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Highly textured $Ba_{0.85}Ca_{0.15}Ti_{0.90}Zr_{0.10}O_3$ ceramics prepared by reactive template grain growth process

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ABSTRACT

⟨001⟩ textured $Ba_{0.85}Ca_{0.15}Ti_{0.90}Zr_{0.10}O_3$ (BCZT) lead-free ceramics were fabricated by reactive-templated grain growth (RTGG) method using plate-like $BaTiO_3$ templates. Aligned plate-like $BaTiO_3$ particles grew at the expense of equiaxed matrix powder with random orientation with increasing sintering temperature, resulting in development of texture. Highly textured (texture fraction $f=0.98$) BCZT ceramics were prepared at first with a RTGG process. Development of texture was investigated during sintering. The influence of templates content on texture fraction and electrical properties of the textured BCZT ceramics were also studied. The textured ceramics shows high piezoelectric coefficient ($d_{33}=455$ pC/N) and dielectric properties ($\varepsilon_r \sim 3500$, $\tan\delta \sim 0.010$).

1. Introduction

Lead-free piezoelectric ceramics have been actively studied in recent years as alternatives to lead-based piezoelectric ceramics due to the toxicity of Pb [1]. Some methods have been used to improve piezoelectric properties of lead-free piezoelectric ceramics, such as compositional engineering [2,3] or microstructural engineering (e.g., texturing [4]). Lead-free $Ba(Zr_{0.2}Ti_{0.8})O_3$–$x(Ba_{0.7}Ca_{0.3})TiO_3$ (BZT–BCT) system have attracted great attention and been investigated widely by adding dopants (e.g., CuO, ZnO, Y_2O_3 etc.) [3,5,6]. However, few studies of the texturing effects on the electrical properties of BZT–BCT ceramics have been carried out to date.

Anisotropy of the piezoelectric response is a common phenomenon in the field of perovskite piezoceramics [7,8]. Giant piezoelectric coefficients d_{33} (exceeds 2500 pC/N) were obtained in $Pb(Zn_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ and $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ single crystals along (001) direction [9]. However, the process of single crystals growth is complex and time-consuming [10]. Textured ceramics, which have a simpler prepare process compared with single crystals, take advantage of anisotropic nature of the piezoelectric response of single crystals.

The templated grain growth (TGG) technique and reactive-templated grain growth (RTGG) method are commonly used to obtain textured ceramics [4,11]. In the TGG process, anisometric templates are aligned in matrix powder and act as nucleation sites for the preferential matrix grain growth during sintering. However, it is hard to synthesize anisometric templates which possess the same composition with matrix powder (e.g. $Ba(Zr_{0.2}Ti_{0.8})O_3$–$x(Ba_{0.7}Ca_{0.3})TiO_3$). Therefore, heterotemplates are often used in TGG method. Heterotemplates materials can cause compositional deviation or inhomogeneity, which is undesirable make the degradation of ceramic performance [12,13]. In the RTGG process, anisometric templates are aligned in matrix powder and react in-situ with matrix powder to form the composition of piezoelectric ceramics during sintering. Thus, RTGG method avoid the shortcomings of TGG method.

In this work, a simple RTGG mothed ($BaTiO_3$, $BaZrO_3$, $CaTiO_3$ powder were used as raw material directly rather than synthesized BCZT powder) was used to prepare texture BCZT. Highly textured $Ba_{0.85}Ca_{0.15}Zr_{0.1}Ti_{0.9}O_3$ (BCZT) ceramics were prepared at first with a RTGG process. The effect of sintering condition on the grain orientation of the samples was investigated. In addition, the influence of templates content on texture fraction and electric properties of the textured BCZT ceramics were studied systematically.

2. Experimental procedure

Ceramics with ⟨001⟩ texture were prepared using tape-casting with $BaTiO_3$ (BT) templates. Plate-like $Bi_4Ti_3O_{12}$ particles were used as precursor to synthesize plate-like $BaTiO_3$ templates. The detailed experimental procedures of plate-like $Bi_4Ti_3O_{12}$ precursor

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and $BaTiO_3$ particles are described in Refs. [14,15], respectively. $BaTiO_3$ (99.0%), $BaZrO_3$ (99.0%), $CaTiO_3$ (99.0%) powder were used as raw materials. $Ba_{0.85}Ca_{0.15}Ti_{0.90}Zr_{0.10}O_3$ (BCZT) ceramics were obtained based on following reaction:

x($BaTiO_3$ templates)+(0.75−x)($BaTiO_3$ powder)+0.10$BaZrO_3$+0.15$CaTiO_3$→BCZT, where x range from 0.05 to 0.20. Tape-casting slurries were prepared in solvents (the azeotropic mixture of butanone and ethanol) and dispersant (polyvinyl butyral) by ball milling for 4 h, then added binder (polyvinyl butyral) and plasticizer (dibutyl phthalate and polyethylene glycol) for an additional 24 h. The BT templates were added and mixed for 2 h by ball milling. The mixed slurry cast at 70 cm/min using a 150 μm blade gap to form films. The films were laminated under a pressure of 80 MPa at 90 °C for 15 min to form green compacts with a thickness of 1.5 mm then cut into 10 mm disks. The binder and plasticizer were burned out at 600 °C for 1.5 h, then consolidated by isostatic pressing at 200 MPa. The samples with x=0.10 were sintered at 900–1300 °C for 5 h. The samples (0.05 ≤ x ≤ 0.20) were sintered at 1400 for 5 h.

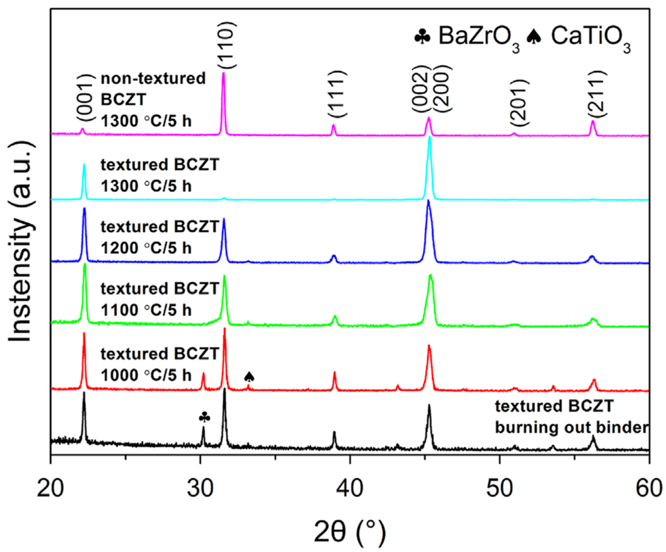

Fig. 1. XRD patterns of the BCZT ceramics sintered at different temperatures.

The phase structure were analyzed by X-ray diffractometer (XRD, Rigaku D/Max 2500, Tokyo, Japan). The texture fraction (f) was calculated from XRD pattern by Lotgering's method [16]. Scanning electron microscopy (SEM, S4800, Hitachi, Japan) was used to obtain the microstructure patterns of the ceramics. Silver electrodes were painted and fired at 600 °C for 15 min for dielectric and piezoelectric characterization. Sintered samples were poled in silicone oil at room temperature under 3.0 kV/mm for 20 min. Dielectric properties were measured by an LCR meter (Agilent E4980A, Santa Clara, CA). The piezoelectric constant d_{33} was measured by quasistatic d_{33} meter (ZJ-3AN, Institute of Acoustics Academic Sinica, Beijing, China).

3. Results and discussion

Fig. 1 shows the XRD patterns of the textured BCZT ceramics sintered at different temperatures, as well as the non-textured BCZT sintered at 1300 °C. The solid solution formation of Zr^{4+} was finished at 1100 °C, as judged from the disappearance of the

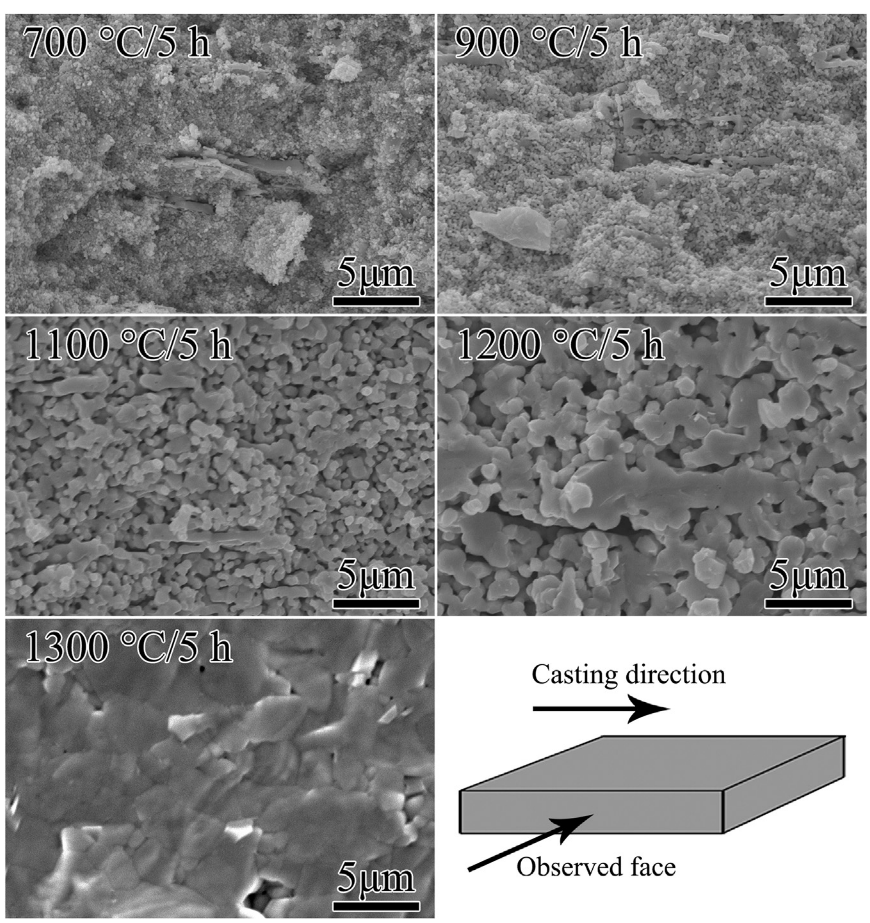

Fig. 2. SEM images of the textured ceramics (x=0.10) sintered at different temperatures.