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$BaTiO_3$ ceramics toughened by dispersed coarse particles

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Abstract

A new approach was proposed and investigated for toughening $BaTiO_3$ ceramics, where coarse $BaTiO_3$ particles were incorporated and dispersed into the fine-grained matrix. The fracture toughness was enhanced significantly by increasing the content of coarse $BaTiO_3$ particles and reached the maximum of 2.0 $MPam^{1/2}$, while that of the matrix was 1.1 $MPam^{1/2}$. The piezoelectric properties showed slight variation in comparison to the reference $BaTiO_3$ ceramics.

Keywords: B. Grain size; C. Toughness and toughening; C. Piezoelectric properties; $BaTiO_3$ ceramics

1. Introduction

Barium titanate ($BaTiO_3$) ceramics are extensively used for positive temperature coefficient resistors, capacitors, gas sensor and other electronic devices. During end termination and soldering, a stress of 30–50 MPa is generated in $BaTiO_3$, which can measurably decrease the fracture toughness [1]. Recently, electronic devices have been miniaturized and used under quite severe atmospheres, and improved mechanical properties of $BaTiO_3$ ceramics are required. So far, some approaches [2–6] have been investigated to improve the mechanical performance of $BaTiO_3$ ceramics, but the piezoelectric properties are inevitably degraded in such approaches.

In the present work, a new approach is proposed to toughen $BaTiO_3$ ceramics without degrading piezoelectric properties, in which coarse $BaTiO_3$ particles were incorporated and dispersed into the fine $BaTiO_3$ matrix. The microstructures, mechanical and piezoelectric properties of toughened $BaTiO_3$ ceramics were investigated together with the discussion on the toughening mechanism.

2. Experimental

The fine and coarse end member powders of $BaTiO_3$ were calcined from high purity $BaCO_3$, TiO_2 powders at 1150 and 1300 °C in air for 2 h, and milled in ethanol using zirconia media for 48 and 24 h, respectively. After fine and coarse $BaTiO_3$ powders were milled for 24 h and dried, the powders were pressed at 98 MPa into disk samples of 20 and 10 mm in diameter, respectively, for measuring the piezoelectric and mechanical properties. These compacts were sintered at 1325 °C for 3 h, and cooled at a rate of 100 °C/h. Samples of 20 mm in diameter were prepared and poled in silicon oil at 120 °C for 30 min with an electric field of 3 kV/mm.

SEM observation was carried out for microstructure characterization. A Coulter LS-230 laser particle size analyzer determined the particles size of calcined $BaTiO_3$ particles, and the data is shown in Fig. 1. The average grain size and grain size distribution of $BaTiO_3$ samples were measured by linear intercept method [7,8] from SEM micrographs. Fracture toughness was evaluated for five tests each sample through the indentation method [9] under a load of 98 N.

$$\left(K_{IC}\phi/Ha^{1/2}\right)(H/E\phi)^{2/5} = 0.142(c/a)^{-1.56} \quad (1)$$

where K_{IC} is the fracture toughness, H is the hardness, E is the Young's modulus, ϕ is a constant factor (approx.

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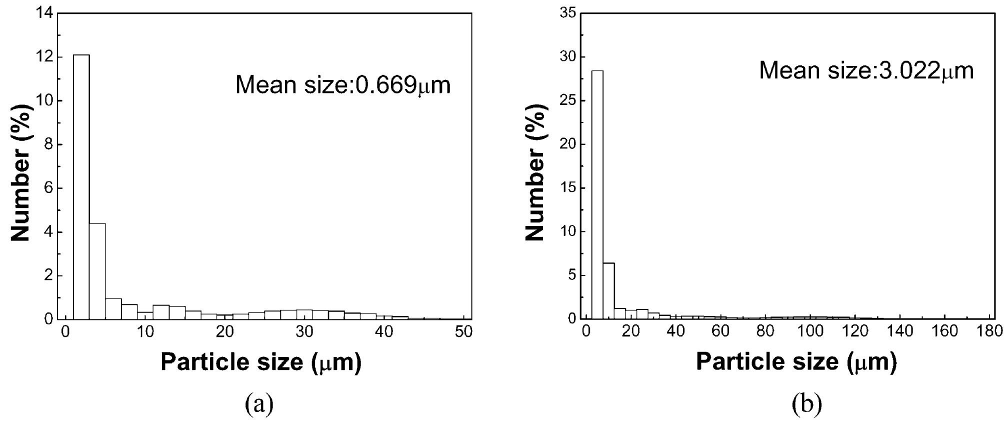

Fig. 1. Particle size distribution of calcined $BaTiO_3$ powders: (a) fine $BaTiO_3$, (b) coarse $BaTiO_3$.

3), a is the half-diagonal of the Vickers indent and c is the length of the crack.

The relative dielectric constant $\varepsilon/\varepsilon_o$ of unpoled specimens was measured using an LCR meter (HP4284A) at 1 MHz and the relative dielectric constant $\varepsilon_{33}^{T}/\varepsilon_o$ and dissipation factor tanδ of poled specimens were measured at 1 kHz. Planar electromechanical coupling factor K_P and mechanical quality factor Q_m were determined by a routine resonant technique.

3. Results and discussion

As shown in Fig. 2, the coarse $BaTiO_3$ grains disperse in the fine-grained $BaTiO_3$ matrix with some porosity. There is a narrow grain size distribution in the specimen without coarse $BaTiO_3$ (Fig. 3), whereas the grain size distribution of other specimens is broader, and more large grains are observed with increasing incorporated coarse grain content x.

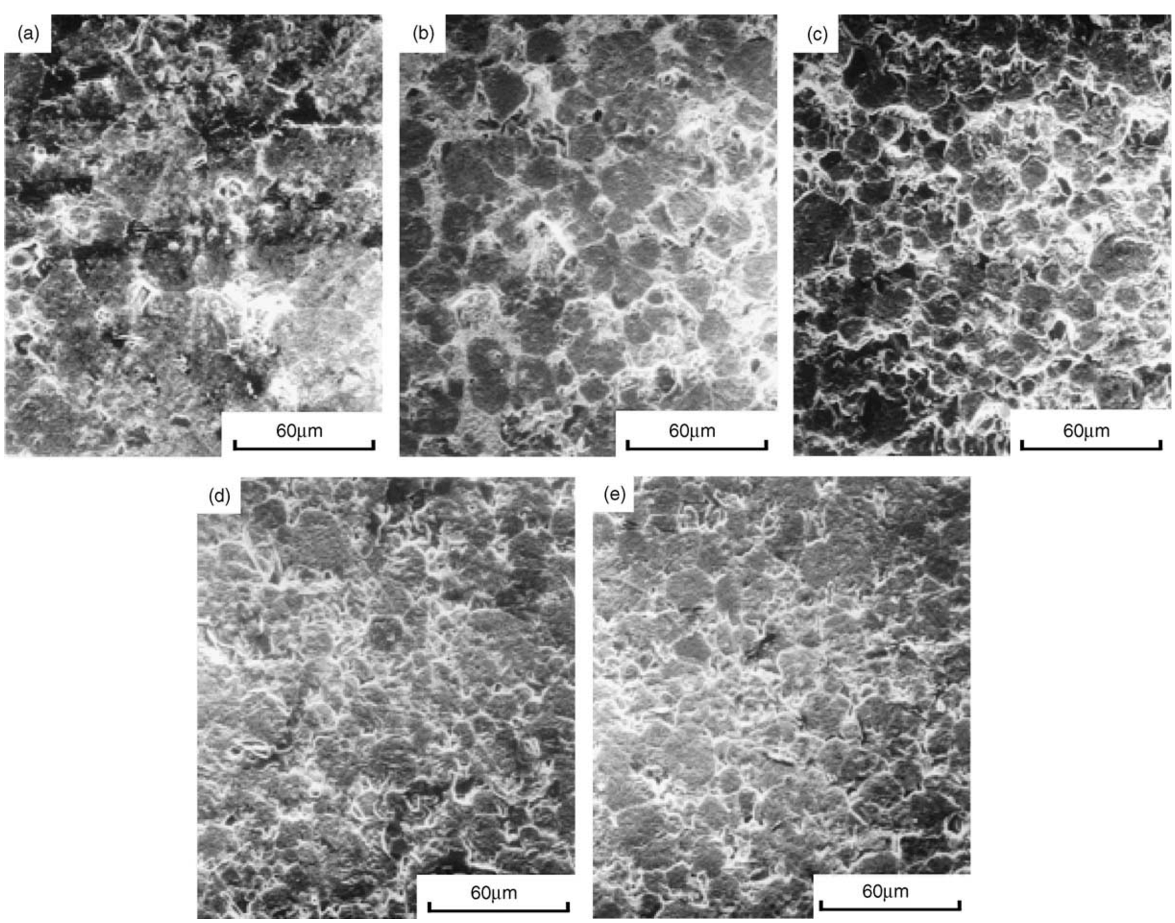

Fig. 2. SEM micrograph of $BaTiO_3$ ceramics for (a) $x=0$; (b) $x=5$; (c) $x=10$; (d) $x=15$; (e) $x=20$ wt.%.

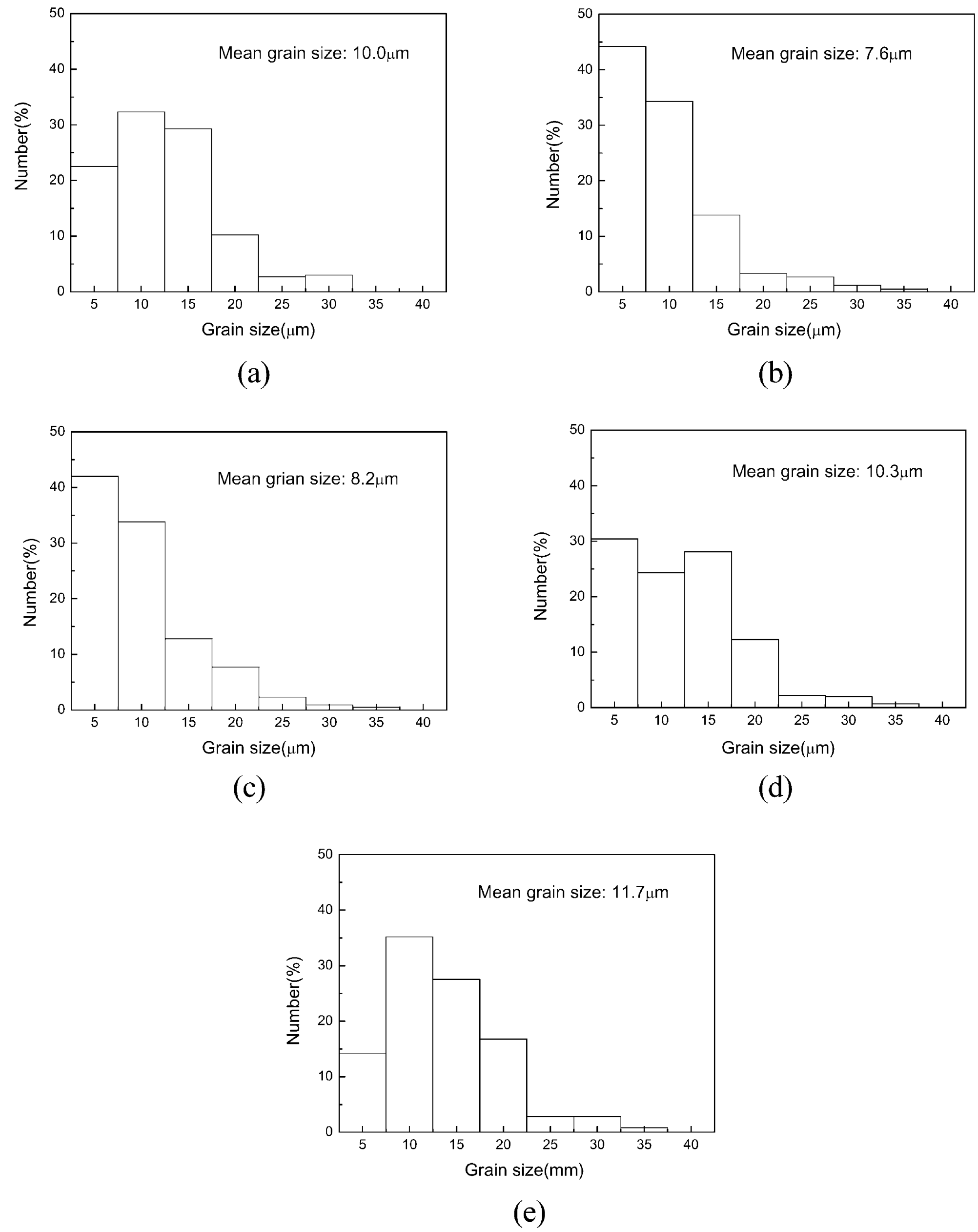

Fig. 3. Average grain size and grain size distribution in $BaTiO_3$ for (a) $x=0$; (b) $x=5$; (c) $x=10$; (d) $x=15$; (e) $x=20$ wt.%.

As shown in Table 1, the fracture toughness K_{IC} of $BaTiO_3$ ceramics is enhanced significantly by incorporating coarse $BaTiO_3$ grains, and reaches the maximum of 2.0 $MPam^{1/2}$ at $x=15$ wt.%, while that of the matrix is 1.1 $MPam^{1/2}$. The fracture mode of these specimens was almost transgranular fracture, which results in rough surfaces of the cleaved grains (see Fig. 4). In the toughened ceramics the fracture is dominated by the coarse grains instead of the pre-coexisted processing flaws. At the room temperature, the thermal expansion anisotropy due to $BaTiO_3$ perovskite structure leads to residual stresses between different orientations of grains

Table 1
Mean grain size, mechanical and piezoelectric properties of toughened $BaTiO_3$ ceramics

The incorporated coarse $BaTiO_3$ content (wt.%)	Mean grain size (μm)	K_{IC} ($MPam^{1/2}$)	ϵ/ϵ_O	$\epsilon^T_{33}/\epsilon_O$	tanδ	K_P (%)	Q_m
0	10.0	1.1±0.2	2118	2085	0.02	31	276
5	7.6	1.3±0.3	2064	2024	0.04	31	280
10	8.2	1.7±0.2	2027	1990	0.03	30	275
15	10.3	2.0±0.4	1989	1965	0.01	30	267
20	11.7	1.4±0.4	1961	1933	0.02	30	271

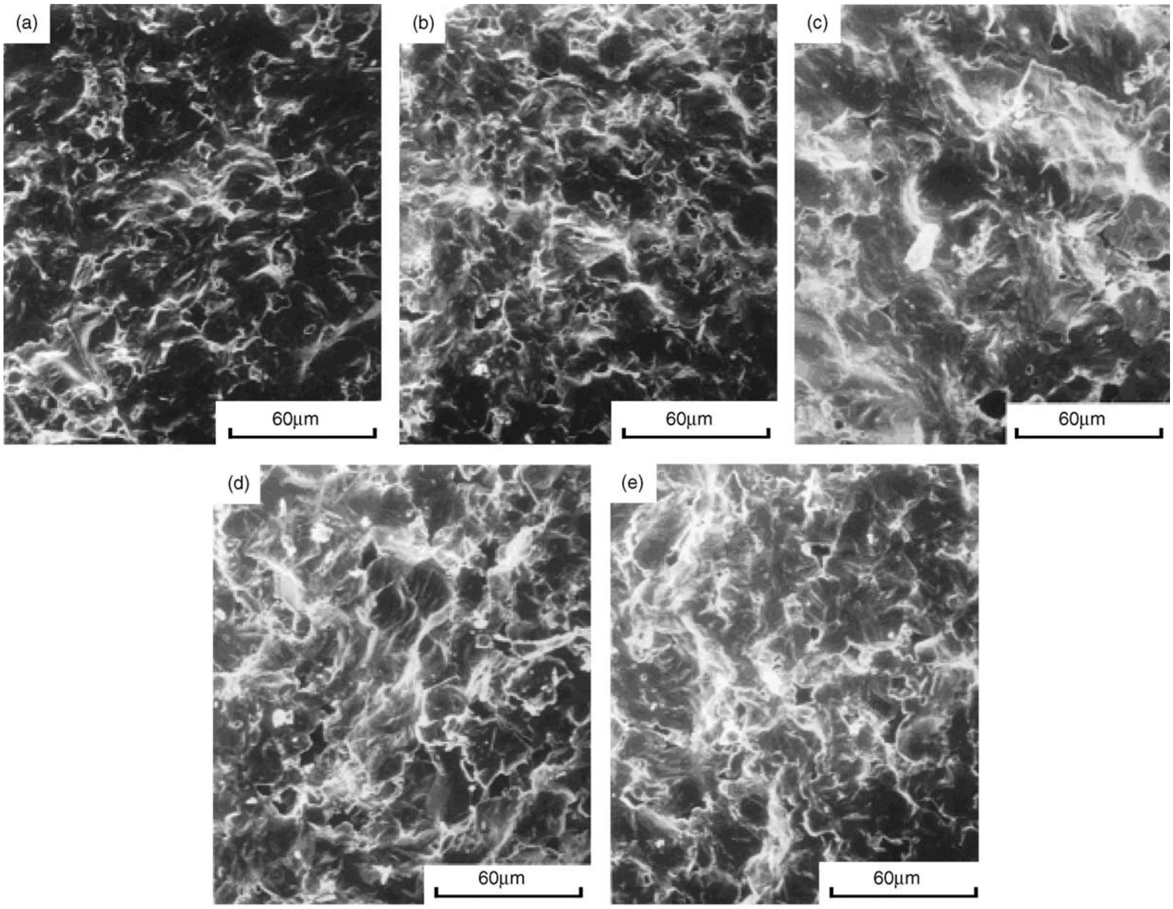

Fig. 4. SEM micrograph of $BaTiO_3$ ceramics fracture surface for (a) $x=0$; (b) $x=5$; (c) $x=10$; (d) $x=15$; (e) $x=20$ wt.%.

during cooling, and the mismatch of residual stresses brings to microcracks. The grain size is larger, and more microcracks occur [10]. Then the contribution of microcracks to fracture energy increases with increasing grain size and the incorporated coarse grains. On the other hand, the fracture energy of transgranular fracture is proportional to the grains cross-sectional area. The porosity reduces the fracture energy of primary crack growth by decreasing the grain cross-sectional area. The fracture toughness of toughened $BaTiO_3$ is attributed to two countertrends of fracture energy as incorporated coarse grains content increase: (1) an increase in fracture energy due to the increasing number of microcracks from the effects of thermal expansion stresses and (2) a decrease in fracture energy due to the decreasing grain cross-sectional area caused by porosity. Under the integrated influences, the fracture toughness rises to a maximum, and then decreases with increasing incorporated coarse grains.

Table 1 lists the piezoelectric properties of the toughened $BaTiO_3$ ceramics. Planar electromechanical coupling factor K_P, mechanical quality factor Q_m and dissipation factor tanδ vary little with x. The dielectric constant generally decreases with increasing x, and this can be clarified by the variation of grain size with x. In $BaTiO_3$ ceramics, it is known that the dielectric constant depends strongly on grain size [11]. The size of domain shows a parabolic scaling relation with the grain size[12]:

$$\text{domain size} \propto (\text{grain size})^m \quad (2)$$

where the index m is positive. The twin wall decreases with increasing grain size, which leads to the decrease of dielectric constant. The grain size generally increases with increasing x, and subsequently decreases the dielectric constant.

4. Conclusions

$BaTiO_3$ Ceramics were significantly toughened by incorporating coarse $BaTiO_3$ particles into the fine-grained $BaTiO_3$ matrix. The fracture toughness K_{IC} reached the maximum of 2.0 $MPam^{1/2}$. The transgranular fracture was the primary fracture mode and the microcrack was the main toughening mechanism. The piezoelectric properties varied little and the dielectric constant decreased with increasing x since the larger grain size led to the decreased twin wall.

Acknowledgements

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