

EFFECT OF BALL-MILLING ROTATION SPEED ON ELECTRICAL PROPERTIES OF BCZT CERAMIC

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Abstract

Multi-layer ceramic capacitors (MLCCs) are typically found in electronic applications. They are utilized when a device needs higher capacitance and additional energy storage capabilities. MLCCs are installed in the input or output filters of power supplies. They can also be integrated in capacitor banks. The leaded configurations can be customized according to their application. Lead-free $0.5\text{Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3-0.5(\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3$ ceramic (BCZT) was successfully fabricated by conventional solid state sintering method. The effect of ball-milling rotation speed (0, 350, 400, 450 and 500 rpm) on the particle size and electrical properties such as dielectric and piezoelectric properties of BCZT ceramic were systematically investigated. The varies of ball- milling rotation speed changes the particle size and the electrical properties. The particle size was depended by increasing the rotation speed. Moreover, the BCZT ceramic with rotation speed of 400 rpm exhibited good piezoelectric and dielectric properties with the piezoelectric coefficient (d_{33}) of 370 pC/N and a dielectric constant at room temperature of 2,794.

Keywords: BCZT ceramic, dielectric, ball milling, particle size

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Introduction

In recent years, significant progress has been made in the field of ceramic materials, particularly in the development of functional ceramics for various applications. Barium Calcium Zirconate Titanate (BCZT) is a promising material that possesses extraordinary electrical properties, including a high dielectric constant, ferroelectric behavior, and piezoelectric properties (Mahajan *et al.*, 2008; Pisitpipathsin *et al.*, 2013; Pisitpipathsin and Kantha, 2018). BCZT ceramics are attractive for use in electronic devices, energy storage, and sensors due to these properties.

To optimize the efficacy of BCZT ceramics, numerous processing techniques, including the widely used ball milling method, have been investigated. The mechanical process of ball milling involves the grinding and mixing of powders, which reduces particulate size and creates a homogenous mixture. This technique offers a number of benefits, including improved material uniformity, enhanced reactivity, and increased surface area, all of which can considerably affect the properties of the resulting ceramics (Boonlakhorn and Thongbai, 2020; Zhou *et al.*, 2007).

Among the many variables that influence the ball-milling procedure, the rotational speed of the milling container is crucial. The rotational speed determines the kinetic energy transferred to the milling balls, which influences the effectiveness of particulate size reduction and the overall mixing procedure. Therefore, rotation speed can affect the electrical properties of BCZT ceramics, such as their dielectric constant, conductivity, and polarization behavior.

This investigation seeks to provide valuable insights for optimizing the fabrication process and augmenting the performance of BCZT-based electronic and energy devices. This study's findings can contribute to the advancement of functional ceramic materials, allowing their incorporation into a wide variety of technological applications.

Materials and Methods

The $0.5\text{Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3\text{-}0.5(\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3$ (BCZT) ceramics were successfully synthesized through the conventional ceramic fabrication technique. High-purity powders of BaCO_3 , CaCO_3 , ZrO_2 and TiO_2 , each with a purity exceeding 99.0%, served as the initial raw materials. The individual powders were accurately weighed according to the specified composition and subjected to thorough mixing through the ball-milling method in an ethanol medium, utilizing zirconia balls for a duration

of 24 h. Subsequently, the resulting mixtures underwent drying and calcination at $1,350^\circ\text{C}$ for 2 h. X-ray diffraction (XRD) analysis was conducted to investigate the phases present in the prepared powder. Then, the calcined powder is partitioned into five distinct groups, each subsequently subjected to grinding procedures at varying speeds specifically, at 350, 400, 450, and 500 rpm for 24 h. The original, non-reground powder is a reference in this comparative analysis denoted as 0 rpm. After the powder preparation, a particle size analyzer was employed to assess the dimensions of particulate matter and the microstructure of the powder was scrutinized utilizing scanning electron microscopy (SEM). Then the BCZT powders underwent uniaxial pressing to form pellet-shaped specimens with a 10 mm diameter. These pressed samples were subjected to conventional sintering at $1,450^\circ\text{C}$ in an air atmosphere for 2 h, under controlling a heating/cooling rate of $5^\circ\text{C}/\text{min}$, facilitating the production of dense BCZT ceramics.

To characterize the dielectric and piezoelectric properties, the two circular surfaces of all BCZT ceramics were precisely ground to achieve parallel faces. Subsequently, these surfaces were coated with silver paste to serve as electrodes for facilitating electrical contact. The ceramic specimens designated for piezoelectric measurements were poled at room temperature within a silicone oil bath, subject to a direct current (DC) electric field of $3.0\text{ kV}/\text{mm}$ for 30 min. Subsequently, a 24-hour aging period preceded the commencement of testing. The dielectric properties and the piezoelectric constant value (d_{33}) of BCZT ceramics were quantified using an LCZ meter and a d_{33} meter, respectively.

Results and Discussion

The XRD pattern of the BCZT powder is shown in Figure 1. It reveals no secondary phases, indicating the formation of a pure BCZT phase. This result is consistent with our previous work (Kantha *et al.*, 2017).

Figure 2 shows SEM image of BCZT powder that was calcined at 1,350 degrees Celsius and processed through a ball mill operating at various rotational speeds. The image reveals a distinct contrast between the untreated powder, which appears as lumps, and the ball-milled powder, which exhibits the characteristics of a finely processed particulate.

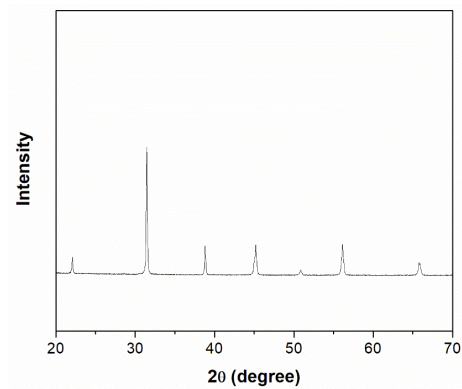


Figure 1. X-ray diffraction pattern of the BCZT powder

This heightened energy fosters more efficient size reduction and homogenization of the powder particles, culminating in a decrease in particle size. This reduction is particularly pronounced up to a specific rotational speed. Optimal grinding efficiency is achieved at a particular rotational speed, where the interplay of impact and attrition forces is optimized. This results in effective particle size reduction and homogenization. However, beyond this optimum speed, further increases in rotational speed may yield diminishing returns in terms of size reduction. Excessive speeds can induce particle agglomeration, diminishing the efficiency of the grinding process and potentially leading to an increase in particle size. During the higher rotational speeds ball milling process, the initial

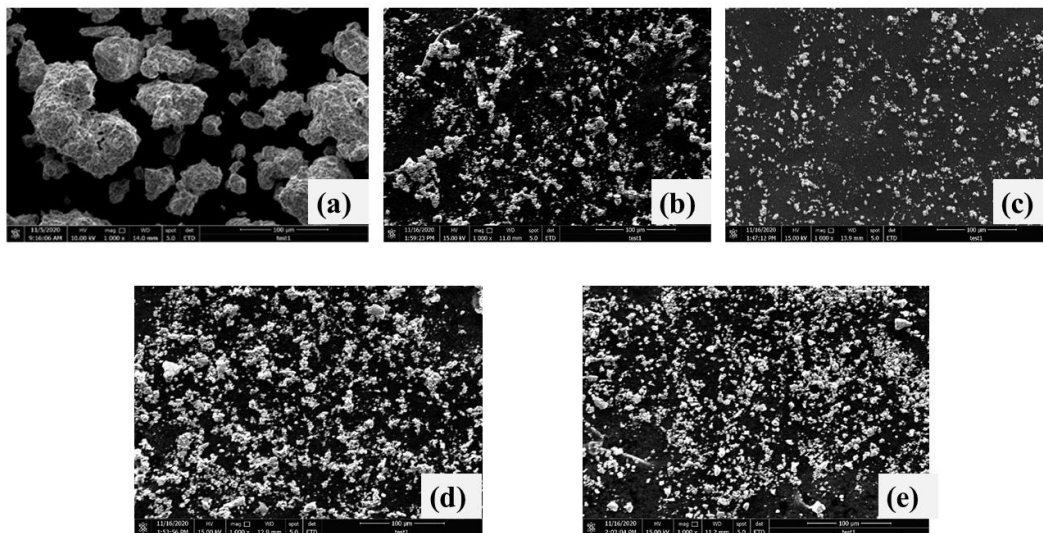


Figure 2. SEM image of BCZT powder calcined at 1,350°C under difference ball mill speeds of (a) 0, (b) 350, (c) 400, (d) 450, and (e) 500 rpm

Upon the analysis of powder particle size distribution, as shown in Figure 3, it was observed that the untreated powder (0 rpm) exhibited the largest average particle size of 4.24 μm . Moreover, there was a discernible trend wherein the powder particle size diminished with escalating rotational speeds, reaching a minimum average particle size of 0.12 μm at 400 rpm. Subsequently, as the rotational speed continued to increase, there was an observed increment in the average powder particle size. The influence of the rotational speed of a ball mill on the dimensions of powder particles during the grinding process is considerable. The relationship between the rotational speed and particle size is intricate and can be explicated through distinct stages. Initially, with an increase in rotational speed, there is an augmentation in the kinetic energy transferred to the milling balls.

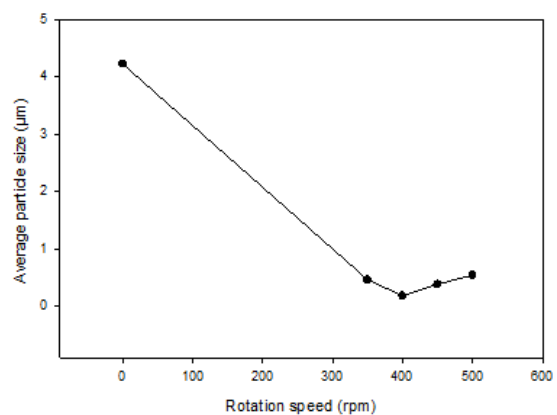


Figure 3. Average powder particle size as a function of rotation speed

powder was repeatedly fractured, achieving a finer particle size. However, this refinement also increased the particles' surface energy and their attraction to each other. Additionally, the enclosed environment prevented adequate heat dissipation, which caused these fine particles to clump into larger aggregates (Siheng *et al.*, 2020). Consequently, careful control and understanding of the rotational speed are imperative for achieving the desired characteristics of the ground powder particles. The particle size of powders in ferroelectric materials assumes a pivotal role in shaping their dielectric properties (Murali *et al.*, 2010; Hannachi *et al.*, 2015; Alhajiet *et al.*, 2021). These properties depend upon the material's capacity to respond to an applied electric field, are subject to multifaceted influences exerted by particle size. In particular, finer particles exhibit a greater surface area in comparison to their larger counterparts. This augmented surface area fosters heightened interaction with the surrounding environment, thereby augmenting the dielectric constant.

Figure 4 depicts the observed relationship between rotational speed and dielectric constant. As the rotational speed increased, the dielectric constant initially exhibited a positive correlation with reduced powder size. This trend culminated in a maximum value of 2,794 at 400 rpm. Beyond this peak, the dielectric constant demonstrated an inverse relationship with increasing rotational speed, meaning it decreased as the rotational speed continued to rise. Interestingly, Figure 5 reveals a captivating correlation between the piezoelectric coefficient (d_{33}) of the BCZT ceramic and their respective rotation speeds, mirroring the observed trends in dielectric properties. The coefficient exhibits a pronounced peak value of 370 pC/N at a rotational speed of 400 rpm. This suggests a potential interplay between dielectric behavior and piezoelectric response within the material under varying processing conditions.

From the experimental results mentioned above, it can be seen that, controlling the ball mill rotational speed is indispensable for achieving the targeted particle size distribution. Achieving an optimal equilibrium is essential to leverage the benefits of enhanced grinding efficiency while simultaneously minimizing detrimental effects such as agglomeration. Consequently, optimizing the rotational speed stands out as a crucial parameter for attaining the desired characteristics of the milled powder particles. In summary, the particle size of ferroelectric materials serves as a critical determinant influencing their dielectric properties. The meticulous adjustment of particle size enables the fine-tuning of

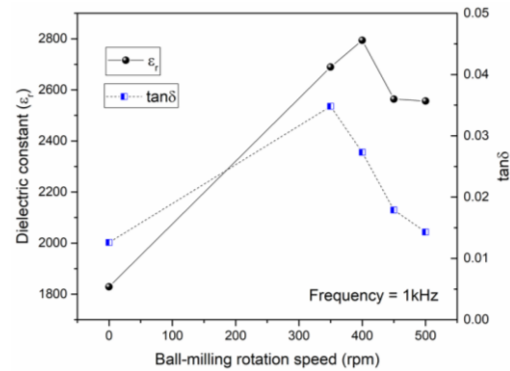


Figure 4. Dielectric properties of BCZT ceramics as a function of rotation speed

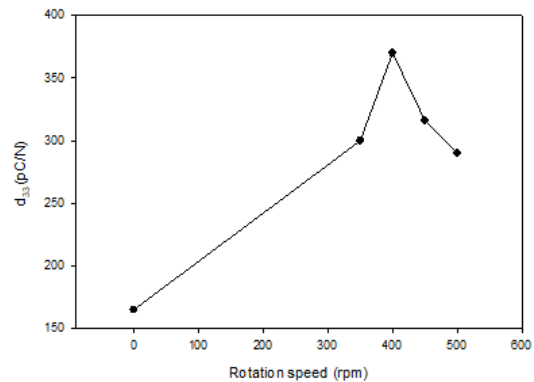


Figure 5. Piezoelectric coefficient of BCZT ceramics as a function of rotation speed

dielectric constants, and losses, underscoring its indispensability in the strategic design and engineering of ferroelectric materials for diverse applications in electronics and energy storage.

Conclusions

The effect of ball-milling rotation speed on the structure and electrical properties, including dielectric and piezoelectric properties, of $0.5\text{Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3-0.5(\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3$ ceramic was investigated systematically. Variations in the rotational speed of a ball mill alter the particle size and electrical properties. The particulate size was determined by the rotational speed. The rotational speed that produces the finest powder particle size is 400 rpm. In addition, the BCZT ceramic with a rotation speed of 400 rpm exhibited excellent piezoelectric and dielectric properties, with a piezoelectric coefficient of 370 pC/N and a dielectric constant at room temperature of 2,794.

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