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Dielectric and electromechanical properties of modified cement/polymer based 1–3 connectivity piezoelectric composites containing inorganic fillers

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Here the modified cement/polymer based 1–3 piezoelectric composites containing inorganic fillers were prepared. The influences of filler content and composite thickness on dielectric and electromechanical properties of the composites were investigated. The results indicate that the composites with fillers of graphite and strontium ferrite (Sr-ferrite) have larger static relative dielectric constant and smaller dielectric loss than those with silicon powder. The strontium ferrite can improve the thickness electromechanical coupling property of the composites. When the strontium ferrite content is 5 wt.% of the cement/polymer matrix, the piezoelectric composite has the largest thickness electromechanical coupling coefficient of 64.27% and the smallest mechanical quality factor 8.64. The graphite filler shows a certain suppression effect on the planar resonance of the composites. The coupling effects between the planar and thickness modes of the composites were reduced by decreasing the composite thickness, and pure thickness mode resonance can be obtained at a small composite thickness.

1. Introduction

Presently, it is gaining increasingly popular to employ various intelligent materials, systems and structures to perform the real-time health monitoring or nondestructive evaluation of the civil engineering structures [1–3]. Piezoelectric ceramic is one of the most important intelligent materials due to its positive and negative piezoelectric effects, which are being widely used in many fields such as mechanical engineering, aeronautical and space engineering and medical engineering. However, the traditional piezoelectric ceramic materials encountered problems when they were used in concrete structures, such as poor interfacial matching ability, poor acoustic impedance mismatch between piezoelectric ceramic and concrete materials. In the last decade, cement based piezoelectric composite was developed by using piezoelectric ceramic as active phase and cement paste as passive matrix phase [4–7], which exhibits potential application prospect in health monitoring field of concrete engineering structures due to its good piezoelectric properties and acoustic impedance matching ability with concrete.

Recently, various cement based piezoelectric composites have been developed based on the connectivity of piezoelectric phase and matrix phase in the two-phase piezoelectric composite [8], such as 0–3 [4,7,9–11], 2–2 [12–14] and 1–3 [6,15]. The 1–3 connectivity cement based piezoelectric composite was especially concerned due to the superior piezoelectric, electromechanical coupling and acoustic impedance matching properties. Lam et al. [6] fabricated 1–3 cement based piezoelectric composite and investigated the electromechanical properties of the composite at a low volume fraction of piezoelectric ceramic. Potong et al. [15] fabricated the 1–3 cement based piezoelectric composite by using non-Lead Barium Zirconate Titanate as active phase, and they also analyzed the dielectric and piezoelectric properties of this composite. In our previous study [16], we fabricated 2–2 and 1–3 piezoelectric composites with cement matrix and cement/polymer matrix, and discussed their ultrasonic application in monitoring of cement hydration reaction process. Presently, cement was usually used as matrix of the cement based piezoelectric composites due to it similar properties with concrete materials. However, it is known that there exists a poor interfacial bonding ability between cement matrix and piezoelectric ceramic due to the hydration shrinkage effects, which therefore results in the performance deterioration of the cement based piezoelectric composite. Furthermore, because the acoustic impedance of cement paste is

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close to that of concrete, it needs to decrease the volume faction of piezoelectric ceramic in the composites to meet the acoustic impedance matching requirement between the cement based piezoelectric composite and concrete materials, thus the properties of the composites will also correspondingly be reduced due to the low volume faction of piezoelectric ceramic. Therefore, it is important to modify the matrix composition of the traditional cement based piezoelectric composites for improving the interfacial, piezoelectric and acoustic properties of the composite.

Here the epoxy resin which has superior binding ability and low acoustic impedance value was especially considered as the modifying material of cement matrix, and graphite powder, silicon powder and strontium ferrite were used as inorganic fillers of cement matrix to improve the properties of the composites. The influences of filler content and composite thickness on dielectric and electromechanical coupling properties of the novel cement/polymer based piezoelectric composites were mainly discussed in this research. The significance of this research is to explore an effective method for solving the disadvantage of traditional cement based piezoelectric composites in SHM (i.e., structural health monitoring) application of concrete.

2. Experimental method

PZT (Lead Zirconium Titanate) ceramic was employed as the active phase, and the ordinary Portland 42.5R cement and epoxy resin were selected as the matrix phase of the cement/polymer based piezoelectric composites. The relative dielectric constant and dielectric loss of cement/polymer matrix are about 11 and 0.35, respectively. Additionally, three types of inorganic powder of low relative dielectric constant were used as the filler of the piezoelectric composites, that is, graphite powder (carbon content $\geqslant$ 99.85, particle size $\leqslant 30\ \mu m$, relative dielectric constant $\approx$ 10), silicon powder (silicon content $\geqslant$ 99.0, particle size = 74um, relative dielectric constant $\approx$ 3), and strontium ferrite (particle size = 1.5 μm, relative dielectric constant $\approx$ 7). The main properties of PZT ceramic are listed in Table 1.

The piezoelectric composites were fabricated by secondary dicing and filling method through the following procedures. The diamond cutting machine with a blade thickness of 0.35 mm was used to cut the piezoelectric ceramic block. A casting mould with dimensions of 12 mm $\times$ 12 mm $\times$ 10 mm was used for casting matrix materials, and a vacuum chamber was used to eliminate the air induced into the matrix of the composite. First, series of piezoelectric ceramic sheets with a width of 1 mm were cut accurately by using the diamond cutter along a direction parallel to the polarization axis of piezoelectric ceramic block. The spacing between piezoelectric ceramic sheets is 0.5 mm, and a ceramic foundation with a height of 0.5 mm was left to maintain the upper ceramic sheets. The piezoelectric ceramic body was then put into the ultrasonic cleaner for 10 min to clean the ceramic residue. After drying in the air, the ceramic body was fixed into the mould for the next casting. Cement, epoxy resin and hardener were weighed and mixed according to a mass proportion of 4:4:1. Then, the graphite powder, silicon powder and strontium ferrite powder with mass percentages of 1 wt.%, 5 wt.%, 10 wt.%, 15 wt.% of the mixture of cement and polymer were added into the cement/polymer mixture, respectively. The cement/polymer mixture containing inorganic fillers was put into the vacuum chamber for about 10 min to eliminate the pores, and then was poured into the piezoelectric ceramic body. After solidifying for 48 h in the air, the piezoelectric composite bodies were taken out of the mould, and the incision procedure was performed again along the direction perpendicular to the first incision direction. The processes of casting, vacuumizing and solidifying were repeated. Finally, the 1–3 cement/polymer based piezoelectric composites were fabricated after cutting off the ceramic foundation. The dimensions of the piezoelectric composites were 10 mm (±0.2 mm) in length, 7.5 mm (±0.2 mm) in width. The composite thickness was 10 mm, 8 mm, 6 mm and 4 mm, respectively.

Table 1
Properties of the piezoelectric ceramic.

Ceramic	k_p (%)	k_t (%)	d_{33} (pC·N^{-1})	ε_r	tan δ	Q_m	ρ (10^3 kg m^{-3})
PZT	58	48	260	1050	<0.3%	1000	7.5

(*Note:* k_p-planar electromechanical coupling coefficient; k_t-thickness electromechanical coupling coefficient; d_{33}-piezoelectric strain constant; ε_r-relative dielectric constant; *tan* δ-dielectric loss; Q_m-mechanical quality factor; ρ-density.)

3. Results and discussion

3.1. Effects of filler content on properties of the composite

3.1.1. Dielectric properties

For testing purpose, the upper and lower surfaces of the cement/polymer based piezoelectric composite perpendicular to the poling direction were polished using the Al_2O_3 grinding medium and cleaned by acetone, and then a thin layer of silver paint was coated on the surfaces as electrodes. An impedance phase analyzer (Agilent 4294A, America) was used to measure the dielectric properties of the composites. The relative dielectric constant (ε_r) can be calculated in terms of the following equation,

$$\varepsilon_r = C_p t/(A\varepsilon_0) \quad (1)$$

where C_p is capacitance; t and A are the thickness and electrode area of the composites, respectively; ε_0 is vacuum permittivity with value of 8.85×10^{-12} Fm^{-1}.

In order to investigate the influence of filler content on dielectric properties of the composites, the piezoelectric composites with a thickness of 4 mm were especially discussed here. The relative dielectric constant and dielectric loss of the composites as a function of frequency are illustrated in Fig. 1. It can be seen that there exist two obvious dielectric peaks in the relative dielectric constant vs. frequency spectra, which are mainly contributed by the piezoelectric ceramic. The weak dielectric peak around 120 kHz is mainly due to the planar mode resonance of the piezoelectric composites, and the strong dielectric peak around 420 kHz is due to the thickness mode resonance of the composites. Above phenomena can be explained by the piezoelectric effect and polarization effect of the piezoelectric composites. It is known that piezoelectric material can generate polarization effect under the external electric field. When the frequency of external electric field is similar to the resonance frequency of the piezoelectric composites, there will generate amounts polarization charge in the composites, which results in the increase of the relative dielectric constant around the resonance frequency. The thickness mode resonance of the composites is stronger than the planar mode resonance, therefore, the dielectric peak at the thickness mode is higher than that at the planar mode. In addition, it also can be observed from Fig. 1(a)–(c) that the relative dielectric constant of the piezoelectric composites are dependent of the inorganic fillers. The relative dielectric constant of the composites containing graphite powder shows the decreasing trend with increasing the graphite powder content, while that of the composites containing Sr-ferrite powder shows the increasing trend with increase of the Sr-ferrite content. Nevertheless, the influence of silicon powder on dielectric constant of the composites is not obvious.