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Piezoelectric cement-based 1-3 composites

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ABSTRACT This paper presents a new functional material for smart structure applications. Piezoelectric PZT/cement 1-3 composites that have good compatibility with civil engineering structural materials have been studied. The composites with different volume fractions of PZT ranging from 0.25 to 0.77 were fabricated by the dice-and-fill method. It was found that the 1-3 composites have good piezoelectric properties that agreed quite well with theoretical modeling. The thickness electromechanical coupling coefficient could reach 0.55 in the composite with a ceramic volume fraction of 0.25. Those composites have potential to be used as sensors in civil structure health monitoring systems.

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1 Introduction

Effective structural health monitoring using sensors based on smart materials and systems has gained considerable importance in civil engineering [1–3]. In these smart systems, sensors and actuators are key components. The materials used in civil engineering, e.g., concrete and cement etc., have characteristics that are different from materials used in mechanical systems. Hence the sensors and actuators used in smart mechanical systems may not be applicable in civil engineering structures. The aim of this work is to develop smart materials that have good compatibility with civil engineering structural materials such as concrete which is commonly used in civil engineering structures [4].

Piezoelectric materials have been widely used in the application of smart structures [5, 6]. Among different piezoelectric materials, piezoelectric ceramics such as lead titanate (PT) and lead zirconate titanate (PZT) are commonly used because they have high electromechanical coupling factor k_t and piezoelectric d coefficients.

Piezoelectric composites have been widely studied in recent years in order to obtain properties superior to a single phase material. There are ten connectivity patterns according to the connectivity of the active element and passive matrix phase [7]. Piezoelectric 1-3 ceramic/polymer composites consist of piezoelectric rods embedded in a polymer phase have been widely used in transducer applications [8–10]. Connectivity is defined as the number of dimensions in which a phase is self-connected [11] while 1-3 is one of ten connectivities in biphasic composites. Those composites which combine the advantages of the ceramics and polymer phases have played an important role in the field of medical ultrasound. The 1-3 connectivity enhances the electromechanical coupling in the thickness mode effectively. Besides, it maintains the high piezoelectric characteristics of ceramics and provides low acoustic impedance because of the incorporation of the passive phase. The characteristics of 1-3 composites can be adjusted by tailoring the ceramic volume fraction to meet the specific requirements of various applications. In the past, many models have been introduced for investigating the physical and electrical properties of 1-3 composites [12, 13]. In these models, a larger electromechanical coupling coefficient k_t can be obtained that approached the longitudinal coupling factor k_{33} of the ceramic rods in the composites.

To develop 1-3 composites for use in civil engineering applications, cement paste has been used as the passive matrix of the composites. In this study, piezoelectric ceramic/cement 1-3 composites with 0.25 to 0.77 volume fractions of PZT prepared by a dice-and-fill method have been fabricated. The properties of the 1-3 composites were determined by the resonance techniques and compared with theoretical modeling.

2 Experimental

In this study, the active ceramic phase was PZT fabricated using PKI 802 PZT powder supplied by Ultrasonic Powders Ltd., USA. PZT ceramic discs of 30 mm diameter and 5 mm thick were fabricated by dry pressing and sintering at 1325 °C for two hours. Before poling, air-dried silver was applied to the two flat surfaces of the discs as electrodes. They were poled in silicone oil along their thickness direction by applying a (dc) field of 6 kV/mm at 120 °C for 30 minutes. The electric field was maintained until the sample was cooled to 50 °C. After poling, the ceramic discs were short-circuit at 40 °C to remove the injected charges.

PZT/cement 1-3 composites were fabricated using a dice-and-fill technique [14]. In order to avoid mode coupling, the aspect ratio (thickness to width ratio) of the PZT rods inside the composite samples was higher than two. Poled ceramic disc was cut in one direction using a diamond saw (Buehler Isomet 2000) with a 0.5 mm thick blade. Cement paste (cement : water = 10 : 5) was used to fill the grooves in the

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		PZT 8 ceramics	Cement (C:W ∼ 10:5)	Normal Concrete (Sand stone) [17]
Piezoelectric coefficient	$d_{33}(10^{-12}$ C/N)	215		
Electromechanical coefficient	k_t	0.40		
Density	ϱ (kg/m^3)	7563	1961	2400
Longitudinal wave velocity	υ_l (m/s)	4575	2740	3730
Elastic compliance	s_{33} (10^{-12} m^2/N)	13	69	30
Thermal expansion coefficient	α (10^{-6} K^{-1})	6	14	9–12
Acoustic impedance	Z_a (MRayl)	34.6	6.1	9.0

TABLE 1 Properties of PZT PKI 802 ceramic, cement paste and normal concrete

diced disc. After the cement paste was dried at a temperature of 50 °C under a condition of relatively high humidity for five days, a second set of cuts perpendicular to the first direction was made. Since the ceramic rods are fragile, the cuts in both directions cannot be processed simultaneously. After filling the second set of cuts with cement paste, the composite was dried under the same condition mentioned above and then cured at room temperature for 28 days. Excess cement was ground away using ultra fine wet silicon carbide abrasive papers. The composites were repoled in case they were depoled during dicing.

Density, ϱ, of the samples was measured based on the Archimedes' principle using an electronic balance. The piezoelectric strain coefficient, d_{33}, was measured by a d_{33} meter (ZJ-3B), which was supplied by the Beijing Institute of Acoustics, Academia Sinica. The impedance and phase of the samples were measured using an impedance/gain phase analyzer (Agilent 4294A). The electromechanical properties of the samples were determined at room temperature following the IEEE standard on piezoelectricity [15]. Electromechanical coupling coefficient in the thickness direction k_t, elastic stiffness, c_{33}^D and acoustic impedance Z_a were determined by measuring the resonant frequency, f_r and anti-resonant frequency, f_a of the thickness mode resonance in the samples. To model the performance of the 1-3 composites, materials parameters of the passive matrix were characterized. The acoustic velocity and the elastic properties of a cured bulk cement plate were determined using the ultrasonic immersion method [16].

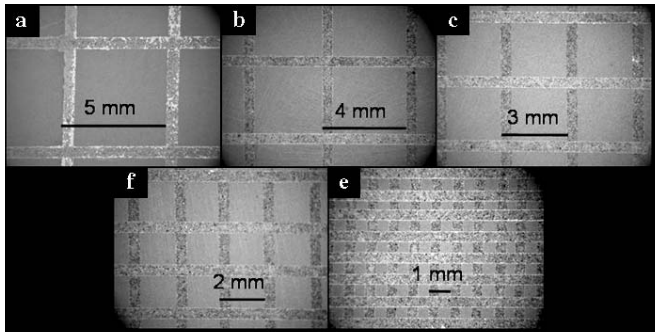

FIGURE 1 Photographs of PZT/cement 1-3 composites with different ϕ

Density	ϱ	(kg/m^3)	1961
Longitudinal wave velocity	υ_l	(m/s)	2740
Shear wave velocity	υ_s	(m/s)	1641
Longitudinal stiffness	c_{33}	(10^{10} N/m^2)	1.66
Shear stiffness	c_{44}	(10^{10} N/m^2)	0.47
Stiffness in x-y plane	c_{12}	(10^{10} N/m^2)	0.60
Longitudinal compliance	s_{33}	(10^{-12} m^2/N)	68.90
Shear compliance	s_{44}	(10^{-12} m^2/N)	168
Compliance in x-y plane	s_{12}	(10^{-12} m^2/N)	−15.20
Shear modulus	G	(10^{10} N/m^2)	0.60
Bulk modulus	K	(10^{10} N/m^2)	0.87
Young's modulus	Y	(10^{10} N/m^2)	1.45
Acoustic impedance	Z_a	(MRayl)	6.1
Mechanical quality factor	Q_M		20
Poisson's ratio	σ		0.22

TABLE 2 Room-temperature material properties of cement paste with a water : cement ratio of 0.5

3 Results and discussion

To show the feasibility of compatibility of the piezoelectric cement-based composites, the properties of the PZT, cement paste and normal concrete were measured or found in the literature and are listed in Table 1. It shows that the acoustic impedance of the ceramics is very different to that of the concrete. If a PZT sensor is embedded in concrete, the energy transfer between the PZT and the host concrete materials would be degraded by such mismatching. By introducing the cement as a passive matrix, the properties of the 1-3 composites, such as acoustic impedance, can be tailored.

Five PZT ceramic/cement 1-3 composites with ceramic volume fractions ϕ of 0.25 to 0.77 have been fabricated and are shown in Fig. 1. Due to the blade vibration during dicing, the resultant cement width in the composites varied from 0.5 mm to 0.6 mm. The measured electrical impedance and phase vs. frequency spectra of (a) a PZT ceramic disc and (b) a PZT/cement 1-3 composite are shown in Fig. 2. A strong thickness resonance with its third harmonics can be observed in the 1-3 composite.

The piezoelectric, electromechanical and elastic properties of the PZT ceramics and the cement paste with the water : cement ratio of 0.5 are shown in Tables 1 and 2, respectively. The properties of those two elements are used to calculate the theoretical properties of the 1-3 composites by the modified series and parallel model [12, 13]. The theoretical density ϱ and piezoelectric coefficient d_{33} of the composites are determined by (1) and (2):

$$\varrho = \phi \varrho_{\text{ceramic}} + (1-\phi)\varrho_{\text{cement}} \quad (1)$$

$$d_{33} = \frac{\phi\, d_{33,\text{ceramic}}\, s_{11,\text{cement}}}{\phi\, s_{11,\text{cement}} + (1-\phi)\, s_{33,\text{ceramic}}^{E}} \quad (2)$$

where s is the elastic compliance. It is seen that the density ϱ and piezoelectric coefficient d_{33} of the composites increase

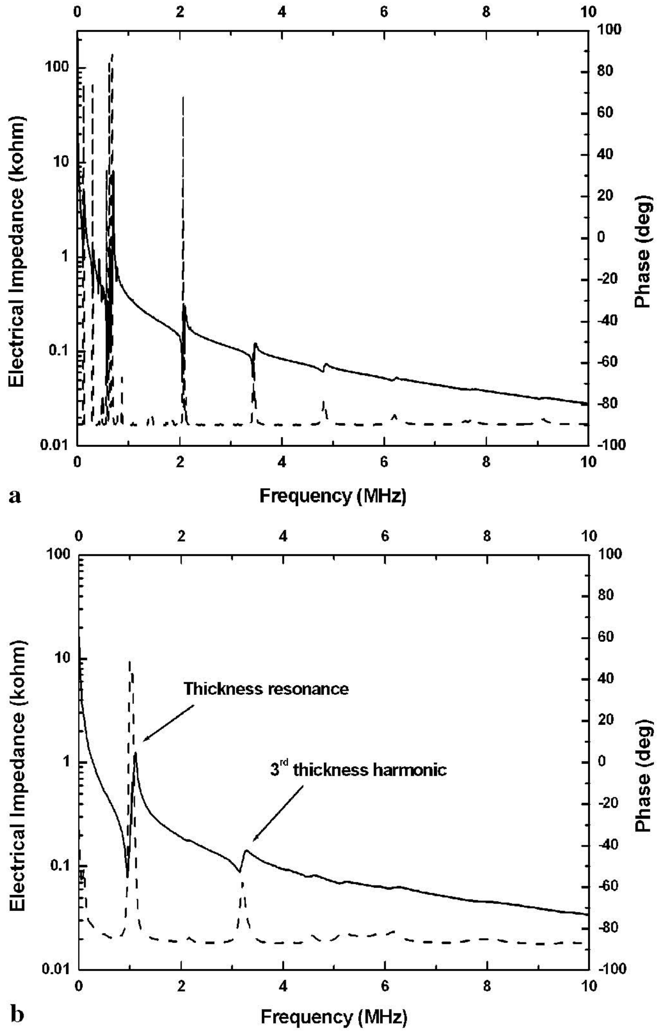

FIGURE 2 Electrical impedance and phase vs. frequency spectra for (**a**) a PZT PKI 802 bulk disc and (**b**) a PZT 802/cement 1-3 composite with properties of PZT PKI 802 ceramic, cement paste and normal concrete $\phi = 0.56$

monotonically as shown in Figs. 3 and 4. As mentioned before, the electromechanical coupling factor of the thickness mode can be enhanced effectively using the 1-3 connectivity.

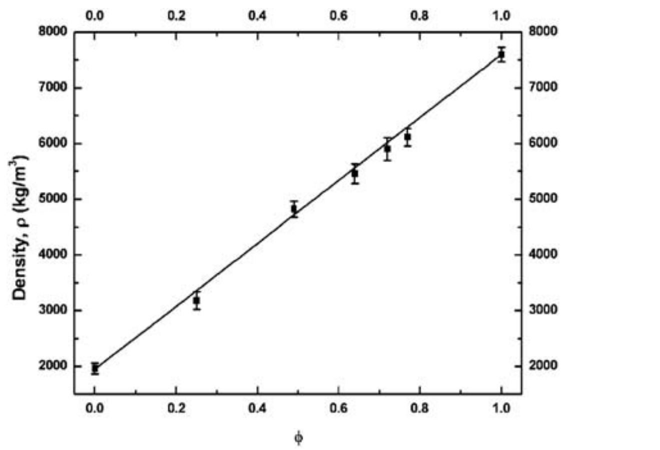

FIGURE 3 Density ϱ of the PZT/cement 1-3 composites as a function of ϕ. (*symbols*: experimental data; *line*: theoretical prediction)

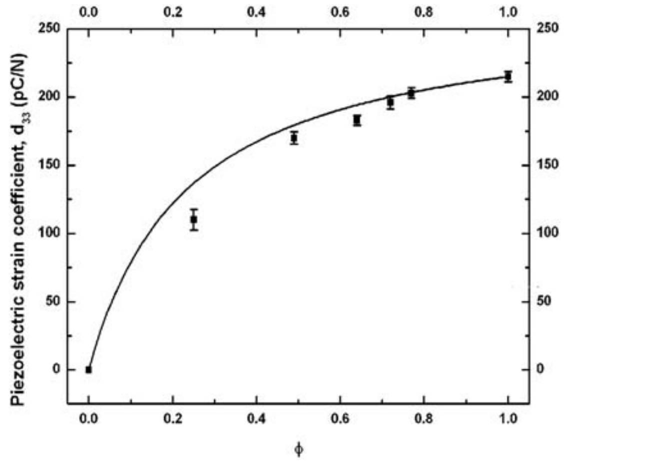

FIGURE 4 Piezoelectric strain coefficient d_{33} of the PZT/cement 1-3 composites as a function of ϕ. (*symbols*: experimental data; *line*: theoretical prediction)

The theoretical electromechanical coupling coefficient k_t in the composite can be determined by (3):

$$k_t = \sqrt{1 - \frac{c_{33}^E}{c_{33}^D}} \tag{3}$$

where

$$c_{33}^E = \phi \left[c_{33,\text{ceramic}}^E - \frac{2\left(c_{13,\text{ceramic}}^E - c_{12,\text{cement}}\right)^2}{C(\phi)} \right] + (1-\phi)\; c_{11,\text{cement}} \tag{4}$$

$$c_{33}^D = c_{33}^E + \frac{\phi^2 \left[e_{33,\text{ceramic}} - \frac{2\, e_{31,\text{ceramic}} \left(c_{13,\text{ceramic}}^E - c_{12,\text{cement}}\right)}{C(\phi)} \right]^2}{\phi \left[\varepsilon_{33,\text{ceramic}}^S - \frac{2 e_{31,\text{ceramic}}^2}{C(\phi)} \right] + (1-\phi)\; c_{11,\text{cement}}} \tag{5}$$

$$e_{33,\text{ceramic}} = k_{t,\text{ceramic}} \sqrt{c_{33,\text{ceramic}}^D \varepsilon_{33,\text{ceramic}}^s} \tag{6}$$

$$e_{31,\text{ceramic}} = \frac{\left(\varepsilon_{33,\text{ceramic}}^T - \varepsilon_{33,\text{ceramic}}^s - d_{33,\text{ceramic}} e_{33,\text{ceramic}}\right)}{2 d_{31,\text{ceramic}}} \tag{7}$$

$$C(\phi) = \left(c_{11,\text{ceramic}}^E + c_{12,\text{ceramic}}^E\right) + \frac{\phi}{1-\phi}\left(c_{11,\text{cement}} + c_{12,\text{cement}}\right) \tag{8}$$

where $\varepsilon_{33,\text{ceramic}}^T$ is the constant stress relative permittivity measured at 1 kHz and $\varepsilon_{33,\text{ceramic}}^S$ is the constant strain relative permittivity measured at high frequency ($\sim 2^* f_a$). Figure 5 shows that k_t of the composites can reach 0.55 even with $\phi \sim 0.25$ which is higher than that of a PZT802 ceramic disc ($k_t \sim 0.40$) and approaches to its k_{33} value ($k_{33} \sim 0.56$). Since the cement matrix exerts some lateral clamping on the ceramic rods, k_t of the composites cannot reach as high as the k_{33} value of a free ceramic rod. In Figs. 6 and 7, c_{33}^D and Z_a of the composites increase almost linearly with ϕ. The elastic stiffness

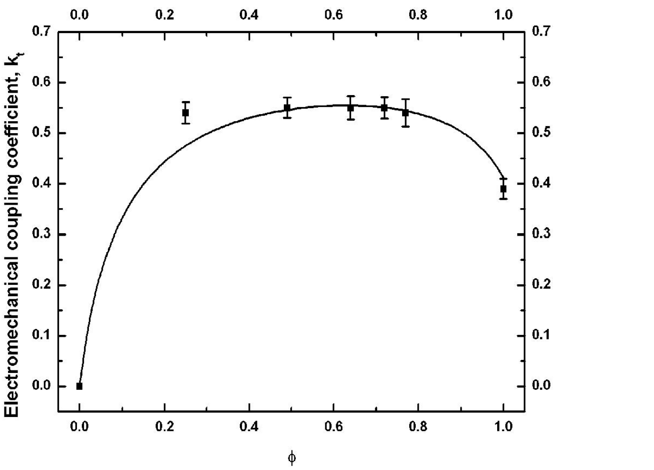

FIGURE 5 Electromechanical coupling coefficient k_t of the PZT/cement 1-3 composites as a function of ϕ. (*symbols*: experimental data; *line*: theoretical prediction)

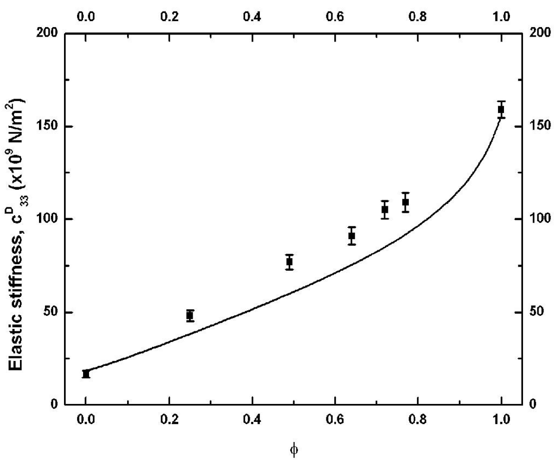

FIGURE 6 Elastic stiffness c^D_{33} of the PZT/cement 1-3 composites as a function of ϕ. (*symbols*: experimental data; *line*: theoretical prediction)

c^D_{33} can be determined by (5) and acoustic impedance Z_a can be determined by (9):

$$Z_a = \sqrt{\varrho c^D_{33}} . \quad (9)$$

The acoustic impedance of the composites can be tailored to better match that of the civil engineering structural materials by changing the ceramic volume fraction. As shown in Figs. 3–7, the experimental data agree quite well with the modified series and parallel model. The model shows that the parameters of the 1-3 composites (except k_t) increase with ϕ because of the increase contribution of the active PZT phase.

4 Conclusion

The PZT/cement 1-3 composites with various PZT volume fractions have been fabricated successfully using the dice-and-fill technique. The performance of the composites was characterized by the resonance method. The experimental data were compared and agreed quite well with the prediction of the modified series and parallel model. The composites have high k_t value of 0.55 which is comparable to the k_{33} value of a free ceramic rod. By changing the ceramic volume fraction, the properties of the composites can be tailored to match that of the civil engineering structural materials, i.e., concrete. These composites will be used as sensors in civil structure health monitoring systems.

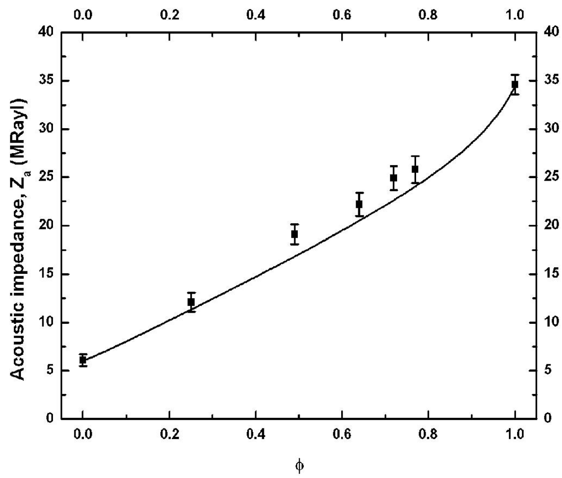

FIGURE 7 Acoustic impedance Z_a of the PZT/cement 1-3 composites as a function of ϕ. (*symbols*: experimental data; *line*: theoretical prediction)

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REFERENCES

1 B. Culshaw: Artech House, Boston London
2 S. Aizawa, T. Kakizawa, M. Higasino: Smart Mater. Struct. **7**, 617 (1998)
3 C.K. Soh, K.K-H Tseng, S. Bhalla, A. Gupta: Smart Mater. Struct. **9**, 533 (2000)
4 Z.J. Li, D. Zhang, K. Wu: J. Am. Ceram. Soc. **85**, 305 (2002)
5 J. Kim, B. Ko: Smart Mater. Struct., **7**, 801 (1998)
6 P. Janker, M. Christmann, F. Hermle, T. Lorkowski, S. Storm: J. Eur. Ceram. Soc. **19**, 1127 (1999)
7 R.E. Newnham, D.P. Skinner, L.E. Cross: Mater. Res. Bull. **13**, 525 (1978)
8 C.G. Oakley: Applications of Ferroelectrics, IEEE 7th Inter. Symp. **6–8**, 233 (1990)
9 W.A. Smith: Applications of Ferroelectrics, Proc. of the 7th IEEE Inter. Symp. **6–8**, 145 (1990)
10 J.A. Hossack, G. Hayward: IEEE Trans. Ultrason. Ferroelect., Freq. Contr., **38**, 618 (1991)
11 R.E. Newnham, D.P. Skinner, L.E. Cross: Mater. Res. Bull. **13**, 525 (1978)
12 H.L.W. Chan, J. Unsworth: J. Appl. Phys. **65**, 1754 (1989)
13 W.A. Smith, B.A. Auld: IEEE Trans. Ultrason. Ferroelect., Freq. Contr. **38**, 40 (1991)
14 H.L.W. Chan, J. Unsworth: J. Appl. Phys. **65**, 1754 (1989)
15 IEEE standard on piezoelectricity, ANSI/IEEE Std. 176-1987
16 B.E. Read, G.D. Dean: *The Determination of Dynamic Properties of Polymers and Composite* (Adam Hilger, 1978)
17 A.M. Neville: *Properties of concrete* (Wiley, New York 1996)