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Study of piezoelectric fibre/cement 1–3 composites

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Abstract To improve the compatibility between the sensor material and civil engineering structural material, a new functional cement-based composite for smart structure applications has been studied. Piezoelectric lead zirconate titanate (PZT) fibres, fabricated using a slurry method, are embedded in a cement matrix to form PZT/cement 1–3 composites. By incorporating PZT fibres into the cement matrix, composites with low PZT volume fractions ranging from 0.05 to 0.22 have been fabricated. The 1–3 composites have good piezoelectric properties that agree quite well with theoretical modeling. The thickness electromechanical coupling coefficient of the composites could reach ~0.5 even for low volume fraction of PZT. These composites have potential to be used as sensors in civil structure health monitoring systems.

Keywords PZT · Cement · 1–3 Composites

1 Introduction

With the development of smart civil engineering structures, structural health monitoring has gained considerable interest [1–3]. In these smart systems, sensors and actuators based on smart materials are the key components. Since civil engineering materials, for example concrete and cement, have different characteristics compared to materials used in mechanical systems (e.g. aluminium and steel), the sensors and actuators used in smart mechanical systems may not be applicable in civil engineering structures. To better cater for civil engineering applications, smart materials with good compatibility to civil engineering structural materials such as concrete should be developed.

Piezoelectric materials have been widely studied and utilized in the applications of smart structures [4, 5]. Piezoelectric ceramics such as lead titanate (PT) and lead zirconate titanate (PZT) are commonly used because they have high electromechanical coupling coefficient k_t and high piezoelectric d coefficients. However, they have extremely high acoustic impedance Z_a.

To overcome the limitation of a single phase material, piezoelectric composites have been widely studied in recent years. Piezoelectric 1–3 ceramic/polymer composites consisting of active piezoelectric rods embedded in a polymer phase, which combine the advantages of both the ceramic and polymer phases, have been widely used in transducer applications [6–8]. Piezoceramic/polymer 1–3 composites have played an important role in the field of medical ultrasound. The 1–3 connectivity enhances the electromechanical coupling in the thickness mode effectively by decoupling the planar mode vibrations. Besides, it maintains the high piezoelectric characteristics of ceramics and provides low acoustic impedance because of the incorporation of the passive phase. By tailoring the ceramic volume fraction of 1–3 composites, the materials characteristics can be adjusted 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 [9, 10]. A higher electromechanical coupling coefficient k_t of the composites can be obtained that approached the longitudinal coupling coefficient k_{33} of the ceramic rods.

To develop 1–3 composites as smart materials in civil engineering applications, cement paste has been used as the

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Table 1 Properties of PZT 802 ceramics, cement paste and normal concrete.

		PZT 8 ceramics	Cement (C:W ~ 10:5)	Normal concrete (sand stone)[15]
Piezoelectric coefficient	d_{33} (10^{-12} C/N)	215		
Electromechanical coefficient	k_t	0.40		
Density	ρ (kg/m^3)	7563	1961	2400
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

passive matrix of the composites. In a previous study [11], piezoelectric ceramic/cement 1–3 composites were fabricated by a dice-and-fill method. To study the composites with low ceramic volume fraction, ceramic fibres prepared by a powder mixing method [12] were embedded in a passive cement matrix. The ceramic fibre/cement 1–3 composites with 0.05 to 0.22 volume fractions of PZT ceramic fibres have been fabricated. Properties of the 1–3 composites were determined by the resonance technique and compared with theoretical modeling.

2 Experimental

In this study, the active ceramic phase was PZT fibres fabricated using PKI 802 PZT powder supplied by Ultrasonic Powders Ltd., USA. Since the fibres can only be drawn using a slurry with appropriate viscosity, the amount of binder plays an important role in the fibre fabrication. The binder used was poly(acrylic acid) (PAA, 25% aqueous solution). After weighing the powder and binder with the appropriate weight ratio (6:1), the mixture was stirred until the slurry became homogenous. The slurry was then put into the mould of a fibre spinning machine (One Shot III from Alex James & Associate Inc. U.S.A.) for drawing the fibres. At around 70°C, PZT fibres were extruded through a mould containing a 0.6 mm diameter hole. The fibres were collected on a spindle. They were then kept at the air ambient in air for 1 day for drying. PZT ceramic fibres of ~ 0.5 mm diameter were obtained after pyrolysis and sintering at 600 and 1325°C, respectively.

A designated number of sintered PZT ceramic fibres were chosen and aligned parallelly in a special holder. The ceramic volume fraction of the composites can be varied by adjusting the quantity of fibres. Cement paste (cement: water=10:5) was used to fill the empty space between the fibres. The composite cube was then degassed in vacuum for 1 h and dried at 50°C under a condition of relatively high humidity for 5 days. To stabilize the properties of the cement matrix, the samples were cured at room temperature for 28 days. The composite cube was then sliced to give ~0.8 mm thick disks by a diamond saw (Buehler Isomet 2000). Excess cement was ground away using ultra fine wet silicon carbide abrasive papers. Before subsequent electrical characterization, the composites should be poled. Air-dried silver was applied to the two flat surfaces

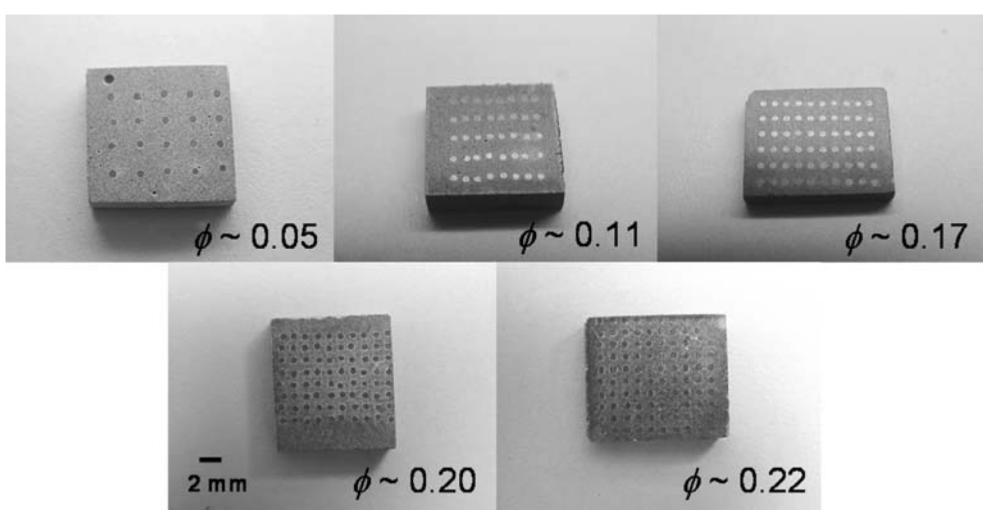

Fig. 1 Photographs of PZT fibre/cement 1–3 composites with different ϕ

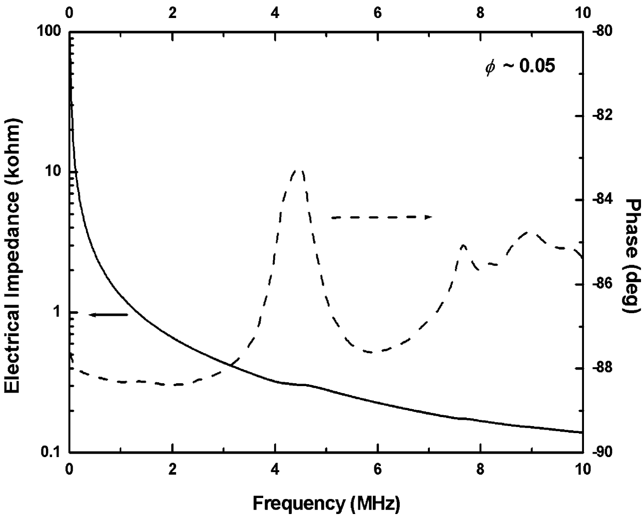

Fig. 2 Electrical impedance and phase vs frequency spectra for a PZT fibre/cement 1–3 composite with ϕ=0.05

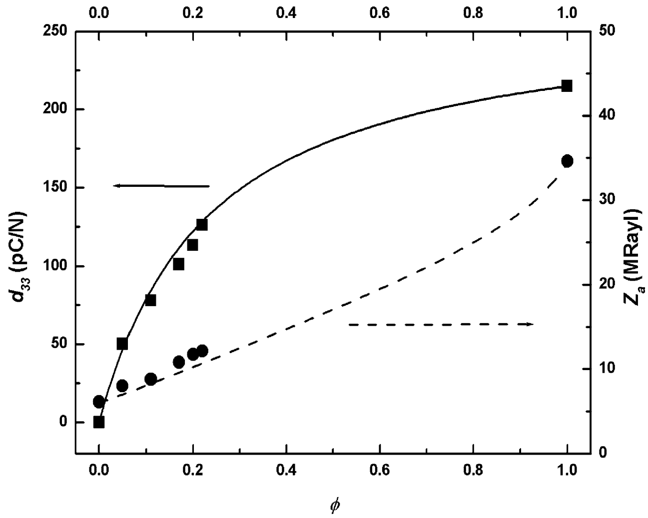

Fig. 3 Piezoelectric coefficient d_{33} and acoustic impedance Z_a of the PZT fibre/cement 1–3 composites as a function of ϕ. (*Symbols*: experimental data; *Line*: theoretical prediction)

of the samples as electrodes. The samples were poled in silicone oil along their thickness direction by applying a d.c. field of 6 kV/mm at 120°C for 30 min. The electric field was maintained until the sample was cooled to 50°C. After poling, the samples were short-circuit at 40°C to remove the injected charges.

The piezoelectric coefficient, d_{33}, was measured by a d_{33} meter (ZJ-3B) 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 [13]. 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 an ultrasonic immersion method [14].

Table 2 Room-temperature elastic properties of cement paste with a water:cement ratio of 0.5.

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

3 Results and discussion

Table 1 shows the properties of the PZT, cement paste and normal concrete measured or found in the literature. It is found 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 varied.

Five PZT ceramic/cement 1–3 composites with volume fractions of 0.05 to 0.22 have been fabricated and are

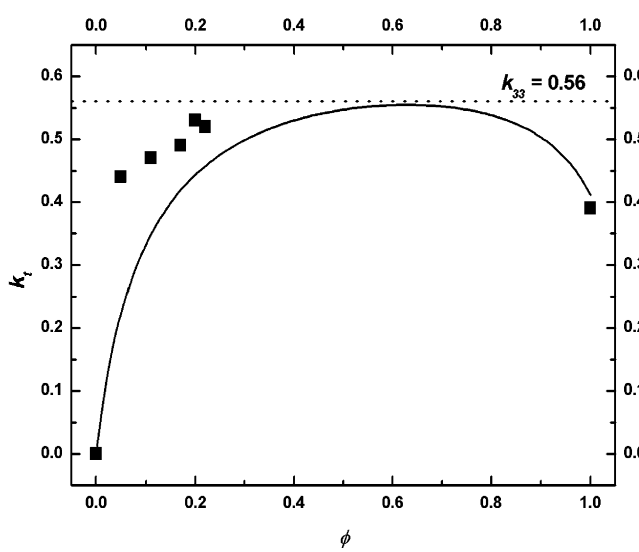

Fig. 4 Electromechanical coupling coefficient k_t of the PZT fibre/cement 1–3 composites as a function of ϕ. (*Symbols*: experimental data; *Line*: theoretical prediction)

shown in Fig. 1. The solid dots on the surface of the composites are the cross-sectional area of the ceramic fibres. Even the ceramic volume fraction ϕ is low ($\phi\sim$ 0.05), the resonance mode of the PZT fibre/cement 1–3 composites can still be observed in the measured electrical impedance and phase vs frequency spectra as shown in Fig. 2.

To compare the experimental data of the PZT fibre/cement 1–3 composites with the modified series and parallel model [9, 10], the piezoelectric, electromechanical and elastic properties of the PZT ceramics and the cement paste with the water: cement ratio of 0.5 shown in Tables 1 and 2 have been used in the calculation. The piezoelectric coefficient d_{33} and acoustic impedance Z_a of the composites is determined by equations (1) and (2):

$$d_{33} = \frac{\phi\, d_{33,\,\text{ceramic}}\, s_{11,\,\text{cement}}}{\phi\, s_{11,\,\text{cement}} + (1-\phi)\, s^E_{33,\,\text{ceramic}}} \tag{1}$$

$$Z_a = \sqrt{\rho \cdot c^D_{33}} \tag{2}$$

where s is the elastic compliance and c is the stiffness constant. It is seen that both the piezoelectric coefficient d_{33} and acoustic impedance Z_a of the composites increase monotonically with ϕ because of the increase contribution of the active ceramic phase as shown in Fig. 3. One of the advantages of the 1–3 connectivity is the effective enhancement of the electromechanical coupling coefficient in the thickness direction k_t and its theoretical value can be determined from the stiffness constants c using Eq. (3):

$$k_t = \sqrt{1 - \frac{c^E_{33}}{c^D_{33}}} \tag{3}$$

where

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

$$c^D_{33} = c^E_{33} + \frac{\phi^2\left[e_{33,\,\text{ceramic}} - \frac{2\, e_{31,\,\text{ceramic}}\left(c^E_{13,\,\text{ceramic}} - c_{12,\,\text{cement}}\right)}{C(\phi)}\right]^2}{\phi\left[\varepsilon^S_{33,\,\text{ceramic}} - \frac{2\, e_{31,\,\text{ceramic}^2}}{C(\phi)}\right] + (1-\phi)\, c_{11,\,\text{cement}}} \tag{5}$$

$$e_{33,\,\text{ceramic}} = k_{t,\,\text{ceramic}}\sqrt{c^D_{33,\,\text{ceramic}}\varepsilon^s_{33,\,\text{ceramic}}} \tag{6}$$

$$e_{31,\,\text{ceramic}} = \frac{\left(\varepsilon^T_{33,\,\text{ceramic}} - \varepsilon^s_{33,\,\text{ceramic}} - d_{33,\,\text{ceramic}} e_{33,\,\text{ceramic}}\right)}{2 d_{31,\,\text{ceramic}}} \tag{7}$$

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

where $\varepsilon^T_{33,\,\text{ceramic}}$ is the constant stress relative permittivity measured at 1 kHz and $\varepsilon^S_{33,\,\text{ceramic}}$ is the constant strain relative permittivity measured at high frequency (~2*f_a). Figure 4 shows that k_t of the composites can reach 0.50 even with $\phi\sim0.05$ which approaches to its k_{33} value ($k_{33}\sim0.56$).

By changing the ceramic volume fraction, the acoustic impedance of the composites can be tailored to better match that of the civil engineering structural materials. Because of the low ceramic volume fraction, the acoustic impedance of the composites can approach to that of the normal concrete (~9 MRayl) at $\phi\sim0.15$. As shown in Figs. 3 and 4, the experimental data agree quite well with the theoretical prediction. Because of the increase contribution of the active PZT phase, d_{33} and Z_a of the 1–3 composites increase with ϕ. It is also seen that the enhancement of k_t is effective even the matrix is cement and ϕ is low.

4 Conclusion

PZT fibre/cement 1–3 composites with low PZT volume fractions have been fabricated successfully. The performance of the composites was characterized by the resonance method. PZT fibre/cement 1–3 composites studied in this work have good performance and their properties agree quite well with the prediction of the modified series and parallel model. At a low ceramic volume fraction of ~0.15, the acoustic impedance of the composites approached that of concrete.

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