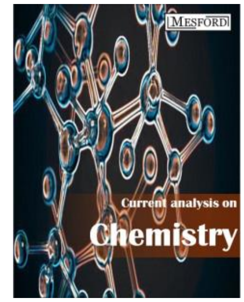

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Sol-gel Synthesis of $(Ba_{0.85}Ca_{0.15})(Zr_{0.1}Ti_{0.9})O_3$ Ceramics and their Dielectric Temperature Properties

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Abstract:

$(Ba_{0.85}Ca_{0.15})(Zr_{0.1}Ti_{0.9})O_3$ piezoelectric ceramics were prepared by a simple sol-gel method. The surface morphology and structure were characterized by Scanning Electron Microscopy (SEM) and X-ray diffraction (XRD). Furthermore, based on the analysis of dielectric temperature spectrum with different frequency, $(Ba_{0.85}Ca_{0.15})(Zr_{0.1}Ti_{0.9})O_3$ ceramics have been found to exhibit promising dielectric temperature performance, which can be attributed to their unique structure. This research may provide new insights into engineering application.

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Keywords:
BCZT, dielectric temperature properties, sol-gel, SEM, XRD.

1. INTRODUCTION

Over the past several years, a series of piezoelectric materials, BCZT lead-free piezoelectric ceramics have extensively studied, which is an important material in different areas, such as sensors, ultrasonic motors, buzzers and so on [1-4]. They have drawn significant attentions due to superior piezoelectric performance [5, 6].

BCZT powders are usually synthesized by solid-state reaction [7-9] and then densified to prepare BCZT ceramics. Although this method is relatively simple, it has some problems such as high calcination temperature and insufficient reaction or introduction of impurities [10, 11]. Sol-gel method is a low-cost, environmental friendly method to prepare materials in different areas [12]. Moreover, we set to seek more effective dielectric temperature properties. Both dielectric temperature constant and dielectric loss of BCZT ceramics are two crucial factors [13, 14].

In this work, we report the two important dielectric temperature properties of BCZT ceramics, in which BCZT ceramics were synthesized by sol-gel method. The morphology and structure of the samples is investigated by Scanning Electron Microscopy (SEM) and X-ray Diffraction (XRD). Furthermore, the dielectric temperature performance of BCZT ceramics, which shows a superb dielectric temperature constant, is also investigated.

2. EXPERIMENTAL

BCZT ceramics were prepared by the sol-gel method, in which stoichiometric ratios of $ZrOCl_2 \cdot 8H_2O$, $Ca(NO_3)_2 \cdot 4H_2O$, $Ba(CH_3OO)_2$, $C_{12}H_{28}O_4Ti$ were mixed in anhydrous ethanol and acetic acid, to obtain the gel of BZT-BCT system. Precursor were mixed intensively. With thermal insulation for a period of time, the sol becomes milky white. Calcination temperatures were 650°C for 4 h. Then the calcined powders were pressed into pellets (using polyvinyl butyral as a binder) and sintered for 2 h in air with 1420℃ sintering temperatures.

The as-prepared products were characterized by X-ray diffraction (XRD, PANalytical, Cu Kα λ=1.5406 Å) and Scanning electron microscopy (SEM, TFSEM-6330).

The dielectric temperature properties were measured by impedance analyzer under different frequency spectrum (heating rate were both 10 °C/min, temperature range were from 20°C to 180°C).

3. RESULTS AND DISCUSSION:

Fig. (**1**) shows XRD patterns of the BCZT ceramics with different molar ratio of BCT-BZT. As seen in Fig. (**1**), when molar ratio ranged from 46:54 to 54:46, most raw materials have been transformed into a single perovskite crystal structure (JCPDS No.74-1968). When the molar ratio of BCT-BZT was

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less than 50:50, a little CaZrO3 appeared. Meanwhile, a few BaZrO3 appeared when the molar ratio of BCT-BZT was greater than 50:50. When the molar ratio of BCT-BZT was 50:50, a completely single crystal of perovskite structure was obtained. Therefore, we use the molar ratio of BCT-BZT (50:50) as following experiment.

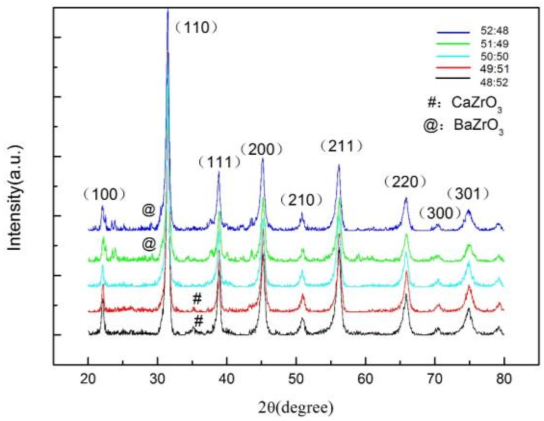

Fig. (1). XRD patterns of different molar ratio of BCZT ceramics

Fig. 2 shows SEM images of BCZT ceramics. As seen in Fig. (**2**), the particles are stacked tightly between each other, and the holes between the particles basically disappear. Moreover, the surface of the ceramics is smooth. The average size of ceramic particles is approximately 8 μm. However, larger grain size can result in a decrease for the matching of ceramic grains. The increased defects of ceramics can decrease the quality of ceramics.

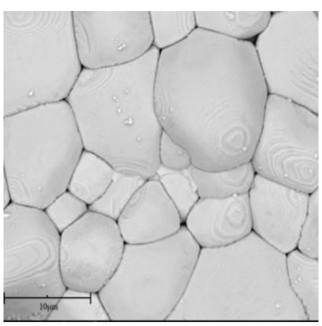

Fig. (2). The SEM images of BCZT ceramics.

When the materials were situated in the Curie temperature point, the electric dipole moment is very large. The phase transition is completed as exceeded the Curie temperature. Therefore, the change of the dielectric constant is satisfied with Curie's law according to the following the equation:

$$\varepsilon_r = \frac{C}{T - T_0}$$

where ε_r is relative dielectric constant, C is Curie constant (K), T_0 is the paraelectric Curie temperature (K) and T is the absolute temperature (K).

Fig. (**3**) shows BCZT ceramic dielectric temperature spectra of 10 KHz alternating electric field. As the temperature increasing, dielectric constant of BCZT ceramic rises rapidly at 40°C. BCZT ceramics has a process of transformation from orthorhombic phase to tetragonal at this time. With increasing temperature, the dielectric constant of BCZT ceramics began to rise sharply, then a relatively steady rise. The dielectric constant of BCZT ceramics change dramatically at 100℃, which the point is the Curie point. When crystal is cooled from a high temperature to Curie point, it passes through a phase transition from a non-ferroelectric phase to a ferroelectric phase. When the temperature is lower than Curie point, the crystal exhibits ferroelectric. In many cases, the dramatic change of material properties is not allowed, while it is often necessary to avoid Curie point in engineering application. Therefore, Curie point plays an important role in practical application.

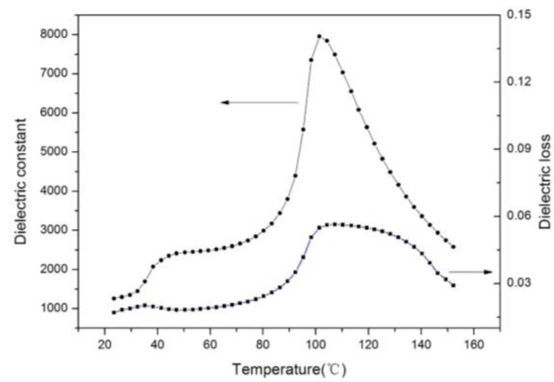

Fig. (3). BCZT ceramic dielectric temperature spectra of 10KHz alternating electric field

The dielectric temperature spectrum of BCZT ceramics under different electric field frequencies are shown in Fig. (**4**). We can see the variation trend of the dielectric constant of ceramics is consistent under different electric field frequencies. With the increase of electric field frequency, the dielectric constant decreases, and the temperature peak is wider, which shows the characteristics of the relaxation ferroelectric. The phase transition between the dispersion phase and the paraelectric ferroelectric phase is a gradual process, without a certain Curie temperature. When the crystal is in a high temperature environment, the crystal situates in the direct current phase. According to the diffusion phase transition theory, in the crystal structure of perovskite, the distribution of the B-bit ion causes the component fluctuation, so that the single Curie temperature point becomes the temperature range.

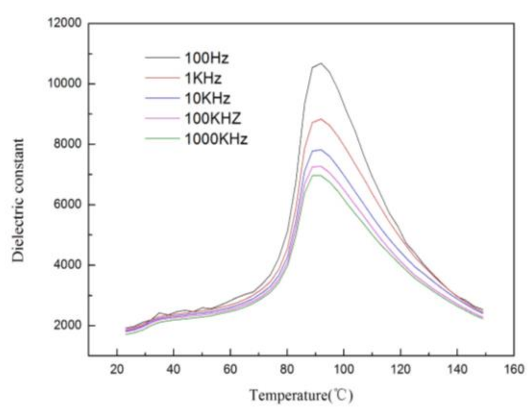

Fig. (4). The dielectric temperature spectrum of BCZT ceramics under different electric field frequencies.

CONCLUSIONS

In summary, we describe a simple method to synthesize BCZT ceramics. The surface morphology and phase structure are presented. The perovskite crystal structure mainly consists of rhombohedral phase and tetragonal phase. The dielectric temperature constant of BCZT ceramics can achieve at about 11000 in certain electric field frequency. Results of this work suggest that the synthesized BCZT ceramics is favorable in the development of engineering application.

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CONFLICT OF INTEREST

The authors declare no conflicts of interest.

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