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Short communication

Preparation of LTCC materials with adjustable permittivity based on $BaO–B_2O_3–SiO_2/BaTiO_3$ system

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ABSTRACT

This paper studied the preparation and characterization of LTCC (low temperature co-fired ceramics) materials based on $BaO–B_2O_3–SiO_2/BaTiO_3$ glass–ceramics, where the sintering temperature was about 900 °C and dielectric constant was effectively adjustable from 5 to 30 by changing the $BaTiO_3$ fraction from 60 wt% to 90 wt%. X-ray diffractometer (XRD), scanning electron microscopy (SEM) were used to examine the effect of different amounts additive on the dielectric properties of this LTCC system and the crystal structure change. The results indicated that $BaTiO_3$ can be used as a dielectric additive aim to adjust the permittivity of $BaO–B_2O_3–SiO_2$ glass, which the main influence factors on dielectric are the contents of the secondary phase, the $BaTiO_3$ phase fraction and the porous structure of the sintered body. Therefore, the microstructure and dielectric property of $BaO–B_2O_3–SiO_2/BaTiO_3$ glass–ceramics composites could be controlled by adjusting the content of $BaTiO_3$ additive.

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1. Introduction

Low temperature co-fired ceramic (LTCC) technology has been playing an important role in modern wireless communication systems, which sintering temperature of LTCC should be below about 900 °C [1]. As is generally known, glass ceramics and glass/ceramic composites are applied widely to LTCC multi-layer structures due to their low sintering temperatures and tailored dielectric properties [1–5]. However, the manufactures of most of glass materials is usually a complex process including melting, cooling and crystallization [6]. As LTCC applications, a single phase ceramic materials are more favorable to achieve the high reliability and a good thermal stability. Therefore, glass plus ceramic is a favorable method to tailor dielectric properties for the fabrication of low-temperature co-fired ceramics [7]. Many new ceramic/glass compositions have recently been developed for microwave purposes, especially low permittivity materials using alumna and suitable glass combinations, but also higher permittivity (ε = 20–100) materials [8].

Barium titanate ($BaTiO_3$) is a well-know ferroelectric materials and widely used as a dielectric for multilayer ceramic capacitors (MLCCS), electro-optic devices and thermistor because of its highly dielectric characteristics. Research on the development of co-fired dielectrics with different dielectric constants has been going for years to fill the need for RF range. 30 wt%BaO–60 wt%B_2O_3–10 wt% SiO_2 glass has been demonstrated as an effective sintering aid to reduce the sintering temperature of $BaTiO_3$ from 1350 °C to 900 °C and still showing good dielectric properties by Jeon et al. [9]. Hsiang et al. studied the effect of different modifiers (BaO, PbO and ZnO) in borosilicate glass on the sintering behavior and the chemical reaction between glasses and $BaTiO_3$. The dissolution of BaO and TiO_2 into $21.43BaO–67.06B_2O_3–11.51SiO_2$ glass led to a noticeable $BaTi(BO_3)_2$ and $Ba_2TiSi_2O_8$ phases formation occurs while the temperature increases [10]. It was found that rutile additive could not be wet the $32BaO–15B_2O_3–53SiO_2$ glass matrix under high-temperature conditions but could accelerate the phase separation and crystallization of the glass phase during the sintering process by Cui et al. [11,12].

The above-mentioned study have focused on obtaining high-density dielectrics by low temperature sintering in the presence of an optimal amount of liquid phase without any undesirable phase formation, and high dielectric ceramics, the most important step is to control the amount of glass additive for obtaining a high densification and to reduce the volume fraction of low dielectric secondary phases and porosity.

In this work, we aimed at developing some adjustable middle-permittivity dielectric ($\varepsilon \approx$ 10–30) material systems sintered at 900 °C for LTCC by mixing $BaTiO_3$ and $BaO–B_2O_3–SiO_2$ (BBS) glass which was carefully designed [13]. In the work reported here, the $BaTiO_3$ is selected as a basic dielectric material because of its excellent dielectric properties. In addition, the glass–ceramics system often introduces certain secondary phases and pores [14].

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Although a porous microstructure has great influence on mechanical property of ceramics materials, it may be beneficial for low temperature co-fired ceramics (LTCC) when it is used in low and middle-permittivity devices [15].

2. Experimental

The $BaO–B_2O_3–SiO_2$ (BBS) glass matrix was prefabricated via the pre-molten method using a mixture of $Ba(OH)_2$ (AR, Guangdong Xilong Chemical Company, China), H_3BO_3 (AR, Guangdong Xilong Chemical Company, China) and SiO_2 (AR, Guangdong Xilong Chemical Company, China) powders with a mole ratio of $BaO/B_2O_3/SiO_2$ = 0.28:0.18:0.54. The powders were ball-milled in ethanol for 12 h, followed by molten processing at 1250 °C for 2 h in air and quenching in water. The obtained glass was subsequently ball-milled in water for 24 h. First, milled glass powders were uniformly mixed with 0, 10, 20, 30, 40, 50, 60, 70, 80 and 90 wt% $BaTiO_3$ (AR, Guangdong Xilong Chemical Company, China) powders and pressed into pellets, respectively. And then measure their shrinkage and dielectric constant after sintering the pellets at 780–1040 °C for 0.5 h in air. Finally, the glass/ceramic samples were fabricated by sintering the pellets which mixed with a new suitable ratio of $BaTiO_3$ to the glass powders at 900 °C for 0.5 h in air.

The average particle size of the glass powders and $BaTiO_3$ powders was measured with a laser granularity analyzer (Winner 2000) is around 4.5 μm and 0.2 μm, respectively. The sinterability of the glass powders should be much poor than fine powders. But in this study, we promote the sinterability of the coarse glass powders by adding $BaTiO_3$ to the glass matrix. The density of samples was measured by the Archimedes method. The body microstructures of samples were examined by scanning electron microscope (SEM, LEO1530, Oxford Instruments). The relative dielectric constant ε and the dissipation factor ($\tan\delta$) of the sintered samples were measured by Agilent E4991A impedance analyzer (Agilent Technologies Company, USA) at the frequency of 100 MHz at room temperature.

3. Results and discussion

First of all, in accordance with the sintering temperature requirements of LTCC systems, in a relatively large range screened the composition of $BaO–B_2O_3–SiO_2/BaTiO_3$ glass–ceramics in the sintering temperature was below 900 °C. Fig. 1 shows the change of the lowest sintering temperature of the samples with the amount of $BaTiO_3$ additive when its line shrinkage reached about 10%. It was found that the lowest sintering temperature was below 920 °C in the composition range of 60–90 wt% or 0–30 wt%. Because our research aim mainly focused on mid and high dielectric constant (10–30) LTCC, therefore, the suitable $BaTiO_3$ additive fraction for obtaining mid and high dielectric constant LTCC materials should be located between 60 wt% and 90 wt%. Fig. 2 shows that the dielectric constant (ε) of the sintered $BaO–B_2O_3–SiO_2/BaTiO_3$ glass/ceramics (the sintered samples with line shrinkage of 10%) changed from 5 to 6 and the dielectric losses ($\tan\delta$) was lower than 0.05 at 100 MHz before the amount of $BaTiO_3$ additive exceeded 60 wt%. The effect of the $BaTiO_3$ additive became noticeable by a substantial increase of the dielectric constant (from 6 to 30 at 100 MHz) in the increment from 60 wt% to 90 wt% fractions. The results indicated that $BaTiO_3$ which possesses superior dielectric properties may act as additional dielectric aid to tailor dielectric constant for the fabrication of the middle-permittivity LTCC material and the sintering temperature for the fabrication can be controlled below 900 °C.

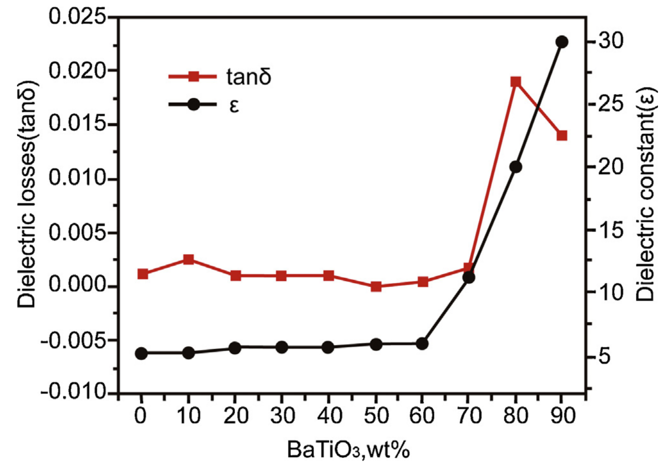

Fig. 2. Effects of the various amounts of added $BaTiO_3$ on the dielectric characterization of the samples with the shrinkage of 10%.

The bulk densities and porous of the samples with 66–88 wt% fraction of $BaTiO_3$ additives sintered at 900 °C for 0.5 h are shown in Fig. 3. It was observed that the porous increased gradually with the increase of the $BaTiO_3$ addition [11]. Their bulk densities were 4.21, 4.31, 4.25, 4.23, 4.12 and 4.04, respectively. Fig. 4 shows the

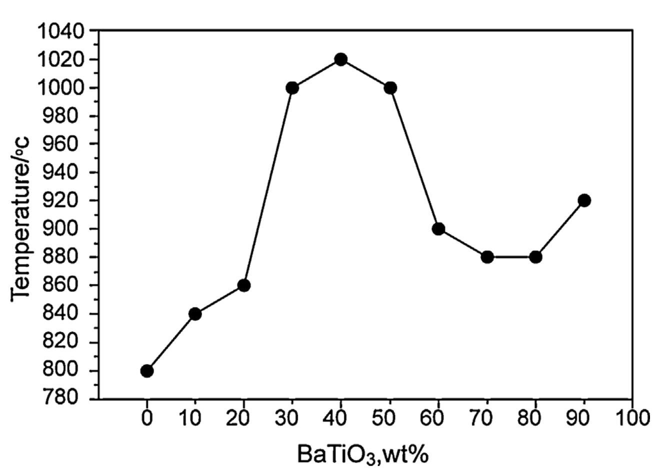

Fig. 1. The change of the lowest temperature of the samples when shrinkage reached about 10%.

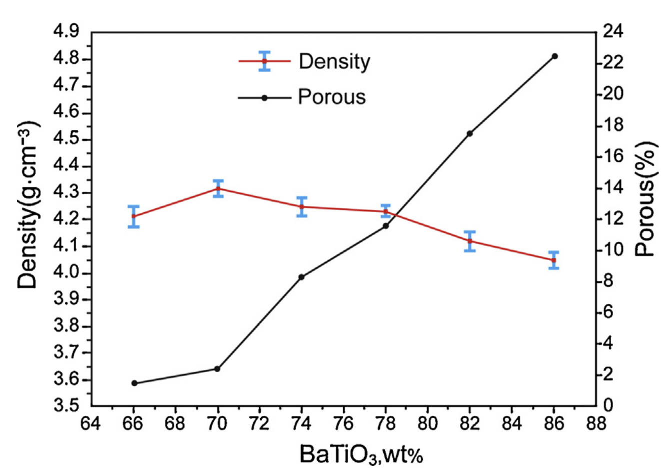

Fig. 3. Porous and density of the samples with various amounts of added $BaTiO_3$ sintered at 900 °C for 0.5 h.

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