum temperatures are given in Table 1. In Fig. 2(a), the densities increased with increasing sintering temperatures and then decreased after reaching their respective maximum values. With increasing x from 0.05 to 0.15, the bulk densities increased from 3.40 g/cm^3 to 3.54 g/cm^3 and displayed a similar

tendency with theoretical density (seen in Table 1). The theoretical density (D) of sintered samples can be calculated using the equation [28]:

$$D = \frac{W_1 + W_2 + W_3}{W_1/D_1 + W_2/D_2 + W_3/D_3} \quad (2)$$

Where W_1, W_2 and W_3 are the weight percentage of LMT, ST and LiF respectively; D_1, D_2 and D_3 are the theoretical density of LMT, ST and LiF, respectively. The increasing in density was attribute to higher theoretical density of $SrTiO_3$ (5.12 g/cm^3) than that of $Li_2Mg_3TiO_6$-4wt%LiF (3.44 g/cm^3). As shown in Fig. 2(b), LiF-doped LMT-ST samples displayed higher relative density of 95% ~ 97% compared with other LMT-based ceramics [18-19].

Fig. 3

Fig. 3 shows XRD patterns of (1-x)LMT-xST-4wt% LiF ($0.05 \leq x \leq 0.15$) ceramics sintered at 900 °C as a function of x. XRD pattern of LMT powders calcined at 1000 °C displayed a cubic rock salt structure $Li_2Mg_3SnO_6$-like phase (JCPDS #39-0932, *Fm-3m* (225)), which was consistent with reported [20]. All sintered specimens displayed a two-phase system with a LMT phase and a perovskite $SrTiO_3$ (JCPDS #79-0176). No LiF phase was detected due to the substitution of smaller F^- ion (R = 1.33 Å) for O^{2-} (R = 1.4 Å), similar phenomenon was reported in LiF-doped LMT system. With increasing x, the diffraction peak intensity of ST phase varied dramatically.

Fig. 4

The SEM micrographs of (1-x)LMT-xST-4wt% LiF ($0.05 \leq x \leq 0.15$) ceramics sintered at 900 °C were displayed in Fig. 4(a~d). Well-dense microstructures were

observed for all samples at 900 °C. The sintered samples were composed of larger grains (marked as A) and smaller grains (marked as B), and the content of smaller grains increased with increasing *x*. Based on the EDS spectrum results (seen in Fig. 4(e and f)), larger grains and smaller grains were identified as $Li_2Mg_3TiO_6$ and $SrTiO_3$, respectively.

Fig. 5

Fig. 5 illustrates ε_r values of (1-*x*)LMT-*x*ST-4wt% LiF ($0.05 \leq x \leq 0.15$) ceramics as functions of *x* and sintering temperatures. As shown in Fig. 5, the variations in ε_r values with sintering temperatures and *x* were consistent with that of bulk density, suggesting that density is the dominating factor to control ε_r values in LMT-ST systems. Moreover, ε_r values of LMT-ST composite ceramics increase from 16.5 to 19.5 with increasing *x* from 0.05 to 0.15. The relative permittivity of the composite can be calculated using the mixture rule [29]:

$$\ln \varepsilon_r = v_1 \ln \varepsilon_{r1} + v_2 \ln \varepsilon_{r2} \quad (2)$$

Where ε_r is the relative permittivity of the composite; v_1, v_2 are the volume fractions of LMT and ST, respectively; ε_{r1}, ε_{r2} are the relative permittivity of LMT and ST, respectively. Since the relative permittivity of ST (ε_r = 290) is very larger than that of LMT (ε_r = 15), the relative permittivity of LMT-ST composite ceramics is expected to increase with increasing *x*. Similar phenomenon was observed in other LMT-based ceramics [21-22]. In $Li_2Mg_{3-x}Ca_xTiO_6$ ceramics [22], the relative permittivity increased with increasing Ca content due to the increased amount of $CaTiO_3$ (ε_r = 170). As presented in Table 1, the calculated relative permittivity values (ε_{cal}) were

generally higher than the measured ones (ε_{meas}) and they behave in a similar tendency. Since relative density of sintered samples were in the range of 95.7%~97.2%, there were some porosity of 3%~4.3% in composite ceramics. When air was considered as a third phase and would effect into the relative permittivity of composite due to its lower ε_r value (ε_r ~1), which would result in a lower relative permittivity compared with the calculated ones.

Fig. 6

Table 2

Fig. 6 demonstrates $Q\times f$ values of (1-x)LMT-xST-4wt% LiF ($0.05 \leq x \leq 0.15$) ceramics as functions of x and sintering temperatures. It is well known that $Q\times f$ of a composition depends on intrinsic parameters such as structural characteristics and extrinsic parameters such as densification/porosity, secondary phases, lattice defects, impurity, and microstructure characteristics [30]. The increase in $Q\times f$ with sintering temperature was attributed to the increase in the density as well as grain growth. Nevertheless, $Q\times f$ values of LMT-ST composite ceramics decreased linearity from 86200 GHz to 48870 GHz as x increased from 0.05 to 0.15. The decreasing of $Q\times f$ values with increasing x was associated with much lower $Q\times f$ values of ST (4800 GHz) than that of LMT. Although $Q\times f$ values of LMT-ST composite ceramics are somewhat lower than other LMT-based ceramic system (seen Table 2), but ceramics shows a significantly low sintering temperature of 900 °C.

Fig. 7

Fig. 7 illustrates τ_f values of (1-x)LMT-xST-4wt% LiF ($0.05 \leq x \leq 0.15$)

composite ceramics as functions of x and sintering temperatures. In general, τ_f is influenced by the composition, the additive and the second phase of the materials [31]. It is seen that the variation in τ_f values with respect to sintering temperature was not significant. However, τ_f values continuous increased from -32 ppm/°C to 46 ppm/°C as increasing x from 0.05 to 0.15. This implies that a near zero of τ_f value could be obtained by tuning $SrTiO_3$ composition (x). Similar effect was also observed in $MgTiO_3$-$SrTiO_3$ system [25]. Compared with LiF-doped LMT ceramics [20], LMT-ST-4wt% LiF composite ceramics displayed a 6.5 ppm/°C of τ_f value when $SrTiO_3$ content is x = 0.1.

Fig. 8

The microwave dielectric materials used in LTCC field are required co-fired with metal electrodes (such as Ag, Cu) at < 960 °C. The chemical compatibility with metal electrodes Ag was investigated by co-firing the 0.9LMT-0.1ST-4wt% LiF compounds with pure Ag powders at 900 °C for 2 h. Fig. 8 displays XRD pattern, SEM image and EDS analyses of 10 wt% Ag-doped 0.9LMT-0.1ST-4wt% LiF ceramics. As shown in XRD pattern, multiphase of LMT, ST and respective metals (Ag) were observed and no additional peak could be detected. In SEM and EDS analyses, the spot belong to the Ag powders phase, and no other chemical elements were detected. The analysis results of Fig.8 implied that no chemical reaction was happened between Ag and 0.9LMT-0.1ST-4wt% LiF ceramics. Similar phenomenons were observed in other Ag addition of LTCC materials [6,9,12-13]. Based on the above data therefore, 0.9LMT-0.1ST-4wt% LiF composite ceramics possessed

optimum dielectric properties of ε_r = 19.5, $Q \times f$ = 64290 GHz, τ_f = 6.5 ppm/°C at 900 °C, which suggests the possibility of utilizing it as an LTCC material.

4. Conclusion

New temperature stable (1-*x*)LMT-*x*ST-4wt% LiF ($0.05 \leq x \leq 0.15$) ceramics were achieved in this work. The influences of ST on the crystal compositions, microstructures and microwave dielectric properties of LMT-based ceramics were studied. Well-densified microstructures were obtained at 900 °C for LMT-ST composite ceramics by doping 4wt% LiF. The microwave dielectric properties of LMT-ST composite ceramics were mainly controlled by the $SrTiO_3$ composition (*x*). A typical 0.9LMT-0.1ST-4wt% LiF sample was prepared with optimum dielectric properties of ε_r = 19.5, $Q \times f$ = 64290 GHz, τ_f = 6.5 ppm/°C. Furthermore, the chemical compatibility of 0.9LMT-0.1ST-4wt% LiF composition with Ag powders was also investigated. Well chemical compatibility was obtained co-fired at 900 °C for 2 h, indicating that low-temperature sintering ceramics powders were suitable for the LTCC applications.

Acknowledgment

This work is supported by the National Natural Science Foundation of China (Grant nos. 51572162 and 51602005), the Natural Science Research Project of High Education Institutions in Anhui Province of China (Grant no. KJ2017A090).

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Table 1. The density, and microwave dielectric properties of 4 wt% LiF-doped LMT-ST ceramics sintered at optimum temperatures

x	0.05	0.08	0.10	0.15
Ts (°C)	900	900	900	900
ρ (g/cm^3)	3.40	3.45	3.51	3.54
D (g/cm^3)	3.52	3.57	3.61	3.69
Relative density (%)	96.6	96.6	97.2	95.7
ε_{cal}	17.6	19.2	20.4	23.7
ε_{meas}	16.5	17.1	17.7	19.5
$Q\times f$ (GHz)	86200	75840	64290	48870
τ_f (ppm/°C)	-27	-15.4	6.5	46.6

D: the theoretical density

Table 2. Comparison of the proposed dielectrics with other similar ceramic systems.

Ceramics composition	ε_r	$Q\times f$ (GHz)	τ_f (ppm/°C)	S.T. (°C)	Ref.
$Li_2Mg_3TiO_6$	15.2	152000	-39	1260	[15]
Li_2O-MgO-TiO_2	14.42	153000	-11.07	1320	[16]
$Li_2(Mg_{0.95}Zn_{0.05})_3TiO_6$	14.6	158000	3.2	1275	[18]
$Li_2Mg_{2.88}Ca_{0.12}TiO_6$	17.8	102246	-0.7	1280	[19]
$Li_2Mg_3TiO_6$-4 wt%LiF	16.2	131000	-44	950	[17]
0.9 $Li_2Mg_3TiO_6$-0.1$SrTiO_3$-4wt%LiF	17.7	64290	6.5	900	*

*This work

Figure captions

Fig. 1. TG/DTA curves of 0.9LMT-0.1ST-4 wt% LiF mixtures

Fig. 2. The density and relative density of 4 wt% LiF-doped LMT-ST ceramics as functions of *x* and sintering temperatures

Fig. 3. XRD patterns of 4 wt% LiF-doped LMT-ST ceramics sintered at 900 °C as a function of *x*

Fig. 4. (a~d) SEM images of 4 wt% LiF-doped LMT-ST ceramics sintered at 900 °C; (e~f) EDS spectrums of Area A and B in the Fig. 4(d)

Fig. 5. ε_r values of 4 wt% LiF-doped LMT-ST ceramics as functions of *x* and sintering temperatures

Fig. 6. $Q \times f$ values of 4 wt% LiF-doped LMT-ST ceramics as functions of *x* and sintering temperatures

Fig. 7. τ_f values of 4 wt% LiF-doped LMT-ST ceramics as functions of *x* and sintering temperatures

Fig. 8. XRD pattern, SEM image and EDS spectrum of 0.9LMT-0.1ST-4 wt% LiF ceramic doped with 10 wt% Ag and sintered at 900 °C for 2 h

Figures and captions

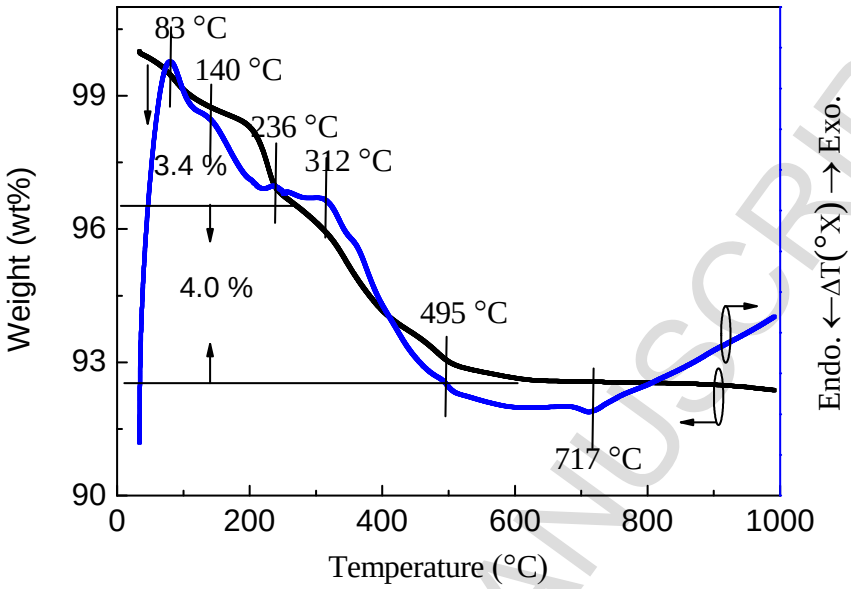

Fig. 1. TG/DTA curves of 0.9LMT-0.1ST-4 wt% LiF mixtures

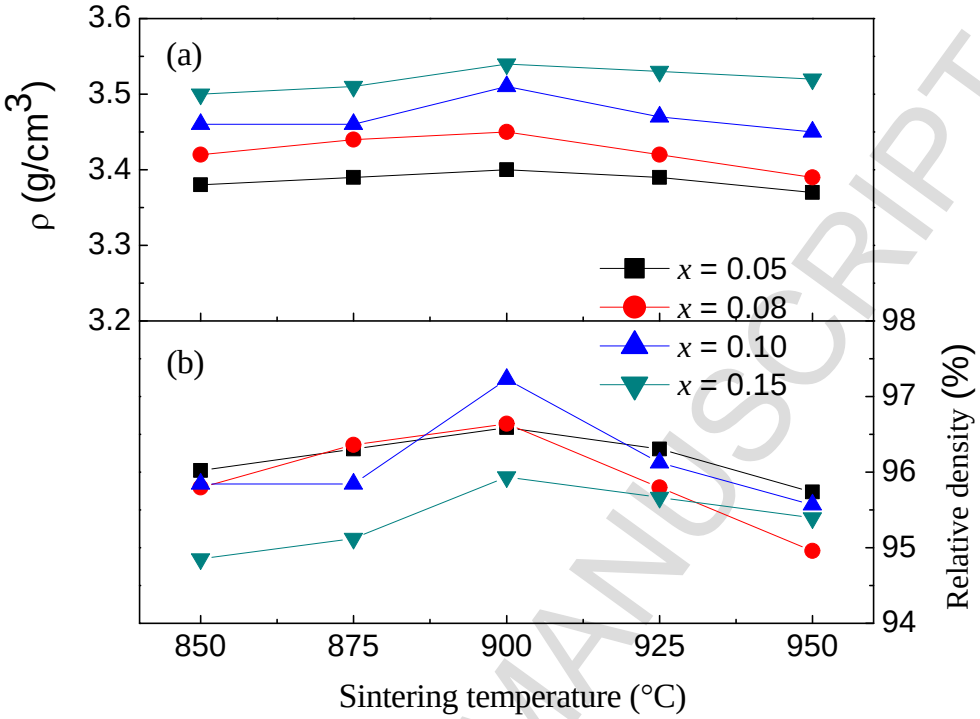

Fig.2. The density and relative density of 4wt% LiF-doped LMT-ST ceramics as functions of *x* and sintering temperatures

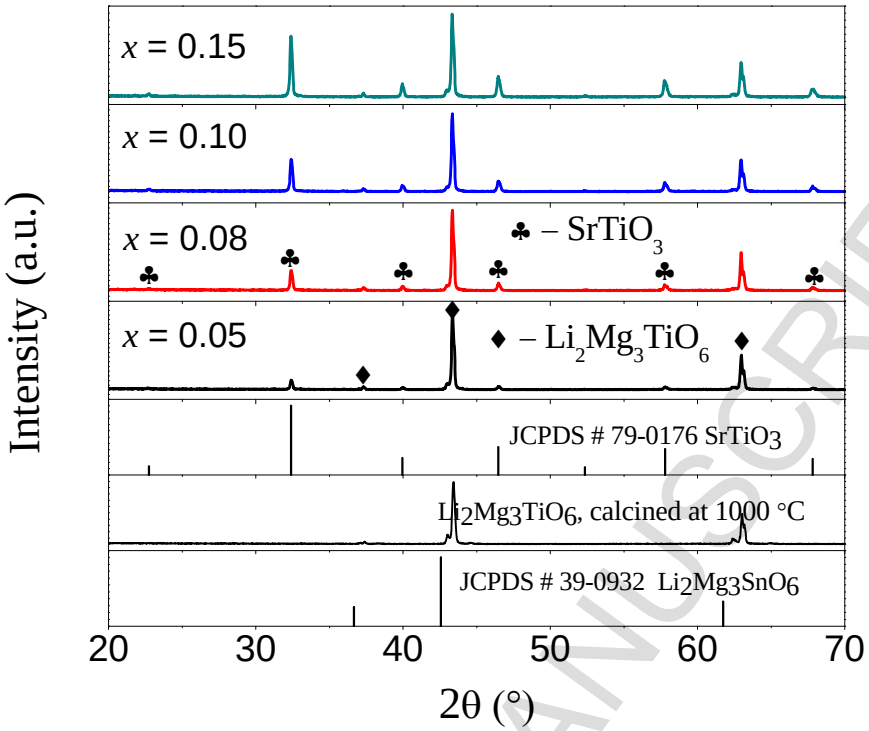

Fig.3. XRD patterns of 4wt% LiF-doped LMT-ST ceramics sintered at 900 °C as a function of *x*

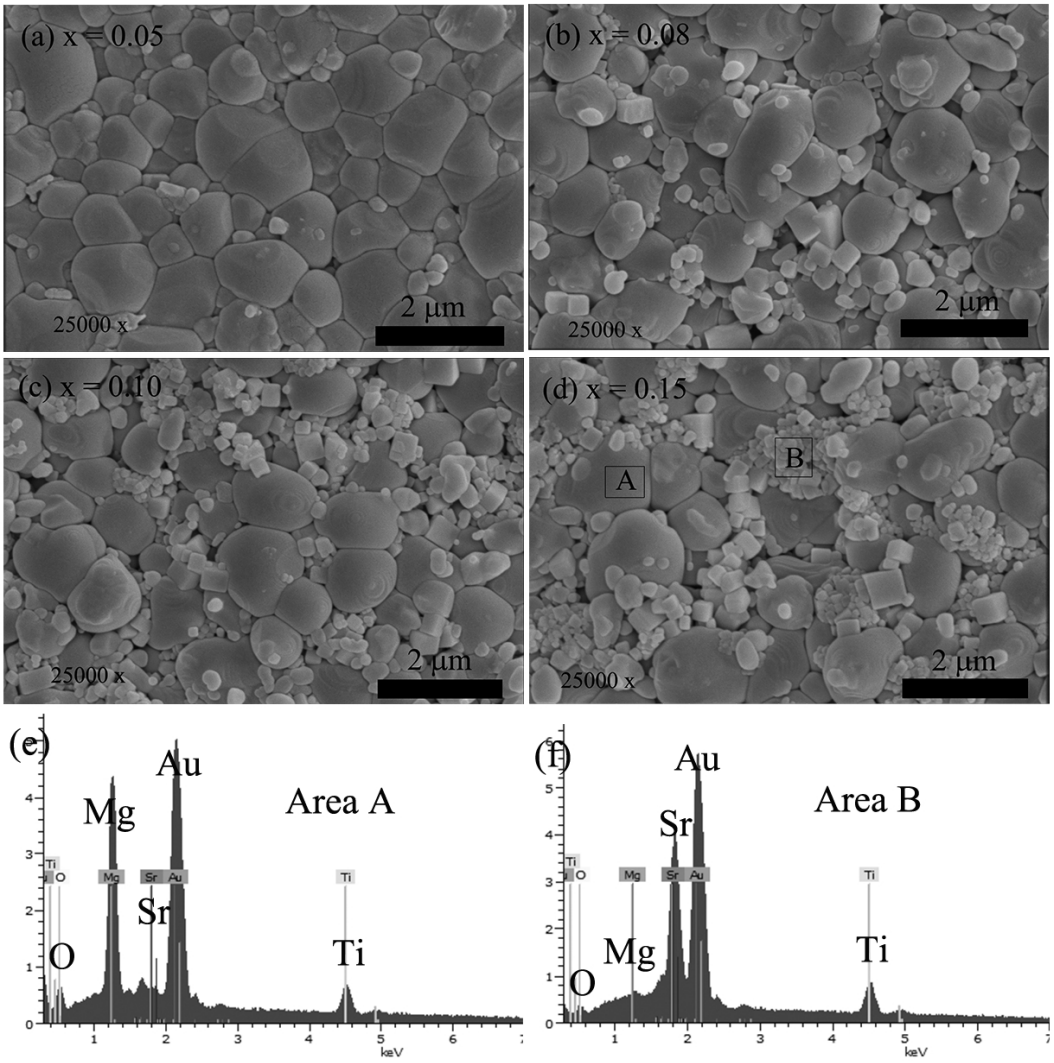

Fig.4. (a~d) SEM images of 4 wt% LiF-doped LMT-ST ceramics sintered at 900 °C;

(e~f) EDS spectrums of Area A and B in the Fig. 4(d)

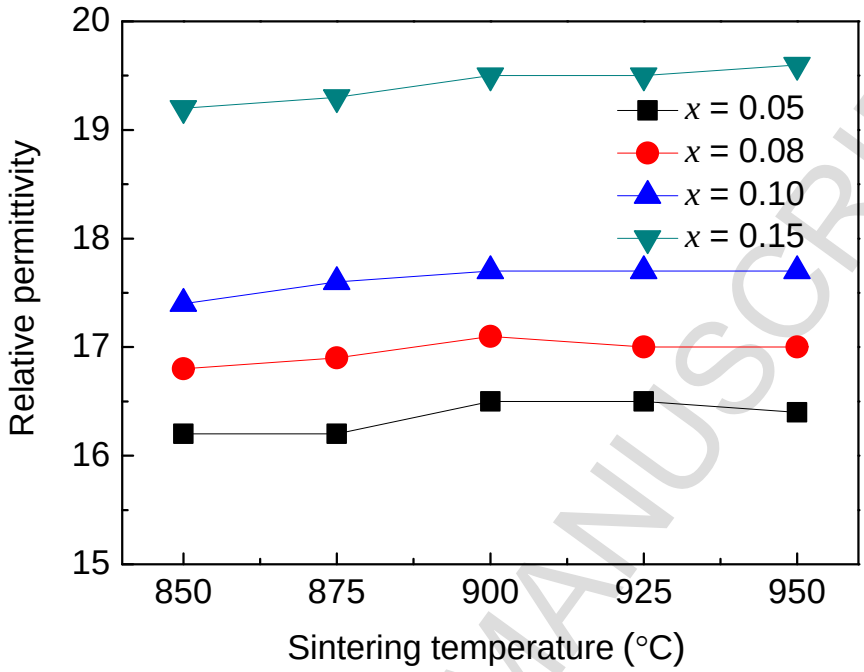

Fig.5. ε_r values of 4wt% LiF-doped LMT-ST ceramics as functions of x and sintering temperatures

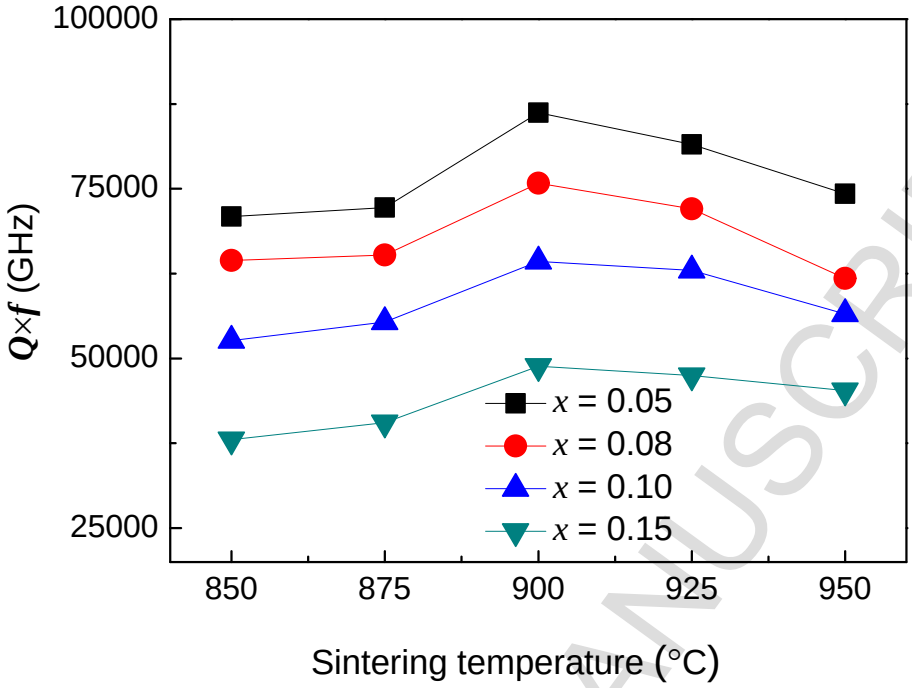

Fig.6. *Q*×*f* values of 4wt% LiF-doped LMT-ST ceramics as functions of *x* and sintering temperatures

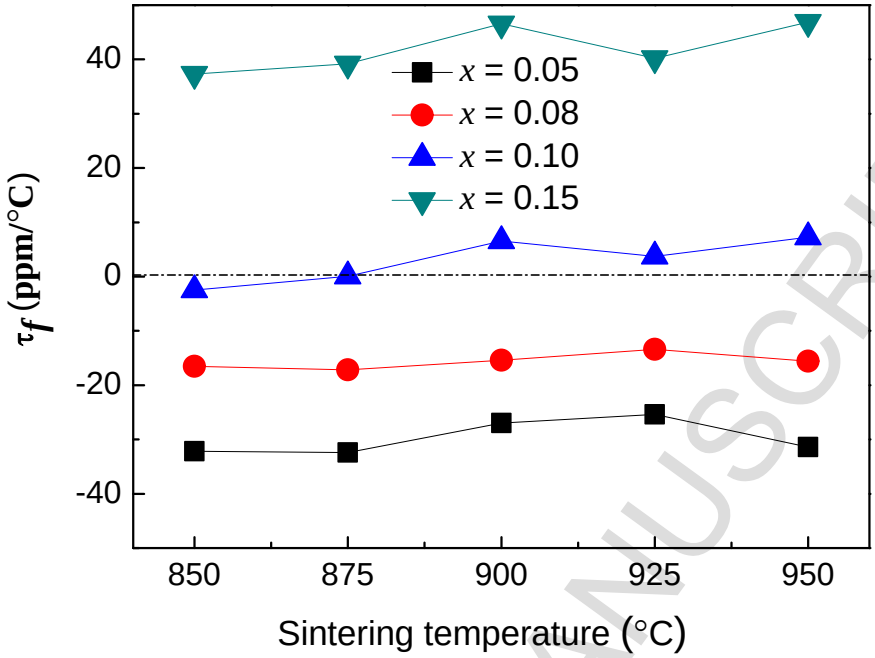

Fig.7. τ_f values of 4wt% LiF-doped LMT-ST ceramics as functions of x and sintering temperatures

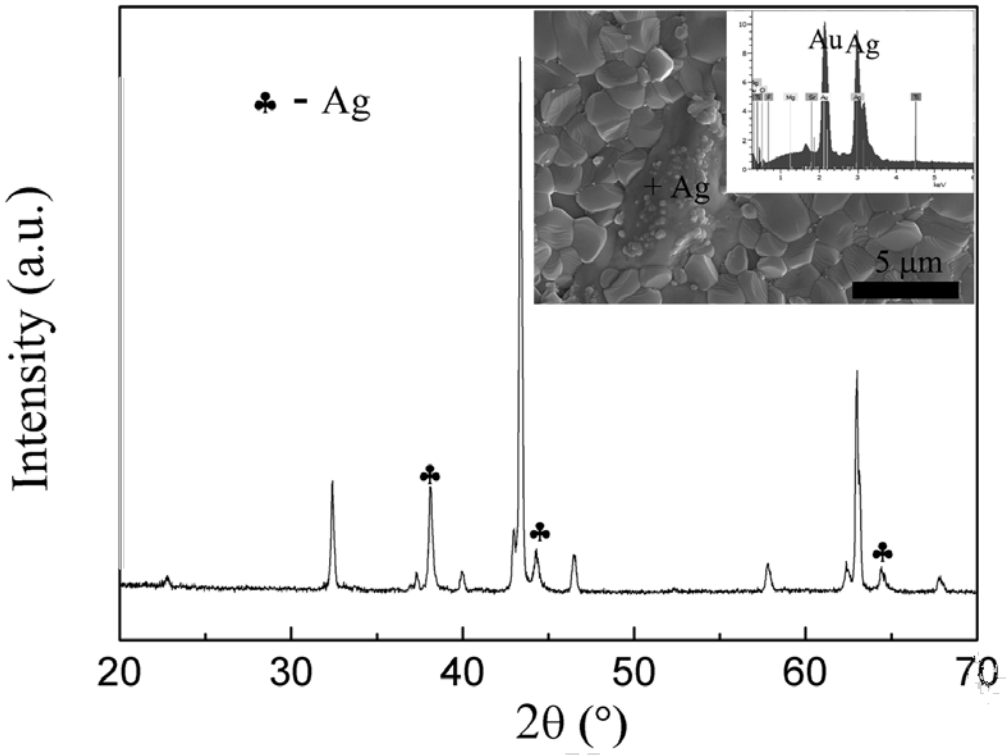

Fig.8. XRD pattern, SEM image and EDS spectrum of 0.9LMT-0.1ST-4 wt% LiF ceramic doped with 10 wt% Ag and sintered at 900 °C for 2 h

- Temperature stable $Li_2Mg_3TiO_6$-$SrTiO_3$ composite ceramics with high Q were prepared.
- LiF-doping lowered sintering temperature of LMT-ST ceramics to 900 °C.
- The temperature stability was improved by adding ST.
- A near zero τ_f values of 6.5 ppm/°C is obtained for samples for x = 0.1.
- Ceramics with well dielectric properties are suitable for LTCC applications.