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Piezoelectric and dielectric behavior of 0-3 cement-based composites mixed with carbon black

Hongyu Gong[a,*], Zongjin Li[b], Yujun Zhang[a], Runhua Fan[a]

[a] *Key Laboratory for Materials Structure and Heredity of Education Ministry, Shandong University, Jingshi Road 73, Jinan 250061, China*
[b] *Department of Civil Engineering, the Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong*

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Abstract

Cement-based 0-3 type piezoelectric composites, fabricated from white cement, lead zirconate titanate (PZT) powder and small amounts of carbon black, are exploited for potential applications in civil engineering. The nature of the aging and poling behavior of the carbon black/PZT/cement composites is investigated. It is shown that the use of a conductive carbon black phase can facilitate the poling process at room temperature. The influences of the carbon black on the piezoelectric and dielectric properties are studied as well. It is demonstrated that the piezoelectric sensitivities of the composites can be dramatically enhanced by incorporation of small amounts of carbon black, due to its conductive properties. With a poling field of 4 kV/mm and poling time of 20 min, the d_{33}, ε_r, $\tan\delta$ and K_t values of the composite containing 70 vol.% PZT and 1.0 vol.% carbon black are 28.5 pC/N, 202.6, 0.19 and 12.2%, respectively.

Keywords: Cement-based composites; PZT; Carbon; Piezoelectric properties; Dielectric properties

1. Introduction

In recent years, new types of cement-based piezoelectric composites have been exploited as promising materials for sensors and actuators in the field of civil engineering.[1–3] Their performances show some advantages over traditional piezoelectric materials, such as piezoelectric ceramics and polymer-based piezoelectric composites. The piezoelectric ceramics, mainly lead zirconate titanate (PZT), exhibit high dielectric constant and density, which lead to a high acoustic impedance when compared with that of concrete, the most common host material in civil infrastructure. The significant difference in acoustic impedance between a transducer and its environment can cause considerable loss of the signal transmission at the boundaries. By integrating ceramic powders with cement, Li's group at HKUST developed, for the first time, 0-3 cement-based piezoelectric composites, which had reasonable piezoelectric activities as well as a similar acoustic impedance to concrete.[4,5] Compared with polymer-based piezo-composites, which also have low acoustic impedances, the cement-based composites have the advantage of excellent interface compatibility with civil engineering structures because of the utilization of cement as matrix.

Piezoelectric composites have been designed with many different connectivity patterns, among which the 0-3 connectivity is the simplest.[6] A 0-3 cement-based piezoelectric composite consists of a three dimensionally connected cement loaded with active piezoelectric ceramic particles connected in zero dimensions. In the pioneering work of Li et al.[1] the 0-3 piezoelectric composites were fabricated by embedding PZT ceramic particles into hydrated white cement, using a normal mixing and spreading method. The composites were poled under an electric field of 2 kV/mm at 160 °C for 1 h. The piezoelectric strain factors (d_{33}) of the composites with PZT contents from 35 to 70% by volume were in the range of 7–33 pC/N. Using a pressing method, Huang et al.[7] developed 0-3 PZT/sulphoaluminate composites containing up to 85% PZT by weight (∼70% by volume). The highest d_{33} value was reported to be 16 pC/N for the 80% PZT composite, which was polarized under 4 kV/mm at 130 °C for 45 min. Chaipanich[8] used a similar pressing method to produce a 0-3 composite from PZT and Portland cement under a poling regime of 2 kV/mm, 130 °C and 45 min. The

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[*] Corresponding author. Tel.: +86 531 88392439; fax: +86 531 88392439.
E-mail addresses: hygong@sdu.edu.cn, gong_hongyu@yahoo.com.cn (H. Gong).

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composites had d_{33} values of 14–43 pC/N for PZT contents of 30–90 vol.%.

The properties of the PZT/cement composites have been found to depend on many factors such as the poling conditions, pressure of formation, PZT particle size, etc. Among these, the poling efficiency of the PZT particles is crucial for improving the properties of the composites as the piezo-active ceramic phase in the composite contributes substantially to the overall piezoelectric properties. The ceramic particles, however, do not form a continuously connected structure across the inter-electrode dimension. Therefore, how to perform an effective poling is a challenging issue for the development of 0-3 cement-based composites.

For a piezoelectric composite with 0-3 connectivity, the electric field (E_1) acting on the ceramic particles is related to the externally applied electric field (E_0) by $E_1 = 3\varepsilon_2 E_0/(\varepsilon_1 + 2\varepsilon_2)$, where ε_1 and ε_2 are the dielectric constants of the piezoelectric filler and the matrix, respectively.[9] Because the dielectric constants of PZT ceramics are much higher than those of cements, the electric field acting on the piezoelectric particles is quite low and insufficient for poling of the ceramics. Therefore, to facilitate the poling of the ceramic particles in the composites, one has to increase the poling field and the poling temperature. Nevertheless, too high a poling voltage can break down the samples and high poling temperature can weaken the mechanical properties of the cement. Alternatively, by adding a small amount of a conductive additive such as graphite, germanium or silicon, the electrical conductivities of the composite can be adjusted and more continuous electrical flux paths established between ceramic particles. Then the poling process can be carried out much more easily.[10]

The piezoelectric properties of ferroelectric materials after poling may degrade with time, which is defined as aging. That is because of the release of internal stresses and rearrangement of ferroelectric domains. But, so far, studies of the aging behavior of cement-based composites have been limited. Therefore, it is highly desired to investigate the aging characteristics of the cement-based piezo-composites and find ways to produce more stable piezoelectric materials for practical application.

In this paper we report a novel carbon black/PZT/cement system which consists of white cement as a matrix, homogenously distributed PZT particles as a ferroelectric phase, and carbon black added as a conductive phase. The aging and poling conditions of the composites are investigated. The influences of carbon addition on the dielectric and piezoelectric properties of the composites are then examined.

2. Experimental procedures

It has been reported that the ceramic particle size can affect the ferroelectric properties of 0-3 piezoelectric composites. The composites with larger PZT particles had higher d_{33} values, but also higher dielectric losses.[11] In the present study, PZT powders were obtained by ball milling commercial PZT ceramic pieces (Hong Kong Piezo Co. Ltd.) and then sieving through nylon meshes to get a particle size fraction from 38 to150 μm. The remained larger and smaller particles were discarded. The white cement used in the composites was from the Guangdong First Building Materials Factory, China. The mean particle size is about 16 μm. The basic properties of the PZT ceramic and hardened white cement are listed in Table 1. The conductivity enhancer was carbon black (Vulcan XC-72 from Cabot) of density 1.8 g/cm^3 and average particle size 70 nm.

Table 1
Properties of the PZT ceramic and the hardened white cement.

Parameters	PZT ceramic	White cement
Piezoelectric strain factor, d_{33} (10^{-12} C/N)	650	–
Piezoelectric voltage factor, g_{33} (10^{-3} Vm/N)	19.3	–
Dielectric constant, ε_r (at 1 kHz)	3800	56
Electromechanical coupling coefficient, K_p	0.63	–
Mechanical quality factor, Q_m	70	–
Density, ρ (10^3 kg/m^3)	7.6	3.15

To prepare the composite specimens, the PZT, cement and carbon black powders were firstly well mixed using acetone as solvent. The volume fraction of the PZT powder in the composites was 70%, and the carbon black additions were from 0 to 1.7 vol.%. After the volatilization of the acetone, weighted mixture was compressed in a stainless steel mound under a pressure of 100 MPa to form a disc of diameter 14 mm and thickness 1.5 mm. The discs were then put into a curing chamber at 60 °C and relative humidity 100% for 24 h to speed up the hydration process of the cement matrix. After drying the specimens, silver paste was coated on both sides of the discs to form electrodes. Finally, for piezoelectric activation, the samples were poled along the thickness direction in a silicon oil bath under a poling filed of 2–6 kV/mm for 20–100 min at ambient temperature.

The piezoelectric strain factors (d_{33}) were measured by a d_{33} meter (Model ZJ-3B, Institute of Acoustics Academia Sinica, China). The impedance spectra were determined by an impedance/grain-phase analyzer (Model 4294A, Hewlett Packard, Tokyo, Japan). The capacitance and dielectric loss were examined by the same apparatus at 1 kHz. The relative dielectric constants (ε_r) and piezoelectric voltage factors (g_{33}) were calculated using the relationships:

$$\varepsilon_r = \frac{C_p t}{\varepsilon_0 A}$$

$$g_{33} = \frac{d_{33}}{\varepsilon_0 \varepsilon_r}$$

where C_p is the sample capacitance at 1 kHz, t is the thickness, A is the electrode area, and ε_0 is the vacuum dielectric constant (8.85×10^{-12} F/m). The thickness electromechanical coupling coefficients (K_t) were calculated according to the following equations[12]:

$$K_t^2 = \frac{\pi}{2}\frac{f_s}{f_p} \tan\left(\frac{\pi}{2}\frac{f_p - f_s}{f_p}\right)$$

where f_s and f_p are the series resonant frequency and the parallel resonant frequency, respectively, which can be replaced by the frequencies at the minimum and maximum impedances (f_n and f_m) in the fundamental resonant region of impedance

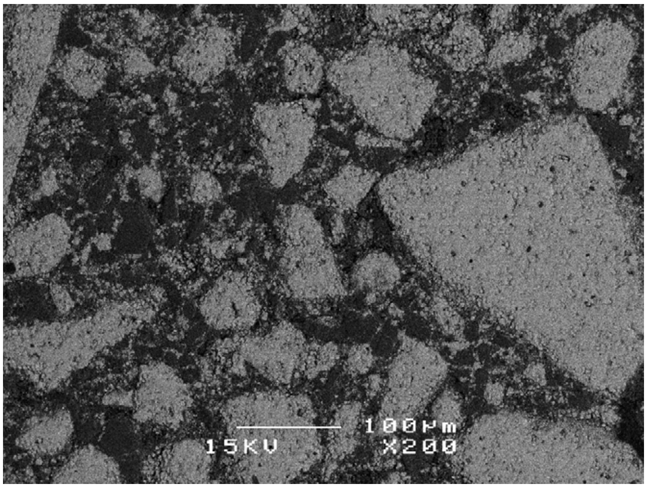

Fig. 1. Backscattered SEM image of polished surface of the carbon black/PZT/cement composite.

spectrum. The planar electromechanical coupling coefficient (K_p) was obtained by using the curve of K_p versus $\Delta f/f_n$. The microstructure of the samples was observed by scanning electron microscopy (SEM, JEOL-6300F).

3. Results and discussions

Fig. 1 shows the backscattered SEM image of the polished surface of the PZT/cement composite with 1.0 vol.% carbon. It can be seen that PZT particles in light color with irregular shapes are dispersed evenly within the cement matrix in dark color. The nano-sized carbon black particles, however, are too small to be identified in this photo.

3.1. Aging behavior

The variation of piezoelectric strain factor, d_{33}, of the carbon black/PZT/cement composites as a function of aging time is shown in Fig. 2. The samples were all polarized under a DC electric field of 4 kV/mm for 20 min at room temperature. It can be seen that the piezoelectric coefficients of the composites were sensitive to the carbon black addition. For the composite without carbon, the d_{33} values were low and stable. At the carbon black contents of 0.2–1.0 vol.%, the d_{33} fluctuate in the first 10 days after poling and then decrease slowly over the following 10 days. In order to accelerate the aging process, all specimens were annealed at 70 °C for 72 h under a short-circuit condition on the 23rd day. The d_{33} values of the composites fell dramatically during the heating period, then increased slowly and approached a relatively stable state after the 41st day. It is also observed that the d_{33} values of the composite with 1.7 vol.% carbon black are lowest and show little change during the whole aging period.

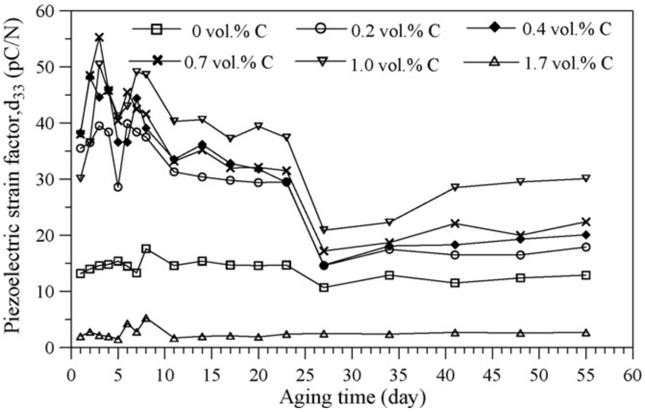

Fig. 2. Variations of piezoelectric strain factor (d_{33}) of the composites with the aging time.

Aging is an intrinsic property of piezoelectric ceramics and mainly relates to the rearrangement of ferroelectric domains. Under an externally applied high voltage, the 90° and 180° ferroelectric domains of piezoelectric ceramics are forced to rotate so as to be parallel to the direction of the electric field. The orientation of the 90° domains will generate great internal stress on domain walls, which in turn means excites the material to a high-energy non-equilibrium state. When the poling field is removed, the 90° domains have a tendency to revert to random states to release the residual internal stresses, after which the piezoelectric properties degrade gradually with time.[13] However, the PZT/cement piezo-composites exhibit a very different aging behavior from the piezo-ceramics. As demonstrated in Fig. 2, there is no reduction of d_{33}. On the contrary, an increase of d_{33} values from 13.2 pC/N on the 1st day to 14.6 pC/N on the 23rd day was observed. The slight increase of d_{33} value of cement-based piezo-composite with aging has been reported by Li et al.[5] and Cheng et al.[2] Cheng believed it might be due to the increase of interfacial stresses between cement and ceramic particles as cement undergoing hydration proceeds.

Carbon black has a strong effect on the aging of the composites. With the addition of 0.2–1.0 vol.% carbon black the d_{33} value showed a remarkable increase but considerable fluctuations for several days after poling (Fig. 2). On one hand, the significant increase of d_{33} values indicates that more ferroelectric domains of the piezoelectric ceramic fillers were orientated. On the other hand, the fluctuation of d_{33} values is supposed to be related to the structural characteristics of cement. Hydrated cement is generally considered as a porous inorganic material. Many kinds of ions, such as Ca^{2+}, $SO_4{}^{2-}$, Na^+ and Al^{3+} are released during the cement hydration process. These ions are dissolved in the electrolyte and adsorbed on to the solid surfaces of the pores. They present opposite charges to that of the solid surface on to which they are absorbed. Due to the presence of ion pairs in this double-electric layer, electric dipoles are formed in the cement paste.[14,15] Furthermore, the mobility of these ions may be accelerated by the introduction of the conducting carbon black phase. Therefore, the possibility of reorientation of the electric dipoles within the cement matrix during poling may be enhanced. Very high d_{33} values of about 40–50 pC/N are measured for the composites with carbon black in the first several aging days. However, such polarization of the ion dipoles in cement is believed to be less stable than that of the ferroelectric domains in PZT ceramic. As a result, the d_{33} values of the composites would fluctuate and irreversibly decrease during the aging process. After the accelerated aging process, the remanent charges and the temporary alignment of dipoles in the cement

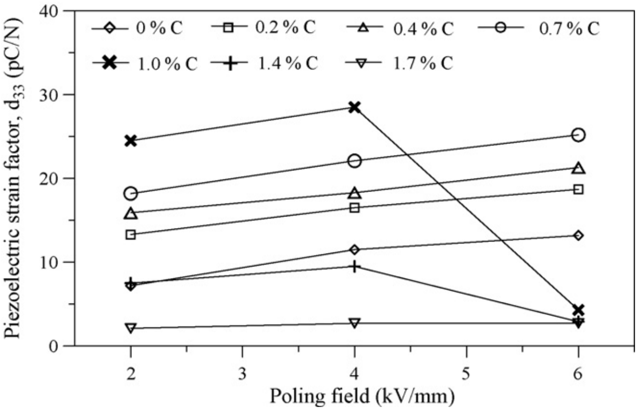

Fig. 3. Dependence of the piezoelectric strain factor (d_{33}) of the carbon black/PZT/cement composites on the poling field.

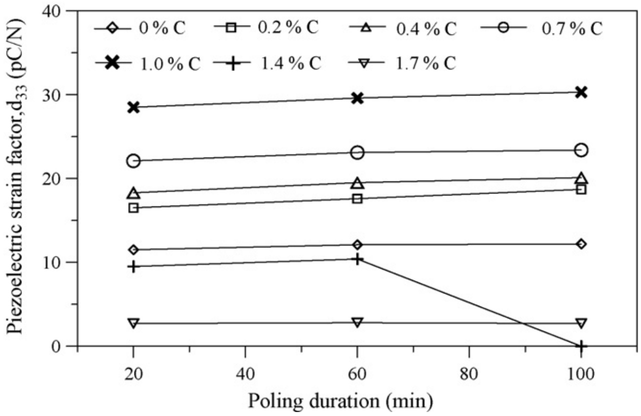

Fig. 4. Dependence of the piezoelectric strain factor (d_{33}) of the carbon black/PZT/cement composites on the poling duration.

matrix might be lost, giving a corresponding decrease in d_{33} followed by longer term constant values.

The composite with 1.7% carbon black showing very low d_{33} value may due to the excessive carbon black amounts. The addition of carbon black can improve conductivity of the composites and facilitate the poling process, but too much carbon black may breakdown the insulation of the composites. With 1.7% carbon black, the composite cannot endure the applied high voltage any more. Therefore, the embedded PZT particles would not be polarized effectively, and then the composite shows inferior piezoelectric properties.

3.2. *Poling effects*

As mentioned before, the piezoelectric ceramic particles embedded in the 0-3 cement-based piezoelectric composites are difficult to be polarized because only part of external electric field acts on the piezoelectric phase due to the large differences in resistivity and permittivity between the ceramic powders and the cement matrix. In order to improve the poling efficiency of the composites, elevated poling temperatures (above 120 °C) and higher poling fields are normally required. Unfortunately, the physical and mechanical properties of cement may be damaged when it is subjected to such a temperature rise. It has been reported that cracks appeared and total porosity increased in cement materials when heated up to 100 °C because of the dehydration of C–S–H gel, which was the main product in the hydrated cement and was thus mainly responsible for its mechanical properties.[16] In this study, it is found that the poling process can be facilitated by the introduction of carbon black, which allowing poling to be conducted at room temperature.

In order to investigate the influence of the poling field on the value of d_{33}, the poling time was fixed at 20 min, and the d_{33} values were measured when the composites were in a stable state, that is, on the 41st day after poling. The dependence of d_{33} on poling field is shown in Fig. 3. It can be seen that d_{33} increases with increasing poling field, as expected. The poling electric field is the driving force which causes domains in PZT particles to switch and reorientate, causing the composites to exhibit piezoelectric properties. A higher poling field generally yields higher d_{33} values. However, as the adding amounts of carbon black increase, the leakage current in the composite would increase and the insulation of the composites decrease accordingly. When being polarized at 6 kV/mm, the composites containing carbon black equal to or larger than 1.0% cannot tolerate such high external electric field, and dielectric breakdowns take place. Thus, the d_{33} values decrease dramatically.

Fig. 4 shows values of d_{33} for the composites as a function of poling time under a poling field of 4 kV/mm. The values of d_{33} increase slightly with increasing poling time. In general, 180° domain switch is easer than 90° domain switch, and takes place over a short period of time, during the initial period of poling. By prolonging the poling time, more reorientations of 90° domains can be initiated and the piezoelectric properties are improved accordingly. In this work, the increase of d_{33} with poling time from 20 to 100 min is very limited, indicating that a poling time of 20 min was long enough for the ferroelectric domains to be oriented. Meanwhile, too long a poling time increases the possibility of dielectric breakdown for the composite with higher amounts of carbon black.

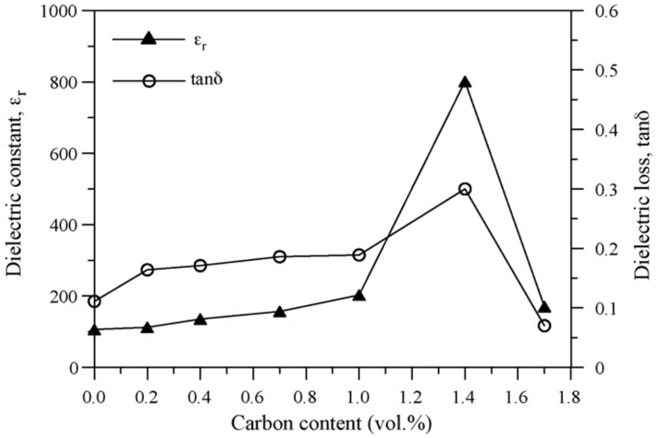

Fig. 5. Dependence of the dielectric constant (ε_r) and dielectric loss ($\tan\delta$) of the carbon black/PZT/cement composites on the carbon content.

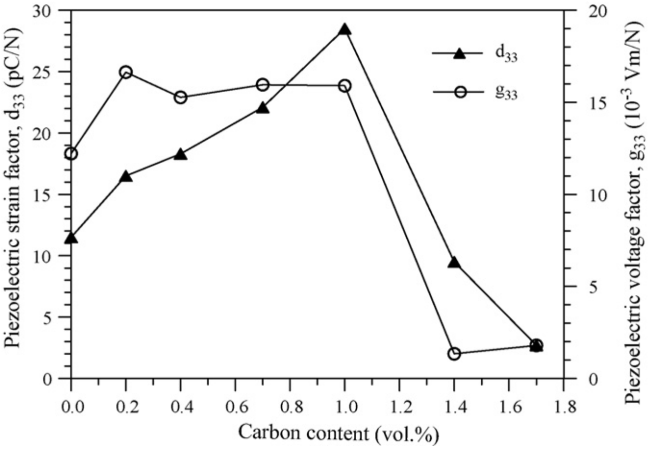

Fig. 6. Dependence of the piezoelectric strain factor (d_{33}) and piezoelectric voltage factor (g_{33}) of the carbon/PZT/cement composites on the carbon content.

3.3. Dielectric and piezoelectric behavior

The influence of carbon content on the dielectric constant (ε_r) and dielectric loss (tan δ) at 1 kHz are plotted in Fig. 5. The values were obtained on the 41st day after poling. It can be seen that the dielectric constant increases from 106.4 at 0 vol.% of carbon black to 202.6 at 1.0 vol.%, then jumps to 801.1 at 1.4 vol.%, finally decreases to 170.0 at 1.7 vol.%. Basically, the dielectric constant results from permanent dipole motions within a system, indicating the poling efficiency of the composites. By adding a small amount of conductive additives (1.0 vol.%), the electrical conductivities of the composite can be improved. Therefore, the electric field acting on the ceramic phase increases and the ceramic particles can be poled effectively.

A sudden increase of the dielectric constant at 1.4 vol.% carbon black can be explained by using the theory of percolation [17]. From the electrical transport viewpoint, the carbon black/PZT/cement composites can be approximately considered as a two-phase system containing conductive carbon black and insulating PZT/cement mixture matrix, which would exhibit a metal-insulator transition with increasing conductive filler concentration. The metal–insulator transition is usually characterized by an abrupt discontinuity in the conductivity and a rapid change in dielectric constant in the neighborhood of the percolation threshold where the conductive particles begin to coalesce into a network.[17,18]

The dielectric loss (tan δ) demonstrates a similar feature to the dielectric constant (ε_r) as shown in Fig. 5. The enhancement in the dielectric loss at 1.0 vol.% is related to the increase of the electrical conductivity of the composite. With 1.4 vol.% of carbon black, there is a sudden jump at around the percolation threshold just as in the case of the dielectric constants. According to the percolation threshold theory, at the threshold value (about 1.4% in this study), the carbon black particles begin to come into contact in the composite. In this case, the leakage current reaches to its maximum and the composite exhibit very high dielectric loss. After the threshold point, more carbon black particles may connect to form a conductive network. As a consequence, the insulation of the composite is broken, and

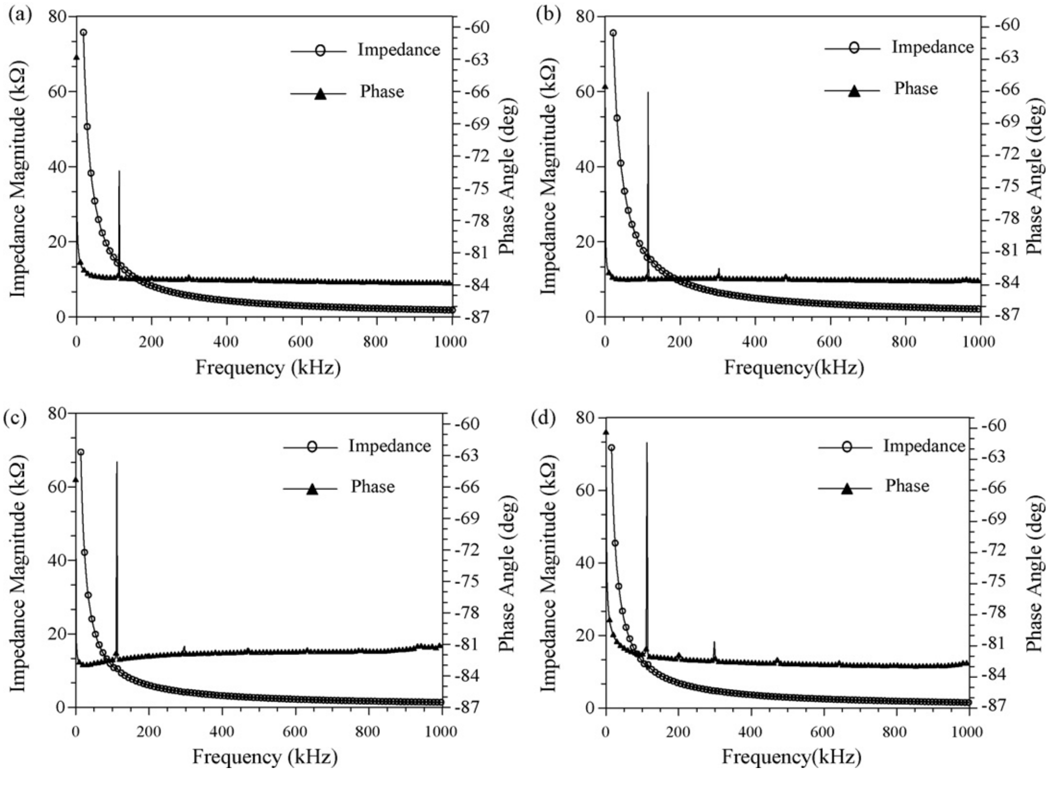

Fig. 7. The impedance/phase versus frequency spectra of the carbon black/PZT/cement composites with (a) 0 vol.% and (b) 0.2 vol.%; (c) 0.7 vol.% and (d) 1.0 vol.% carbon black.

Table 2
The electromechanical coupling coefficients of the carbon black/PZT/cement composites.

Carbon content (vol.%)	K_t (%)	K_p (%)
0	9.47	11.19
0.2	10.51	11.23
0.4	10.79	11.23
0.7	10.46	12.25
1.0	12.16	13.26

the composite becomes a quasi-conductor with low dielectric constant.

The influence of carbon black on the piezoelectric strain factor (d_{33}) of the composites after aging for 41 days is shown in Fig. 6. The d_{33} values of the composites increase with the increase of the carbon black. The highest d_{33} value is 28.5 pC/N for the composite with 1.0 vol.% carbon black, which is more than twice that of the composite without carbon black. Small amounts of carbon black particles can create electric paths between PZT particles and improve the poling efficiency of the PZT phase. However, when the added carbon exceeds 1.4 vol.%, the piezoelectric activity of the composite are greatly reduced, suggesting too much connectivity between the PZT particles in the composites. High dielectric loss at this stage makes it difficult to apply the voltage required for poling PZT particles because the electric energy is dissipated by large leakage currents, hence the piezoelectric activity decreases.

The variation of piezoelectric voltage factor (g_{33}) with carbon black volume fraction is also plotted in Fig. 6. The g_{33} values were calculated using the measured values of d_{33} coefficients and the dielectric constants of the composites. The composites containing carbon black from 0.2 to 1.0 vol.% show high g_{33} values. This is a consequence of the more rapid increase in d_{33} than that in ε_r. The very low g_{33} for the composite with 1.4 vol.% is due to the relatively low d_{33} coefficient and very high dielectric constant.

Fig. 7 shows the impedance/phase versus frequency spectra of the composites. The peaks on the impedance curves correspond to the piezoelectric and reverse piezoelectric effects of the cement-based composites. The impedance peak intensity increase with increasing carbon black content and almost doubled at 1.0 vol.%. For the composites containing more than 1.0 vol.% of carbon black, however, there is no peak observed on the impedance and phase curves. This is in good agreement with the experimental results of the piezoelectric coefficient measurement. That is, at the saturation point of about 1.4 vol.%, excessive carbon black increases too much of conductivity of the composites, leading to a very low d_{33} value and thus no obvious impedance peak can be detected.

Finally, the calculated thickness and planar electromechanical coupling coefficients of the composites are listed in Table 2. It can be seen that both K_t and K_p values increase with increasing carbon volume fraction, indicating that the converse efficiency between electrical and mechanical energy is also improved by the carbon phase.

4. Conclusions

A novel three-phase piezoelectric composite with conductive carbon black and PZT piezoelectric ceramic particles embedded into the cement matrix, has been fabricated. The volume fraction of PZT was fixed at 70% and the carbon additions were in the range from 0.2 to 1.7 vol.%. The influence of carbon black on the dielectric and piezoelectric properties of the 0-3 cement-based piezo-composites was investigated. The d_{33} values of the composites fluctuated at the initial stage of the aging process, possibly due to the temporarily aligned dipoles in the cement matrix whose conductivity was enhanced by the introduced carbon balck. After being short-circuited at 70 °C for 72 h, the piezoelectric coefficients of the composites finally reached stable values after 41 days aging.

The effect of poling conditions on the piezoelectric properties of the carbon black/PZT/cement composites was investigated. The application of carbon black as a conductive phase was able to facilitate the poling process at room temperature. The optimum poling conditions for the composites were a poling field of 4 kV/mm and poling time of 20 min.

The piezoelectric strain coefficient (d_{33}), dielectric constant (ε_r), dielectric loss (tan δ) and electromechanical coupling coefficients (K_t and K_p) were determined as a function of volume fraction of carbon in the composite. It was found that a limited amount of carbon black additive of about 1.0 vol.% was optimum for enhancing the dielectric and piezoelectric properties of the composites. With this amount of carbon black added, the d_{33} coefficient increased by 1.5 times compared with the composite without the conductive filler. Too much of carbon black usage, however, reduced the piezoelectric activities of the composites due to its excess electric conductivity.

Acknowledgments

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