

2014

Design, Fabrication, and Properties of 2-2 Connectivity Cement/ Polymer based Piezoelectric Composites with Varied Piezoelectric Phase Distribution

Xu Dongyu

Cheng Xin

Sourav Banerjee

University of South Carolina, United States, banerjes@cec.sc.edu

Huang Shifeng

Follow this and additional works at: https://scholarcommons.sc.edu/emec_facpub



Part of the [Applied Mechanics Commons](#), [Ceramic Materials Commons](#), and the [Electro-Mechanical Systems Commons](#)

Publication Info

Published in *Journal of Applied Physics*, Volume 116, Issue 24, 2014, pages #244103-.

This Article is brought to you by the Mechanical Engineering, Department of at Scholar Commons. It has been accepted for inclusion in Faculty Publications by an authorized administrator of Scholar Commons. For more information, please contact digres@mailbox.sc.edu.

Design, fabrication, and properties of 2-2 connectivity cement/polymer based piezoelectric composites with varied piezoelectric phase distribution

Xu Dongyu, Cheng Xin, Sourav Banerjee, and Huang Shifeng

Citation: [Journal of Applied Physics](#) **116**, 244103 (2014); doi: 10.1063/1.4904931

View online: <http://dx.doi.org/10.1063/1.4904931>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/jap/116/24?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Dielectrophoretically structured piezoelectric composites with high aspect ratio piezoelectric particles inclusions](#)
J. Appl. Phys. **111**, 124107 (2012); 10.1063/1.4729814

[Preparation and electrical properties of fine-scale 1–3 lead zirconic titanate/epoxy composite thick films for high-frequency ultrasonic transducers](#)
J. Appl. Phys. **103**, 084119 (2008); 10.1063/1.2903456

[Piezoelectric, dielectric, and ferroelectric properties of 0–3 ceramic/cement composites](#)
J. Appl. Phys. **101**, 094110 (2007); 10.1063/1.2730559

[Coupled magnetic–electric properties and critical behavior in multiferroic particulate composites](#)
J. Appl. Phys. **94**, 5930 (2003); 10.1063/1.1614866

[Piezoelectric properties of 3-X periodic Pb \(Zr x Ti 1-x \) O 3 –polymer composites](#)
J. Appl. Phys. **92**, 6119 (2002); 10.1063/1.1513202

A promotional banner for the Journal of Applied Physics. It features the AIP logo and the journal title at the top. Below this, the text 'Meet The New Deputy Editors' is centered. At the bottom, there are three circular headshots of the new deputy editors, each with their name written to the right: Christian Brosseau, Laurie McNeil, and Simon Phillpot. The background is a vibrant orange with a pattern of small, colorful dots.

Design, fabrication, and properties of 2-2 connectivity cement/polymer based piezoelectric composites with varied piezoelectric phase distribution

Xu Dongyu,^{1,2} Cheng Xin,¹ Sourav Banerjee,² and Huang Shifeng¹

¹Shandong Provincial Key Laboratory of Construction Materials Preparation and Measurement, School of Materials Science and Engineering, University of Jinan, Jinan, Shandong 250022, China

²Department of Mechanical Engineering, University of South Carolina, Columbia, South Carolina 29208, USA

(Received 26 October 2014; accepted 10 December 2014; published online 23 December 2014)

The laminated 2-2 connectivity cement/polymer based piezoelectric composites with varied piezoelectric phase distribution were fabricated by employing Lead Zirconium Titanate ceramic as active phase, and mixture of cement powder, epoxy resin, and hardener as matrix phase with a mass proportion of 4:4:1. The dielectric, piezoelectric, and electromechanical coupling properties of the composites were studied. The composites with large total volume fraction of piezoelectric phase have large piezoelectric strain constant and relative permittivity, and the piezoelectric and dielectric properties of the composites are independent of the dimensional variations of the piezoelectric ceramic layer. The composites with small total volume fraction of piezoelectric phase have large piezoelectric voltage constant, but also large dielectric loss. The composite with gradually increased dimension of piezoelectric ceramic layer has the smallest dielectric loss, and that with the gradually increased dimension of matrix layer has the largest piezoelectric voltage constant. The novel piezoelectric composites show potential applications in fabricating ultrasonic transducers with varied surface vibration amplitude of the transducer. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4904931>]

I. INTRODUCTION

Presently, piezoelectric ceramic is being widely used in many fields due to its characteristics of simple structures, fast response speed, and good sensing/actuating abilities.¹⁻⁴ However, there also exist various compatibility problems when piezoelectric ceramic is used in concrete engineering structures, such as interface and acoustic impedance mismatching between piezoelectric ceramic and concrete material, long-term durability, and reliability problems, which accordingly restrict the further application of piezoelectric ceramic in civil engineering field.

In order to solve these problems, cement based piezoelectric composite consisting of piezoelectric ceramic and cement paste matrix was developed. In 2002, Li *et al.* fabricated a 0-3 connectivity cement based piezoelectric composite by using Lead Zirconium Titanate (PZT) piezoelectric ceramic powder as active phase and cement paste as passive matrix phase.⁵ Their research showed that acoustic impedance of this piezoelectric composite could match with that of concrete material by adjusting the proportion of cement and piezoelectric ceramic powder. In the following years, many scholars showed their interest in the 0-3 connectivity cement based piezoelectric composite.⁶⁻⁸ The fabrication technique of the 0-3 connectivity cement based piezoelectric composite is simple, however, the piezoelectric properties of this composite are usually very weak. Therefore, cement based piezoelectric composites with other connectivity patterns have recently been developed based on the connectivity of bi-phase piezoelectric composite.⁹ The laminated 2-2 connectivity cement based piezoelectric composites, where the first number denotes the connectivity of the ceramics phase and the second refers to that of the matrix phase, are

especially concerned due to the simple architecture and superior sensing and actuating abilities.¹⁰⁻¹² In 2005, Dong *et al.* fabricated a 2-2 cement based piezoelectric composite by using arranging-casting method and demonstrated its sensing and actuating capability as a self-sensing actuator of civil engineering structures. In 2009, Xu *et al.* fabricated a 2-2 cement based piezoelectric composite by employing dicing-filling technique and studied the influences of piezoelectric ceramic volume fraction on composite properties.¹⁰ In 2010, Han *et al.* presented the exact static analysis of 2-2 cement based piezoelectric composites based on the theory of piezo-elasticity.¹¹ In 2012, Potong *et al.* investigated the fabrication technique and properties of 2-2 cement based piezoelectric composite with Portland cement paste as matrix phase and lead-free barium zirconate titanate as active phase.¹² Although the 2-2 connectivity cement based piezoelectric composites with excellent properties have been reported, there still exists an obvious shortcoming for this composite, namely, the poor interfacial bonding between cement matrix layer and piezoelectric ceramic layer caused by the shrinkage effects of cement hydration. Therefore, it becomes important to enhance the interfacial bonding ability of the 2-2 cement based piezoelectric composite in order to improve its stability and reliability in practical engineering application. In addition, it is also of great significance to design the novel piezoelectric composite based on the development of piezoelectric sensor array technology, such as multiple-element piezoelectric composite,¹³ the piezoelectric composites with specific acoustic field.¹⁹

It is well known that epoxy resin has been widely used in civil engineering field to improve the mechanical and durability properties of cement mortar and concrete

materials.^{20–23} Because the epoxy resin has superior binding characteristic and low acoustic impedance value, the mixture of cement powder and epoxy resin instead of conventional cement paste was considered here as matrix phase of the piezoelectric composites. The composite is termed as 2–2 connectivity cement/polymer based piezoelectric composites in this research. The dimensions of both cement/polymer matrix layer and piezoelectric ceramic layer were designed for possible application in fabricating piezoelectric transducer without effects of edge wave or plane wave. The 2–2 cement/polymer based piezoelectric composites are also expected to have potential application in concrete engineering field such as cement hydration monitoring,^{13–15} monitoring of concrete strength development and damage behavior,^{16,17}

II. EXPERIMENTAL PROCEDURES

A. Design of the 2–2 connectivity piezoelectric composites

Usually, the 2–2 connectivity piezoelectric composite is consisted of active piezoelectric ceramic phase and passive matrix phase. As for the classical 2–2 connectivity piezoelectric composite, because the dimensions of piezoelectric ceramic and matrix layer in the composite are unchanged, piezoelectric transducers fabricated by using this composite will also have uniform surface vibration amplitude because of the uniform piezoelectric phase distribution in the composite.¹⁹ Nevertheless, a desirable local distribution of piezoelectric ceramic volume fraction can be obtained by varying dimension and arrangement of piezoelectric ceramic or matrix layer in the composites. The piezoelectric, dielectric, and electromechanical coupling properties of the composite will correspondingly present specific distribution due to the varied piezoelectric phase distribution. Therefore, the piezo-

electric transducers with specific distribution of surface vibration amplitude could also be tailored by adjusting the local distribution of piezoelectric ceramic volume fraction in the composites.

Here, six 2–2 connectivity piezoelectric composites were designed based on the dimensional variation of piezoelectric ceramic or matrix phase. The schematic diagram of the composites in two-dimensional plane (X-Y) is shown in Figure 1. The poling direction is along the composite thickness direction (i.e., Z axis direction). In Figures 1(a)–1(c), the dimensions of matrix layers (i.e., the spacing between the piezoelectric ceramic layers) remain the same, but the dimensions of piezoelectric ceramic layers are varied. Also, in Figures 1(d)–1(f), the dimensions of piezoelectric ceramic layers keep unchanged, but the dimensions of matrix layers are varied. Figure 2 shows the unit cell of piezoelectric composites in X-Y plane. Symbol l stands for the length of both piezoelectric ceramic and matrix layer, t_i and d_i represent the width of the i th layer of piezoelectric ceramic and matrix, respectively.

In this research, the 2–2 connectivity cement/polymer based piezoelectric composites with varied piezoelectric phase distribution were designed based on the following method. As for the 1# piezoelectric composite (Figure 1(b)), the width of piezoelectric ceramic layer in the composite increases based on arithmetic progression. As for the 2# piezoelectric composite in Figure 1(c), the width of the piezoelectric ceramic layer first increases according to arithmetic progression, and then repeats the variation in the composite. As for the 3# piezoelectric composite in Figure 1(d), the width of the piezoelectric ceramic layer first increases in terms of arithmetic progression, and then decreases with the same common difference. The width (x_i) of the i th layer piezoelectric ceramic in 1#, 2#, and 3# piezoelectric composites can be expressed as follows:

$$x_i = x_0 + (i - 1)a; (i = 1, 2, \dots, n; a = \text{constant}), \quad (1)$$

$$\begin{cases} x_i = x_0 + (i - 1)a; (i = 1, 2, \dots, m; a = \text{constant}) \\ x_i = x_0 + (i - m - 1)a; (i = m + 1, m + 2, \dots, 2m; a = \text{constant}) \\ \vdots \\ x_i = x_0 + [i - (p - 1)m - 1]a; (i = (p - 1)m + 1, m + 2, \dots, pm; a = \text{constant}), \end{cases} \quad (2)$$

$$\begin{cases} x_i = x_0 + (i - 1)a; \left(i = 1, 2, \dots, \frac{(n-1)}{2}; a = \text{constant}\right) \\ x_i = x_0 + (n - i)a; \left(i = \frac{(n+1)}{2}, \frac{(n+3)}{2}, \dots, n; a = \text{constant}\right), \end{cases} \quad (3)$$

where n is the number of ceramic layer, a is the common difference, and x_0 is the width of the first piezoelectric ceramic layer. m is the number of piezoelectric ceramic layer in the repeated unit cell of the 2# piezoelectric composite and p is

the repetition time of the unit cell in the 2# piezoelectric composite.

Based on the mathematical expressions, it is known that the distribution of piezoelectric ceramic volume fraction in

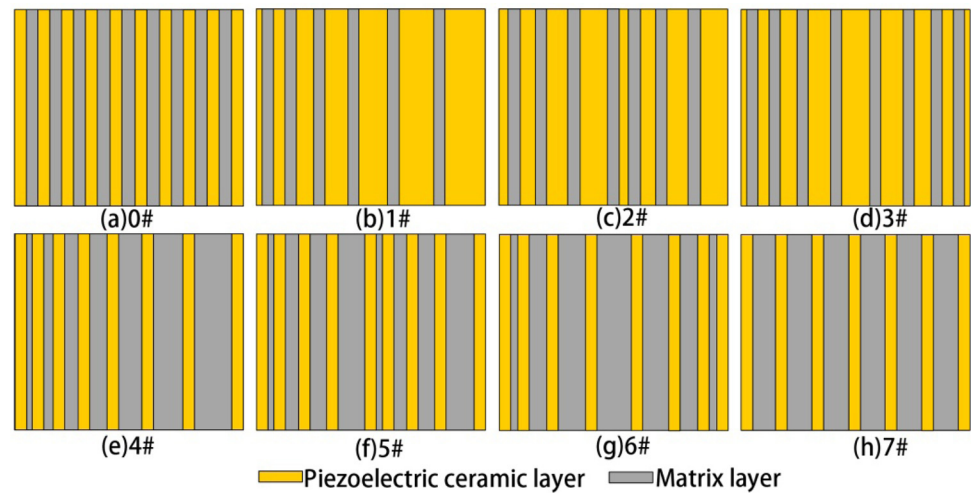


FIG. 1. Two-dimensional (in X-Y plane) schematic diagram of 2-2 connectivity piezoelectric composites with varied distribution of piezoelectric ceramic phase.

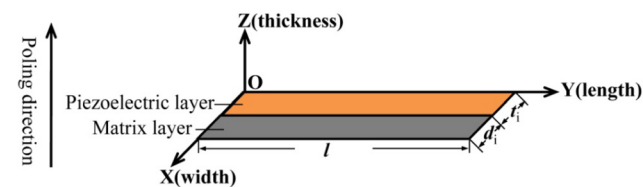


FIG. 2. Unit cell of the 2-2 connectivity piezoelectric composite.

the composite mainly depends on dimension and arrangement of piezoelectric ceramic layer. Also, the 2–2 connectivity cement/polymer based piezoelectric composites with varied piezoelectric phase distribution can also be designed by varying the dimension and arrangement of matrix layer in the composites, as shown in Figures 1(e)–1(g). The mathematical expressions, which are similar to Eqs. (1)–(3) will not be listed here again.

B. Fabrication of 2–2 connectivity cement based piezoelectric composites

The 2–2 connectivity cement/polymer based piezoelectric composites were fabricated by using dicing-filling technique.¹⁰ The polarized PZT piezoelectric ceramic was used as active phase, and mixture of epoxy resin (bisphenol A diglycidyl ether), polyamide hardener, and cement (ordinary Portland 42.5R) was used as matrix phase. The mechanical properties of the solidified epoxy resin are as follows: tensile strength—22.6 MPa, tension modulus—2.165 GPa, and compressive strength—66.4 MPa. The property parameters of the PZT piezoelectric ceramic are shown in Table I.

It can be seen from Table II that because the acoustic impedance value between cement paste and concrete

material is similar, acoustic impedance of the classical 2–2 connectivity cement based piezoelectric composites can match with that of concrete material only when the piezoelectric ceramic volume fraction is very low. This correspondingly results in the poor piezoelectric properties of the composites. Furthermore, the overall performance of the cement based piezoelectric composites also deteriorates due to the shrinkage effects of cement hydration. Therefore, the epoxy resin was considered here to solve above problems.

The detailed fabricating procedure of the 2–2 connectivity cement/polymer based piezoelectric composite is described as follows. The dimensional parameters of the composite were initially designed and calculated based on the mathematical model. Then, series of piezoelectric ceramic sheets were cut accurately by using diamond cutter along a direction parallel to the polarization axis of piezoelectric ceramic, and meanwhile a common ceramic base with a height of 0.5 mm was kept to maintain the upper piezoelectric ceramic sheets. The 2–2 connectivity piezoelectric ceramic body was then put into the ultrasonic cleaner for about 10 min to clear the ceramic residue. Then, the piezoelectric ceramic body was fixed into the mould for the next casting after drying in the air. Cement powder, epoxy resin, and hardener were mixed with a proportion of 4:4:1 by weight. After continuously stirring for about 2 min, the mixture was put into a vacuum pumping system for about 10 min to eliminate the pores induced during stirring, and then it was poured into the piezoelectric ceramic body. The 2–2 connectivity piezoelectric composite body was taken out of the mould after curing in the air for 48 h, and the upper and lower surfaces perpendicular to the polarization direction of the composite were polished using Al₂O₃ grinding medium.

TABLE I. Property parameters of the PZT piezoelectric ceramic. k_p —planar electromechanical coupling coefficient; k_t —thickness electromechanical coupling coefficient; d_{33} —piezoelectric strain constant; ϵ_r^T —relative permittivity; $\tan \delta$ —dielectric loss; Q_m —mechanical quality factor; ρ —density; S_{33}^E —elastic compliance coefficient.

Ceramic type	k_p (%)	k_t (%)	d_{33} (pC·N ⁻¹)	ϵ_r^T	$\tan \delta$	Q_m	ρ (10 ³ kg m ⁻³)	S_{33}^E (10 ⁻¹² m ² N ⁻¹)
PZT-4	58	48	260	1050	<0.3%	1000	7.5	12

TABLE II. Density and acoustic impedance parameters of different materials.

	Hardened cement paste	Hardened epoxy resin	PZT piezoelectric ceramic	Plain concrete
Density ρ (10^3 kg m^{-3})	~ 2	~ 1.2	~ 7.5	~ 2.4
Acoustic impedance Z (M rayl)	~ 7	~ 3	~ 32	~ 9

Finally, a thin layer of conductive silver paste was coated as electrodes on both surfaces perpendicular to thickness direction of the composite. The fabrication flow of the composites is illustrated in Figure 3.

Here, the 2–2 connectivity cement/polymer based piezoelectric composites (numbered as 1#, 2#, 3#, 4#, 5#, and 6#) with varied distribution of piezoelectric ceramic volume fraction were fabricated. Besides, the piezoelectric composites with uniform distribution of piezoelectric ceramic volume fraction (numbered as 0# and 7#) were also fabricated for comparison. As for all the piezoelectric composites, the mass fraction of cement powder in cement/polymer matrix is 44.4 wt.%. The total volume fraction of piezoelectric ceramic in the 0–3# piezoelectric composites is 76.7 vol.%, and the width for each cement/polymer matrix layer is 0.5 mm. The overall dimension of the 0–3# piezoelectric composites is 24.5 mm in length, 21.5 mm in width, and 9 mm in thickness. The width of each piezoelectric ceramic layer is shown in Table III.

As for the 4–7# piezoelectric composites, the total volume fraction of piezoelectric ceramic is 28.6 vol.%, and the width for each piezoelectric ceramic layer is 1 mm. The overall dimension of the 4–7# piezoelectric composites is 24.5 mm in length, 19 mm in width, and 9 mm in thickness. The width of each matrix layer in the composites is shown in Table IV.

C. Performance test

An impedance analyzer (Agilent 4294A, USA) was used to test impedance-frequency spectra of piezoelectric composites in a frequency range of 1 kHz–2 MHz as well as

capacitance at 1 kHz. A d_{33} piezometer (Model ZJ-3A, China) was used to test the piezoelectric strain constant (d_{33}) of the composites under a frequency of 110 Hz. In order to obtain the d_{33} value of the composites as accurately as possible, the average d_{33} value of 10 times measurement at random location was calculated. The relative permittivity (ϵ_r^T) and piezoelectric voltage constant (g_{33}) of the composites were obtained based on the following equations:

$$\epsilon_r^T = C \times t / (\epsilon_0 \times A), \quad (4)$$

$$g_{33} = d_{33} / (\epsilon_r^T \times \epsilon_0), \quad (5)$$

where C is capacitance of the piezoelectric composites at 1 kHz; t and A are thickness and area of the composite, respectively; ϵ_0 is the vacuum permittivity.

III. RESULTS AND DISCUSSION

A. Dielectric properties

The relative permittivity (ϵ_r^T) and dielectric loss ($\tan \delta$) of the 2–2 connectivity cement/polymer based piezoelectric composites were calculated by the parallel model.^{24,25}

$$\epsilon_r^T = \epsilon_{rp}^T \times \phi_p + \epsilon_{rm}^T \times \phi_m, \quad (6)$$

$$\tan \delta = \tan \delta_p \times \phi_p + \tan \delta_m \times \phi_m, \quad (7)$$

where ϵ_r^T , ϵ_{rp}^T , and ϵ_{rm}^T represent the relative permittivity of piezoelectric composite, piezoelectric ceramic, and cement/polymer matrix, respectively; $\tan \delta$, $\tan \delta_p$, and $\tan \delta_m$ are the dielectric loss of piezoelectric composite, piezoelectric ceramic, and cement/polymer matrix, respectively; ϕ_p and ϕ_m stand for the total volume fraction of piezoelectric ceramic and cement/polymer matrix in the composites.

Figure 4 illustrates the ϵ_r^T and the $\tan \delta$ value of different piezoelectric composites. It can be seen from Figure 4(a) that the 0–3# piezoelectric composites have larger ϵ_r^T value than the 4–7# composites; furthermore, the ϵ_r^T value is almost independent of the dimensional variation of piezoelectric ceramic layer, which basically agrees with the theoretical value. However, there exists obvious variation of ϵ_r^T value among the 4–7# piezoelectric composites. The ϵ_r^T value of the 7#

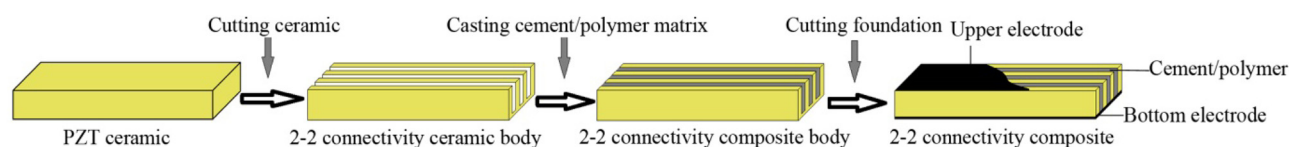


FIG. 3. Fabrication flow of the 2-2 connectivity cement/polymer based piezoelectric composite.

TABLE III. The width of piezoelectric ceramic layer in 0–3# piezoelectric composites.

Composite	Ceramic layer										
	1st	2nd	3rd	4th	5th	6th	7th	8th	9th	10th	11th
0# (mm)	1.50	1.50	1.50	1.50	1.50	1.50	1.50	1.50	1.50	1.50	1.50
1# (mm)	1.00	1.10	1.20	1.30	1.40	1.50	1.60	1.70	1.80	1.90	2.00
2# (mm)	1.00	1.55	2.10	1.00	1.55	2.10	1.0	1.55	2.10	1.00	1.55
3# (mm)	1.00	1.22	1.44	1.66	1.88	2.10	1.88	1.66	1.44	1.22	1.00

TABLE IV. The width of matrix layer in 4–7# piezoelectric composites.

Composite	Matrix layer				
	1st	2nd	3rd	4th	5th
4# (mm)	2.00	2.50	3.00	3.50	4.00
5# (mm)	1.00	3.50	6.00	1.00	3.50
6# (mm)	1.00	3.50	6.00	3.50	1.00
7# (mm)	3.00	3.00	3.00	3.00	3.00

piezoelectric composite with uniform piezoelectric phase distribution is obviously larger than that of the 4–6# piezoelectric composites, and deviates obviously from the theoretical value. In addition, it can be observed in Figure 4(b) that the $\tan \delta$ value of the 0–3# piezoelectric composites is significantly less than that of the 4–7# piezoelectric composites, and basically agrees with the parallel model theory. However, the $\tan \delta$ value of the 4–7# piezoelectric composites is obviously larger than the theoretical value, and a distinct difference also exists among different composites.

It is known that piezoelectric ceramic has larger relative permittivity ($\epsilon_{rp}^T = 1050$) and smaller dielectric loss ($\tan \delta_p = 0.3\%$) than cement/polymer matrix ($\epsilon_{rm}^T \approx 14$, $\tan \delta_m \approx 0.5\%$). Therefore, the dielectric property of the 0–3# piezoelectric composites, which has a large total volume fraction of piezoelectric ceramic, mainly depends on piezoelectric ceramic. Furthermore, the dielectric property of the composites is also hardly influenced by the dimensional variation of piezoelectric ceramic phase; therefore, the experimental value can be in accordance with the theoretical value. However, as for the 4–7# piezoelectric composites, because cement/polymer matrix instead of piezoelectric ceramic makes great contribution on dielectric property of the piezoelectric composites, they have smaller relative permittivity and larger dielectric loss than the 0–3# piezoelectric composites. The obvious deviation of $\tan \delta$ value between the experimental value and theoretical value is probably due to the interfacial polarization effects. Because there exist various interfacial polarization effects in not only inner of cement/polymer matrix but also interfaces of

piezoelectric ceramic and matrix, the dielectric property of the 4–7# piezoelectric composites, which have a large volume fraction of cement/polymer matrix can be more easily influenced by the matrix dimensional variation. It is known that the piezoelectric composite with different permittivity is required in practical ultrasonic nondestructive application based on the operating frequency range. Therefore, the 2–2 connectivity piezoelectric composites with varied distribution of piezoelectric ceramic or cement/polymer matrix have potential application in fabricating the desirable ultrasonic sensor/actuators.

B. Piezoelectric properties

According to the parallel mode theory,^{24,25} the d_{33} value of the 2–2 connectivity cement/polymer based piezoelectric composites can be calculated by the following expressions:

$$d_{33}/s_{33} = \phi_p d_{33(p)}/s_{33(p)} + \phi_m d_{33(m)}/s_{33(m)}, \quad (8)$$

$$1/s_{33} = \phi_p/s_{33(p)} + \phi_m/s_{33(m)}, \quad (9)$$

where $d_{33(p)}$ and $d_{33(m)}$ are the piezoelectric strain constant of piezoelectric ceramic and cement/polymer matrix, respectively. $s_{33(p)}$ and $s_{33(m)}$ are the elastic compliance of the piezoelectric ceramic and cement/polymer matrix, respectively. Because $d_{33(m)} \approx 0$, formula (8) can be simplified as the following equation:

$$d_{33} = d_{33(p)}s_{33(m)}/[(s_{33(m)} - s_{33(p)}) + s_{33(p)}/\phi_p]. \quad (10)$$

Figure 5 shows the d_{33} and the g_{33} value of different piezoelectric composites. There exist differences between the parallel model results and the experimental results, especially for 4–7# piezoelectric composite. The reason is mainly attributed to the difference between assumptions of the parallel model and the real composite structure. It can be seen from Figure 5(a) that the d_{33} value of the 0–3# piezoelectric composites is larger than that of the 4–7# piezoelectric composites. Furthermore, the d_{33} value of the 1–3# piezoelectric composites with varied distribution of piezoelectric ceramic

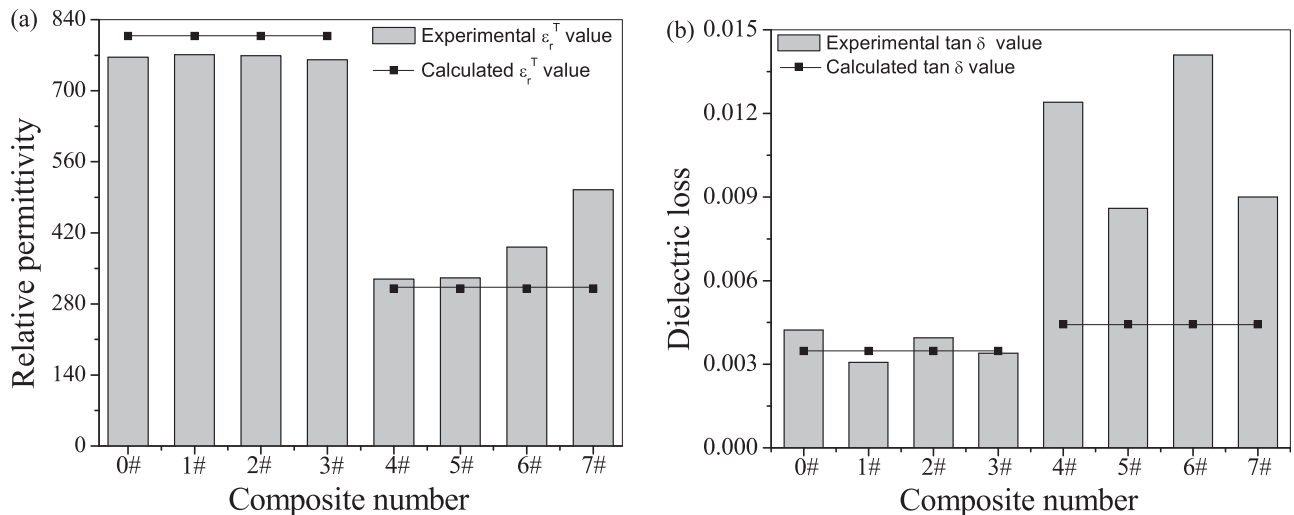


FIG. 4. Dielectric property of the 2-2 connectivity cement/polymer based piezoelectric composites.

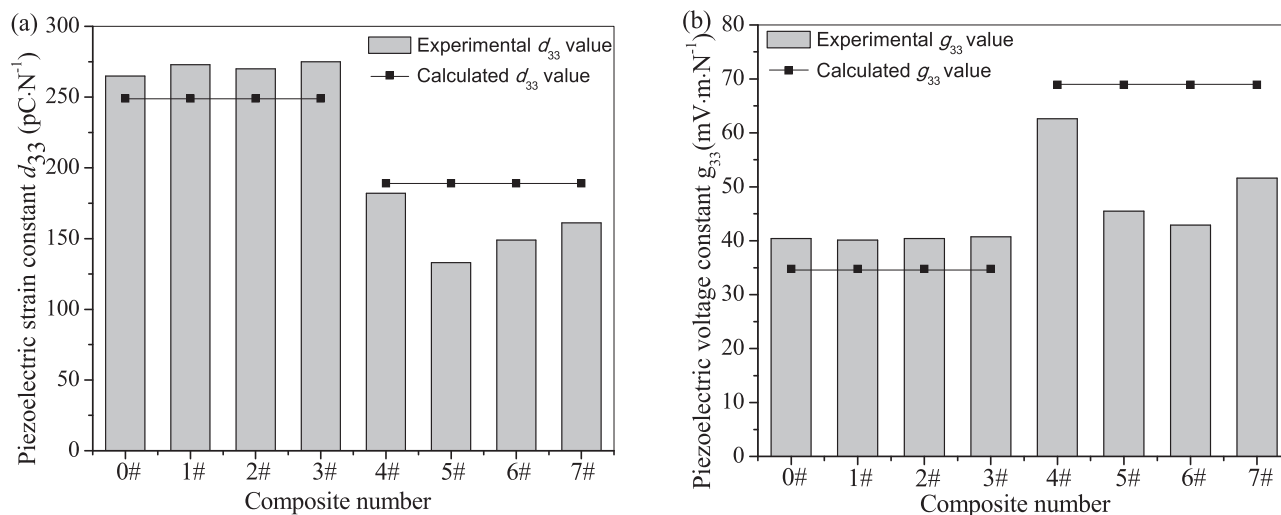


FIG. 5. Piezoelectric property of the 2-2 connectivity cement/polymer based piezoelectric composites.

phase is a little larger than that of the 0# piezoelectric composite. As for the 4–7# piezoelectric composites, there exists an obvious variation of the d_{33} value among different composites. In Figure 5(b), it can be observed that the g_{33} value of the 4–7# piezoelectric composites is obviously larger than that of the 0–3# piezoelectric composites. The influence of dimensional variation of cement/polymer matrix on the g_{33} value of the 4–7# piezoelectric composites is obvious, and the 4# piezoelectric composite has the largest g_{33} value among all composites.

It is known that the piezoelectric property of the composites mainly depends on piezoelectric ceramic phase, and the cement/polymer matrix plays a great role in transferring stress.^{26,27} The 0–3# piezoelectric composites have large d_{33} average value due to the large contribution of piezoelectric ceramic phase. However, as for the 4–7# piezoelectric composites, the d_{33} average value is small because of the weak contribution of piezoelectric ceramic phase. This indicates that the piezoelectric strain constant of the 2–2 connectivity cement/polymer based piezoelectric composites has great dependence on the piezoelectric phase distribution when the total volume fraction of piezoelectric ceramic phase is small in the composite. In addition, it is known based on Eq. (5)

that the g_{33} value of the piezoelectric composite mainly depends on the d_{33} and ϵ_r^T . Although the 4–7# piezoelectric composites has smaller d_{33} value than the 0–3# piezoelectric composites, however, their ϵ_r^T value is larger than that of the 0–3# piezoelectric composites, which is the reason that the 4–7# piezoelectric composites have larger g_{33} value. Therefore, the transmitting transducer with a large d_{33} value and the receiving transducers with a large g_{33} value can be fabricated based on the varied distribution of piezoelectric ceramic phase in the composite.

C. Electromechanical coupling properties

Figure 6 shows the impedance-frequency spectra of 2–2 connectivity cement/polymer based piezoelectric composites. It is known that piezoelectric material has various resonance modes (i.e., resonance peaks) at different frequency range under the excitation of external electric field, such as planar resonance mode and thickness resonance mode. The resonance peak amplitude indicates the resonance ability of the piezoelectric composites. Because of the damping effects of cement/polymer matrix, the larger the volume fraction of cement/polymer matrix is, the less the resonance peak

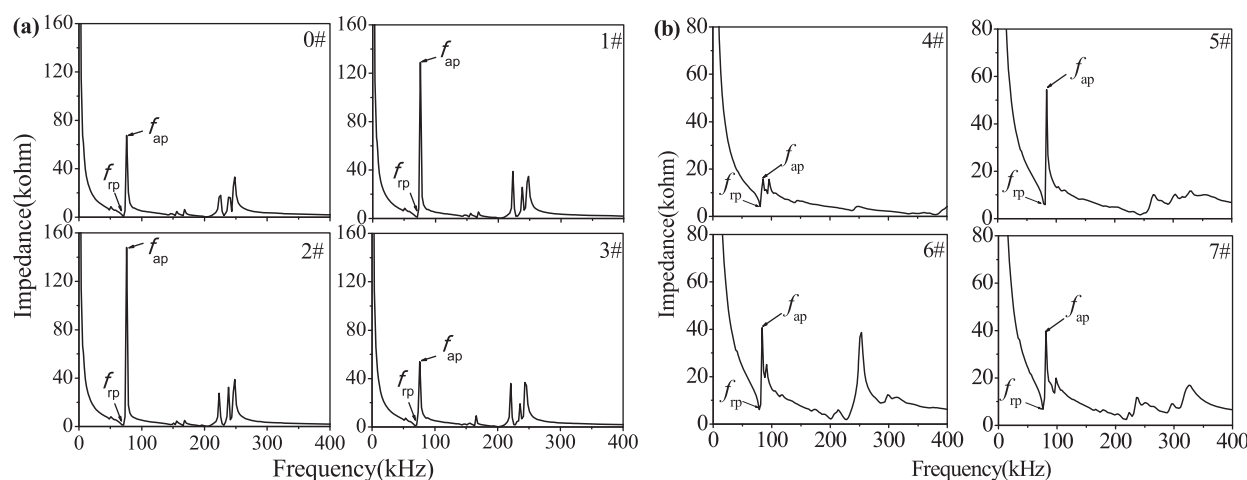


FIG. 6. Impedance-frequency spectra of the 2-2 connectivity cement/polymer based piezoelectric composites.

TABLE V. Planar electromechanical coupling property of the 2-2 connectivity cement/polymer based piezoelectric composites. f_{rp} and f_{ap} represent resonance and anti-resonance frequency at planar mode, respectively; k_p was obtained by referring to the National Standard of China, GB/T 2414.1-1998.

Composite type	0#	1#	2#	3#	4#	5#	6#	7#
f_{rp} (kHz)	70.965	70.965	70.965	70.965	80.960	80.960	78.461	75.963
f_{ap} (kHz)	75.963	75.963	75.963	75.963	85.958	85.958	83.459	80.960
k_p (%)	40	40	40	40	37.9	37.9	38.5	39.0

amplitude of the composite. Therefore, it can be seen that the 0–3# piezoelectric composites have larger planar mode resonance peaks than 4–7# piezoelectric composites. In addition, there also exists an obvious coupling resonance peak around the planar resonance peaks for the 4–7# piezoelectric composite, which might be the stopband resonance caused by Bragg reflections.²¹ Because the composite thickness is not small enough, it also can be clearly observed in Figure 6(a) that the thickness mode resonance peaks which appear at about 200–300 kHz couple with other resonance modes, such as the 3rd order harmonic (around 230 kHz) of the planar resonance mode.^{27–30}

It is known based on the definition of frequency constant (N) that the resonance frequency (f_{rp}) of piezoelectric material at planar mode is inversely proportional to the planar dimension (l).

$$N = f_{rp}l. \quad (11)$$

Because the planar dimension of the piezoelectric composites is the same, f_{rp} , f_{ap} and the planar electromechanical coupling coefficient (k_p) of the composites also keep unchanged, as shown in Table V. Nevertheless, the k_p value of the 4–7# piezoelectric composites is a little smaller than that of the 0–3# piezoelectric composite because of the effects of cement/polymer matrix.

It is known that the electromechanical coupling coefficient is an important parameter of ultrasonic transducer; therefore, the desired piezoelectric ultrasonic transducer can be fabricated in terms of the varied distribution of piezoelectric ceramic phase in the composite. In this research, the novel 2–2 connectivity cement/polymer based piezoelectric composites were designed and fabricated. However, the local distribution characteristics of dielectric, piezoelectric, and electromechanical coupling properties of the composites were not discussed. Besides, further investigation on fabrication technology and properties of the piezoelectric transducers made of this composite should also be performed in the future work.

IV. CONCLUSIONS

The 2–2 connectivity cement/polymer based piezoelectric composites with varied piezoelectric phase distribution were designed based on the dimensional variations of piezoelectric ceramic layer and matrix layer. The piezoelectric composites were also fabricated by using PZT piezoelectric ceramic as active phase and mixture of cement powder and epoxy resin as matrix phase. The piezoelectric ceramic volume fraction shows varied distribution in the composites according to the dimensional variations of piezoelectric ceramic and cement/polymer matrix. Therefore, the desirable

composite properties can be obtained. The novel 2–2 connectivity cement/polymer based piezoelectric composites might have potential applications in fabricating piezoelectric transducers with specific distribution of surface vibration amplitudes.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Grant Nos. 51202089 and 51202090), Natural Science Foundation of Shandong Province (Grant No. ZR2012EMQ010), State Key Laboratory of High Performance Civil Engineering Materials (Grant No. 2012CEM009), and Program for Scientific Research Innovation Team in Colleges and Universities of Shandong Province.

- ¹K. Diamanti, C. Soutis, and J. Hodgkinson, *Comput. Sci. Technol.* **65**, 2059 (2005).
- ²J. B. Ihn and F. K. Chang, *Smart Mater. Struct.* **13**, 609 (2004).
- ³V. Giurgiutiu, A. Zagari, and J. J. Bao, *Struct. Health Monit.* **1**, 41 (2002).
- ⁴G. B. Song, H. C. Gu, and Y. L. Mo, *Smart Mater. Struct.* **17**, 033001 (2008).
- ⁵Z. J. Li, D. Zhang, and K. R. Wu, *J. Am. Ceram. Soc.* **85**, 305 (2002).
- ⁶X. Cheng, S. F. Hang, J. Chang, and Z. J. Li, *J. Appl. Phys.* **101**, 094110 (2007).
- ⁷Z. J. Li, H. Y. Gong, and Y. J. Zhang, *Curr. Appl. Phys.* **9**, 588 (2009).
- ⁸F. Z. Wang, H. Wang, H. J. Sun, and S. G. Hu, *Smart Mater. Struct.* **23**, 045032 (2014).
- ⁹K. H. Lam and H. L. W. Chan, *Appl. Phys. A* **81**, 1451 (2005).
- ¹⁰B. Q. Dong and Z. J. Li, *Comput. Sci. Technol.* **65**, 1363 (2005).
- ¹¹D. Y. Xu, X. Cheng, S. F. Huang, and M. H. Jiang, *Curr. Appl. Phys.* **9**, 816 (2009).
- ¹²R. Han, Z. F. Shi, and Y. L. Mo, *Arch. Appl. Mech.* **81**, 839 (2010).
- ¹³R. Potong, R. Rianyo, A. Ngamjarujana, and A. Chaipanich, *Ceram. Int.* **40**, 8723 (2014).
- ¹⁴J. Zhang, L. Qin, and Z. J. Li, *Mater. Struct.* **42**, 15 (2009).
- ¹⁵T. Voigt, T. Malonn, and S. P. Shah, *Cem. Concr. Res.* **36**, 858 (2006).
- ¹⁶D. Y. Xu, L. Qin, S. F. Huang, and X. Cheng, *Mater. Chem. Phys.* **132**, 44 (2012).
- ¹⁷D. G. Aggelis, *Mater. Struct.* **46**, 519 (2013).
- ¹⁸G. Trtnik and M. Gams, *Cem. Concr. Res.* **43**, 1 (2013).
- ¹⁹M. J. Zhou, M. Sun, M. M. Li, S. H. Xie, and S. F. Huang, *J. Electroceram.* **28**, 139 (2012).
- ²⁰D. F. Liu and M. X. Li, *Appl. Acoust.* **17**, 11 (1998).
- ²¹D. D. L. Chung, *J. Mater. Sci.* **39**, 2973 (2004).
- ²²Y. Ohama, *Cem. Concr. Compos.* **20**, 189 (1998).
- ²³D. W. Fowler, *Cem. Concr. Compos.* **21**, 449 (1999).
- ²⁴V. R. Riley and I. Razl, *Composites* **5**, 27 (1974).
- ²⁵H. L. W. Chan and J. Unsworth, *J. Appl. Phys.* **65**, 1754 (1989).
- ²⁶W. A. Smith and B. A. Auld, *Ultrason. Ferroelectr. Freq. Control* **38**, 40 (1991).
- ²⁷H. L. W. Chan and J. Unsworth, *IEEE Trans. Ultrason. Ferro. Freq. Control* **36**, 434 (1989).
- ²⁸T. Gururaja, W. A. Schulze, L. E. Cross, R. E. Newnham, B. Auld, and Y. Wang, *IEEE Trans. Sonics Ultrason.* **32**, 481 (1985).
- ²⁹H. L. W. Chan, J. Unsworth, and T. Bui, in *IEEE Ultrasonics Symposium* (1987), p. 651.
- ³⁰D. Y. Wang and H. L. W. Chan, *Mater. Sci. Eng., B* **99**, 147 (2003).