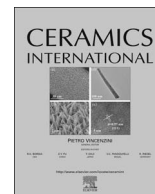




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## Thermal expansion behaviors of 0–3 connectivity lead-free barium zirconate titanate-Portland cement composites

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### ABSTRACT

New study on thermal expansion properties of 0–3 connectivity lead-free barium zirconate titanate (BZT:5%Zr)-Portland cement composites were determined in the temperature range of approximately –100 to +250 °C by dilatometer thermal expansion measurement. All the composites were found to have the three phase transitions of BZT ceramic in the cooling cycle. The physical and mechanical properties of hydration product in cement matrix were changed at elevated temperatures and the results were concluded from the thermal expansion data. When compared to the concrete structure, the thermal expansion coefficient value of the 0–3 composites with ≈ 30–70% of barium zirconate titanate ceramic content can be matched with concrete. The theoretical equations of models were applied for the calculation with the thermal expansion coefficient and the results were found to fit closest to that of the Dougill's model.

### 1. Introduction

Composites of piezoelectric ceramic and cement have been developed to have smart or intelligent properties so that the composites can behave as a sensor for smart concrete structures [1–5]. The main advantage of these composites over other piezoelectric ceramic and polymer-based piezoelectric composites is the acoustic signal matching of the composite with the concrete structure ( $\approx 6.90\text{--}11.23 \times 10^6$  kg/m<sup>2</sup>s) [5–7]. The mismatching would thus lead to a loss transmission of the signal between the two medium [1–5]. Li et al. [1] showed that the acoustic impedance of the 0–3 composite with 40–50% lead zirconate titanate (PZT) and cement can match that of concrete, as opposed to the pure PZT [1].

Lead-based piezoelectric ceramics such as lead zirconate titanate (PZT) and lead niobium lithium zirconate titanate (P(LN)ZT) has been used with cement to fabricate piezoelectric cement based composites [1–5], but the use of lead-based piezoelectric ceramics has caused serious environmental concerns because of the toxicity of lead oxide and its high vapor pressure during sintering [8–12]. Barium zirconate titanate (BaTi<sub>1-x</sub>Zr<sub>x</sub>O<sub>3</sub>; BZT) ceramics are attractive candidates for lead-free piezoelectric ceramics and these ceramics show the promising

potentials of piezoelectric/electrostrictive properties [8–15]. BaTi<sub>1-x</sub>Zr<sub>x</sub>O<sub>3</sub> ceramics with  $x = 0.05$  have the advantages of good piezoelectric properties, low temperature poling and low Curie temperature [11]. These properties are good for piezoelectric-cement based composites and it allows easy poling at low temperature. However, the ideal piezoelectric-cement based composite is not only due to the high piezoelectric characteristic of the ceramics, but it also depends on many factors such as good compatibility to concrete, environmental friendly chemistry, tendency of composite to change in volume in response to a change in temperature. Concrete is the most commonly used in building materials around the world and today concrete is increasingly being used [16]. Some factors during the hydration of cement such as variations in relative humidity or temperature and chemical reactions, will cause shrinkage or expansion of concrete. Under the temperature cycle, the physical and mechanical properties of cement may be damaged during the service life [17–20]. Moreover, the temperature and pressure condition can affect the chemistry of hydration product of cement phase [21]. The thermal expansion coefficient is a property which can be of considerable importance in mechanical and structural design applications of a material [22]. Therefore, thermal expansion of 0–3 barium zirconate titanate-Portland cement composites has been

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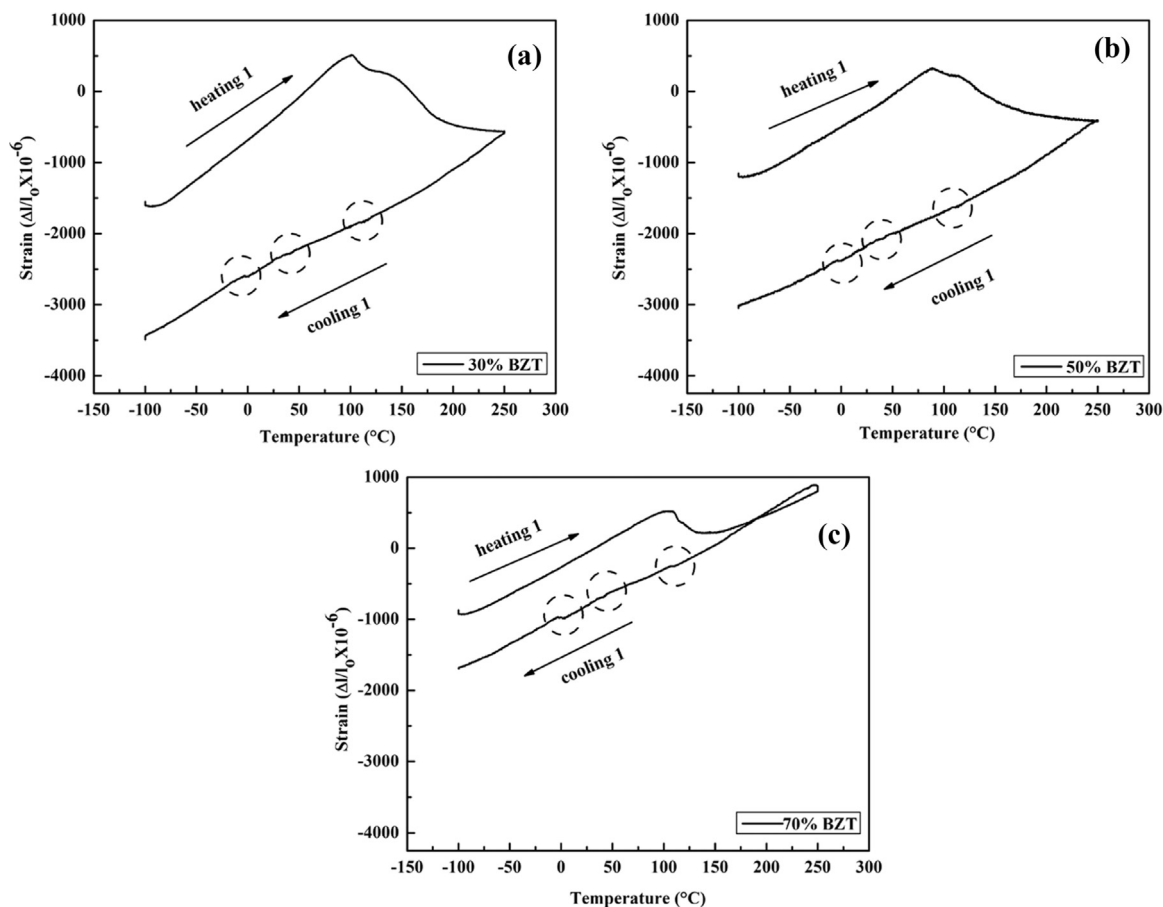


Fig. 1. Thermal expansion as a function of temperature for 0–3 BZT-PC composites for first run; (a) 30% BZT composite, (b) 50% BZT composite and (c) 70% BZT composite.

studied and the transition temperatures of composites system are presented in this paper for the first time.

## 2. Materials and methods

### 2.1. Fabrication of 0–3 connectivity lead free piezoelectric composites

Barium zirconate titanate,  $\text{BaZr}_{0.05}\text{Ti}_{0.95}\text{O}_3$  (BZT), ceramics were prepared using the conventional ceramic fabrication technique. Sintering was carried out at  $\approx 1450^\circ\text{C}$  after the calcination of BZT powder prepared at  $\approx 1200^\circ\text{C}$ . BZT ceramic particles of different BZT volume content of 30%, 50% and 70% and  $425\ \mu\text{m}$  BZT particle size were then mixed with normal Portland cement (PC) to produce 0–3 connectivity composites. The composites were pressed into the model disks using hydraulic press. Thereafter, the composites were put in the curing chamber for 3 days in a controlled  $60^\circ\text{C}$ , 98% relative humidity (RH) chamber.

### 2.2. Testing methods

The thermal expansion of all samples was examined with the use of a linear voltage-differential transformer (LVDT) dilatometer. Bar shape samples of  $\approx 6\ \text{mm}$  long and  $\approx 2\ \text{mm}$  cross-section were placed inside a fused silica holder. They were subjected to the temperature range of  $-100$  to  $250^\circ\text{C}$  at a rate of  $2^\circ\text{C}/\text{min}$  for the heating and cooling. Moreover, the LVDT was used as it able to give a linear output for every unit displacement.

## 3. Results and discussion

### 3.1. The thermal strain

The thermal strain dependences of temperature of 0–3 BZT-PC composites with different BZT content are presented in Fig. 1. The result shows measurement of composites as temperature was increased from  $-100^\circ\text{C}$  to  $250^\circ\text{C}$  at a rate of  $2^\circ\text{C}/\text{min}$ . For the first heating run, from  $-100^\circ\text{C}$  to  $\approx 100^\circ\text{C}$ , the composite expand and then it contracted for the remainder of the temperature range ( $\approx 100^\circ\text{C}$  to  $250^\circ\text{C}$ ). The shrinkage at elevated temperatures has been attributed to a loss of moisture from cement matrix. Many forms of water present in cement matrix at about room temperature such as the water content in a free state (pores and voids) and also bound water in hydration products of cement paste influence the paste strength. A rise in temperature induces the first drive out of the free water from pores and the dehydration would occur in cement matrix if severe enough [21]. Hydration products of cement such as calcium silicate hydrate and calcium hydroxide are known to dehydroxylate at  $\approx 105$ – $120^\circ\text{C}$  and  $\approx 450$ – $480^\circ\text{C}$ , respectively [23–25]. This agrees with the report by Janotka and Nürnberggerov [26] where the loss of bound water in cement paste from the decomposition of the C-S-H is markedly influenced by the temperature at  $100$ – $200^\circ\text{C}$ . Moreover, increasing temperatures also affect the reaction in cement matrix [27] where the loss of evaporable water and bound water occurs at  $30$ – $105^\circ\text{C}$ . At  $120^\circ\text{C}$ , the evaporable water occurs in cement paste is fully eliminated [28]. Ettringite becomes unstable and dehydrate at the temperature range between  $60^\circ\text{C}$  and  $80^\circ\text{C}$  [23–25,28].

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