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The effect of composition on $Li_2(Mg_{0.3}Zn_{0.7})Ti_3O_8$–$x TiO_2$ microwave dielectric ceramics for low temperature co-fired ceramics technology application

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The effect of composition of $Li_2(Mg_{0.3}Zn_{0.7})Ti_3O_8$–$x TiO_2$ ($-0.36 \le x \le 0.36$, for short LMZT) on sinterability, crystalline phase, microstructure and microwave dielectric properties of the ZnO–B_2O_3–SiO_2 (for short ZBS) glass doped samples was systematically investigated. It shows that as x is over zero, rutile phase was detected and the crystalline grain size of LMZT ceramics decrease abruptly. It was also found that the bulk density and the quality factor ($Q \times f$) gradually decreased, dielectric constant (ε_r) increased, the temperature coefficient of resonant frequency (τ_f) shifted to positive value with the increase of x value. When $x=0.12$, LMZT ceramics with 3 wt% ZBS sintered at 900 °C for 3 h show excellent dielectric properties: $\varepsilon_r=25.1$, $Q \times f=19{,}620$ GHz, $\tau_f=-2.3$ ppm/°C. It is compatible with Ag electrodes, which makes it a promising ceramic for low temperature co-fired ceramics technology application.

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1. Introduction

In recent years, much attention has been paid to the development of low temperature co-fired ceramics (LTCC) for its benefits offered to the fabrication of miniature multilayer devices. It is required to lower the sintering temperatures of the dielectric ceramics so that they are capable of being co-fired with low-loss conductors, such as Ag (melting point 960 °C) [1–4]. Low firing microwave dielectrics are also required to have high dielectric constant (ε_r), high quality factor values ($Q \times f$), and temperature coefficients of resonant frequency (τ_f) approaching to zero [5,6].

Recently, microwave dielectric properties of cubic spinel $Li_2(Mg_xZn_{1-x})Ti_3O_8$ ($x=0$–1) were reported by George et al. [7,8] and Zhou et al. [9]. The ceramics exhibit good dielectric properties of ε_r about 25.6–27.5, $Q \times f$ up to 72,000 GHz and τ_f in the range of $-15 \sim 3.2$ ppm/°C. Furthermore these materials are very cheap and can be sintered at relatively low temperature (1150 °C). Several researches focused on decreasing the sintering temperature of $Li_2ATi_3O_8$ (A=Mg, Zn) ceramics with different sintering aids. George et al. [10] reported that 3 wt% LMZBS (lithium magnesium zinc borosilicate) glass can lower the sintering temperature of $Li_2ZnTi_3O_8$ to 900 °C and the composition shows dielectric properties with $\varepsilon_r=23.2$, $Q \times f=31{,}300$ GHz and $\tau_f=-15.6$ ppm/°C. Li et al. [11] and Zhang et al. [12] investigated the effect of ZnO–B_2O_3 (ZB) frit and H_3BO_3 on sinterability and microwave dielectric properties of $Li_2ZnTi_3O_8$ respectively. 0.25 wt% ZB or 1% H_3BO_3 can decrease the sintering temperature of $Li_2ZnTi_3O_8$ to 950 °C and 880 °C respectively without obvious degradation of microwave dielectric properties. All their studies show that it is easy to lower the sintering temperature of $Li_2ZnTi_3O_8$ dielectrics. However, the grain size of $Li_2(Mg_xZn_{1-x})$-Ti_3O_8 dielectrics is normally big as 30–50 μm in spite of different sintering aids were added or not [7–11]. It is well known that insulation resistance of dielectrics at a high electric field strongly depends on the microstructures such as porosity and grain size [13]. It may have problem to fabricate miniature multilayer microwave components using such dielectrics with so big crystalline grain, because it has the proper short circuit risk in the case of dielectric layer thinner than 100 μm is used.

In the present work, the composition of $Li_2(Mg_{0.3}Zn_{0.7})Ti_3O_8$–$x TiO_2$ ($-0.36 \le x \le 0.36$, for short LMZT) was studied and it was found that the composition adjustment can decrease the grain size of dielectric ceramics. 3 wt% ZnO–B_2O_3–SiO_2 (for short ZBS) glass was used as sintering aid to decrease the sintering temperature of LMZT ceramics to 900 °C for LTCC use. The effect of x value on sinterability, crystalline phase, microstructure and microwave dielectric properties of the ZBS glass doped LMZT samples were systematically investigated.

2. Experimental procedure

The LMZT compositions were synthesized by the conventional solid-state reaction method using high-purity Li_2CO_3 (98%), MgO

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(98%), ZnO (99%) and TiO_2 (99.8%). The starting materials were mixed for 24 h in a ball mill with zirconia balls according to the desired stoichiometry and ground in ethyl alcohol to prevent dissolution of Li_2CO_3 in water. The mixtures were dried and calcined in an alumina crucible at 900 °C for 3 h in air. The calcined powder was remilled with 3 wt% home-made ZnO–B_2O_3–SiO_2 glass for 24 h. After drying, polyvinyl alcohol (PVA) was added to the mixed powder as a binder. Pellets of 20 mm in diameter and 10–11 mm in thickness were prepared under a pressure of 80 MPa by uniaxial pressing. These pellets were sintered in air at 880–960 °C for 3 h at a heating rate of 2.5 °C/min.

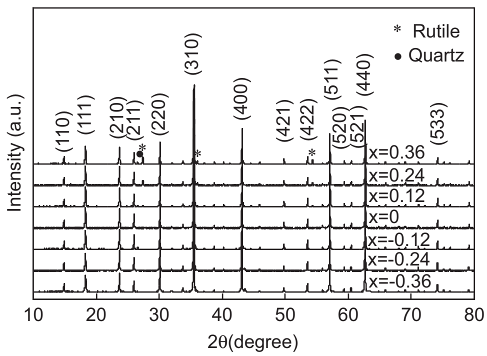

Fig. 1. XRD patterns of 3 wt% ZBS added $Li_2(Mg_{0.3}Zn_{0.7})Ti_3O_8$–$x$$TiO_2$ ($x=-0.36$ to 0.36) ceramics sintered at 920 °C.

The bulk density was measured by the Archimedes method. The crystalline structures of the samples were examined by X-ray diffraction analysis (XRD, Rigaku D/max-RA) with CuKa radiation. The surface microstructures of as-fired samples were observed by scanning electron microscope (SEM, FEI SIRION-100). Microwave dielectric properties were measured by the Hakki–Coleman dielectric resonator method [14], using a network analyzer (Agilent 8719 Et). The resonant frequency was measured in the temperature range of 25–80 °C, and τ_f was calculated from the following equation:

$$\tau_f = \frac{f_{80}-f_{25}}{f_{25}\times 55}\times 10^6 (\mathrm{ppm}/^\circ\mathrm{C})$$

where f_T is the resonant frequency of the dielectric resonator at temperature T (°C).

3. Results and discussion

Fig. 1 shows XRD patterns of 3 wt% ZBS added LMZT ceramics sintered at 920 °C. The main diffraction peaks of all the samples can be indexed according to $Li_2CoTi_3O_8$-type cubic spinel structure (matched with JCPDF #49-0449). When x is over zero, rutile phase was detected and it enhanced in intensity with the increasing of x value. The weak diffraction peak at around 26.6° could be indexed according to quartz phase (JCPDF #46-1045). It indicated the crystallization of SiO_2 from ZBS glass was occurred in the sample of $x=-0.12$, 0.12 and 0.36.

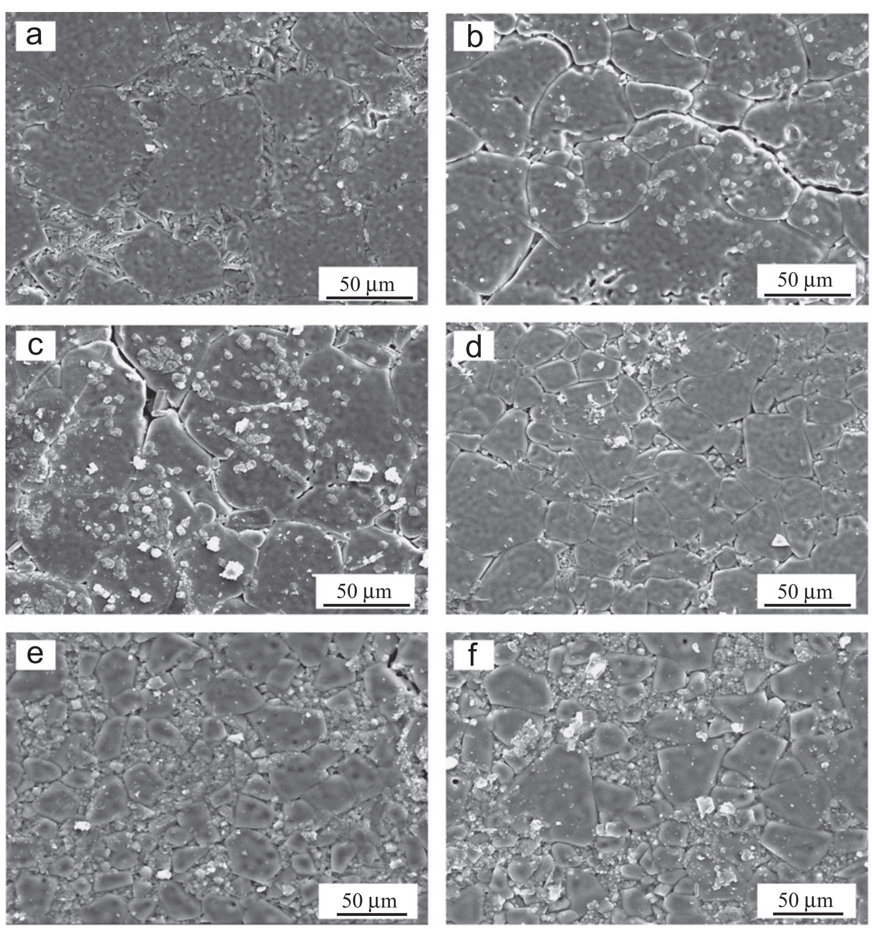

Fig. 2. Scanning electron microscopy of the surfaces of the 3 wt% ZBS added $Li_2(Mg_{0.3}Zn_{0.7})Ti_3O_8$–$x$$TiO_2$ with (a) $x=-0.36$, (b)$x=-0.24$, (c) $x=-0.12$, (d) $x=0$, (e) $x=0.12$ and (f) $x=0.24$ ceramics sintered at 920 °C.

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