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Short communication

Effect of particle size on the dielectric and piezoelectric properties of 0–3BCTZO/cement composites

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Abstract

0–3 $Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$ (BCTZO)–cement composites were prepared by normal mixing and pressing of Ordinary Portland cement and BCTZO particles with average sizes of 569.8 and 8.9 μm. Three different composites were prepared with BCTZO to cement ratios of 30%, 50%, and 70% by volume. The effect of BCTZO particle size on the dielectric and piezoelectric properties of cement based 0–3 piezoelectric composites was investigated. The dielectric constant increases with both increasing BCTZO particle content and increasing BCTZO particle size. The piezoelectric properties were found to increase with the increasing BCTZO particle content and decreasing BCTZO particle size. It was found that a 70% BCTZO composite with an average particle size of 8.9 μm produced good dielectric constant, tan δ and piezoelectric properties of 107, 0.09 and 52 pC/N, respectively.

Keywords: C. Dielectric properties; C. Piezoelectric properties; Cement composites; Particle size

1. Introduction

Recently, cement based-piezoelectric composites (0–3, 1–3, 2–2 etc.) have been of interest because of their promising applications in civil engineering structures such as sensors and actuators [1–5]. In particular, cement composites of 0–3 connectivity, cement–piezoelectric material composites, were first reported by Li et al. [1]. They fabricated the lead zirconate titanate (PZT)–cement composites using normal mixing and spreading methods and have shown that the composites have compatibility with cement and have potential to be used for sensor applications. Xin et al. [2] investigated 0–3 lead magnesium niobate (PMN)– sulphoaluminate cement composites using a ball milling and pressing method and found good performances of both piezoelectricity and electromechanical coupling. Chaipanich's group et al.[3–5] investigated the piezoelectric and dielectric properties of PZT–cement-based composites prepared by normal mixing and compressing. However, these piezoelectric–cement composites are made using PZT or PMN and are toxic on the environment. As an alternative, $BaTiO_3$ (BTO)–cement composites of 0–3 connectivity were fabricated using a normal mixing and compassing method by Rianyoi et al. [6]. However, the piezoelectric properties of these composites are lower than those of the PZT–cement composites. Therefore, they tried to form the piezoelectric–cement composites in 1–3 types [7], which have good piezoelectric properties than those of 0–3 type cement composites.

Today, $BaTi_{0.8}Zr_{0.2}O_3$–$Ba_{0.7}Ca_{0.3}TiO_3$ (BZT–BCT) piezoelectric ceramics, with a composition close to the morphotropic phase boundary (MPB), exhibit large piezoelectric response, large dielectric constant and high spontaneous polarization [8–11], and are replacing PZT in sensor applications. Therefore, it is expected that this piezoelectric will play a more active role than those of 0–3 lead free piezoelectric–cement composites.

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In this study, we report the $Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$ (BCTZO)–cement composites with high piezoelectric coefficient, produced by the normal mixing and compressing of white cement and BCTZO ceramic particles. The dielectric and piezoelectric properties of these composites were investigated.

2. Experimental

$Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$ (BCTZO) ceramic particles were prepared by a solid state reaction method. $BaCO_3$ (99%), $CaCO_3$ (96%), TiO_2 (99%) and ZrO_2 (99%) were used as the

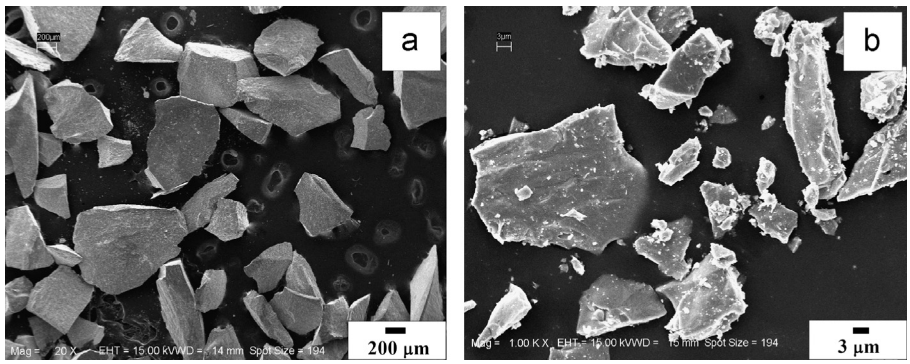

Fig. 1. The SEM images of BCTZO particles with the average particle size (a) 569.8 μm and 8.9 μm.

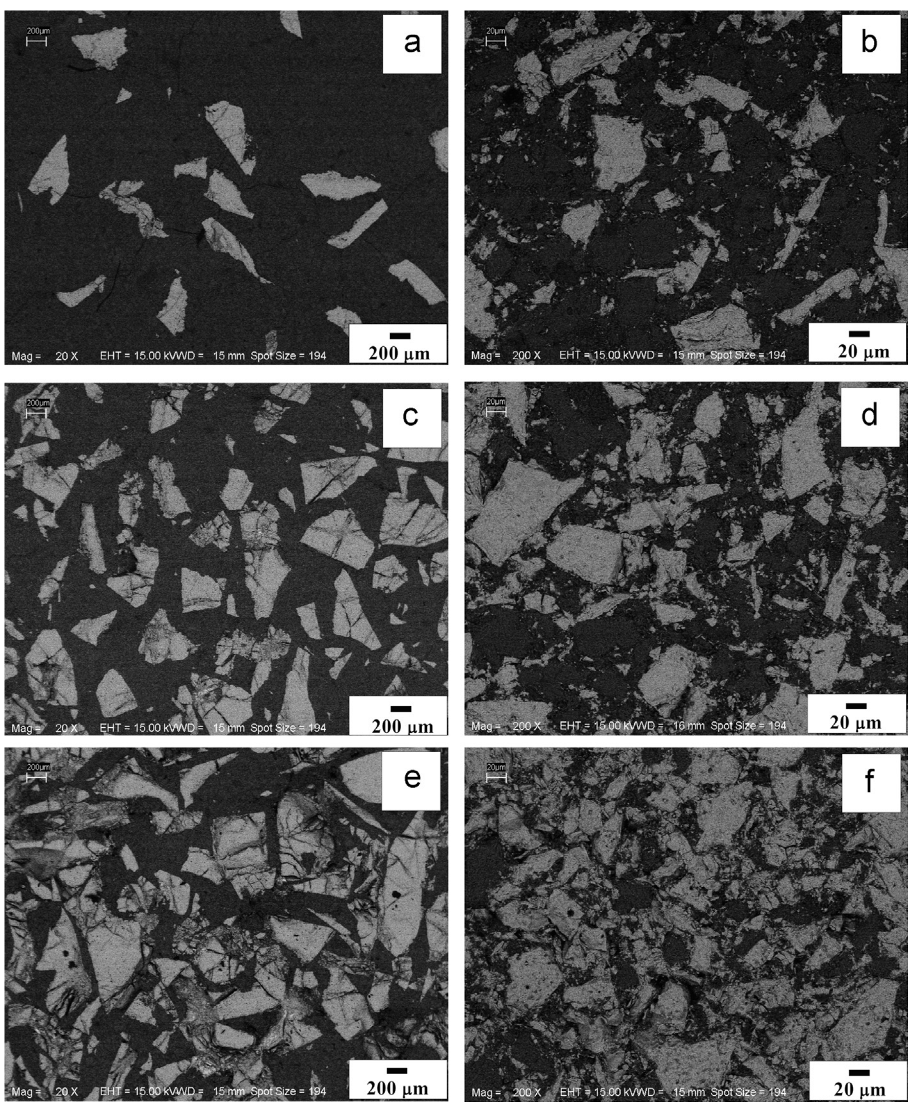

Fig. 2. The backscattered electrons SEM images of BCTZO–cement composites (a) 30%BCTZO, (b) 50%BCTZO and (c) 70%BCTZO for the average particle size as 569.8 μm, and (d) 30%BCTZO, (e) 50%BCTZO and (f) 70%BCTZO for the average particle size as 8.9 μm.

starting materials. Initially, all of these materials were mixed with 2-propanol and were ball milled for 24 h. After drying, the mixture precursor powders were calcined at 1300 °C for 3 h. The calcined BCTZO powders were compacted into disk-shaped pellets with a diameter of 13 mm and thickness of 1.5 mm with 221 MPa pressure. The green pellets were sintered at 1450 °C for 3 h. The sintered BCTZO ceramics were ground and sieved to select the size of BCTZO particles. To prepare the BCTZO–cement composites, the sintered BCTZO particles and Ordinary Portland cement were mixed and then pressed under a pressure of 146 MPa to form discs with a diameter of 16 mm and a thickness of about 2.5 mm. The disc specimens were cured in water vapor at 60 °C for 3 days and then dried.

The morphology of these samples was characterized by scanning electron microscopy (SEM, LEO VP1450, UK). Silver paint was used as the electrode and the dielectric properties of the composite discs were measured at room temperature by using an Agilent 4294A Precision impedance analyzer. The dielectric constant (ε_r) was calculated from the following equation:

$$\varepsilon_r = \frac{C_p t}{\varepsilon_0 A}$$

where C_p is the sample capacitance, t is the thickness, ε_0 is the permittivity of free space constant (8.854×10^{-12} F/m) and A is the electrode area.

The piezoelectric properties of composite discs were polarized at room temperature in silicon oil for 45 min under an electric field of 1 kV/mm. After being held for 24 h at room temperature, the piezoelectric strain factor (d_{33}) of the composite discs was measured by using a d_{33} meter (Model 90-2030, APD International, Ltd.).

3. Results and discussion

The morphology of the BCTZO particles was studied by SEM as shown in Fig. 1(a and b). The ground BCTZO particles have plate-like sharpness with sizes ranging from 294.8 to 1113.8 μm (Fig. 1(a)) or with sizes ranging from 1.6 to 36.9 μm (Fig. 1(b)). The average particle sizes of the ground BCTZO particles were evaluated to be 569.8 and 8.9 μm. SEM images of the composites, with 30%, 50% and 70% by volume of BCTZO particles, are shown in Fig. 2(a)–(f). In the backscattered electron SEM images, the black phase is cement and the white phase is BCTZO particles. It can be seen that using low content BCTZO particle results in their being isolated in the cement matrix, whereas using high content BCTZO particle results in their contact. However, with high content of BCTZO particles many cracks are observed, which are due to high compression during the formation of composite bulk.

The dielectric constants and tan δ, for the BCTZO–cement 0–3 composites of 30%, 50 %and 70% by volume and average particle size of 569.8 μm, are shown, respectively in Fig. 3(a)–(b). The dielectric properties were measured at 1 kHz and it was found that the dielectric constant increases with increasing BCTZO particle content. The values of dielectric constant were found to be 46.1, 80.7 and 154.0 and tan δ was found to be 0.12, 0.1 and 0.11 correspondingly. It was found that the dielectric constant of composites with the average particle size of 8.9 μm also increases with the increasing BCTZO particle content. The values of dielectric constant were found to be 38.7, 64.1 and 107.0 and tan δ was found to be 0.11, 0.12 and 0.09 correspondingly (Fig. 4). The dielectric constants and tan δ of the BCTZO–cement 0–3 composites, for the average particle size of 569.8 and 8.9 μm, are summarized in Table 1. From these results, it can be seen that all of the samples behave similarly to those in previous studies which show increasing dielectric constant with increasing piezoelectric particle content [1–6]. The dielectric constant increases with the increasing BCTZO particle size because of the influence of the dielectric constant of BCTZO bulks (4270 at 1 kHz), which is higher than that of cement bulk (19.7 at 1 kHz) and is explained by the composites model [1,6,12]. It was also found that the dielectric constant decreases with decreasing BCTZO particle size because of the low polarizations in small particles resulting in a lower dielectric constant in the samples.

The piezoelectric strain factor (d_{33}) values of the BCTZO–cement 0–3 composites of 30%, 50% and 70% by volume for the average particle sizes of 569.8 and 8.9 μm are also summarized in Table 1. It was found that the piezoelectric

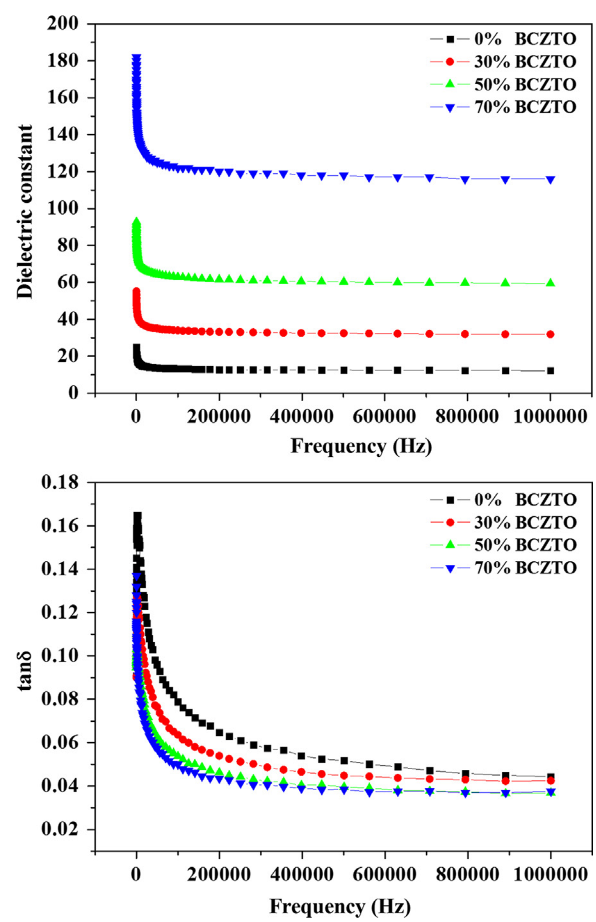

Fig. 3. Dielectric constant and tan δ of various BCTZO–cement composites with the average particle size as 569.8 μm measured at room temperature.

strain factor increases with increasing BCTZO particle content. In the composites with average particle size as 569.8 μm, the values of piezoelectric strain factor were found to be 4, 18 and 44 pC/N correspondingly. For composites with an average particle size of 8.9 μm, it was found that piezoelectric strain factor also increases with increasing BCTZO particle content. The values of piezoelectric strain factor were found to be 10, 32 and 52 pC/N. It is well known that cement is not piezoelectric and the incorporated BCTZO particles play an important role in the piezoelectricity of the composite. And then it can be seen that the piezoelectric strain factor of composites increases with decreasing BCTZO particle size [13,14]. Shifeng et al. [13] reported that piezoelectric properties decreased with decreasing ceramic particle size because the small ceramic particles (1.45–68.5 μm) have a high surface area and the volume ratio of lead lithium niobate to lead zirconate to lead titanate (PLN) leads to low piezoelectric properties or non-piezoelectric properties on the surface layer. On the other hand, because the chances for large ceramic particles (107.08–294.07 μm) to contact each other increase, the connectivity patterns change. In addition, Li et al. found that high piezoelectric properties can be obtained by using high crystalline nano-PZT particle (200 nm) cement composites [14] with a high d_{33} value of 52 pC/N. However, this study shows that smaller BCTZO particles have higher surface areas leading to lower interface space, resulting in better contact between the individual BCTZO particles. This is quite clear from the SEM results. The authors obtained quite good performances compared to those obtained by others at Composites with 70 vol% of piezoelectric particles, the BCTZO–cement composites, with average sizes of 569.8 and 8.9 μm have d_{33} values of 44 and 52 pC/N, respectively, which are higher than that of the BTO–cement composite (16 pC/N) [6] and the PZT–cement composite (43 pC/N) [3]. Therefore, the authors expect that 1–3 will play a more active role than 0–3 lead free piezoelectric–cement composites for sensors and actuators applications in civil engineering structures.

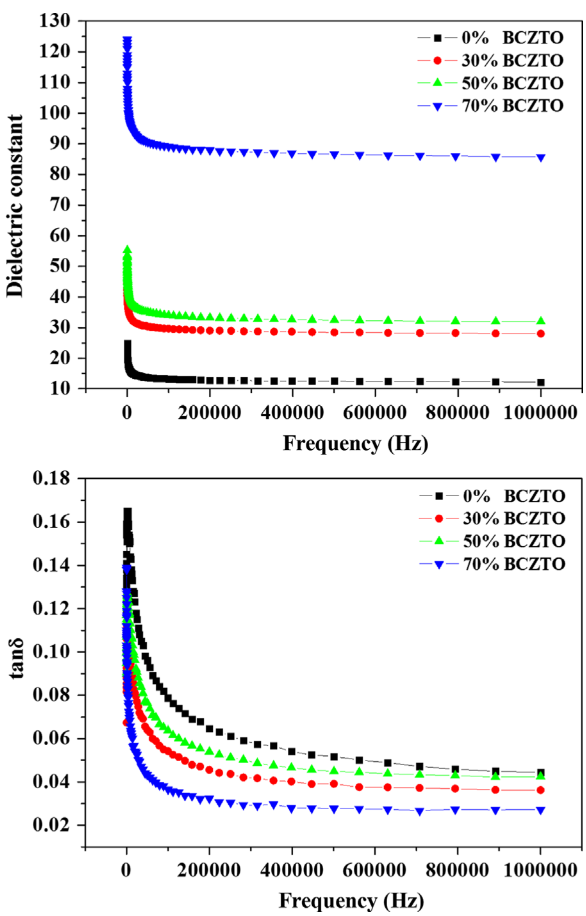

Fig. 4. Dielectric constant and tan δ of various BCTZO-–cement composites with average particle size as 8.9 μm measured at room temperature.

4. Conclusion

In this study, $Ba_{0.85}Ca_{0.15}Ti_{0.9}Zr_{0.1}O_3$ particles were prepared by the solid state reaction method. The cement based 0–3 piezoelectric composites were fabricated by normal mixing and pressing of Ordinary Portland cement and BCTZO particles and curing in water vapor at 60 °C for 3 days. It was found that the dielectric constant and piezoelectric strain factor increase with increasing volume percentage of BCTZO particles. Good values for dielectric constant, dielectric loss and piezoelectric strain factor were obtained (107, 0.19 and 52 pC/N, respectively) for 70 vol% BCTZO particles with an average particle size of 8.9 μm. The piezoelectricity of the composites containing small BCTZO particles was high due to the high surface area and low interface space between the individual BCTZO particles, leading to good connection between those particles.

Table 1
Dielectric constant, tan δ and piezoelectric strain factor of various BCTZO–cement composites with the average particle size 569.8 and 8.9 μm measured at room temperature.

BCTZO particle content (vol%)	Dielectric constant ε_r measured at 1 kHz		tan δ, measured at 1 kHz		Piezoelectric charge constant, d_{33} (pC/N)	
	569.8 μm	8.9 μm	569.8 μm	8.9 μm	569.8 μm	8.9 μm
30	46.1	38.7	0.12	0.11	4	10
50	80.7	64.1	0.10	0.12	18	32
70	154.0	107.0	0.11	0.09	44	52

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References

[1] Z.J. Li, D. Zhang, K. Wu, Cement-based 0–3 piezoelectric composites, Journal of the American Ceramic Society Advanced Materials 85 (2) (2002) 305–313.

[2] C. Xin, H. Shifeng, C. Jun, X Ronghua, L. Futian, L. Lingchao, Piezoelectric and dielectric properties of piezoelectric ceramic–sulphoaluminate cement composites, Journal of the European Ceramic Society 25 (2005) 3223–3228.

[3] A. Chaipanich, Dielectric and piezoelectric properties of PZT–cement composites, Current Applied Physics 7 (2007) 537–539.

[4] N. Jaitanong, A. Chaipanich, T. Tunkasiri, Properties of 0–3 PZT–Portland cement composites, Ceramics International 34 (2008) 793–794.

[5] A. Chaipanich, G. Rujiganagul, T. Tunkasiri, Properties of Sr- and Sb-doped PZT–Portland cement composites, Applied Physics A 94 (2009) 329–337.

[6] R. Rianyoi, R. Potong, N. Jaitanong, R. Yimnirun, Dielectric, ferroelectric and piezoelectric properties of 0–3 barium titanate–Portland cement composites, Applied Physics A 104 (2011) 661–666.

[7] R. Potong, R. Rianyoi, A. Ngamjarurojana, A. Chaipanich, Dielectric and piezoelectric properties of 1–3 non-lead barium zirconate titanate–Portland cement composites, Ceramics International 39 (2013) S53–S57.

[8] W.F. Liu, X.B. Ren, Large piezoelectric effect in Pb-free ceramics, Physical Review Letters 103 (2009) 257602.

[9] P. Wang, Y.X. Li, Y.Q. Lu, Enhanced piezoelectric properties of $(Ba_{0.85}Ca_{0.15})(Ti_{0.9}Zr_{0.1})O_3$ lead-free ceramics by optimizing calcination and sintering temperature, Journal of the European Ceramic. Society 31 (2011) 2005–2012.

[10] J.G. Wu, D.Q. Xiao, W.J. Wu, J.G. Zhu, J. Wang, Effect of dwell time during sintering on piezoelectric properties of $(Ba_{0.85}Ca_{0.15})(Ti_{0.90}Zr_{0.10})O_3$ lead-free ceramics, Journal of Alloys and Compounds 509 (2011) L359–L361.

[11] D.Z. Xue, Y.M. Zhou, H.X. Bao, C. Zhou, J.H. Gao, X.B. Ren, Elastic, piezoelectric, and dielectric properties of $Ba(Zr_{0.2}Ti_{0.8})O_3$–$50(Ba_{0.7}Ca_{0.3})TiO_3$ Pb-free ceramic at the morphotropic phase boundary, Journal of Applied Physics 109 (2011) 054110.

[12] R. Potong, R. Rianyoi, A. Chaipanich, Dielectric properties of lead-free composites from 0–3 barium zirconate titanate–Portland cement composites,, Ferroelectrics. Letters 38 (2011) 18–23.

[13] H. Shifeng, L. Lingchao, C. Jun, X. Dongyu, F. Futian, C. Xin, Influence of ceramic particle size on piezoelectric properties of cement-based piezoelectric composites, Ferroelectrics. Letters 332 (2006) 187–194.

[14] Z.J. Li, H.Y. Yu, Y.J. Zhang, Fabrication and piezoelectricity of 0–3 cement based composite with nano-PZT powder, Current Applied Physics 33 (2009) 588–591.