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Influence of barium titanate content and particle size on electromechanical coupling coefficient of lead-free piezoelectric ceramic-Portland cement composites

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Abstract

The 0–3 lead-free piezoelectric ceramic-Portland cement composites were prepared by mixing and pressing the Portland cement (PC) and barium titanate (BT) ceramic powder. The influences of BT particle size and BT content on the electromechanical coupling coefficient (K_t) of the composites were investigated. The results indicate that the particle size of BT used to produce the composite under the conditions of the same BT content (at 50% BT) and fabrication technique has an influence on the K_t values. The electromechanical coupling coefficient was found to increase with the particle size of BT used where the values of K_t are found to be at 10.8% and 14.1% for composites with median particle size of 75 μm and 425 μm, respectively. Furthermore, K_t of composites increase with increasing content of BT (at the same particle size of 425 μm) when the content of BT reaches 70%, K_t is 16.6%. In addition, the acoustic impedance of the composite also increase with an increase of BT content.

Keywords: B. Composites; C. Electrical properties; C. Piezoelectric properties; D. $BaTiO_3$ and titanates

1. Introduction

Piezoelectric–cement based composite is effective and applicable both in piezoelectric properties and compatibility (similar acoustic impedance as that of concrete, $\approx 9 \times 10^6\ \mathrm{kg\,m^{-2}\,s^{-1}}$ [1]), and it can be utilized to fabricate piezoelectric–cement based sensors for applications in concrete structures [1]. Therefore, piezoelectric–cement based composites have been widely studied and developed for use as smart or intelligent structure [1–7].

Lead-based piezoelectric material is one of the most widely used materials as active piezoelectric phase to fabricate composites of high piezoelectric coefficient and high electromechanical coupling [8]. However, the toxic lead oxide from processing of lead-based piezoelectric materials vaporizes during processing, and it remains in the environment for a long time. This toxic material accumulates in the organisms, causing damage to the brain and nervous system [9]. To solve the environmental problem caused by lead oxide in lead-based piezoelectric ceramics, it is necessary to replace the lead-based piezoelectric ceramics in the composites. One of the superior candidates for lead-free piezoelectric ceramics is barium titanate $BaTiO_3$ (BT) where it has a relatively high electromechanical coupling factor and has been used for piezoelectric applications [10].

In our previous study [11], we investigated the effect of the BT concentration on the dielectric, ferroelectric and piezoelectric properties of 0–3 barium titanate–Portland cement composites. The results of this study indicate that all the properties were improved when the concentration of BT was increased. Moreover, the properties of cement based piezoelectric composites were found to depend on many factors, such as content of piezoelectric ceramic, particle size of piezoelectric ceramic, method of fabrication, poling temperature and poling time [1–7]. The piezoelectric properties of cement based piezoelectric composites are mainly due to the piezoelectric ceramic, moreover the content and particle size of piezoelectric ceramic have great influence the piezoelectric properties of composites.

In this work, the influence of BT content and BT particle size on the electromechanical coupling coefficient and acoustic

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impedance of lead-free piezoelectric cement-Portland composites were studied in order to obtain the optimum value of the composites properties.

2. Experimental

Barium Titanate, $BaTiO_3$ (BT), ceramic was produced from the mixed oxide method using barium carbonate and titanium oxide as the starting precursors followed by calcination at 1200 °C, and the BT powder was sintered at 1400 °C. In order to study the effect of particle size on the electrical properties of composite, BT ceramics were ground and sieved to various median particle sizes (75 μm, 212 μm and 425 μm) and the BT content was kept constant at 50%. For the effect of BT content, BT ceramic particles of 425 μm size were used at 30, 40, 50, 60 and 70 vol%. BT ceramics of the different size and volume were mixed with normal Portland cement, PC (American Society for Testing and Materials, type I cement) to produce 0–3 connectivity BT–PC composites. The mixed composition was axially pressed to form disk samples of ~15 mm in diameter and ~1.5 mm in thickness.

Thereafter, the composites were put in a curing chamber with 98% relative humidity at 60 °C for 3 days. The density and porosity of the composites was measured using the Archimedes method. The measured densities of the composite samples with BT particle size of 75 μm, 212 μm and 425 μm were 3.60×10^3 kg/m^3, 3.72×10^3 kg/m^3 and 3.83×10^3 kg/m^3, respectively; while the porosities of the same composite samples were 10.52%, 10.16% and 9.22%, respectively. Moreover, the densities of the composite samples, by volume percentage (vol%) of BT: 30%, 40%, 50%, 60% and 70%, were 3.22×10^3 kg/m^3, 3.51×10^3 kg/m^3, 3.83×10^3 kg/m^3, 4.12×10^3 kg/m^3 and 4.40×10^3 kg/m^3, respectively; while the porosities of the same composite samples by vol% of BT were 8.52%, 8.77%, 9.22%, 9.25% and 9.30%, respectively.

After drying the composites, silver paste was coated onto both sides of the discs as electrodes. The composites were poled for piezoelectric measurement at 60 °C in silicone oil bath with polarization voltage 1 kV/mm for ~45 min. An impedance meter (Hewlett Packard 4194A) was then used to obtain the electric impedance and the phase of the composites. However, it should be note that it would become very difficult to pole samples with 30% vol% BT, because of the higher non-piezoelectric cement matrix content in the composite. The thickness electromechanical coupling coefficient (K_t) was calculated from a plot of electric impedance against frequency using the following Eq. (1) [12].

$$K_t^2 = \frac{\pi f_m}{2 f_n} \tan\left(\frac{\pi}{2}\frac{f_n - f_m}{f_n}\right) \tag{1}$$

where f_m and f_n are the frequencies at minimum and maximum electric impedance, respectively.

The velocities (V_c) were measured by a TM-8812 ultrasonic thickness meter. The experimental values of the acoustic impedance (Z_c) were obtained by multiplying the densities (ρ_c) with the velocities. The acoustic impedance (Z_c) equation can be written as [1]

$$Z_c = \rho_c V_c \tag{2}$$

The velocity (V_c), is related to the mechanical properties of the medium through the expression

$$V_c = \left(\frac{E_c}{\rho_c}\right)^{0.5} \tag{3}$$

where E is the elastic bulk modulus of the composite and ρ_c is the density [1].

The density of composites can be expressed as:

$$\rho_c = v_1 \rho_1 + v_2 \rho_2 \tag{4}$$

where v_1 and v_2 are the volume percentages of the ceramic phase and the cement phase, respectively, and ρ_1 and ρ_2 are the density of the ceramic phase and the cement phase, respectively [1]. The elastic modulus of the composites E_c can be written as Eq. (5) by using a series model

$$E_c = \frac{1}{(v_1/E_1) + (v_2/E_2)} \tag{5}$$

where E_1 and E_2 are elastic moduli of the ceramics phase and the cement phase, respectively [1].

3. Results and discussion

3.1. Influence of BT particle size on the composites properties

3.1.1. Influence of BT particle size on the electromechanical properties

The impedance magnitude and phase spectra of composites with difference frequency under different BT particle size can be seen in Fig. 1(a–c). It can be seen that the impedance value of the composites became lower when the composite was made using the larger BT particle size. Moreover, the peaks spectra of composites with larger BT particle size are higher than those of composites with smaller BT particles, which mean that larger particle size brings a higher piezoelectric effect to the composites [1]. It may be due to the greater overall contact surface area between the cement matrix and the BT particles, of the same volume content, when BT particle size is smaller therefore would decrease the poling efficiency and weaken the bonding between the two phases of composites [5–7].

Moreover, the calculated values of K_t for the composites can be seen in Fig. 1(d). It is seen that the K_t increased with increasing particle size of BT used. When the median particle size of composites are 75 μm and 425 μm, K_t are 10.8% and 14.1%, respectively.

3.1.2. Influence of BT particle size on the acoustic impedance

The acoustic impedance (Z_c) and the porosity of the composites with different BT particle size are presented in Fig. 2. It can be seen that the values of Z_c of the composite

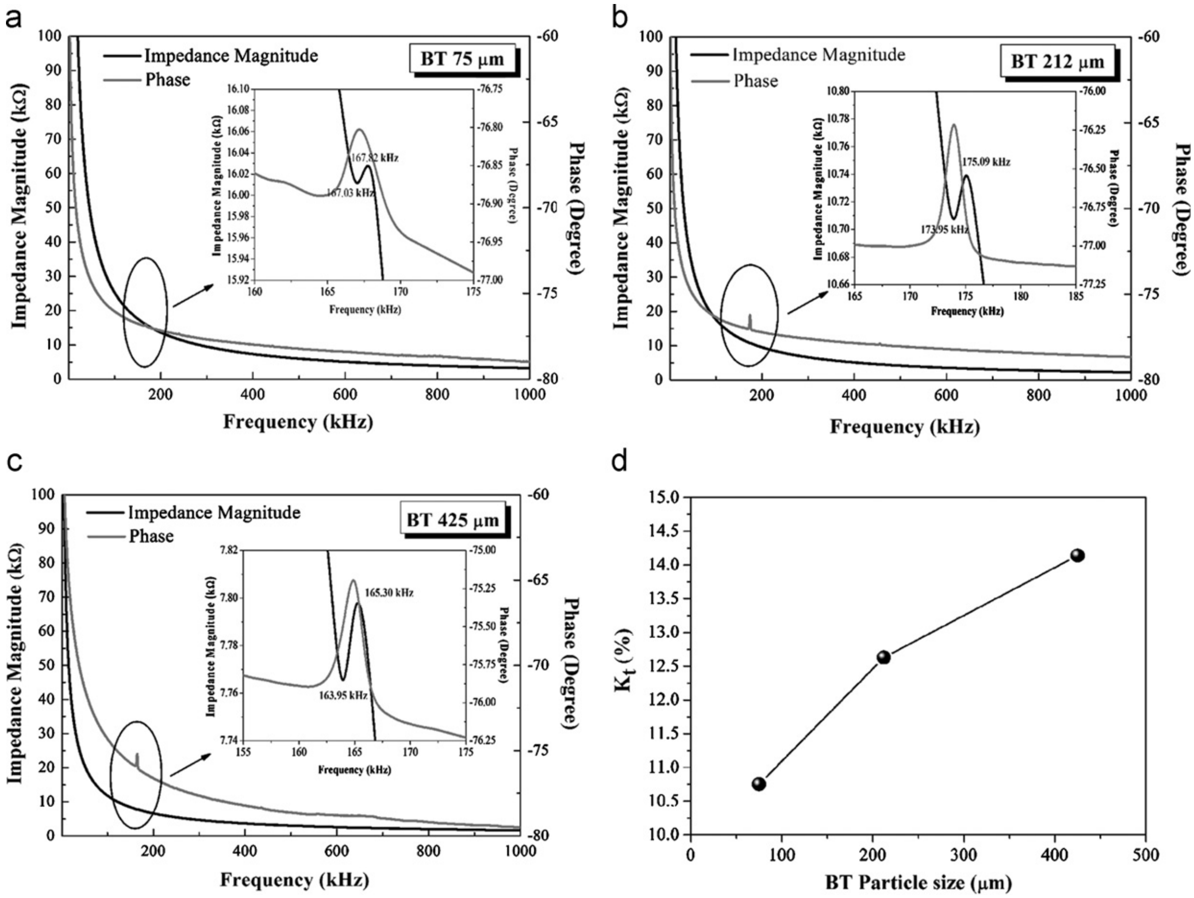

Fig. 1. Impedance spectra versus frequency of the 0–3 BT–PC composites at different BT median particle size of (a) 75 μm, (b) 212 μm, (c) 425 μm and (d) K_t versus particle size at the BT content of 50%.

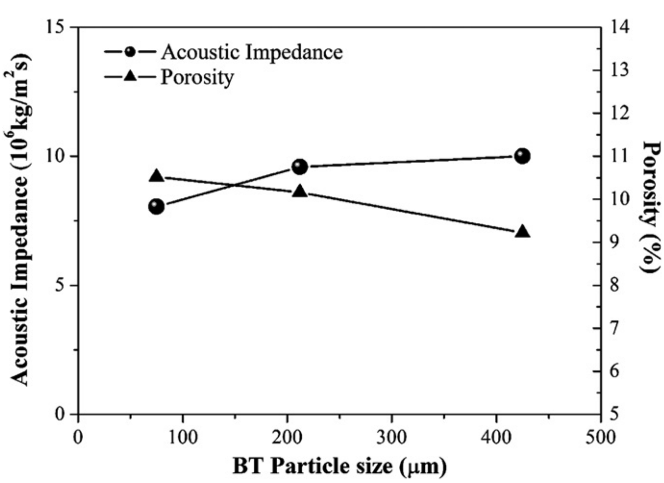

Fig. 2. Effect of BT particle size on the acoustic impedance and porosity of 0–3 BT–PC composites.

increase with increasing BT particle size. The reason for the reduction in Z_c with smaller particle size was the presence of possible defects such as porosity in the different composites. Thus, smaller particle size will result in an overall increase in the particle contact surface areas at the same volume. At the particle interface, part of wave will be transmitted and part of wave will be reflected [13].

3.2. Influence of BT ceramic content on the composites

3.2.1. Influence of BT ceramic content on the electromechanical properties

Fig. 3 shows the variation of impedance and phase with frequency under different BT content. As can be seen, the impedance of the composites decreases as the BT content increase. Moreover, the BT content has also influenced the resonance and anti-resonance frequency. Furthermore, the phase spectra of the composites show that the peaks increase with the increase in BT content. This peak indicates that the addition of BT induces an electromechanical coupling to the composites, caused by the piezoelectric effect and reverse piezoelectric effect [12].

The calculated electromechanical coupling coefficients of the composites are shown in Fig. 4. It can be seen that the values of K_t for all the composites range from 10.6% to 16.6%. Furthermore, it is observed that the value of K_t increases with increasing BT content, which means that an efficient conversion between electrical and mechanical energy is remarkably influenced by the BT content.

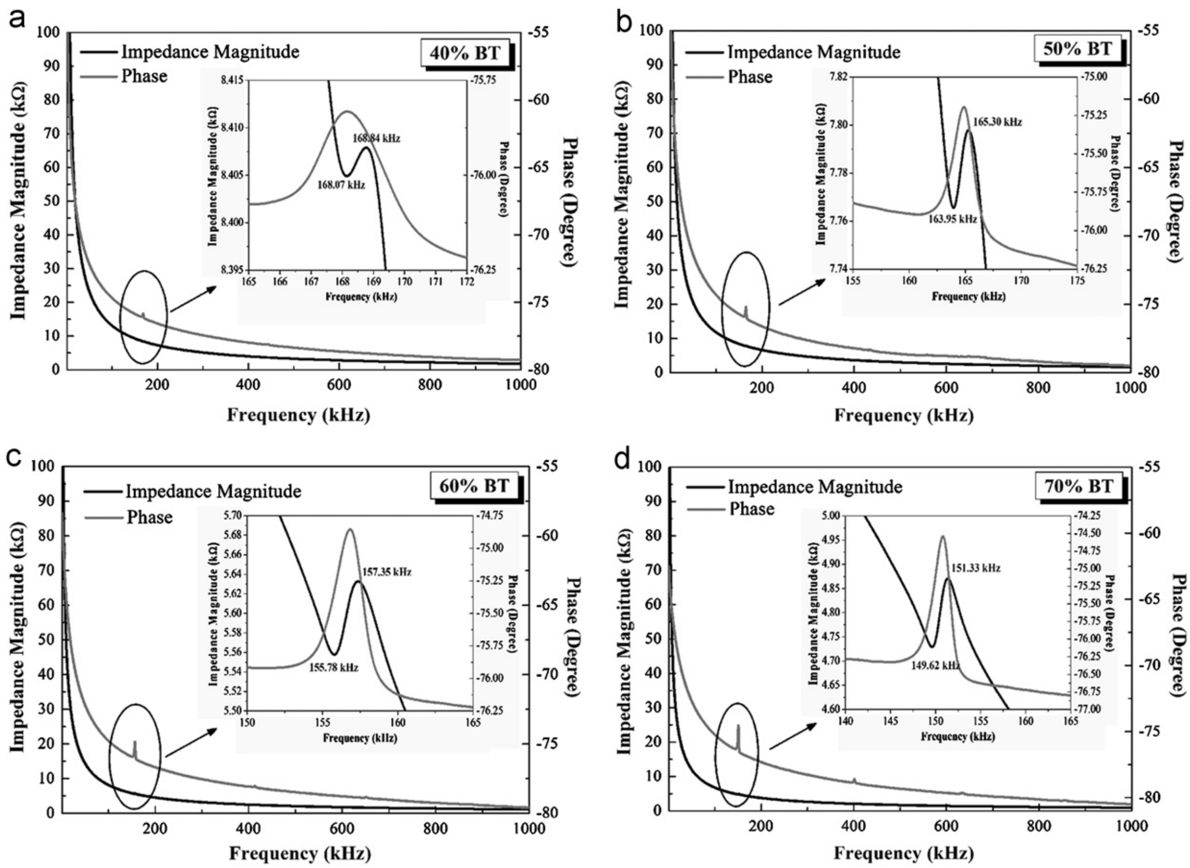

Fig. 3. Impedance spectra of the 0–3 BT–PC composites with frequency at different BT content of (a) 40 vol%, (b) 50 vol%, (c) 60 vol% and (d) 70 vol%.

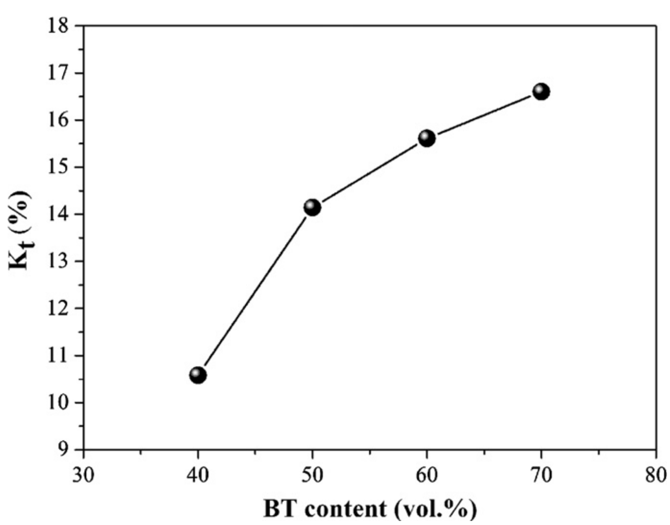

Fig. 4. Effect of BT content on the K_t values of 0–3 BT–PC composites.

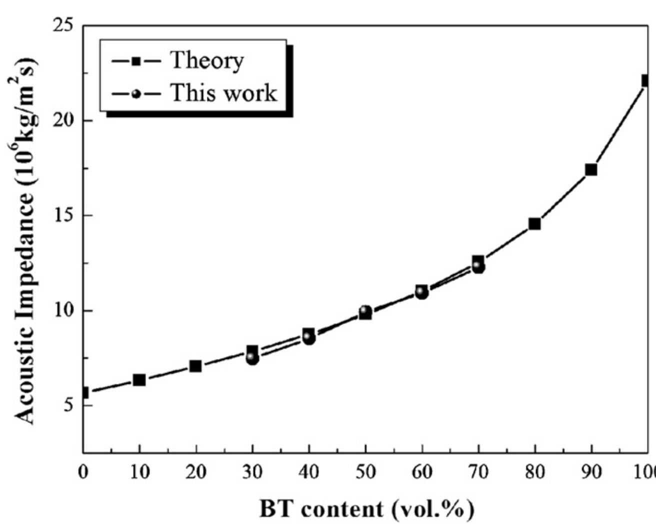

Fig. 5. Effect of BT content on the acoustic impedance of 0–3 BT–PC composites.

3.2.2. Influence of BT ceramic content on the acoustic impedance

Fig. 5 shows the acoustic impedance (Z_c) of the composites with different BT content, together with the series theory. The Z_c values of composites were obtained by multiplying the density (ρ_c) with the velocity (V_c). It can be seen that the acoustic impedance of the composite exhibits the increasing trend with increasing BT content. The composite with ∼40–60% of BT ceramic particles has an acoustic impedance close to that of concrete. According to this, the BT ceramic content in the composites should be adjusted to a proper proportion to match with the acoustic impedance of the concrete (9×10^6 kg m^{-2} s^{-1}).

4. Conclusions

The 0–3 lead-free cement based piezoelectric composites from BT and Portland cement were successfully fabricated. The results showed that the particle size of BT has an influence on the electromechanical coupling coefficient (K_t). In addition, the electromechanical coupling properties can be improved by increasing the BT content in composite. Furthermore, the acoustic impedance of the composite can also be changed to match the host structure material, i.e., concrete. According to the acoustic impedance from our experimental data, 40–60 vol% is the optimal BT content for an acoustic impedance match between lead-free cement based piezoelectric composites and concrete.

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References

[1] Z. Li, D. Zhang, K. Wu, Cement-based 0–3 piezoelectric composites, Journal of the American Ceramic Society 85 (2002) 305–313.
[2] B. Dong, Z. Li, Cement-based piezoelectric ceramic smart composites, Composites Science and Technology 65 (2005) 1363–1371.
[3] N. Jaitanong, A. Chaipanich, T. Tunkasiri, Properties 0-3 PZT-Portland cement composites, Ceramics International 34 (2008) 793–795.
[4] A. Chaipanich, N. Jaitanong, R. Yimnirun, Effect of carbon addition on the ferroelectric hysteresis properties of lead zirconate–titanate ceramic–cement composites, Ceramics International 37 (2011) 1181–1184.
[5] S.F. Huang, L. Lingchao, C. Jun, X. Dongyu, L. Futian, C. Xin, Influence of ceramic particle size on piezoelectric properties of cement-based piezoelectric composites, Ferroelectrics 332 (2006) 187–194.
[6] A. Chaipanich, N. Jaitanong, Effect of PZT particle size on the electromechanical coupling coefficient of 0–3 PZT-cement composites, Ferroelectrics Letters 36 (2009) 37–44.
[7] Z. Li, H. Gonga, Effects of particle size on the piezoelectric properties of 0–3 PZT/cement composites. In: AIP Conference Proceedings on Multiscale and Functionally Graded Materials, 2006, Oahu Island, Hawaii, 973 (2008), pp. 538–543.
[8] S. Schwarzer, A. Roosen, Tape casting of piezo ceramic/polymer composites, Journal of the European Ceramic Society 19 (1999) 1007–1010.
[9] T. Takenaka, H. Nagata, Current status and prospects of lead-free piezoelectric ceramics, Journal of the European Ceramic Society 25 (2005) 2693–2700.
[10] E. Aksel, J.L. Jones, Advances in lead-free piezoelectric materials for sensors and actuators, Sensors 10 (2010) 1935–1954.
[11] R. Rianyoi, R. Potong, N. Jaitanong, R. Yimnirun, A. Chaipanich, Dielectric, ferroelectric and piezoelectric properties of 0–3 barium titanate–Portland cement composites, Applied Physics A 104 (2011) 661–666.
[12] S.F. Huang, J. Chang, F.T. Liu, L.C. Lu, Z.M. Ye, X. Cheng, Poling process and piezoelectric properties of lead zirconate titanate/sulphoaluminate cement composites, Journal of Materials Science 39 (2004) 6975–6979.
[13] M.G. Grewe, T.R. Gururaja, Thomas R. Shrout, Robert E. Newnham, Acoustic Properties of particle/polymer composites for ultrasonic transducer backing applications, IEEE Transactions on Ultrasonics, Ferroelectrics and Frequency Control 37 (1990) 506–514.