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Authors: Weiqiong Liu, Ruzhong Zuo

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A novel Li_2TiO_3–Li_2CeO_3 ceramic composite with excellent microwave dielectric properties for low-temperature cofired ceramic applications

Weiqiong Liu, Ruzhong Zuo*

Institute of Electro Ceramics & Devices, School of Materials Science and Engineering, Hefei University of Technology, Hefei, 230009, P.R. China

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* the corresponding author
Email: rzzuo@hotmail.com
Tel: 86-551-62905285; Fax: 0086-551-62905285

ABSTRACT

A novel low-temperature sinterable (1-x)Li_2TiO_3-xLi_2CeO_3 (x=0.08-0.16 in molar) microwave dielectric ceramic was successfully prepared by a conventional solid-state reaction method. The X-ray diffraction and scanning electron microscopy analysis revealed the coexistence of two phases with different structures owing to their good chemical stability. Their relative content was easily adjusted to achieve near-zero temperature coefficient of the resonant frequency (τ_f) according to the mixing rule of dielectrics. The low-temperature sintering and desirable microwave dielectric properties can be simultaneously achieved by adding Li_2CeO_3 to the Li_2TiO_3 matrix owing to its low-firing characteristic and opposite-sign τ_f. The composite ceramics with x=0.14 could be well sintered at 850 ºC and exhibited excellent microwave dielectric properties of ε_r~21.2, Qxf~ 59,039 GHz and τ_f~-7.4 ppm/ºC. In addition, no chemical reaction was identified between the matrix phase and Ag, suggesting that the Li_2TiO_3-Li_2CeO_3 ceramics might be promising candidates for low-temperature co-fired ceramic applications.

Keywords**:** LTCC; Microwave dielectrics properties; Co-firing; Near-zero τ_f

1. Introduction

In the past few years, microwave telecommunication technology has made great achievements. Miniaturization of microwave components is essential for the production of multilayer or chip devices in wireless communications. Low temperature cofired ceramic (LTCC) technology is imperative for the fabrication of modern electronic devices [1]. Dielectric materials for LTCC applications require high relative permittivity (ε_r >20 for miniaturization), low sintering temperature (<960 ºC, the melting temperature of Ag electrode), high quality factor (Qxf>10,000GHz), and a near-zero temperature coefficient of resonant frequency ($|\tau_f| \leq$ 10 ppm/°C) [2]. These merits are the key requirements for practical applications. For this purpose, LTCC with suitable dielectric properties has been widely investigated.

Many ceramics have been reported so far because of their good microwave dielectric properties, such as Mg_2SiO_4, $Ba(Mg_{1/3}Ta_{2/3})O_3$, $Mg_4Nb_2O_9$, $ZnAl_2O_4$, etc [3-7]. However, the high sintering temperature (>1000 °C) and undesirable τ_f values impeded their practical applications. Addition of low-melting-point additives, such as V_2O_5, Bi_2O_3 and B_2O_3, was considered to be the simplest way to lower the sintering temperature [8-10]. The usage of the opposite-τ_f materials was a common way to tailor τ_f values [11-12]. However, these methods usually tended to cause the deterioration in microwave dielectric properties. Hence, searching for new materials with excellent microwave dielectric properties and low sintering temperatures is still an arduous task for scientific research.

Li_2TiO_3 ceramic with a rock salt structure has attracted extensive attention for its good microwave dielectric properties [13]. Pure Li_2TiO_3 ceramic cannot be well densified at high sintering temperatures (>1200 °C) because of serious volatilization of lithium. A series of low-melting-point additives such as $ZnO-B_2O_3$, LiF, $Li_2O-MgO-B_2O_3$ has successfully improved the density of Li_2TiO_3 ceramic and boosted the Qxf value at the same time. Good microwave dielectric properties ($\varepsilon_r = 20-24$, Qxf = 15,000-70,000 GHz and $\tau_f = 30 \sim 37$ ppm/°C) were obtained at a relatively low sintering temperature (900 °C-950 °C) [14-16]. However, the undesirable τ_f value is another important factor impeding its practical application. It was reported that a few metal oxides (MO, M=Mg, Zn, Ni) can form complete solid solutions with Li_2TiO_3, and the replacement mechanism can be considered as $3M^{2+} \leftrightarrow 2Li^{+} + Ti^{4+}$ [17, 18]. The addition of MO increased the cation ordering degree of Li_2TiO_3, thus resulting in the achievement of high Qxf values. Meanwhile, the change in the tilting angle of oxygen octahedra successfully tailored the τ_f value to near zero. Furthermore, LiF and BCB were added in Li_2TiO_3-MO (LTM) systems to further decrease the sintering temperature. Good microwave dielectric properties were achieved by Li_2TiO_3-MgO-4wt% LiF (ε_r=15.8, Qxf=64,500 GHz, and τ_f=-0.2 ppm/°C) and Li_2TiO_3-NiO-1wt% BCB (ε_r=19, Qxf=62,252 GHz, and τ_f=-1.65 ppm/°C) at 850 °C for 4 h [18, 19].

The addition of new materials with a low sintering temperature, an opposite-sign τ_f value and a good dielectric performance may be the simplest and effective way for simultaneously solving LTCC-oriented problems. Li_2CeO_3 with a CeO_2 structure was reported to have a large τ_f value of -123 ppm/°C, a high Qxf value of ~143,700 GHz and a relative permittivity of ε_r ~15.8 when sintered at an extremely low temperature (720 °C) [20]. Compared with other τ_f-modifiers and sintering aids, relatively good microwave dielectric properties would make Li_2CeO_3 become a more suitable one. Hence, one can expect that a new diphasic composite material with a near-zero τ_f value, a low sintering temperature and a high Qxf value could be obtained by combining Li_2TiO_3 with Li_2CeO_3. The sintering behavior, microstructure, microwave dielectric properties and chemical compatibility with Ag were investigated in detail.

2. Experimental

The (1-x)Li_2TiO_3-xLi_2CeO_3 (x=0.08~0.16 in molar) ceramics were prepared by a conventional solid-state reaction method using high-purity starting powders of Li_2CO_3 (98.0 %), TiO_2 (99.0 %) and CeO_2(99.0 %). The raw materials were weighed according to the above formula and then ball-milled for 4 h using zirconia balls and alcohol as the medium on a planetary milling machine. The resulting slurries were then rapidly dried and calcined at 800 °C for 6 h in air. The calcined powders were re-milled for 6 h and then mixed together with 5 wt% PVA as a binder. The granulated powders were subsequently pressed into cylinders with dimensions of 10 mm in diameter and 7-8 mm in height. The specimens were first heated at 550 °C in air for 4 h to remove the organic binder, and then sintered at 800 ºC ~900 ºC for 4 h.

The crystal structure of the fired ceramics was identified via an X-ray diffractometer (XRD, D/Max2500 V, Rigaku, Japan) using Cu $K\alpha$ radiation. The bulk densities of the sintered ceramics were measured by the Archimedes method. The microstructure of the pellets was observed using a field-emission scanning electron microscope (FE-SEM; SU8020, JEOL, Tokyo, Japan) equipped with an energy dispersive spectrometer (EDS). Before the SEM observation, the sample was polished and thermally etched at 650 ºC for 30 min. Microwave dielectric properties of sintered ceramics were measured using a network analyzer (N5230C, Agilent, Palo Alto, CA) and a temperature chamber (GDW-100, Saiweisi, Changzhou, China). The τ_f values of the samples were measured in the temperature range from 20 ºC to 80 ºC and calculated by the following equation:

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

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

3. Results and discussion

Fig. 1(a) presents the normalized XRD patterns of (1-x)Li_2TiO_3-xLi_2CeO_3 ceramics sintered at optimum temperatures for 4 h. The powder diffraction standard file for Li_2CeO_3 was considered to be the same with CeO_2 [20]. All the main peaks can be well indexed by Li_2TiO_3 (JCPDS #33-0831) and CeO_2 (JCPDS #81-0792). No other phases can be observed in the whole composition range. The XRD results suggest that no chemical reaction occurred between Li_2TiO_3 and Li_2CeO_3.The Li_2TiO_3 crystallized in a monoclinic structure with the space group C2/c, and the Li_2CeO_3 crystallized in a cubic structure with the space group Fm-3m [13, 20].

The big difference in their crystal structures might be responsible for the formation of composite ceramics. Moreover, Figs. 1(b) and (c) locally present the strongest diffraction peaks of Li_2TiO_3 and Li_2CeO_3 phases, which were found to slightly shift toward higher angles as x increased from 0.08 to 0.10, and then remain at the same degrees with furthering increasing x. This phenomenon might be related to the ionic diffuse between Li_2TiO_3 and Li_2CeO_3. The smaller Ti ions could be easier to dissolve into the crystal lattice of Li_2CeO_3, such that the reduced content of Ti^{4+} ions would result in the decrement of the unit cell volume of Li_2TiO_3, corresponding to the diffraction peak shift toward higher angles in Fig. 1(b). At the same time, the replacement of smaller Ti^{4+} for Ce^{4+} would also result in the decrement of the unit cell volume in the crystal lattice of Li_2CeO_3, corresponding to the diffraction peak shift toward higher angles in Fig. 1(c). The completely different crystal structures between Li_2TiO_3 and Li_2CeO_3 determined the limited solid solubility. As a result, the diffraction peak position did not exhibit obvious changes any more as x was more than 0.08.

The SEM image of the x=0.14 ceramic sintered at 850 ºC for 4 h is shown in Fig. 2. It can be noted that two kinds of grains were distributed in the specimen, such as large irregular grains (marked as a), and small roundish grains (marked as b). The EDS analysis was also used to distinguish these two kinds of grains. The EDS results of x=0.14 sample are listed in Table 1. It can be clearly seen that the large irregular grains mainly contained Ti and O elements, meaning that they belonged to the Li_2TiO_3 phase, and small roundish grains were dominantly composed of Ce and O elements, which should belong to the Li_2CeO_3 phase. Li elements were not detected by EDS because of its light atomic mass. It should be noted that small roundish grains also contained a certain amount of Ti, but large irregular grains contained little Ce, which means that a small amount of Ti^{4+} ions were dissolved in the Li_2CeO_3 crystal lattice, while Ce^{4+} could hardly dissolve in the Li_2TiO_3 crystal lattice. The EDS result kept a good consistency with the above-mentioned XRD result. The basic reason seems to be attributed to the fact that relatively small Ti^{4+} ions (0.61 Å) compared with Ce^{4+} ions (0.87 Å) would be easily dissolved in the crystal lattice of the other phase [21].

Fig. 3 shows the variation of the sample density, ε_r and Qxf of (1-x)Li_2TiO_3-x Li_2CeO_3 ceramics as a function of sintering temperature. As shown in Fig. 3(a), the density of each composition firstly increased with increasing sintering temperature, then reached the maximum value at 850 °C, and finally decreased with further increasing sintering temperatures. Fig. 3(b) indicates that the variation of ε_r showed a similar trend to that of the bulk density as a function of sintering temperature. It was reported that the composition, grain size, and the density should primarily contribute to the relative permittivity. Among them, the density of the samples usually affected the relative permittivity markedly [22]. Therefore, the ε_r value increased with increasing sintering temperature up to 850 ºC and then decreased thereafter. As shown in Fig. 3(c), the improvement in quality factor of each composition from 800 ºC to 850 ºC should be attributed to the increase in sample density. The deterioration of Qxf values might be due to the degradation in densification caused by the evaporation of lithium at elevated temperatures [23, 24].

Fig. 4 shows lnε_r, $(Qxf)^{-1}$ and τ_f values of Li_2TiO_3-Li_2CeO_3 ceramics as a function of the volume fraction of Li_2CeO_3. According to the mixing rule, the microwave dielectric properties of a dielectric composite can be described by the following equations [25]:

$$\ln\varepsilon_r = V_1\ln\varepsilon_{r1} + V_2\ln\varepsilon_{r2} \qquad (2)$$

$$(Qxf)^{-1}=V_1(Qxf)_1^{-1}+V_2(Qxf)_2^{-1} \qquad (3)$$

$$\tau_f = V_1\tau_{f1} + V_2\tau_{f2} \qquad (4)$$

where V is the volume fraction of each phase. It can be seen that $\ln\varepsilon_r$, $(Qxf)^{-1}$ and τ_f in current work have a nearly linear relationship with the volume percentage of the Li_2CeO_3 phase. Fig. 4(a) shows that the $\ln\varepsilon_r$ values of (1-x)Li_2TiO_3-xLi_2CeO_3 ceramics decreased with increasing the volume fraction of Li_2CeO_3, which could be attributed to a lower ε_r value of Li_2CeO_3 (~15.8) than that of Li_2TiO_3 (~23.3). In addition, the measured $\ln\varepsilon_r$ values were found to be close to the calculated $\ln\varepsilon_r$ values, indicating that the mixing rule is applicable. However, the measured Qxf values were found to be lower than calculated values (Fig. 4(b)). This is probably because the quality factor is more easily influenced by other factors such as lattice defects, grain size, and porosity and so on. For τ_f values (Fig. 4(c)), the similar difference between measured and calculated values could be also found in other studies [26, 27]. Nevertheless, optimal microwave dielectric properties of $\varepsilon_r \sim 21.2$, Qxf ~ 59,039 GHz, and a near-zero $\tau_f \sim -7.4$ ppm/°C were obtained in 0.86Li_2TiO_3-0.14Li_2CeO_3 ceramics as sintered at 850 °C for 4h.

Table 2 presents a comparison of microwave dielectric ceramics with similar permittivities and low sintering temperatures (≤850 °C). By comparison, $PbWO_4$, Li_2TiO_3-NiO-1 wt% BCB, 0.45$(Na_{0.5}La_{0.5})MoO_4$-0.55$(Na_{0.5}Bi_{0.5})MoO_4$, and 0.86Li_2TiO_3-0.14Li_2CeO_3 were reported to have relatively small τ_f values. And relatively high Qxf values of 62,252 GHz and 59,309 GHz were obtained in Li_2TiO_3-NiO-1wt% BCB and 0.86Li_2TiO_3-0.14Li_2CeO_3, respectively. Compared with the former composition, the addition of Li_2CeO_3 can play a crucial role in tailoring undesirable τ_f values and decreasing relatively high sintering temperatures at the same time. A relatively simple processing of the Li_2TiO_3-Li_2CeO_3 composite ceramic might make it be an ideal candidate for practical LTCC applications.

In order to identify the suitability of LTCC applications, the chemical compatibility of the Li_2TiO_3-Li_2CeO_3 composite ceramic with Ag was examined. 30 wt% Ag powder was added into the $0.86Li_2TiO_3$-$0.14Li_2CeO_3$ powder, and the mixture was co-sintered at 850 ºC for 4 h. Fig. 5 shows the XRD pattern and SEM image of the co-fired sample at 850 °C. It is noted that all diffraction peaks could be well indexed to Li_2TiO_3, Li_2CeO_3 and Ag, suggesting that no any chemical reaction occurred between the composite ceramic and the Ag particle. This could be further confirmed by the SEM image of the cofired sample, as shown in the inset of Fig. 5. Distinct boundary between the ceramic and the Ag particle could be observed. These results demonstrated an excellent chemical stability between the Li_2TiO_3-Li_2CeO_3 ceramic and the Ag electrode at 850 ºC and a large potential of the studied composite for LTCC applications.

4. Conclusions

The $(1-x)Li_2TiO_3$-xLi_2CeO_3 composite ceramics were fabricated by a conventional solid-state reaction method. The phase composition, microstructure, sintering behavior and microwave dielectric properties of $(1-x)Li_2TiO_3$-xLi_2CeO_3 ceramics were investigated as a function of sintering temperature. The low-temperature sintering, high Qxf value and the temperature stability of resonant frequency can be simultaneously achieved by adding Li_2CeO_3 to the Li_2TiO_3 matrix. The X-ray diffraction and scanning electron microscopy analysis revealed the stable coexistence of two phases with different structures owing to their good chemical stability. The x=0.14 composite ceramic could be well sintered at 850 ºC and exhibited excellent microwave dielectric properties of $\varepsilon_r \sim 21.2$, Qxf ~ 59,039 GHz, and $\tau_f \sim -7.4$ ppm/ºC. Besides, the composite ceramics exhibited a good chemical compatibility with Ag, showing great potentials for practical applications in various LTCC devices.

Acknowledgements

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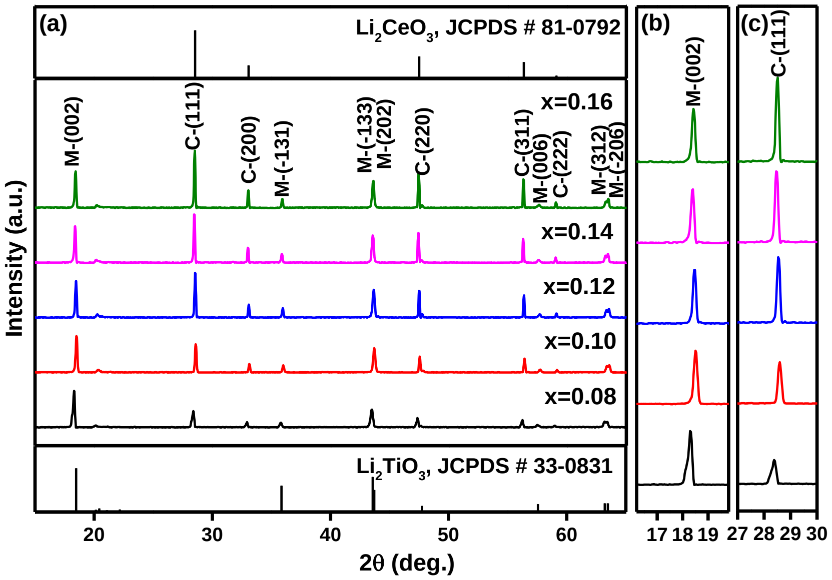

Fig. 1. (a) XRD patterns of $(1\text{-}x)Li_2TiO_3\text{-}xLi_2CeO_3$ ceramics sintered at optimum temperatures, (b) and (c) the locally enlarged diffraction peaks for the Li_2TiO_3 and Li_2CeO_3 phases, respectively.

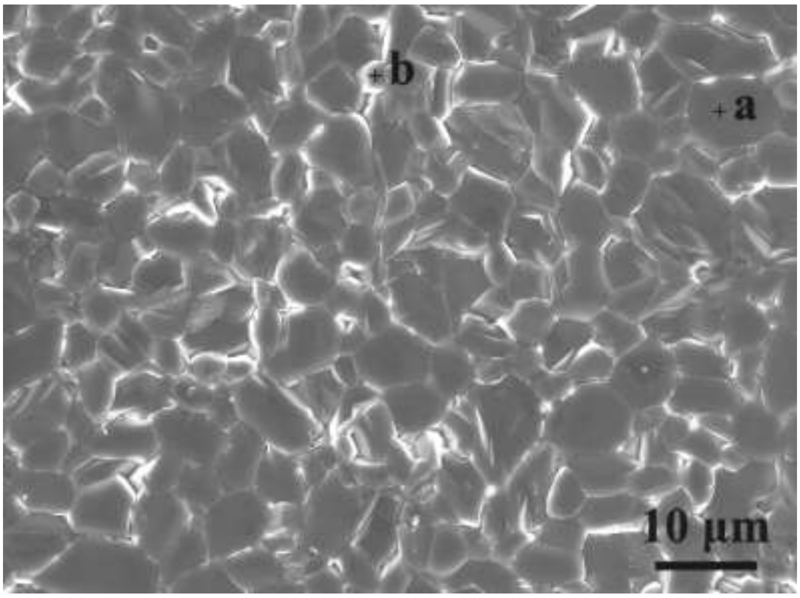

Fig. 2. SEM image on the polished and thermally surface of the x=0.14 ceramic sintered at 850 °C for 4 h.

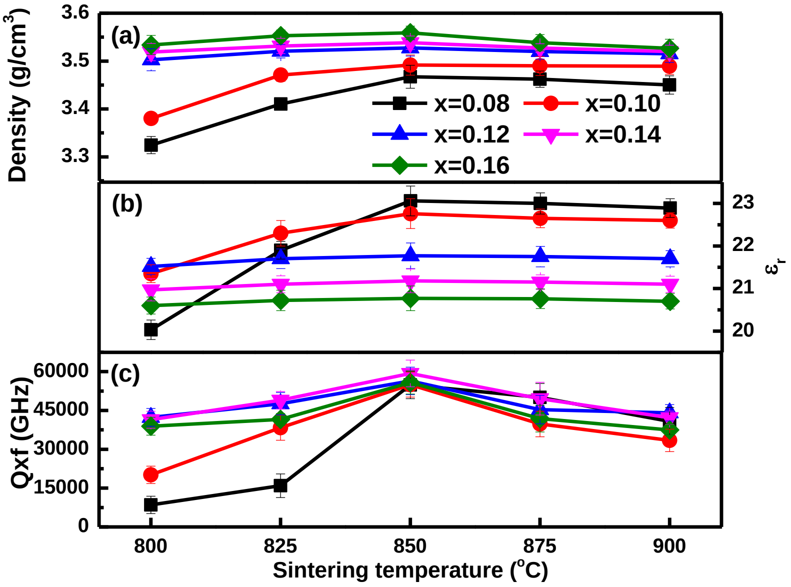

Fig. 3. (a) The sample density, (b) ε_r and (c) Qxf values of (1-x)Li_2TiO_3-xLi_2CeO_3 ceramics as a function of sintering temperature.

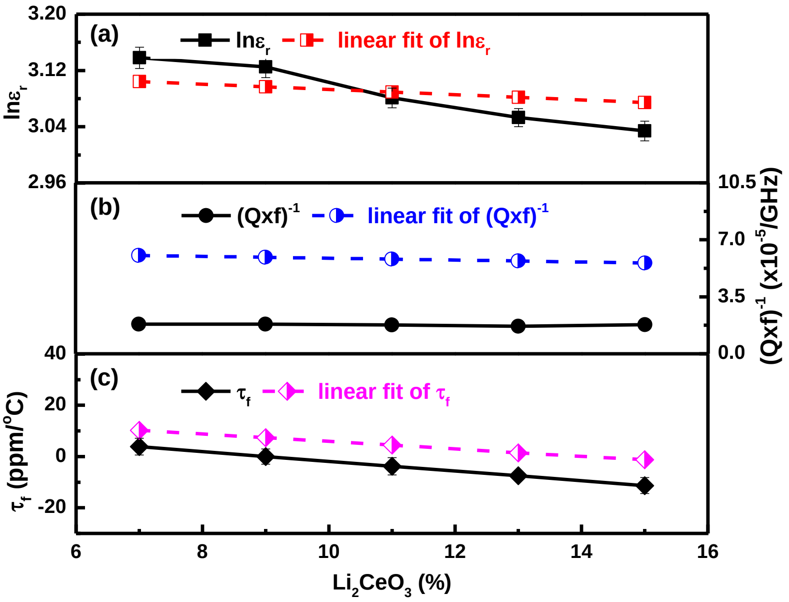

Fig. 4. (a) The lnε_r, (b) $(Qxf)^{-1}$ and (c) τ_f values of (1-x)Li_2TiO_3-xLi_2CeO_3 ceramics as a function of the volume fraction of Li_2CeO_3.

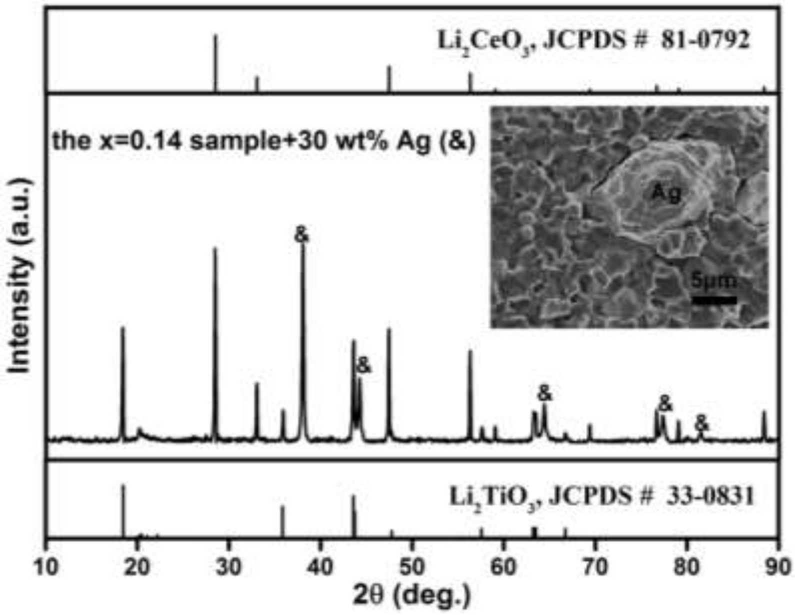

Fig. 5. XRD pattern and SEM image of the x=0.14 ceramic sample co-fired with 30 wt% Ag at 850 ºC for 4 h.

Table 1. EDS results for the *x*=0.14 sample sintered at 850 ºC for 4 h.

	Elements (atom%)			Total (%)
Points	Ti	Ce	O	
a	18.2	0.4	81.4	100.0
b	12.8	28.0	59.2	100.0

Table 2. Comparisons of microwave dielectric ceramics with similar permittivities and low sintering temperatures (≤850°C).

composition	S.T.(°C)	ε_r	Qxf (GHz)	τ_f(ppm/ºC)	Ref.
Li_2TiO_3-5wt% Li_2O-MgO-B_2O_3	850	21.4	64100	+26.9	16
Li_2TiO_3-NiO-1wt% BCB	850	19	62252	-1.65	18
$(Li_{0.5}Ln_{0.5})MoO_4$ (Ln=Nd, Gd, Sm, Ce)	580-750	19.5-20.6	1990-4570	+209-+235	28
$Na_2BiZn_2V_3O_{12}$	600	22.3	19960	+15.5	29
$PbWO_4$	850	21.6	34500	-22.2	30
Pb_2MoO_5	610	19.1	21960	-60	31
0.86Li_2TiO_3-0.14Li_2CeO_3	850	21.18	59309	-7.4	This work