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Influence of water-to-cement ratio on piezoelectric properties of cement-based composites containing PZT particles

Huang Hsing Pan [a,*], Chi-Kuei Wang [a], Mang Tia [b], Yu-Min Su [a]

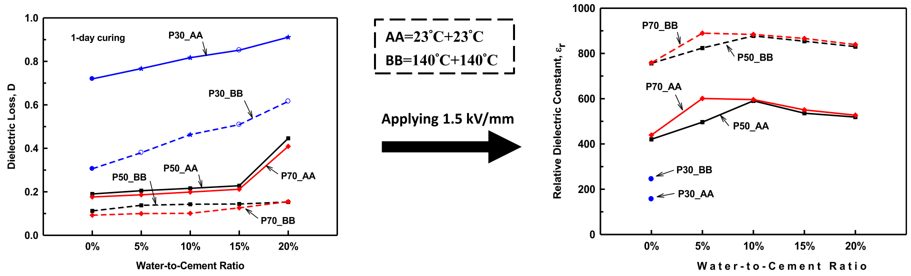
[a] Department of Civil Engineering, Kaohsiung University of Science and Technology, Kaohsiung, Taiwan
[b] Department of Civil and Coastal Engineering, University of Florida, Gainesville, FL, USA

HIGHLIGHTS

- Piezoelectric cement with a favorable w/c can enhance its piezoelectric properties.
- A criterion for the feasibility of the poling process is provided for PZT/cement composites.
- Piezoelectric cement containing 50% PZT and w/c = 10%, with no admixtures, has the d_{33} value of 133 pC/N.
- The thickness electromechanical coupling coefficient K_t is strongly dependent on w/c.

GRAPHICAL ABSTRACT

Dielectric loss of PZT/cement composites before polarization. Relative dielectric constant ε_r versus w/c at 100 days.



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ABSTRACT

To enhance piezoelectric properties, the effect of water-to-cement ratio (w/c) on 0–3 type cement-based composites containing 30–70% lead zirconate titanate (PZT) ceramic as the inclusion is investigated, where the w/c varying from 0% to 20%. Dominate factors that affect the facilitation of polarization and piezoelectric properties are also discussed. Results indicate that the composites with a higher w/c have higher values of porosity and dielectric loss, resulting in longer trigger times during the polarization. To facilitate polarization, the dielectric loss and the resistivity of the composites had better be less than 0.75 and 100 kΩ-m, respectively. Piezoelectric cement with a favorable w/c can improve its piezoelectric properties. The optimum w/c of the composites for 50% PZT (P50) and 70% PZT (P70) to obtain the highest piezoelectric strain factor d_{33} and relative dielectric constant ε_r values are 10% and 5%, respectively, and the values at the age of 100 days are $d_{33} = 133$ pC/N and $\varepsilon_r = 878$ for P50, and $d_{33} = 143.1$ pC/N and $\varepsilon_r = 890$ for P70. Moreover, the thickness electromechanical coupling coefficient (K_t) strongly depends on w/c, and the effect of temperature treatment on K_t becomes prominent with the increase in w/c.

1. Introduction

Cement-based piezoelectric composites have been developed and used as alternative piezoelectric sensors to monitor the health of concrete structures for two decades [1–10]. Lead zirconate titanate (PZT) ceramic is commonly used as the inclusion material in cement-based piezoelectric composites because of its effective piezoelectric properties compared with other piezoelectric materials such as barium titanate ($BaTiO_3$). The 2–2 and 1–3 types of cement-based piezoelectric composites comprising plate-like and bar-like PZT inclusions, respectively, have been developed and

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[*] Corresponding author.
E-mail address: pam@nkust.edu.tw (H.H. Pan).

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potentially used as cement-based piezoelectric sensors [1–2,7–11], exhibiting a high piezoelectric strain factor d_{33}. Similar to the 2–2 type cement–matrix composite, a smart aggregate comprising a PZT platelet embedded in the cement matrix [12] acting as a transducer has been applied for traffic monitoring in highways [13], crack propagation monitoring in RC beams [14], monitoring the effects of temperature and loading on concrete structures [15], and strength gain in concrete [16].

The 0–3 type of cement-based piezoelectric composite (also called piezoelectric cement), which has been developed and intends to apply as a piezoelectric transducer since 2002, is a two-phase composite with PZT particles randomly distributed in a cement matrix for diminishing the mismatch in acoustic impedance and volume compatibility between PZT inclusions and the cement matrix [3–5]. However, the 0–3 type composite has innately less piezoelectric behavior than the 2–2 type and 1–3 type composites at the same amounts of PZT inclusions because of the geometric alignments and distributions of PZT in the cement, for instance, d_{33} < 50 pC/N for piezoelectric cement containing 50% PZT and d_{33} > 150 pC/N for the 2–2 type and 1–3 type composites. For the applications of piezoelectric cement sensor, piezoelectric cement via frequency, mechanical-electric response and acoustic emission technology have been proposed applied for structural health monitoring (SHM) [17–22] in concrete structures. However, piezoelectric cement as sensors applied for SHM in conjunction with electromechanical impedance (EMI) technique are seldom reported due to its low piezoelectric properties.

Many studies [23–37] on the manufacturing process, polarization conditions, and admixtures have reported the enhancement of the piezoelectric strain factor d_{33}, dielectric constant ε, and thickness electromechanical coupling coefficient K_t of piezoelectric cements. For example, piezoelectric cement admixtures such as silica fume [38], carbon [39], carbon black [30,40], carbon nanotube [31], slag and fly ash [33,41], and kaolin [42] have been reported to be effective in increasing d_{33} and ε_r, when used in suitable amounts. During the poling process, specimens with a higher poling voltage and poling temperature [24,25,43] and appropriate poling time [23–25,44] typically yield higher d_{33} and K_t values than other specimens.

Adequate fabrication processing of samples of piezoelectric cement is another method of improving its piezoelectric properties, particularly d_{33} and ε_r. Previous studies have reported that a higher curing temperature [25,43,45] and forming pressure [8,46] can improve the piezoelectric properties of specimens. Those subjected to temperature pretreatment prior to the polarizing process exhibited lower values of dielectric loss (D), which enables specimens to be easily poled, thus imparting superior piezoelectric and dielectric properties [44,47]. Even though no mineral admixtures, the value of d_{33} for piezoelectric cement containing 50% PZT raised from 55 pC/N to 106 pC/N if pretreatment temperature on the specimen is 150 °C [47]. Compared with specimens cured for longer durations, those cured for 1 day at 90 °C and 100% relative humidity exhibited a higher polarization efficiency and higher d_{33} and ε_r; however, the opposite curing effect was found in the thickness electromechanical coupling coefficient K_t [48]. In addition, the specimens of piezoelectric cement subjected to a 140 °C pretreatment temperature and 1-day curing have been used as piezoelectric cement sensors, with d_{33} = 101 pC/N, that were embedded in concrete to monitor the strength development and detect the damage by using electromechanical impedance (EMI) technique [49]. EMI technique is one of monitoring methods for SHM based on electric impedance–frequency spectra variation of piezoelectric sensors [50,51]. Results indicate the piezoelectric cement sensor has the ability of concrete health monitoring, and the sensitivity of monitoring is even better than that of PZT sensors because the effective frequencies of monitoring for the piezoelectric cement sensor are broader than for PZT sensors.

Piezoelectric cement sensors with higher piezoelectric properties have more sensitivity of structural health monitoring, especially for d_{33} greater than 100 pC/N. To fabricate higher piezoelectric properties of piezoelectric cement are still ongoing. Except that the pretreatment temperature technique [47] is capable of increasing tremendously the d_{33} and ε_r, the mixing of cement and PZT particles with solvent has also been trying to improve piezoelectric properties. Specimens have been fabricated by combining PZT inclusions and cement with water and superplasticizers or with solvent (e.g., acetone and ethyl alcohol) and casting the mixture (composite) by using a spreading paste method [3,8,24] or pressing methods [30–32,36,38,46,52–54]. Chaipanich et al. [53] evaluated PZT/cement composites and highlighted that specimens with PZT content greater than 50 vol% must be fabricated using pressure forming and not the spreading paste method because of the lack of sufficient cement paste to bind the PZT particles. Pan and Chen [45] compared specimens fabricated using pressure forming method with those fabricated using a normal mixing and compacting method (spreading paste method) and found that although specimens fabricated using the paste mixing had slightly greater piezoelectric characteristics than specimens obtained using the pressing method, the pressure-forming method was recommended for PZT/cement specimens because of the lower presence of variations and voids.

In the past decade, most specimens of piezoelectric cement have been fabricated using pressure forming method [28–48,52–54] to investigate their piezoelectric and dielectric properties. The cement is prepared by combining PZT inclusions and admixtures with solvent or water and superplasticizers followed by casting through compression. The manufacturing of specimens (PZT/cement composite) with proper water is beneficial to enhance piezoelectric properties of PZT/cement composites. However, few reports have explored the effect of water-to-cement ratio (w/c) on the piezoelectric properties of 0–3 type PZT/cement composites, except for a study on 2–2 type PMN/cement piezoelectric composite fabricated by a cut-filling technique [8] that the PMN ceramic plates were put into the mold for casting cement with w/c from 0.3 to 0.5 (spreading paste method). Therefore, this study investigated the influence of w/c on 0–3 type cement–matrix composites and determined a suitable w/c that yielded greater piezoelectric behavior. In addition, more water in the mixing of cement and PZT particles induces more pores in the composite, especially for 0–3 type PZT/cement composites. The composite with more pores having higher dielectric loss D values that make the poling difficult, always leading to lower piezoelectric properties after the polarization. Hence, in this study three piezoelectric cements with 30%, 50%, and 70% PZT, by volume, were analyzed. Specimens with w/c from 0 to 0.20 were cast by compressing at 80 MPa and double heating at 140 °C [48] to enhance the d_{33} and ε_r values; these specimens were compared with those fabricated at 23 °C.

2. Experiments

2.1. Materials and mixture proportions

The piezoelectric cement comprised ASTM type I cement as the matrix and PZT ceramic as the inclusion material, with no admixtures. The specific gravity and fineness of cement were 3.15 and 349 m^2/kg, respectively. A Ka type of PZT ceramic with specific gravity = 7.9, d_{33} = 470 pC/N, ε_r = 2100, K_t = 0.72, dielectric loss (D) = 1.5%, and piezoelectric voltage factor $g_{33} = 24 \times 10^{-3}$V·m/N (tested using IEEE Standard 176–1987) was captured from Elecceram Technology Co. Ltd. (Taiwan). The PZT ceramic without being polarized, originally a flat dish, was pulverized to 75–150 μm particles (#100–#200 ASTM sieve). Distilled water was added to the piezoelectric cement.

The 0–3 type PZT/cement composites with 30%, 50%, and 70% PZT inclusions by volume, respectively, are labeled as P30, P50, and P70. Five w/c's, namely 0%, 5%, 10%, 15%, and 20% by weight, were labeled with a prefix "w" on each composite. For example, w0 and w15 represent composites with w/c = 0% and 15%, and P50w10 denotes piezoelectric cement with 50% PZT and w/c = 10%.

2.2. Specimen fabrication and pores

To prepare specimens, fresh cement and PZT particles were first mixed without adding water, and then the mixture was placed in a solar-planetary mill by applying a clockwise and counterclockwise rotation for 5 min to ensure that the raw materials were thoroughly distributed. Subsequently, an amount of water corresponding to the individual w/c from 0 to 0.2 was added to the mixture in a small jar, and the mixture was stirred into a paste by using a tamping bar for 2 min. Fig. 1 presents mixtures with w/c = 0% (upper right) and 20% (upper left), respectively. The mixture was then divided into three portions, and each portion was placed in a 15-mm-diameter cylindrical steel mold with peening. Finally, the mixture in the mold was pressed together at 80 MPa for 5 min to form a disc-like specimen (Fig. 1) with w/c = 0% (lower right) and 20% (lower left). The original thickness of PZT/cement composite disk before polishing is 2.5 mm. The specimens were immediately cured in a controlled chamber at 90 °C and 100% relative humidity for 24 h to ensure that the hydration produced suitable strength in these specimens.

After curing, the specimens were polished to a thickness of 2 mm. The pores of the specimens were monitored through optical microscopy (OM), and the porosity for each specimen was determined through an image analysis with pixel threshold criteria. The polishing process affects the surface porosity of the samples inevitably. To diminish experimental errors, the porosity of piezoelectric cement is the average of three specimens, which was measured at six positions of the specimen.

2.3. Specimen treatment and measurement

Specimens were subjected to the pre-treatment and post-treatment, both at 140 °C, for 40 min (called the BB treatment) by following the pretreatment temperature technique, and specimens maintained at 23 °C (called the AA treatment) were used as a reference. The pretreatment temperature technique is double-heating that the heating before and after the manufacturing of electrodes on the specimen [48]. The electrodes of the specimen were prepared by coating both sides of the specimen with silver paint (SYP-4570) according to the manufacturer's instructions and then curing them at 150 °C for 30 min to ensure that the paint adhered firmly. After the temperature treatment process, the specimens were cooled to 23 °C, and an impedance phase analyzer (Model 6520A) was adopted to measure the dielectric loss (D) and resistivity (ρ) of each specimen at 1 kHz and 1 V. Because the specimens were not polarized by outer electric field at this stage, no piezoelectric property was induced.

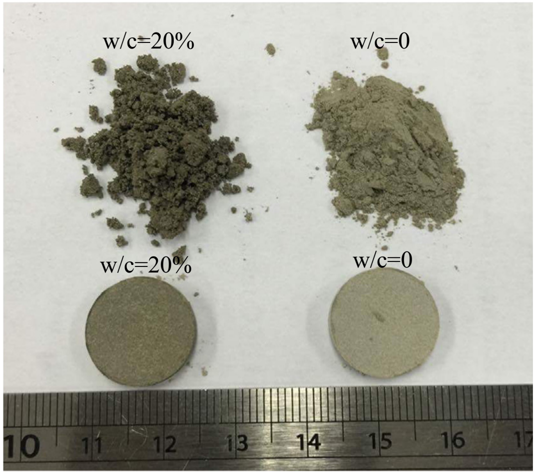


Fig. 1. The mixture with w/c = 20% (at upper left-side) and w/c = 0 (upper right), and the formed specimen with w/c = 20% (lower left) and w/c = 0 (lower right).

Subsequently, specimens were polarized using a poling voltage of 1.5 kV/mm at 150 °C (poling temperature) for 40 min (poling time) to induce piezoelectric properties. During the poling process, specimens subjected to breakdown (shock-burned holes) or trigger time (a poling duration counted from 0 to poling voltage) of more than 3 h were considered as polarization failures. After the specimens were successfully polarized, piezoelectric properties were measured using a d_{33} piezometer (Model P/N 90-2030) and an impedance phase analyzer. All experimental data were measured from 1 h (day 0) to 100 days after polarization completion.

Piezoelectric properties such as the piezoelectric strain factor (d_{33}), relative dielectric constant (ε_r), and thickness electromechanical coupling coefficient (K_t) of the specimens (piezoelectric cement) were measured and determined at controlled conditions of 23 °C and 50% relative humidity. The piezoelectric strain factor d_{33} was directly measured using the d_{33} piezometer at 110 Hz at nine positions on each specimen. The ε_r value was calculated using the thickness t and electrode area A of the specimen, and the capacitance C was measured at 1 kHz as follows [24]:

$$\varepsilon_r = \frac{C\,t}{A\,\varepsilon_0} \tag{1}$$

where $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m is the vacuum permittivity.

K_t was determined using the resonance frequency at the minimum impedance f_m and that at the maximum impedance f_n obtained using the impedance–frequency spectra of specimens. The K_t value was calculated using the following equation [23]:

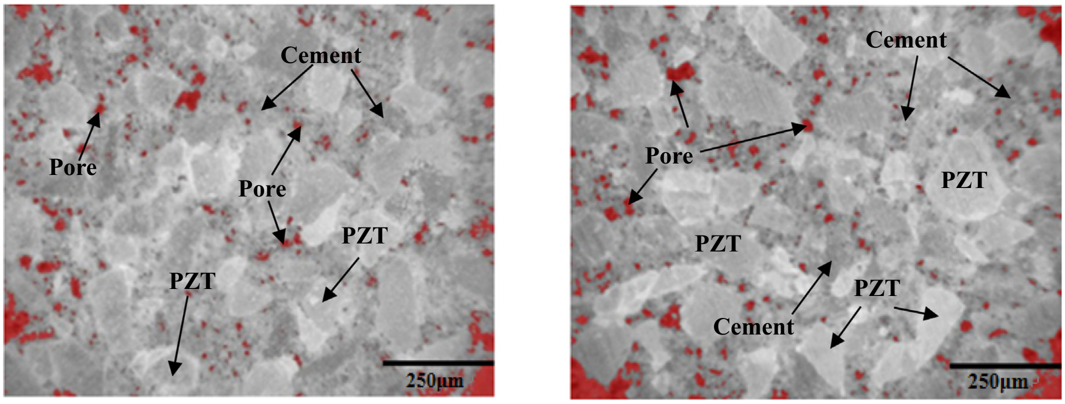
$$K_t^2 = \frac{\pi}{2}\frac{f_m}{f_n}\tan\left(\frac{\pi}{2}\frac{f_n - f_m}{f_n}\right) \tag{2}$$

Experimental results are presented as the average of three specimens.

3. Results and discussion

3.1. Porosity

Specimens were monitored through OM at 350× magnification after curing for 24 h. From the OM images, the pores of specimens were located using the image analysis software (PIA), which highlights the pores in red on the images by selecting a division value of 90 (pixel threshold criteria) to distinguish between black and white in the OM images. Fig. 2 presents the pores for the composite P50 with w/c = 5% and 20%. After the image analysis, the porosity of the specimens was determined by calculating the ratio of the red region and entire image area for three PZT/cement composites with w/c; the results are presented in Fig. 3. P50 comprised specimens with porosity values 3.39%, 3.50%, 3.63%, 3.92%, and 4.25% at w/c = 0%, 5%, 10%, 15%, and 20%, respectively. The composites with higher w/c exhibited higher porosity values (Fig. 3). Porosity increments in the specimens were apparently caused by the



(a) P50w5 (b) P50w20

Fig. 2. OM images for (a) P50w5 with the porosity of 3.50% and (b) P50w20 with 4.25%. (The red marks on the OM image are pores.) (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

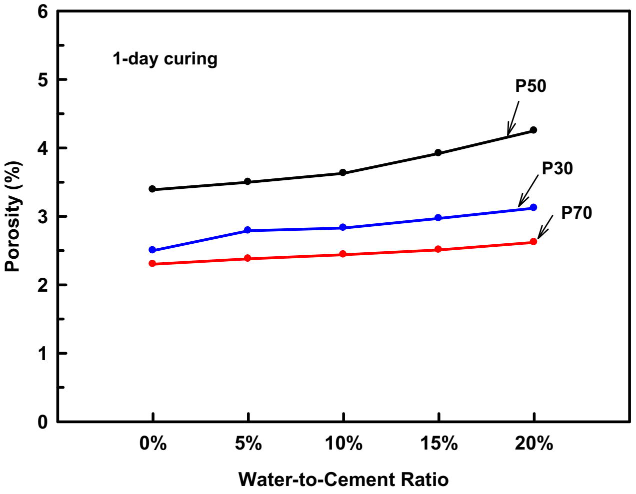
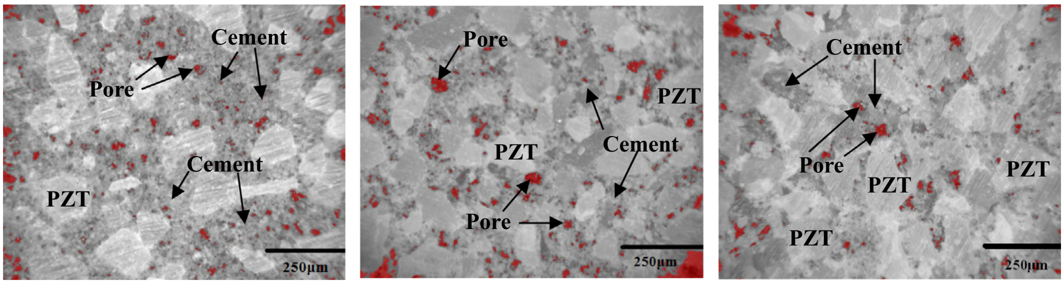


Fig. 3. Porosity of PZT/cement composites versus water-to-cement ratio.

volume occupied by the increasing quantity of water added during the fabrication of specimens.

The OM images indicated that pores were typically present within the cement matrix, and a few pores were located on the interfacial boundary of the cement and PZT particles. The higher the cement content is, the higher the number of pores in PZT/cement composites is. However, P50 exhibited higher porosity than P30 and P70 (Fig. 3) for all w/c. To address this, the OM images of P30w0, P50w0, and P70w0 (Fig. 4) indicate that although P30 (30% PZT and 70% cement) had more cement matrix content than P50, the pore size in P30 was smaller because the higher air content in the cement matrix was easily expelled by applying 80-MPa compression during the forming process. This was verified by measuring, for instance, the pore size from the OM images in Fig. 4, where the average pore size in P30w0, P50w0 and P70w0 is 16.4 μm, 19.1 μm and 23.2 μm respectively. Fig. 5 displays the pore size distribution of three PZT/cement composites, with the exception of one pore of P50w0 at the size between 140 mm and 150 mm. The total pore number for P30w0, P50w0 and P70w0 is 126, 129 and 83, respectively. P70 (70% PZT and 30% cement) had lower cement matrix content than P50 and P30, leading to difficulties in air expulsion during the pressing that causes larger pore size but with the least number of pores, thus P70 has the lowest porosity (Fig. 3). Moreover, P50 has 129 pore numbers that close to P30 (126 pore numbers), and the average pore size (19.1 μm) is greater than that in P30 (16.4 μm). Both factors result in the highest porosity to be observed in P50 (Fig. 3), that is, the porosity in P50 is higher than in P30. It seems that the coupling effect of cement matrix content and air expulsion in compression governs the porosity of specimens. Some experiments for porous PZT ceramics were also conducted to discuss the effect of pore size and orientation on piezoelectric properties [55] and the effect of porosity on the polarization switching behavior of ferroelectrics [56]. The effect of porosity, pore size and orienting anisometric pores on dielectric and piezoelectric properties of 0–3 type cement-based piezoelectric composites remains poorly understood.



(a) P30w0 (b) P50w0 (c) P70w0

Fig. 4. OM images for (a) P30w0 (2.5% porosity), (b) P50w0 (3.39% porosity) and (c) P70w0 (2.3% porosity).

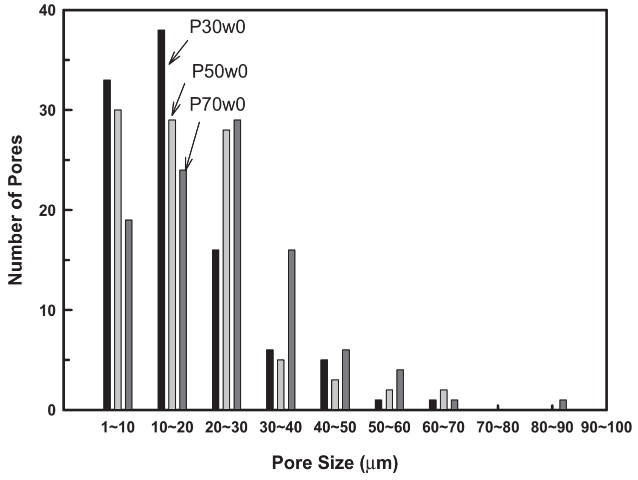


Fig. 5. The size distribution of pores for P30w0, P50w0 and P70w0 respectively. (Note: One pore of P50w0 having the size between 140 μm and 150 μm does not display here.)

3.2. Dielectric loss and resistivity

Previous experimental results [47,48] indicated that PZT/cement composites with lower dielectric losses before polarization tended to be easily poled when the specimens were fabricated by pressing with no water added (w/c = 0). When PZT/cement specimens were prepared by adding water at a w/c from 0% to 20%, the dielectric loss D was measured (Fig. 6). The label "AA" (solid lines) refers to the specimens that were subjected to double 23 °C treatment and "BB" (dash lines) represents specimens subjected to double 140 °C treatments. For the P30 material, the dielectric losses increased with increasing w/c, and P30_BB had a lower D value than P30_AA because of the higher temperature treatment in the manufacturing process. When the PZT inclusions in the composites were increased, the D values for P50_AA and P70_AA gradually increased with w/c and then significantly increased when the w/c was greater than 15%. When the double 140 °C treatment was applied to the specimens, the effect of w/c on the dielectric loss of P50_BB and P70_BB was low, although the D values slightly increased. The w/c had a significant influence on the dielectric loss of P30, P50w20_AA, and P70w20_AA because of the higher cement matrix content in P30 and higher water content in P50 and P70 with the 23 °C treatment at w/c = 20%. When the w/c raises from 15% to 20%, some gel water, free water and capillary water still retain in the composite after 24 h curing, especially for P50 and P70 under the 23 °C treatment (AA treatment). Their inner water can motivate the leakage current through the composite more easily during the poling process, leading to higher current loss. The character of dielectric loss D can somehow reflect the degree of current loss. This is the reason why the D values in P50 and P70 under AA treatment increase evidently at the w/c from 15% to 20%.

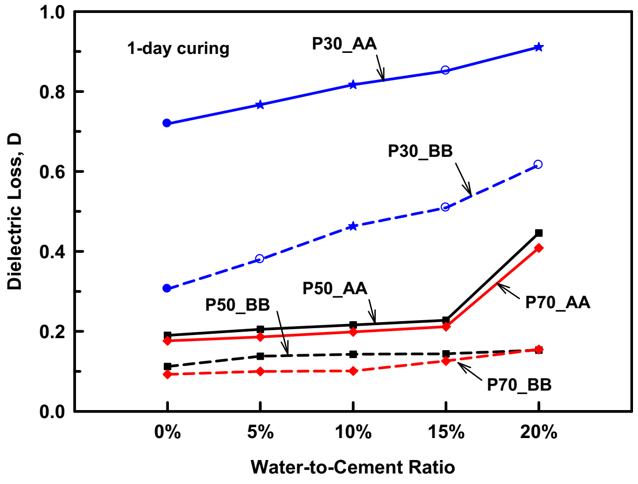


Fig. 6. Dielectric loss of PZT/cement composites versus water-to cement ratio before polarization.

Longer trigger times generally make the poling process difficult. Table 1 lists the trigger times of the specimens during the poling process, where the data with the superscript of the star and hollow circle represent the polarization failure of specimens, which indicates that no piezoelectric property of PZT/cement composites was induced by outer voltage (poling voltage). The trigger time in Table 1 has a good correlation with the dielectric loss D in Fig. 6, which confirms the tendency of higher D values to yield higher trigger times. This connection was also reported for PZT/cement composites by considering curing time and heating condition [48]. The D values of P50 and P70 are quite small (D < 0.22), except for P50w20_AA and P70w20_AA, which facilitates the poling process because of the trigger time ≤13 min. In addition, the P30 material contains a higher cement matrix content than P50 and P70, which leads to higher D values, indicating that only P30 at w/c = 0 can be poled (Table 1). This observation reveals that PZT/cement composites containing ≤30% PZT may cause difficulty in polarization, particularly for the specimens with water. From the experiments related to Fig. 6 and Table 1, one can conclude that, to induce piezoelectricity, the values of dielectric loss should be less than 0.75 for PZT/cement composites before the polarization. Similar result was also concluded for PZT/cement composites containing 10 to 50% slag and fly ash as the substitutions of cement, which the D values less than 0.73 ensure the feasibility of polarization [35].

To assess the influence of w/c on the resistivity ρ of PZT/cement composites, ρ was measured at 1 V and 1 kHz before polarization (Fig. 7). Apparently, higher PZT content and temperature treatment on the specimens result in lower ρ values, and the specimens with a higher w/c and more cement matrix always exhibit higher ρ values. The w/c and heating process prior to polarization affect the resistivity of PZT/cement composites, which implies that water molecules in the cement matrix dominate the resistivity.

The resistivity values were higher in P30 specimens fabricated by adding water than the other composites, which typically experience polarization failure. Moreover, although the resistivity values for P30w0_AA (ρ = 104 kΩ-m), P30w0_BB (ρ = 92 kΩ-m), P50w20_AA (ρ = 66 kΩ-m), and P70w20_AA (ρ = 33 kΩ-m) were high, they could be poled successfully with trigger times of 141.6, 62.3, 70.1, and 25.7 min, respectively. The resistivity at 100 kΩ-m can also be considered as a threshold for PZT/cement composites related to the feasibility of the poling process. P50w20_AA had lower resistivity than P30w0_BB (Fig. 7), but

Table 1
Trigger time for specimens of PZT/cement composites. (Unit: min).

Material	w0	w5	w10	w15	w20
P30_AA	141.6	$290.7^{\bigstar}$	$212.0^{\bigstar}$	$36.5^{\bigcirc}$	★
P30_BB	62.3	$102.3^{\bigcirc}$	$199.8^{\bigstar}$	$66.6^{\bigcirc}$	$20.6^{\bigcirc}$
P50_AA	9.6	10.4	10.6	11.0	70.1
P50_BB	3.1	3.9	3.7	3.9	13.0
P70_AA	5.8	7.2	9.0	12.0	25.7
P70_BB	1.3	1.8	2.5	4.1	6.6

Note: ★ = trigger time >3 h; ○ = time at the occurrence of current breakdown.

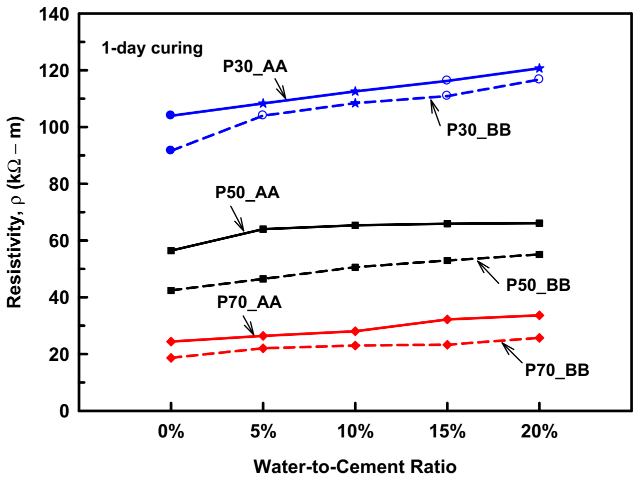


Fig. 7. Resistivity of PZT/cement composites versus water-to-cement ratio at 1 V.

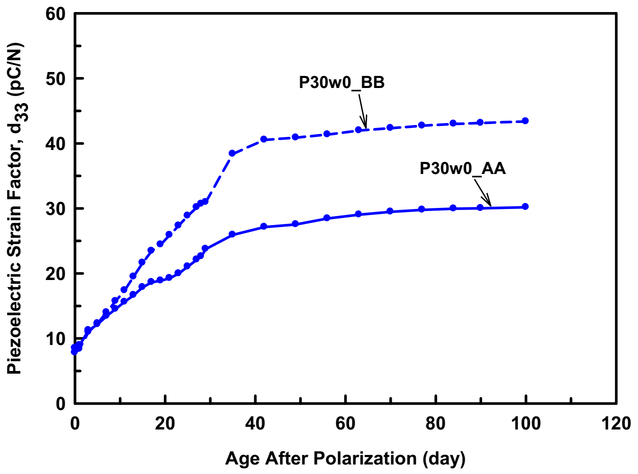


Fig. 8. Piezoelectric strain factor d_{33} of 30% PZT/cement composites versus age.

slightly higher trigger time (Table 1), probably because of higher porosity in P50w20 than in P30w0 (Fig. 3).

Huang et al. [25] pointed out that the interface pore, the cement matrix pores and their inner water have a significant influence on the degree of the poling. The comparisons in Table 1 and Figs. 3, 6, and 7 indicate that a higher w/c during the forming process causes more porosity in specimens, leading to higher dielectric loss and resistivity, and hence longer trigger times to excite poling voltage is required during the poling process. Besides, the resistivity ρ for ceramic materials always has the tendency of decrease with increasing temperature. PZT particles and cement matrix in general belong to the class of ceramics, so that PZT/cement composites has lower ρ values if the composites are applied to higher temperature treatment. Similar to the resistivity, when the temperature treatment (higher temperature) applied to the composite, inner water of the specimen diminishes gradually. At this moment, the composite appears lower D values causing less leakage current through the matter. Hence, the application of temperature treatment to the specimens facilitates the poling process because of lower D and ρ values despite the addition of water (increased w/c) in the manufacturing process. Piezoelectric cement fabricated using PZT/cement composites must contain no less than 30 vol% of PZT to ensure efficient piezoelectric properties.

3.3. Piezoelectric properties

After the poling process, piezoelectric behavior was induced in the PZT/cement specimens to make them piezoelectric. To reflect the piezoelectric behavior developing with aging time after the polarization, the piezoelectric properties of the specimens were measured for 100 days.

3.3.1. Piezoelectric strain factor

The piezoelectric strain factors d_{33} for three piezoelectric cements at AA and BB heating conditions are presented in Figs. 8–10. Only the P30 material at w/c = 0 shown in Fig. 8 has the piezoelectric strain factor d_{33}, and the P30 materials with w/c $\neq$ 0 experienced polarization failure (indicated in Table 1) show zero d_{33} value because of higher dielectric loss D (Fig. 6) and resistivity ρ (Fig. 7) before polarization. The d_{33} values of P30w0 increased rapidly and then increased more slowly after 40 days. The heating treatment on the specimens can improve the d_{33} value of P30w0 because dielectric loss D and resistivity ρ values of the specimens are lower before the polarization (Figs. 6 and 7) that make the specimens easier to induce piezoelectricity.

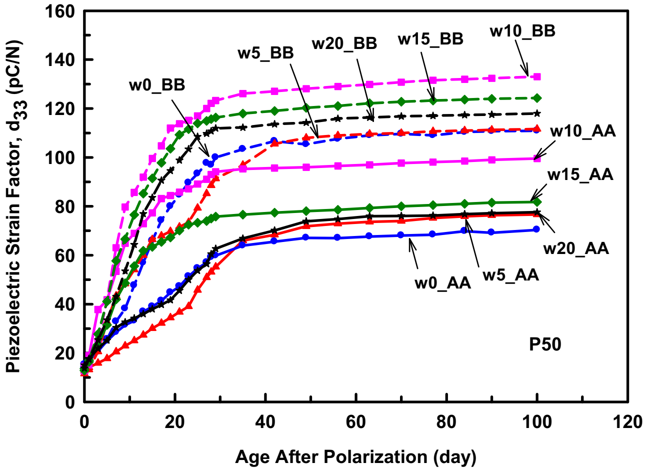


Fig. 9. Piezoelectric strain factor d_{33} of 50% PZT/cement composites versus age.

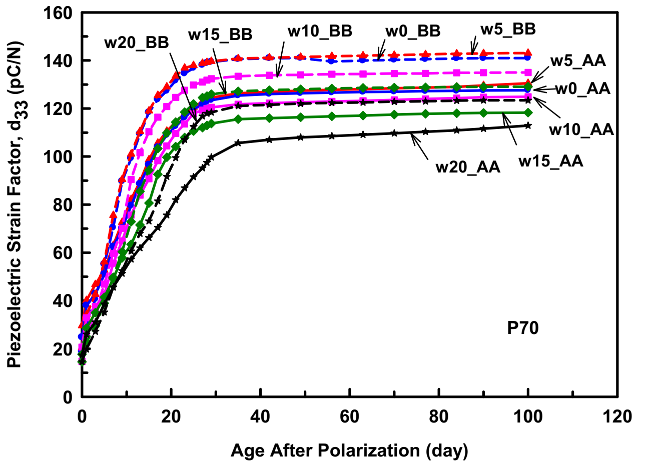


Fig. 10. Piezoelectric strain factor d_{33} of 70% PZT/cement composites versus age.

The double 140 °C treatment applied to the specimens (P30w0_BB) increased the d_{33} value from 30.2 pC/N (P30w0_AA) to 43.4 pC/N at 100 days.

The d_{33} values of P50 with w/c = 0–0.2 are plotted in Fig. 9. For both AA and BB treatments, the order of the effect of w/c was w10 > w15 > w20 > w5 > w0 after 40 days; in other words, the d_{33} value of P50 at w/c = 10% was the highest. The piezoelectric properties directly depend on the degree of poling of the composites. However, the degree of the poling is often restricted by the character of constituents (PZT phase, cement matrix and pore structure), defects formed during the manufacturing, and microstructure [25]. The reason for P50 at w/c = 10% having the highest d_{33} value may be that the specimens at P50 (the equal volume of PZT and cement) with w/c = 10% are easy to be cast by the pressing method, and the structure of the composite and the cement is likely more symmetrical and uniform, compared with the others w/c. The 0–3 type PZT/cement composites with more symmetrical and uniform structures have higher d_{33} values because the orientation of ferroelectrics domain is aligned more effective. For P50 with w/c = 0, the d_{33} value is the lowest, compared with the others w/c, because P50w0 may have the less symmetrical and uniform structure by dry compact. Appropriately increasing the w/c up to 20% can still improve the d_{33} value of P50. The d_{33} value of P50w10_BB at 100 days was 133 pC/N, which is higher than those of P50w0_BB (110.9 pC/N) and P50w0_AA (70.4 pC/N). This result indicates that the application of double 140 °C treatment with w/c = 10% to P50 increased the d_{33} value of P50 by almost 89% from 70.4 pC/N to 133 pC/N. This d_{33} value of 133 pC/N is the highest among all 0–3 type PZT/cement composites containing 50% PZT fabricated until now.

When PZT inclusions increased from 50% to 70%, the highest d_{33} values of P70 occurred at w/c = 5% (Fig. 10) for both AA and BB treatments, in the order w5 > w0 > w10 > w15 > w20. It is noted that the d_{33} development with the age for P70 at w/c = 5% and 0% have no many differences although the d_{33} values at w/c = 5% are slightly higher than at w/c = 0% after 50 days. The volume fraction of cement in P70 is 30% that causes less uniform distribution of cement among PZT by pressing method during the manufacturing of specimens and the cement has less hydration at w/c = 5%, resulting the effect of the cement to the d_{33} values between w/c = 5% and 0% is almost the same. Increasing the w/c will lower the d_{33} values if the w/c of P70 is more than 5%. This is because more hydration in the cement causes higher degree of inhomogeneity in P70 that will hamper the induction of d_{33} during the poling. The highest d_{33} value of piezoelectric cement with 70% PZT was 143.1 pC/N (P70w5_BB), which is a mild increment (12%) compared with d_{33} = 127.6 pC/N for P70w0_AA. For PZT/cement composites with PZT over 50%, the benefit to specimens by increasing w/c to promote d_{33} is limited because only 30% cement binder presents in P70, unlike the 50% cement binder in P50, which contribute to the benefits of w/c on d_{33}.

In Figs. 8–10, the trend of d_{33} for the first 40 days of P30 and P50 depended on temperature treatment (i.e. s-shaped curve for AA and rather smooth curve for BB) while P70 did not. The mechanisms of age-dependent d_{33} on 0–3 type PZT/cement composites are not fully understood. The main hydrated products (CH and CSH) of cement matrix and pore structures (porosity, pore size, pore geometry, orientation and alignment) are influence factors [25,35,47,54] on the development of d_{33} with age. The content of cement matrix in P30, P50 and P70 is different. For P70, the volume of cement matrix is 30%, which is far lower than that of PZT particles, so that the increment variations on d_{33} are less for the first 40 days due to the effect of hydrated products and pores. This make the development of d_{33} curves smooth at the first 40 days (Fig. 10). If the cement matrix increases, for instance P30 and P50, the hydrated products and pores developed are not linear at the first 40 days (Figs. 8 and 9), especially for the composites under AA condition (more inner water than under BB condition), leading to s-shaped curve of d_{33}. The microstructure of the composite and cement matrix is crucial to the study of age-dependent piezoelectricity.

In addition, the relationship to the influence of water-to-cement ratio (w/c) on d_{33} for piezoelectric cement is different from the report by Huang et al. [8] that the d_{33} values of 2–2 type PMN/cement piezoelectric composite increased with increasing the w/c from 0.3 to 0.5. The fluidity of the cement paste in 2–2 type PMN/cement piezoelectric composite compacted surrounding the component of PMN plates easily. The geometric alignment and the distribution of inclusions in cement matrix between the 0–3 type and the 2-2type composite are believed to be the key resulting in the difference.

3.3.2. Relative dielectric constant

The relative dielectric constant ε_r can be calculated from Eq. (1) and is presented in Figs. 11–13. The behavior of ε_r curves for all w/c develops with age and gradually approaches a steady value after approximately 40 days. Similar to the development of d_{33} curves shown in Figs. 8–10, the ε_r with age is also affected by the change of hydrated products and pore structures. Piezoelectric cement with higher d_{33} values always exhibits higher ε_r and the similar trend of d_{33} and ε_r development with age has also been reported [24,35,54]. The specimens subjected to double 140 °C treatment exhibited an increase in ε_r; for example, in Fig. 11, the ε_r values for P30w0_BB and P30w0_AA at 100 days are 245 and 156, respectively, which is an increase of approximately 57%. For P50 and P70 at w/c = 0, the increase in ε_r because of the heating effect was 79% and 73%, respectively at 100 days (Figs. 12 and 13).

The order of the effect of w/c on ε_r is w10 > w15 > w20 > w5 > w0 for P50 (Fig. 12) and w5 > w10 > w15 > w20 > w0 for P70

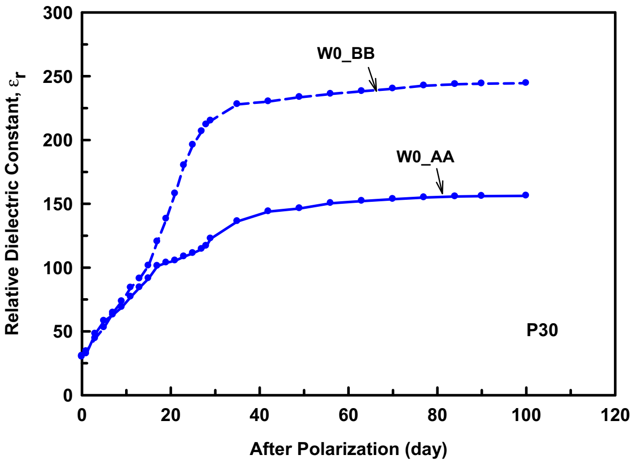


Fig. 11. Relative dielectric constant ε_r of 30% PZT/cement composites versus age.

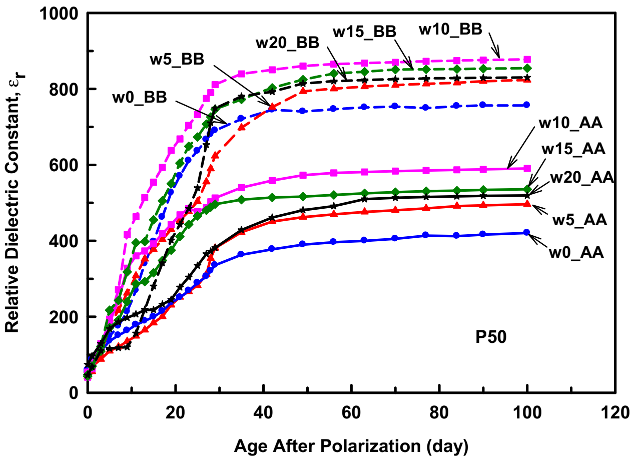


Fig. 12. Relative dielectric constant ε_r of 50% PZT/cement composites versus age.

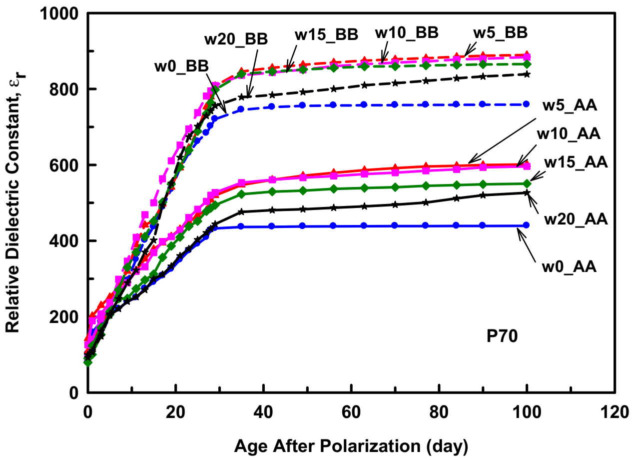


Fig. 13. Relative dielectric constant ε_r of 70% PZT/cement composites versus age.

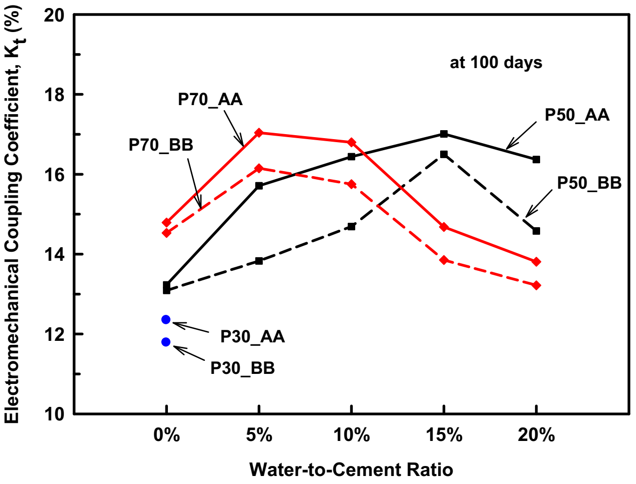


Fig. 15. Thickness electromechanical coupling coefficient K_t versus w/c at 100 days.

(Fig. 13). This w/c effect indicates that suitable adding water to specimens during pressure forming is an effective approach to increase the relative dielectric constant ε_r, probably depending on the degree of heterogeneity in the composite. The optimum ε_r values for P50 and P70 were obtained at w/c = 10% and 5%, respectively, similar to the effect of w/c on d_{33} due to more uniform and homogeneous in the specimens at these amount of water. Fig. 14 presents ε_r values at 100 days for all w/c; the highest ε_r value for P50 was 878 (P50w10_BB) and that for P70 was 890 (P70w5_BB). Except for w/c = 5%, the ε_r of P70 was close to that of P50 for both AA and BB treatments, because although higher PZT amounts in piezoelectric cement provide higher ε_r values, the P50 material has higher porosity than P70, which helps to restore charge (dielectric constant) in the cement binder.

3.3.3. Electromechanical coupling coefficient

The value of the thickness electromechanical coupling coefficient K_t was determined using Eq. (2). The experimental data reveal that the K_t values corresponding to w/c were constant with aging time, and the behavior of age-independent K_t was also reported for P50 at w/c = 0 in reports [35,47]. Fig. 15 presents the relationship between w/c and K_t at 100 days, which indicates that K_t strongly depends on w/c. For P50, K_t first increased with the increase in w/c, but the trend reversed at w/c over 15%, with the optimum K_t values occurring at w/c = 15% for both the AA and BB treatments. Increasing the w/c in P50_AA enhanced the K_t value from 0.132 (at w/c = 0) to 0.17 (at w/c = 15%), which is a 29% increment. The K_t values for P50 at w/c = 20% remained higher than at w/c = 0. When PZT inclusions increased to 70% (P70), the optimum K_t occurred at w/c = 5%; subsequent higher w/c hampered K_t increment. To improve K_t for P70, w/c must be less 15%. In addition, higher PZT content can enhance K_t at w/c = 0 [23,29]. This trend is also true for P50 and P70 at w/c = 5% shown in Fig. 15. As the w/c continue to increases, the K_t values increase for P50 but decrease at w/c = 10% for P70. Apparently, the K_t values for P70 are no longer greater than for P50 when w/c reaches 15% or more. A higher w/c has more influence to pore number and pore size which might be considered as crucial factors needed to be investigated in future. Therefore, a suitable w/c must be used in specimens during the forming process to enhance K_t.

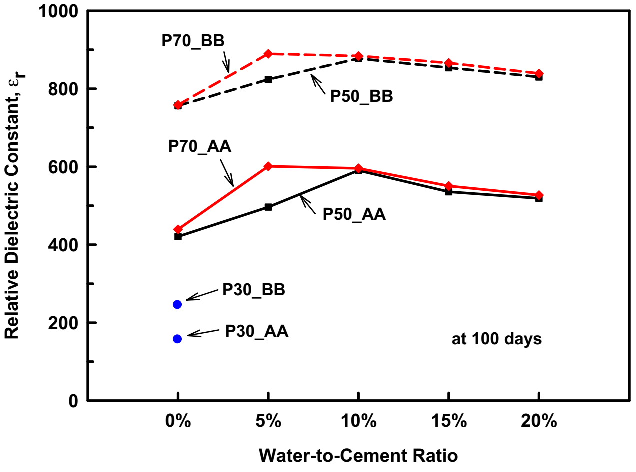


Fig. 14. Relative dielectric constant ε_r versus w/c at 100 days.

Previous studies have reported other approaches for enhancing K_t, including higher poling voltage, temperature, and time [43]; higher PZT content [23,29]; the addition of appropriate admixtures [30,42,57]; and longer curing durations [48]. Using a suitable w/c in piezoelectric cements to promote K_t may be attributed to the influence of the pores because a higher w/c induces more pores in the cement matrix. In addition, Huang et al. [46] evidenced that Kt values increased if the applied forming pressure on the specimen was decreased, leading to more pores in specimens. Notably, although temperature treatment has less effect on K_t for the P50 material with w/c = 0 [48], the differences in K_t between the AA and BB treatments were prominent with the increase in w/c (Fig. 15).

3.4. Age effect

The piezoelectric strain factor d_{33} and relative dielectric constant ε_r of piezoelectric cements are age dependent, and the rate of increase in the curves gradually decreases to 0 after 40 days. Nevertheless, age-dependent behavior of d_{33} and ε_r was not present in PZT ceramics [47,54]. From the d_{33} curves in Figs. 8–10, the age at which asymptotic d_{33} values of P30 were achieved was approximately 90 days or later. For P50, the asymptotic age appears to be earlier than that for P30; for P70, it occurs even earlier, at 50 days.

To investigate the piezoelectric properties developing with aging time in piezoelectric cement, the dielectric loss D was measured after the polarization, and the curves of D versus age were plotted (Figs. 16–18). Before the polarization, the D value for P30w0_AA was 0.72, which was higher than that for P30w0_BB (D = 0.31) (Fig. 6). When the poling process was completed, the initial D values for P30w0_AA and P30w0_BB (Fig. 16) were quite close to each other. The dielectric loss individually developed with

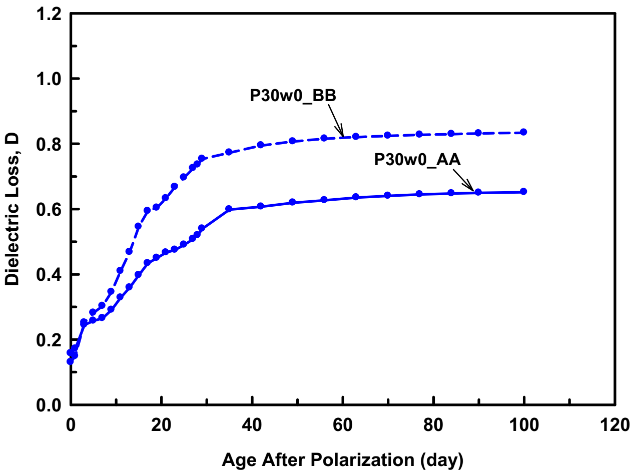


Fig. 16. Dielectric loss D of 30% PZT/cement composites versus age.

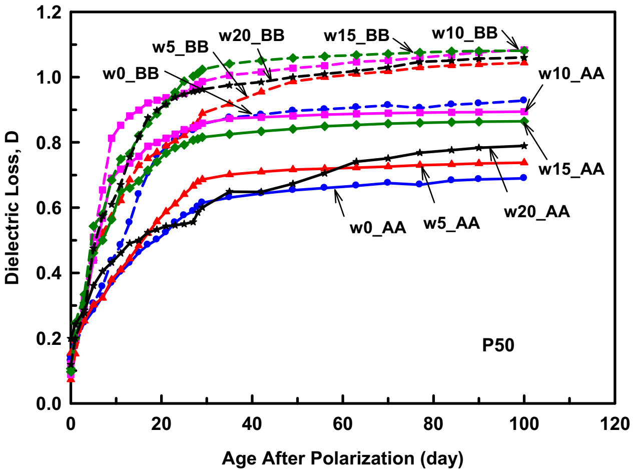


Fig. 17. Dielectric loss D of 50% PZT/cement composites versus age.

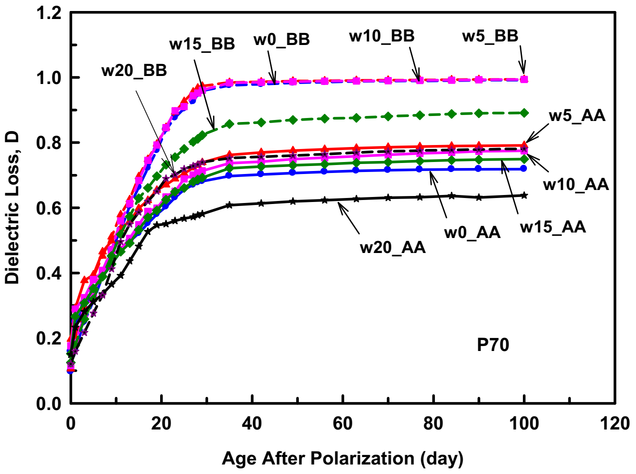


Fig. 18. Dielectric loss D of 70% PZT/cement composites versus age.

aging time, and the values on the BB curve became higher than those on the AA curve after 3 days. The effect of temperature treatment on D is the reverse of that of D before polarization (Fig. 6). The behavior of the ascending curves of D for P30 is similar to that of the d_{33} curves in Fig. 8. In addition, the D curves of P50 shown in Fig. 17 developed with age, and the influence of aging time on D curves was quite similar to the effect on d_{33} curves (Fig. 9). The effects of w/c on these piezoelectric properties are also shown on these figures. In addition, similar behaviors were observed in D curves (Fig. 18) and d_{33} curves (Fig. 10) for P70, with the trend increasing rapidly at an early age and then gradually declining at a later age.

Both D and d_{33} are age dependent and have a mutual interdependent relationship with piezoelectric cement with w/c = 0%–20%. Like d_{33}, the D values after the polarization develop with aging time and approach a stable value at a longer aging time, which could be due to the influence of the cement matrix in the piezoelectric cement. The higher the cement matrix content is, the longer the required aging time is to achieve stable D values. For example, in Fig. 16, stable D values for P30 are obtained after approximately 80 days and those for P50 and P70 (Figs. 17–18) were obtained much earlier. The development of cement binder was affected by material age and w/c, probably causing charge redistribution in the PZT/cement composites. This is one of dominant factors representing the age-dependent d_{33} and ε_r behavior of piezoelectric cement, despite cement not being categorized as a piezoelectric material.

4. Conclusions

Piezoelectric cement with 30–70% PZT and w/c = 0–20% was subjected to polarization with 1.5 kV/mm to investigate the influence of w/c on its piezoelectric properties. Piezoelectric cement with a higher w/c has higher porosity and larger voids in the cement matrix for composites containing ≥50% PZT. The dielectric loss D and resistivity ρ measured before the polarization increased if the composites had higher cement matrix content and higher w/c, which led to an increase in trigger time, thus making the polarization difficult. Higher temperature treatment on the specimens resulted in lower ρ and D values that improved the efficiency of polarization with respect to piezoelectric strain factor d_{33} and relative dielectric constant ε_r, implying that water molecules in the cement matrix are one of the dominant factors. A resistivity of 100 kΩ-m and dielectric loss of 0.75 can be considered as a criterion for the feasibility of the poling process for PZT/cement composites. The optimum w/c for P50 and P70 were 10% and 5%, respectively, for obtaining the highest d_{33} and ε_r values for both AA and BB treatments. The P50w10_BB material exhibited d_{33} = 133 pC/N and ε_r = 878 at 100 days, which are the highest values for P50 manufactured so far. Moreover, P70w5_BB exhibited d_{33} = 143.1 pC/N and ε_r = 890 for P70. The thickness electromechanical coupling coefficient K_t is also strongly dependent on w/c, and P50_AA yielded K_t = 0.17 at w/c = 15%, which is a 29% increment compared with that obtained at w/c = 0. Moreover, the effect of temperature treatment was prominent on K_t for piezoelectric cement that incorporated water. After the polarization, the d_{33}, ε_r, and D curves for all w/c had developed a similar tendency with aging time. The cement matrix is affected by the w/c and age that reflects the age-dependent d_{33} and ε_r behavior on piezoelectric cement. To the end, to achieve an optimized design for 0–3 type PZT/cement composites as piezoelectric sensors and actuators used in concrete SHM, the composite containing 50% PZT and the w/c at 5–10% fabricated by using pressure forming, accompanying with double 140 °C heating treatment, is recommended.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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