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Ferroelectric hysteresis behavior and dielectric properties of 1–3 lead zirconate titanate–cement composites

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Abstract

Lead zirconate titanate (PZT) ceramic was mixed with Portland cement (PC) to form 1–3 connectivity PZT–PC composite using a dice-and-fill technique. Ferroelectric hysteresis behavior and dielectric properties of these composites were investigated using PZT volume content of 60%, 70% and 80%. The results showed that the dielectric constant of the composite materials increased with PZT content and the dielectric constant (ε_r) value is 781 for 80% PZT composite at 1 kHz. The dielectric loss tangent (tan δ) was found to decrease with increasing PZT content and the tan δ value of 80% PZT composite is 0.06. Parallel and series models were also compared to the dielectric measurement results. For the hysteresis measurements, the ferroelectric hysteresis loops can be seen for all composites. The ''instantaneous'' remnant polarization (P_{ir}) was found to increase with increasing PZT content from 3.20 to 4.28 μC/cm^2 at 90 Hz when PZT volume content used was 60% and 80% respectively.

Keywords: B. Composites; C. Dielectric properties; C. Ferroelectric properties; D. PZT

1. Introduction

Lead zirconate titanate (PZT) is the most popular ferroelectric material, which plays a remarkable role in modern electroceramic industry [1]. Moreover, PZT has high dielectric constant, high electromechanical coupling and high piezoelectric coefficient and has been employed as sensors, actuators and transducers [2–6]. Very recently, there have been developments of piezoelectric materials in order to meet the acoustic matching requirement such as those of piezoelectric-cement based composites for smart concrete structures and civil engineering application [7–16]. The first report work on the use of a 0–3 PZT–white Portland cement composite was by Li et al. [14]. Chaipanich [15] investigated the dielectric and piezoelectric properties of 0–3 PZT–cement composite made from ordinary Portland cement (ASTM type I cement) and PZT. Lam and Chan [16] developed 1–3 piezoelectric ceramic/cement composites with 0.25–0.77 volume fractions of PZT were reported and it was found that the 1–3 composites have good compatibility with civil engineering structure materials while being suitable for high sensitivity. The hysteresis study would thus give an indicative of the remnant polarization and the ferroelectricity behavior of the material. In addition, the dielectric results of the composites with 1–3 connectivity were compared to the parallel, series and cubes models and are presented here in this paper.

In this research, it is therefore interesting to study the ferroelectric hysteresis behavior and dielectric properties of 1–3 connectivity PZT-cement composites.

2. Experimental

Lead zirconate titanate ($Pb(Zr_{0.52}Ti_{0.48})O_3$) powder was initially produced from the two-stage mixed oxide method. Starting precursors were lead oxide (PbO), zirconium oxide (ZrO_2) and titanium dioxide (TiO_2) (Riedel-de Haën, >99% purity). PbO and ZrO_2 were thoroughly mixed and milled in ethanol for 24 h. After that, the mixed powder was stirred, dried and calcined in a closed alumina crucible at 800 °C for 2 h to give $PbZrO_3$. $PbZrO_3$ and TiO_2 powder were then calcined at 900 °C with excess PbO to give the PZT powder. In addition,

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PZT ceramics were then produced by sintering PZT powder at 1200 °C for 2 h. 1–3 Lead zirconate titanate–Portland cement composites of ≈2 mm thick and 12 mm in diameter were fabricated using a dice-and-fill method [16,17]. PZT ceramics of different PZT volume content of 60%, 70% and 80% were used. PZT ceramic disc was cut in one direction (*y* axial) using a diamond saw (Buehler ISOMET Low speed saw) with a 0.5 mm thick blade. Portland cement (PC) was cast as paste (water/cement = 0.5) on the matrix phase. Thereafter, the samples were placed for curing at 60 °C for five days under a condition of relatively high humidity and cut in second direction (*x* axial). After filling the second direction of cuts with cement paste, the composites were cured at a temperature of 60 °C and 98% relative humidity for five days before measurements. The dielectric properties of composites were measured using an impedance meter (Hewlett Packard 4194A) at room temperature. The dielectric constant (ε_r) was then calculated from Eq. (1)

$$\varepsilon_r = \frac{Ct}{\varepsilon_0 A} \tag{1}$$

where C is the sample capacitance, t is the thickness, ε_0 is the permittivity of free space constant (8.854×10^{-12} F m^{-1}), and A is the electrode area.

The dielectric constant results were also compared with the composite models as follows:

- parallel model [18]

$$\varepsilon_C = \nu_1\varepsilon_1 + \nu_2\varepsilon_2 \tag{2}$$

- series model [19]

$$\frac{1}{\varepsilon_C} = \frac{\nu_1}{\varepsilon_1} + \frac{\nu_2}{\varepsilon_2} \tag{3}$$

- cubic model [20]

$$\varepsilon_C = \frac{\varepsilon_1 \cdot \varepsilon_2}{(\varepsilon_2 - \varepsilon_1) \cdot \nu_1^{-1/3} + \varepsilon_1 \cdot \nu_1^{-2/3}} + \varepsilon_2 \cdot (1 - \nu_1^{-2/3}) \tag{4}$$

where ε_1 and ε_2 are dielectric constant values of the ceramics phase and the cement phase, respectively, and ν_1 and ν_2 are the volume percentages of the ceramics phase and the cement phase, respectively.

The ferroelectric hyteresis (*P*–*E*) loop behaviors of the composites were measured using a computer controlled modified Sawyer–Tower circuit at room temperature.

3. Results and discussion

The dielectric constant (ε_r) of PZT–PC composites is plotted against the PZT content in Fig. 1. The dielectric constant of the composites can be seen to increase with increasing PZT content and the dielectric constant is highest at 781 for 80% PZT composites at 1 kHz. In addition, dielectric loss (tan δ) of composites showing the effect of PZT content can also be seen in Fig. 1. The dielectric loss (tan δ) is found to reduce with increasing PZT content and the tan δ value of 80% PZT composite is lowest at 0.06 (f = 1 kHz).

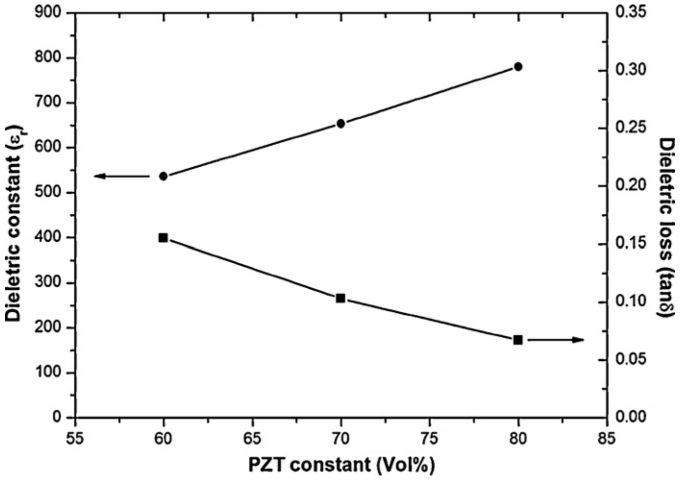

Fig. 1. Dielectric properties results of PZT–Portland cement composites at 1 kHz.

Furthermore, the parallel and series models were compared with the results taken from the measurements as shown in Fig. 2. The dielectric results of 1–3 can be seen to fit closest to that of the parallel model and the results are shown to be higher than 0–3 connectivity composites. This is because the 1–3 connectivity is more closely related to the 2–2 parallel models with the complete phase of PZT ceramic in the *Y* direction. In the case of 0–3 connectivity composites, PZT were randomly distributed particles surrounded by Portland cement matrix and thus the 0–3 composites are more closely related to the series or cube models.

Measured hysteresis loops of the PZT–PC composites are shown in Fig. 3. For comparison, we can define the *y*-axis intercept at a given applied field as the ‘‘instantaneous’’ remnant polarization (P_{ir}) and the *x*-axis intercept is called the ‘‘instantaneous’’ coercive field (E_{ic}) [10]. From Fig. 3, an

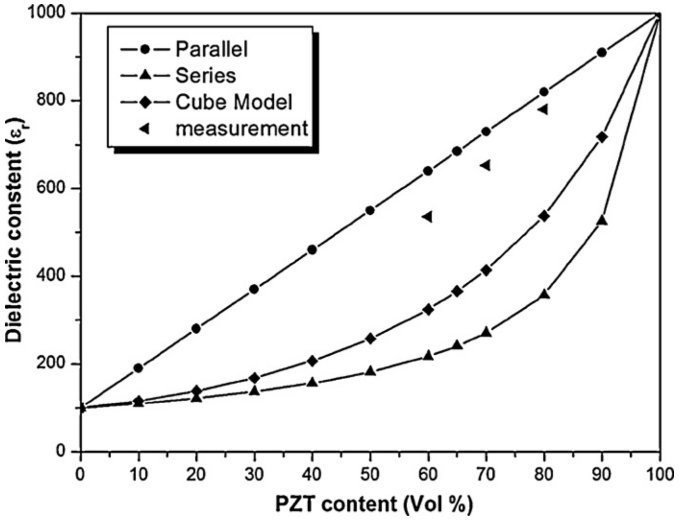

Fig. 2. Comparison of models with the dielectric results of PZT–Portland cement composites.

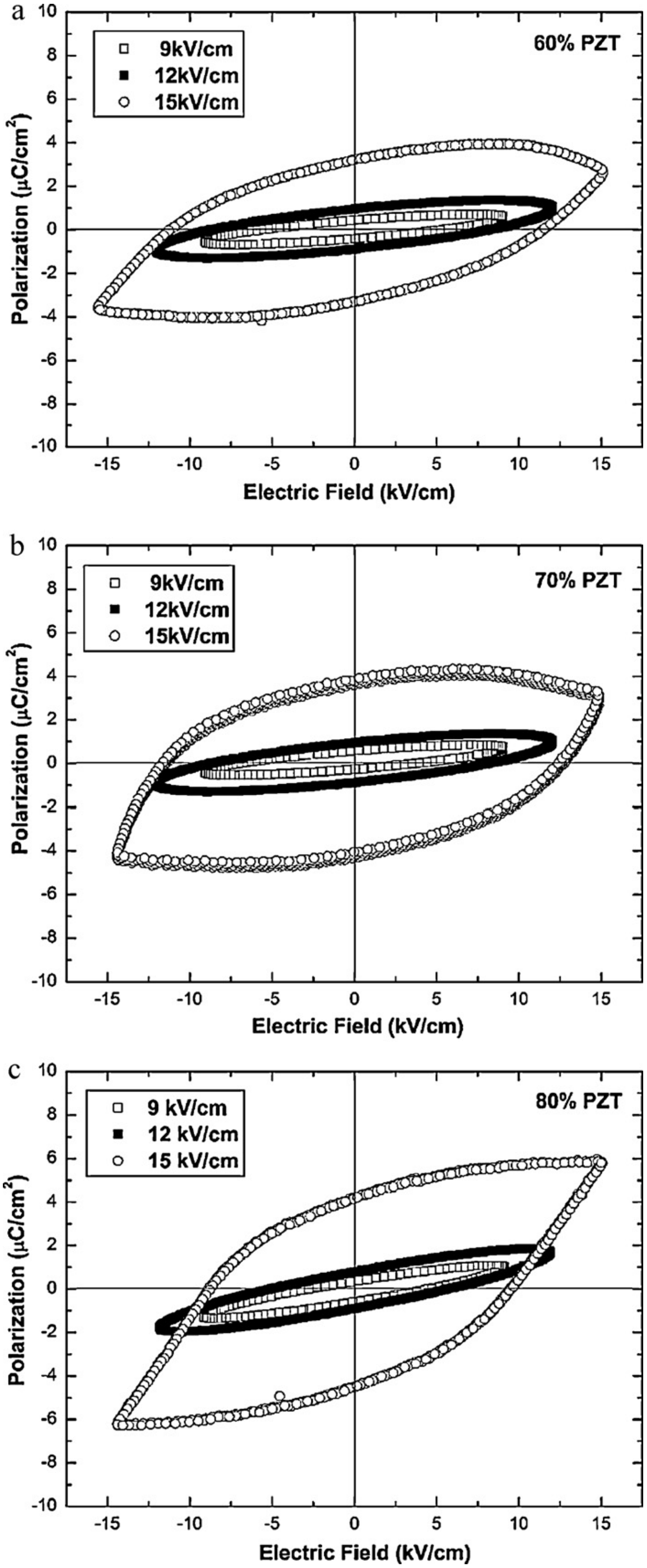

Fig. 3. Effect of external electrical field on the hysteresis loops of the 1–3 connectivity PZT–PC composites at (a) 60% PZT, (b) 70% PZT and (c) 80% PZT.

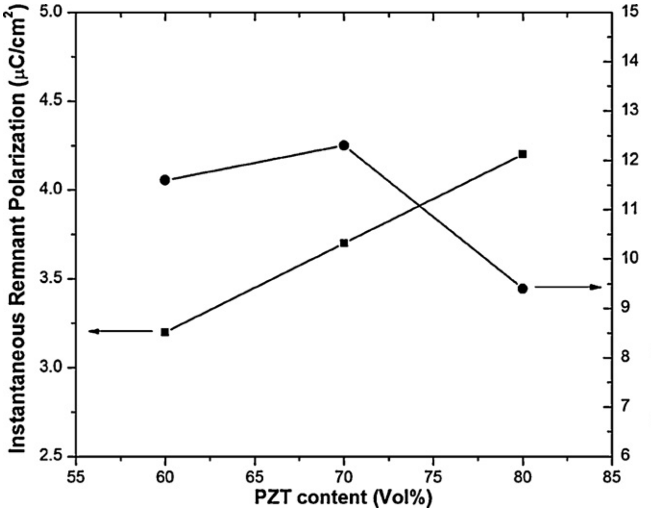

Fig. 4. Effect of PZT contents on instantaneous remnant polarization (P_{ir}) and instantaneous coercive field (E_{ic}) of PZT–Portland cement composites.

increase in the amplitude of the external electrical field make the P–E loops larger and increases its angle to the E axis of inclination [12]. At frequency of 90 Hz, the P_{ir} value can be seen to increase from 0.46 to 3.20 $\mu C/cm^2$ (60% PZT), 0.59 to 3.84 $\mu C/cm^2$ (70% PZT), and 0.41 to 4.28 $\mu C/cm^2$ (80% PZT) when the electric field increases from 9 to 15 kV/cm, respectively. The E_{ic} value can be seen to increase with increasing external electrical field for all composites.

The effect of PZT contents on instantaneous remnant polarization (P_{ir}) and instantaneous coercive field (E_{ic}) of PZT–Portland cement composites is shown in Fig. 4. The P_{ir} value can be seen to increase with increasing PZT content from 3.20 to 4.28 $\mu C/cm^2$ at 90 Hz when PZT content increases from 60% to 80%, respectively. On the other hand, the E_{ic} value is 11.60 kV/cm for the composite with 60% PZT and 9.48 kV/cm for the composite with 80% PZT.

However, the loop shape characteristic also changes with PZT content in that the P_{ir} value of 60% PZT is far lower than that of 80% PZT, while the E_{ic} value of 60% PZT is higher than that of 80% PZT. It is due to the fact that a loss from the cement can be seen to be more sensitive at lower PZT volume content and when an external electrical field acts many weak conducting ions such as OH^-, Ca^{2+} and Al^{3+} in the cement matrix begin to migrate besides the polarization of electron, these ions cause higher conducting loss in the composites [10,12,21–23]. Moreover, cement matrix is non piezoelectric and numerous defect in the composite, such as lattice distortion, and phase boundary and pores of cement matrix, which usually cause a leakage current and distortion of hysteresis loops [10,12,21,22].

4. Conclusions

The dielectric and hysteresis properties of 1–3 PZT–Portland cement composites were investigated. The results obtained from the measurements show the dielectric constant of the composites increased with increasing PZT content and that the dielectric constant was highest at 781 for 80% PZT composites at 1 kHz. The dielectric results of 1–3 were found to fit closest to that of the parallel model and the results are shown to be higher than 0–3 connectivity composites. This is because the 1–3 connectivity is more closely related to the 2–2 parallel models with the complete phase of PZT ceramic in the *Y* direction. Dielectric loss (tan δ) was found to reduce with increasing PZT content and the tan δ value of 80% PZT composites was lowest at 0.06 for composite at 1 kHz. The ''instantaneous'' remnant polarization (P_{ir}) was found to increase with increasing PZT content. The hysteresis loop can be seen for all composites.

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