

Notable grain-size dependence of converse piezoelectric effect in BaTiO₃ ceramics

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ABSTRACT

Converse piezoelectric effect is of critical importance to device applications like actuators, but no systematical investigation concerning the influence of microstructure on it has been reported for BaTiO₃ ceramics so far. Piezoelectric and ferroelectric properties were inclusively investigated for a group of BaTiO₃ ceramics that are fabricated through solid-state reaction route and show various average grain sizes in this study. It was found that the piezoelectric properties of these BaTiO₃ ceramics display significant grain-size dependences. The direct piezoelectric coefficient d_{33} increases with decreasing the average grain size (GS) from 170 pC/N at 40 μm, reaches a maximum value of 413 pC/N at 1.2 μm, and then decreases with a further reduction of GS. Converse piezoelectric effect was characterized by measurement of unipolar strain versus electric field (S – E) curve, and the converse piezoelectric coefficient $d_{33}^*(E)$ was quantitatively calculated from the slope of S – E curve at relatively large E . Interestingly, $d_{33}^*(E)$ is nearly twice as large as d_{33} and shows a quite similar trend of change with GS to d_{33} . It increases largely from 350 pm/V to 870 pm/V when reducing the GS value from 40 μm to 1.2 μm, and then decreases to 480 pm/V with the further GS reduction to 0.7 μm. Meanwhile, the remanent polarization P_r shows an increase with the decreasing of GS, reaches a maximum at 3.3 μm, and then decreases with the further GS reduction. Domain structure is considered to play an essential role in determining the notable grain-size dependence of converse piezoelectric effect.

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1. Introduction

The piezoelectricity of a piezoelectric material includes two aspects, i.e., the abilities to generate an electric charge response when an external mechanical stress is put on or develop a strain response when an external electric field is applied. The former is defined as direct piezoelectric effect and the latter is named as converse piezoelectric effect. Piezoelectric ceramics are an important type of functional materials, which are popularly utilized in sensors, actuators, ultrasonic transducers and other electronic devices. For applications, the direct and converse piezoelectric effects are equally important [1]. Practical device applications may take the advantages of either direct piezoelectric effect or converse piezoelectric effect.

Barium titanate (BaTiO₃) is a fundamental ferroelectric material from viewpoints of both academic studies and industrial applications. Historically, the important physical phenomenon that a poled polycrystalline material can show piezoelectricity was ever

first discovered in its ceramics. For several decades, BaTiO₃-based ceramics had been known to show relatively low d_{33} values of about 190 pC/N [2]. Nevertheless, amazingly high d_{33} values comparable to those of Pb(Ti,Zr)O₃-based ceramics have been obtained in both some pure BaTiO₃ and BaTiO₃-related ceramics in recent years [3–13]. These breakthroughs indicate that BaTiO₃-based ceramics possess a great potential as lead-free piezoelectric materials. On the other hand, a close examination of the literature reveals that BaTiO₃ ceramics show actually a quite diversified relationship between d_{33} and microstructure, according to the raw materials used and the sintering techniques adopted [9,14]. Some BaTiO₃ ceramics display an increase in d_{33} with the decreasing average grain size (GS), passing through a maximum at about 1.0 μm and then decreasing with a further reduction of GS [8,9,14–17]. This phenomenon was named as the piezoelectric grain-size effect in our previous study [15]. However, other BaTiO₃ ceramics display entirely different piezoelectric behaviors, where the change of d_{33} as a function of GS does not obey the rule suggested by the piezoelectric grain-size effect. Instead, relatively high d_{33} values are observed in some middle-sized or coarse-grained BaTiO₃ ceramics [7,9,14,18–20].

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Although converse piezoelectric effect is considered to be of critical importance to the applications such as actuators, systematical investigation concerning the influence of microstructure on converse piezoelectric effect has been rarely reported for BaTiO₃ ceramics so far. In this work, piezoelectric and ferroelectric properties were inclusively studied for a group of BaTiO₃ ceramics that are fabricated through solid-state reaction route and show various average grain sizes. Particular attention was given to the microstructure influence on converse piezoelectric effect. It was found that similarly to direct piezoelectric effect, converse piezoelectric effect is also greatly affected by microstructure.

2. Experimental

The BaTiO₃ ceramics were prepared by the conventional solid-state reaction technique, starting from raw materials of commercial BaCO₃ powder (purity $\geq 99.0\%$, Sinopharm Chemical Reagent Co. Ltd.) and TiO₂ powder (purity $\geq 99.8\%$, Xiantao Zhongxing Electronic Material Co. Ltd.). The preparation procedures are basically the same as those in the previous study [15], except a slight difference in calcination. The raw materials were weighed according to the stoichiometric ratio and ball-milled with zirconia balls in alcohol media for 12 h on a planetary ball mill. The milled slurry mixture was dried, crushed in an agate mortar. The crushed mixture powder was then calcined at 1050 °C for 4 h. This was followed by a second ball-milling procedure with a milling time of 12 h. The obtained fine BaTiO₃ powder after drying was mixed with a 0.5 wt% polyvinyl alcohol (PVA) binder, granulated, and pressed into small disks (15 mm in diameter and 1.5 mm in thickness) at 200 MPa. The PVA binder was then burned out at 650 °C for 0.5 h. Finally, sintering was carried out in air under different temperature conditions from 1190 to 1360 °C for 2 h to 4 h.

In order to characterize the piezoelectric, dielectric and ferroelectric properties, both surfaces of the BaTiO₃ ceramic specimens were coated with silver paint and cured at 570 °C for 0.5 h. Poling was accomplished at 100 °C in silicon oil under 2.0 kV mm⁻¹ for 30 min. Measurements of the piezoelectric and dielectric properties were taken after the passage of 24 h. The d_{33} value was measured by a Berlincourt-type d_{33} meter (YE 2730A) and the permittivity ϵ' was measured using an Agilent 4294A precision impedance analyzer. Temperature control for the dielectric and piezoelectric measurements was accomplished by an Espec SU-261 chamber. The polarization versus electric field (P - E) hysteresis loops and the unipolar strain versus electric field (S - E) curves were measured on Radiant precision workstation (Premier II), to which an MTI optical fiber measurement system is combined for the piezoelectric displacement measurement. To observe the microstructure and domain patterns, some poled BaTiO₃ ceramic specimens were mirror-polished and chemically etched in an aqueous solution of hydrochloric and hydrofluoric acids. The observation was carried out on a HITACH S-4800 scanning electron microscope (SEM). Mass density ρ was obtained by measuring the mass and the dimensions, and relative ceramic density ρ_0 was calculated with the theoretical density of 6.01 g/cm³.

3. Results and discussion

All the BaTiO₃ ceramics prepared in this study have quite dense microstructure with uniform grain-size distribution. Table 1 lists the data of ρ_0 , GS, d_{33} and ϵ' measured at room temperature (of 25 °C) for the poled BaTiO₃ ceramics prepared under various sintering conditions. As can be seen, the values of ρ_0 and GS increase with increasing the sintering temperature. Those BaTiO₃ ceramics

Table 1

Data of the relative density, average grain size, d_{33} and ϵ' for the various poled BaTiO₃ ceramics prepared under conditional method.

| Sinter. Cond. | ρ_0 (%) | g (μm) | d_{33} (pC/N) | ϵ' (at 1 k Hz) |
|---------------|--------------|---------------------|-----------------|-------------------------|
| 1190 °C/2 h | 95.6 | 0.7 | 264 | 2980 |
| 1210 °C/2 h | 96.4 | 0.8 | 325 | 3185 |
| 1220 °C/2 h | 96.5 | 1.0 | 351 | 3310 |
| 1230 °C/2 h | 98.1 | 1.2 | 413 | 3310 |
| 1250 °C/2 h | 98.1 | 3.3 | 310 | 3295 |
| 1250 °C/3 h | 98.2 | 5.6 | 224 | 3210 |
| 1340 °C/2 h | 99.0 | 25 | 198 | 3090 |
| 1330 °C/3 h | 98.4 | 28 | 173 | 2910 |
| 1360 °C/4 h | 99.2 | 40 | 170 | 2800 |

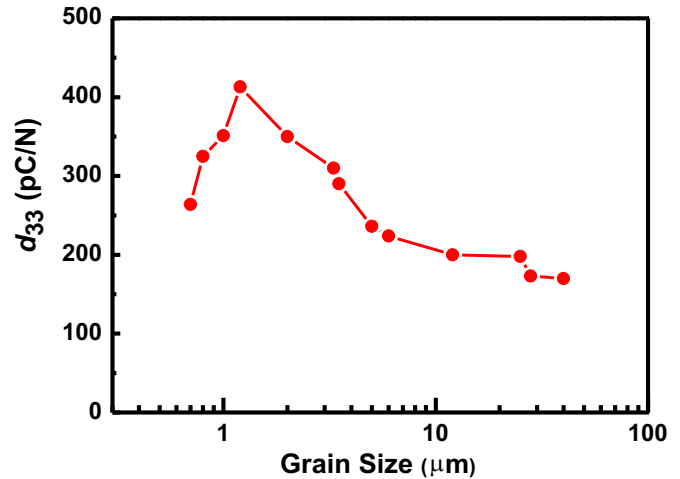


Fig. 1. Grain-size dependence of d_{33} observed at room temperature. Dashed line is inserted for eye's guide.

sintered at 1230 °C higher temperatures show $\rho_0 \geq 98.0\%$ and GS $\geq 1.2 \mu\text{m}$, while the ones sintered at 1190 °C or 1220 °C exhibit the ρ_0 value of 95.6% or 96.5% and the GS value of 0.7 μm or 1.0 μm . Fig. 1 plots the grain-size dependence of d_{33} for the poled BaTiO₃ ceramics. d_{33} shows large change with GS. It increases from 170 pC/N to 413 pC/N with decreasing the GS from 40 μm to 1.2 μm , and then decrease with a further reduction of GS. Meanwhile, ϵ' shows a similar trend of change to d_{33} with GS. Generally, the results observed here are in good consistency with a part of previously reported results [8,9,14–17].

Ferroelectric properties and domain reversal characters were examined for some typical BaTiO₃ ceramics by the measurements of polarization–electric field (P - E) and current–electric field (I - E) loops at room temperature. The result is shown in Fig. 2(a)–(f). Peaks are seen in the I - E loop curves roughly at the E_c values, and the peak sharpness of an I - E loop curve is closely related the squareness of its corresponding P - E loop curve. As shown in Fig. 2 (a) and (f), those BaTiO₃ ceramics with either the smallest GS value of 0.8 μm or the largest one of 40 μm demonstrate the slanted P - E loop curves and the I - E loop curves of broaden peaks, which are similar to that reported in Ref. [21]. Fig. 2(g) summarizes the variations of remanent polarization P_r and coercive field E_c with GS. It is clear from the figure that P_r increases with the decrease of GS from 10.5 $\mu\text{C}/\text{cm}^2$, reaches a maximum value of 13.1 $\mu\text{C}/\text{cm}^2$ at 3.3 μm , and then decreases with the further decreasing GS. The presently observed trends of P_r and E_c changes with GS differs from the previously reported ones for those BaTiO₃ ceramics prepared by techniques of plasma spark sintering or two-step sintering [14,17]. However, it is interesting to note that the BaTiO₃ with GS=3.3 μm has a similar shape of the P - E loop to the one observed for an as-prepared micro-grained BaTiO₃ ceramic

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