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A novel temperature-stable and low-loss microwave dielectric composite ceramics $Li_2Mg_3SnO_6$-$SrTiO_3$

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ABSTRACT

Phase structure, microstructure and microwave dielectric properties of temperature-stable $(1-x)$ $Li_2Mg_3SnO_6$-$xSrTiO_3$ (x = 0.08, 0.12, 0.16, 0.20, 0.24, 0.28) composite ceramics were investigated. The results showed that ceramics contain $Li_2Mg_3SnO_6$ and $SrTiO_3$ phases and well-densified microstructure were obtained at 1375 °C. The microwave dielectric properties strongly depended on $SrTiO_3$ content x. With increasing x, the dielectric constant (ε_r) increased from 11.0 to 17.0, the quality factor ($Q \times f$) decreased from 105,100 GHz to 30,770 GHz, and the temperature coefficient of resonant frequency (τ_f) significantly increased from −35.4 ppm/°C to 37.3 ppm/°C. 0.8$Li_2Mg_3SnO_6$-0.2$SrTiO_3$ composite ceramics displayed a higher relative density of 97.2% and well microwave dielectric properties of ε_r = 14.2, $Q \times f$ = 67,260 GHz, τ_f = −4.8 ppm/°C.

1. Introduction

With developing wireless communication, microwave dielectric ceramics with a low dielectric constant (ε_r), ultrahigh quality factor ($Q \times f$) and near-zero temperature coefficient of resonant frequency ($|\tau_f|$ < 10 ppm/°C) are strongly required [1–3]. Recently, Li-containing compounds such as Li_2CeO_3, Li_2WO_4, Li_2MO_3(M = Ti, Sn, Zr), $Li_2ATi_3O_8$(A = Mg, Zn), Li_3AO_4(A = Nb, Ta, Sb), $Li_2Mg_3BO_6$ (B = Ti, Sn, Zr) have gained many investigated due to their excellent microwave dielectric properties and lower sintering temperatures [4–9]. Among them, $Li_2Mg_3SnO_6$ ceramics, with face centered cubic rock salt structure [10], have been attracted increasing interest because of its out-bound microwave dielectric properties of ε_r = 8.8, $Q \times f$ = 123,000 GHz, τ_f = −32 ppm/°C [9]. However, its larger τ_f value and porous microstructures hindered their practical applications. Some research has made to inhibition Li-volatilization in order to obtain well-densified microstructure by using low melting fluorides, Li-rich or burying the samples in Li-containing sacrificial powder [11–14]. Further studies of $Li_2Mg_3SnO_6$ ceramics about its larger τ_f value were not found. $SrTiO_3$ has been reported as an effectively$_f$ compensator in several diphasic systems due to its large positive $_f$ value (∼+1650 ppm/°C) and good microwave dielectric properties of ε_r = 290, $Q \times f$ = 4800 GHz [15–17]. So, one can be expected that a diphasic composite material with a near-zero $_f$ value and high Q may be obtained by combining $SrTiO_3$ with $Li_2Mg_3SnO_6$.

In this paper, $SrTiO_3$ was employed as an $_f$ compensator for $Li_2Mg_3SnO_6$ ceramics. The effects of $SrTiO_3$ on the crystal compositions, microstructures and microwave dielectric properties of $(1-x)Li_2Mg_3TiO_6$-$xSrTiO_3$ composite ceramics were investigated systematically.

2. Experimental procedure

$Li_2Mg_3SnO_6$ and $SrTiO_3$ powders were prepared by a conventional solid-state reaction route using Li_2CO_3 (98.0%), MgO (99.99%), SnO_2 (99.5%), TiO_2 (99.99%) and $SrCO_3$ (99.9%) powders as starting materials. Stoichiometric Li_2CO_3-MgO-SnO_2 and $SrCO_3$-TiO_2 were mixed according to the formula of $Li_2Mg_3SnO_6$ and $SrTiO_3$, respectively. In order to synthesize $Li_2Mg_3SnO_6$ and $SrTiO_3$ powders, the mixtures were ball-milled in ethanol for 8 h using zirconia balls, and then calcined at 1000 °C for 4 h in an alumina crucible in air. Subsequently, $(1-x)Li_2Mg_3SnO_6$-$xSrTiO_3$ (x = 0.08, 0.12, 0.16, 0.20, 0.24, 0.28) mixtures were prepared by mixing $Li_2Mg_3SnO_6$ and $SrTiO_3$ powders at different weight ratios. The mixtures were re-milled for 8 h using ZrO_2 balls in an alcohol medium. The milled powders were dried, mixed with 5 wt% 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 1350–1400 °C on alumina plates in

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air for 4 h, all the pellets were pretreated at 550 °C in air for 2 h to expel the binder. In order to prevent the evaporation of Li in the sintering process, all the pellets were covered with the sacrificial powders, which were composed by $Li_2Mg_3SnO_6$-$SrTiO_3$ mixing powders.

The crystal structures were analyzed using X-ray diffraction (XRD) with CuK_α radiation (Rigaku D/MAX2550, Tokyo, Japan). The microstructures were investigated using a scanning electron microscope (SEM, Quantax200, FEI Company, Eindoven, Holland) coupled with energy dispersive X-ray spectroscopy (EDS). The bulk densities of the sintered samples were measured by Archimedes method. Microwave dielectric properties of the specimens were measured using a network analyzer (ZVB20, Rohde & Schwarz, Munich, Germany) with the TE_{01} shielded cavity method. The temperature coefficient resonant frequency (τ_f) was calculated with the following formula:

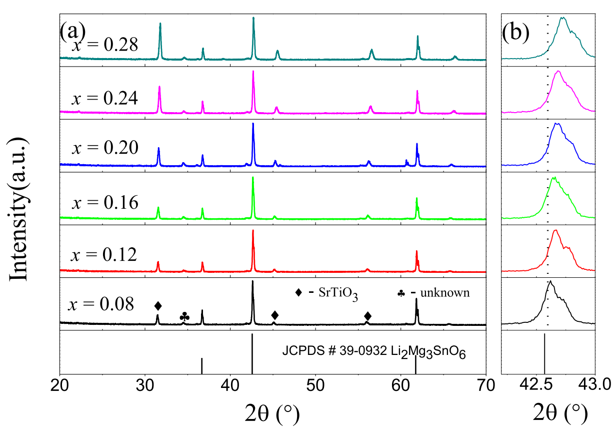

Fig. 1. XRD patterns of $(1-x)Li_2Mg_3SnO_6$-$xSrTiO_3$ ceramics sintered at 1375 °C as a function of x.

$$\tau_f = \frac{(f_2 - f_1) \times 10^6}{f_1(T_2T_1)} \tag{1}$$

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

3. Results and discussion

Fig. 1 shows XRD patterns of $(1-x)Li_2Mg_3SnO_6$-$xSrTiO_3$ ceramics sintered at 1375 °C as a function of x. As shown in Fig. 1(a), a two-phase system with a rock-salt structure $Li_2Mg_3SnO_6$ phase

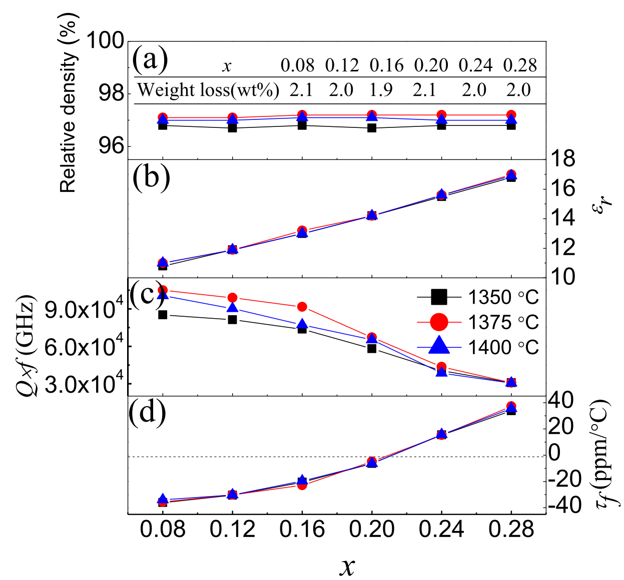

Fig. 3. The relative density, weight loss and microwave dielectric properties in $(1-x)Li_2Mg_3SnO_6$-$xSrTiO_3$ ceramics as a function of x.

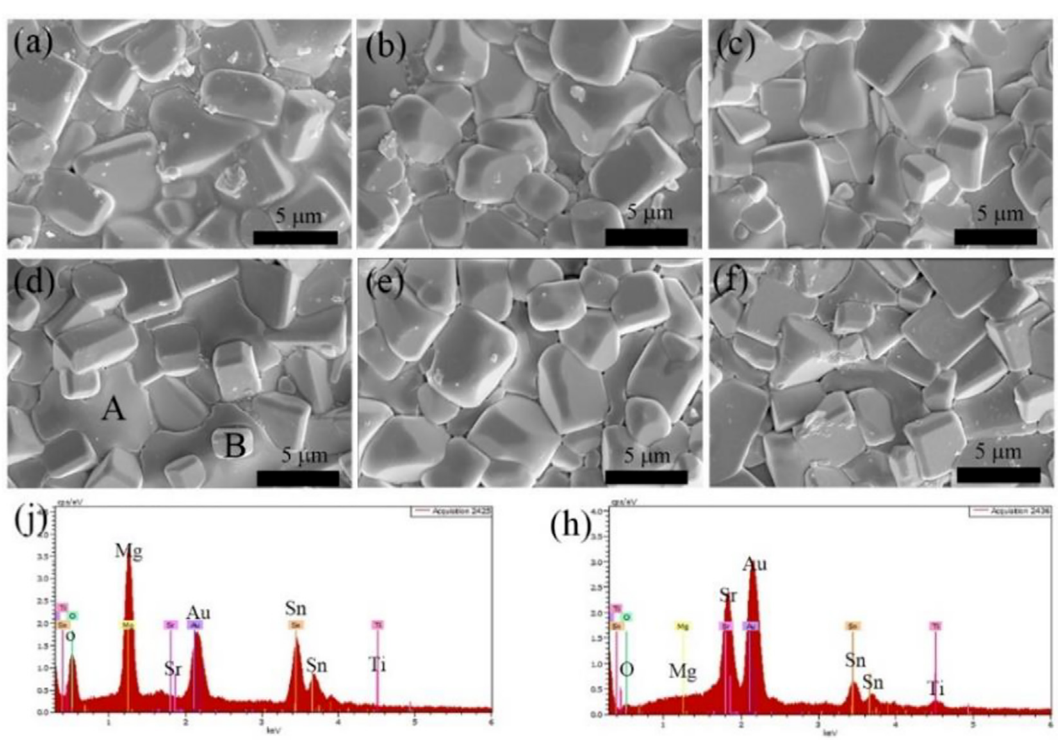

Fig. 2. SEM images of $(1-x)Li_2Mg_3SnO_6$-$xSrTiO_3$ ceramics sintered at 1375 °C: (a) $x = 0.08$, (b) $x = 0.12$, (c) $x = 0.16$, (d) $x = 0.20$, (e) $x = 0.24$, (f) $x = 0.28$.