# Effect of Water Cement Ratio on the Electrical Conductivity and Piezoelectricity of Cement Paste Cured While Applying Direct Current

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## Abstract

Electrical conductivity and piezoelectricity of cement paste are key properties for developing a smart concrete. Applying a static electrical potential on cement paste while curing proved to be effective in altering the aforementioned properties by polarizing the ions in the cement paste. In this study, the effect of the water cement ratio (w/c) on the effectiveness of the static electrical curing technique was tested. Six cement paste specimens (50 × 50 × 50 mm) were self-cured while applying a static electrical potential of 10 V, and another 6 specimens were self-cured without applying electricity to serve as reference. To study the effect of the w/c, 2 different w/c were tested, namely 0.3 and 0.34. The results of the bulk resistivity of the specimens show a reduction of 67% in the resistivity of the specimens that cured while applying electricity when compared to the reference specimens. In addition, the polarized specimens show an improved piezoelectricity behavior. However, there was no significant difference in the behavior of the polarized specimens with different w/c on both the bulk resistivity and the piezoelectricity behavior.

**Keywords**: Cement paste, Conductivity, Piezoelectricity, Self-sensing, Electrical curing, Smart materials.

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# تأثير نسبة خلط الماء على التوصيل الكهربائي والكهرباء الانضغاطية لعجينة الأسمنت المعالجة أثناء تطبيق التيار المباشر

**محمد القرالة***

## ملخص

تعتبر الموصلية الكهربائية والكهرباء الانضغاطية لعجينة الأسمنت من الخصائص الرئيسية لتطوير الخرسانة الذكية. ثبت أن تطبيق جهد كهربائي ثابت على عجينة الأسمنت أثناء المعالجة يغير الخصائص المذكورة أعلاه من خلال استقطاب الأيونات في عجينة الأسمنت. في هذه الدراسة تم اختبار تأثير نسبة الماء إلى الأسمنت (w/c) على فعالية تقنية المعالجة الكهربائية الساكنة. تمت معالجة ست عينات من معجون الأسمنت (50 × 50 × 50 مم) ذاتيًا أثناء تطبيق جهد كهربائي ثابت قدره 10 فولت، وتم معالجة 6 عينات أخرى ذاتيًا دون استخدام الكهرباء لتكون بمثابة مرجع. لدراسة تأثير نسبة (w/c)، تم اختبار نسبتين للخلط، وهما 0.30 و 0.34. أظهرت نتائج المقاومية الكهربائية للعينات انخفاضًا بنسبة 67٪ في مقاومة العينات التي تم معالجتها أثناء تطبيق الكهرباء مقارنةً بالعينات المرجعية. بالإضافة إلى ذلك، تُظهر العينات المستقطبة سلوكًا محسّنًا للكهرباء الانضغاطية. ومع ذلك، لم يكن هناك اختلاف جوهري في سلوك العينات المستقطبة ذات نسب خلط مختلفه من حيث المقاومية الكهربائية وسلوك الكهرباء الانضغاطية.

**الكلمات الدالة:** عجينة أسمنتية، موصلية كهربائية، كهرضغطية، إستشعار ذاتي، معالجة كهربائية، مواد ذكية.

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**Introduction**:

Recently, smart materials of building are catching the interest of the scientific society for what it has of a great potential in many fields. Smart materials in general are those which have the ability to interact with their surroundings whether they are mechanical stresses, temperature fluctuations, or electromagnetic changes. Smart materials are expected to harness the external effects into a useful form of energy and/or sense these effects and report it back in a form of useful information. Concrete is one of the vastly used building materials, hence, improving its properties to harvest energy and/or sense the surrounding effects can be of a great interest for researchers and industry. In general, if the electrical conductivity and piezoelectrical properties of the concrete can be enhanced, its self-sensing properties and energy harvesting capabilities can be improved. In essence, piezoelectric materials can capture all the mechanical excitations and convert it to electricity. Hence, great economical and sustainability benefits can be achieved by harvesting the energy from all the repeated mechanical loadings on the civil infrastructures such as bridges, if we can harness it. On the other hand, enhancing the electrical conductivity of concrete will allow the engineers to monitor the structural health of important infrastructures. The aforementioned applications are only a few of the benefits that can be achieved from enhancing the electrical conductivity and piezoelectricity of concrete.

Electrical conductivity and piezoelectricity of cement paste are key properties for developing a smart concrete (Han et al., 2015). During the past decades, researchers investigated the electrical conductivity of the concrete and concluded that concrete is a good insulator by nature (Wu et al., 2013). A good review about the efforts to enhance the self-sensing capabilities of concrete can be found in the work of Tian et al., (2019). On the other hand, concrete is an intrinsic piezoelectric material for some extent due to the transportation of ions in the diffuse layer (Sun et al., 2000, 2004). Piezoelectricity is a property that some materials exhibit that depends on the internal crystalline arrangement. Generally, two types of piezoelectricity are studied; namely: direct, and indirect piezoelectricity. The direct piezoelectricity defined as producing electrical potential when an external load is applied on the material. While the indirect piezoelectricity is the ability of the material to change its shape or size when an external electrical potential is applied on it (Elahi et al., 2018). Piezoelectric materials have wide range of applications; however, we have a special interest in its application in sensing and structural health monitoring (SHM). Enhancing the piezoelectric properties of the concrete can open the possibilities of self-sensing structures.

Smart concrete should behave in a beneficial fashion to serve multiple purposes, such as: self-sensing of mechanical excitations, converting mechanical energy to useful electrical energy, and self-heating (Chen et al., 2019; Han, et al., 2015; Tian et al., 2019). Other application of smart concrete is to evaluate its physical properties, such as: durability, and compressive strength, without the need of destructive testing methods (Azarsa, & Gupta, 2017; Wei et al., 2012). Investigating the microstructure of the concrete can indeed help to improve the properties of the concrete to make it smarter, an extensive reviews on the microstructure of the concrete can be found in the following studies Han et al., (2015), and Li et al., (2002).

Numerous studies have investigated the possibility to improve the electrical conductivity and piezoelectric properties of concrete, mostly by utilizing electrically conductive fillers such as: carbon or steel fibers (Sun et al., 2000; Yeol et al., 2019), graphite (Al-Bayati et al., 2021), lead zirconate titanate, barium zirconate titanate (Chen et al., 2019), and other additives (Topu et al., 2012). On the other hand, an innovative technique was employed by (Yaphary et al., 2016) to enhance the piezoelectricity of cement paste using electrical curing without any additional fillers. The latter technique showed success in polarizing the microstructure of the cement paste and enhanced its piezoelectric behavior.

## Research significance:

Applying direct current (DC) during the curing process of cement paste proved to alter its electrical conductivity and piezoelectrical properties. However, this phenomenon still needs more investigation to identify the effect of multiple factors on the conductivity and piezoelectrical properties. The effect of factors such as: w/c of the mix, DC curing voltage, curing and ambient temperatures, and electrically conductive fillers are still not known. This research investigates the effect of the w/c of the mix on the piezoelectrical properties of the cement paste when a DC is applied during the curing process. In addition, this research investigates the effect of the mentioned factor on the electrical conductivity characteristics of cement pastes.

## Methodology:

A total of 12 specimens of 50 mm cubes were cast in this experiment. Six specimens were self-cured without applying current to serve as reference, and the other 6 were also self-cured while a DC is applied. In addition to the standard steel molds of 50 mm cubes, cubic molds identical to the standard ones were fabricated from laminated wood to match the standard cubes according to ASTM C109/C109M−20b (2016). The fabricated molds have five sides of wood, from which, two opposite sides were covered by copper plates to act as electrodes. The copper plates were attached to insulated copper wires, then the wires were attached to a rectifier with adjustable voltage capability. Cement paste mix were prepared according to the ASTM C305–06 (2009). The water to cement ratio of the mix that satisfy the normal consistency requirement according to ASTM C187−16 (2011) varied from 0.24 to 0.34. Two values of water to cement ratio by weight were selected for this experiment; namely: 0.3, and 0.34. Three of the reference specimens and 3 of the specimens cured under DC had a water cement ratio of 0.3. Another 3 reference specimens and 3 of the specimens cured under DC had a water cement ratio of 0.34. Only the mixing water was used for curing, which achieved by wrapping the whole specimen by plastic sheets to prevent any loss in the moisture. The mixing and curing processes were implemented in the lab at 23$^{o}$ C temperature and 55% humidity. Two patches of specimens were prepared, with each patch contains 6 cubes. The first patch has a 225g of water and 750g of Portland cement with a mixing ratio of 0.3. The second patch has 255g of water and 750g of Portland cement with a mixing ratio of 0.34. 3 cubes of each patch were cured while applying 10 V of DC and 3 were cured with no electricity applied.

The naming scheme of the specimens contains a letter followed by a number, then a dash followed by 2 numbers. The letter is either R for reference specimens that were cured without electricity, or E for the specimens that were cured while applying electricity. The number that comes directly after the letter denotes the specimen number. The two numbers after the dash denote the water to cement ratio as percentage (i.e. 30 or 34). All the E specimens were cured while applying 10 Volts to induce a DC on them. A schematic diagram for the curing setup of the E specimens is shown in Figure ).

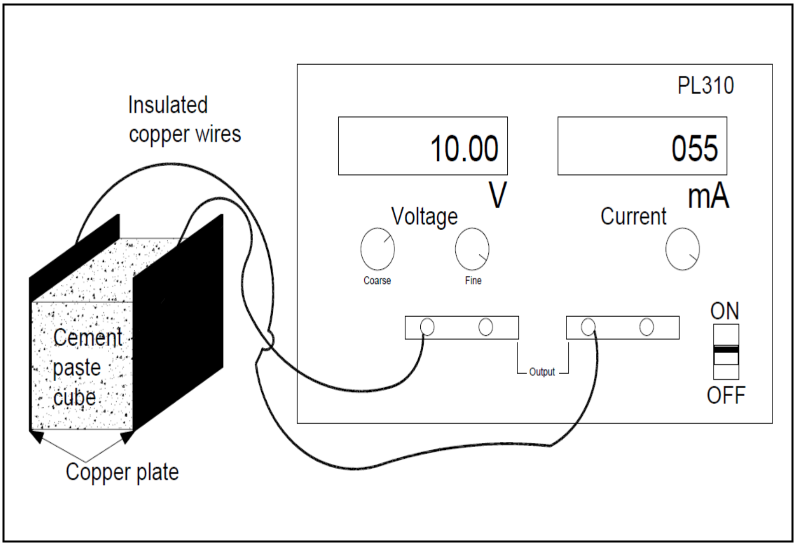


**Figure (1) Setup of cement paste cubes cured while applying DC**

All the specimens were demolded after 7 days from casting. Immediately after the demolding the faces of the cubes that were attached to the electrodes were sanded to remove any irregularities. Then the weight of each specimen was measured and recorded. Then the specimens were put in an oven at 120° C for 24 hours to remove the traces of the moisture from them to simulate the service conditions of the cement paste and eliminate the conductivity of the moisture in the pores (Sun et al., 2004). After drying the specimens, their weights were measured and recorded again to calculate the moisture content using the following formula (1):

$$moisture\ content = \frac{W_{fresh} - W_{dry}}{W_{dry}} * 100\% \quad (1)$$

where; $W_{fresh}$ is the weight of the specimen after demolding, and $W_{dry}$ is the weight of the specimen after oven drying. The electrical bulk resistivity (ρ) of the specimens was measured using a simple setup reported in the literature (Azarsa, & Gupta, 2017). The setup of measuring the bulk resistivity of the specimens contain two stainless steel plates pressed against two opposite sides of the specimens. A wet sponge was used between each plate and the face of the specimen to insure sufficient contact. A solution of water and sodium chloride were utilized to wet the mentioned sponge. The plates were then connected to a bench power supply, which supply an electrical potential and measures the current that passes through the specimen Figure 2(a). The bulk resistivity was then calculated using the formula (2),

$$\rho = \frac{V}{I} * \frac{A}{L} \quad (2)$$

where V: is the applied electrical voltage, I: is the passing current, A: is the cross-sectional area of the specimen, and L: is the distance between the measuring plates. On the other hand, to measure the piezoelectricity of the specimens, the faces that underwent the loading were covered by copper plates that were connected to data logger to record the voltage Figure 2(b). The contact between the loading device and the copper plates were insulated by plastic membrane. The data logger used was the Graphtec midi logger GL220. The faces were the current applied on, were used to apply the loading for the E specimens.

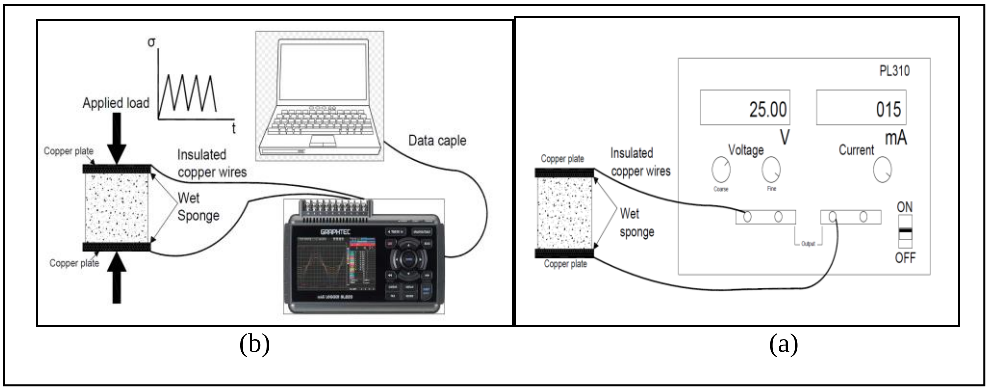


**Figure 1: Testing setup for the specimens (a) electrical conductivity measurement (b) piezoelectricity measurement**

## Results and discussion:

The average moisture content values that have been removed by oven drying from each cube calculated by formula (1) are shown in Table (1), and as average values: 19.35%, and 24.20% for the reference specimens with 0.3, and 0.34 w/c ratio, respectively, and 18.36% and 20.20% for the E specimens with 0.3, and 0.34 w/c ratio, respectively. During the curing of the E specimens, the current that passes the specimens started with high values then dropped with the curing time. This can be attributed to the fact that the mixing water that contains free ions reduces the resistivity of the specimen. Hence, when the cement hardens and the amount of the free water is reduced by the hydration process, the specimens start to develop its resistivity gradually. The measurement of the bulk resistivity of the specimen showed a noticeable difference between the reference specimens and the E specimens. The readings of the passing current were recorded while applying a 25 V electrical potential. The current passed through the reference specimens was 5 mA, while the reading for the E specimens was 15 mA. Hence, the values of the bulk resistivity calculated by formula (2) equals to 25 kΩ.cm, and 8.33 kΩ.cm for the reference and the E specimens, respectively. Which indicates a reduction of 67% in the resistivity of the cement paste due to applying a DC while curing. Increasing the mixing water ratio increases the porosity of the cement paste, hence, increasing its bulk resistivity (Azarsa & Gupta, 2017). However, there was no noticeable difference in resistivity between the E specimens, despite the fact that the w/c ratio were changed. This observation can be attributed such that polarizing of the cement paste using DC eliminates the effect of the conductivity of the pores in the dry condition.

**Table (1) Wet and Dry Weight of the Specimens and its Moisture Content**

| **Sample ID** | **$W_{fresh}$** | **$W_{dry}$** | **Moisture content %** |
|---|---|---|---|
| R1-30 | 306.99 | 257.29 | 19.32 |
| R2-30 | 309.57 | 259.75 | 19.18 |
| R3-30 | 316.12 | 264.39 | 19.57 |
| Average | | | 19.35 |
| R1-34 | 256.31 | 206.02 | 24.41 |
| R2-34 | 257.51 | 207.53 | 24.08 |
| R3-34 | 257.28 | 207.33 | 24.09 |

| Sample ID | $W_{fresh}$ | $W_{dry}$ | Moisture content % |
|---|---|---|---|
| Average | | | 24.20 |
| E1-30 | 248.03 | 211.11 | 17.49 |
| E2-30 | 237.72 | 201.87 | 17.76 |
| E3-30 | 231.59 | 193.26 | 19.83 |
| Average | | | 18.36 |
| E1-34 | 229.91 | 191.38 | 20.13 |
| E2-34 | 236.79 | 196.97 | 20.22 |
| E3-34 | 228.48 | 190.02 | 20.24 |
| Average | | | 20.20 |

All the specimens were tested to measure its piezoelectricity performance. The applied loading was fluctuating between 2.5 kN and 6 kN, which corresponds to stress levels equal to 1 MPa and 2.4 MPa, respectively. The piezoelectrical voltage was recorded and the results are shown in Figure . The specimen E3-34 broke during the initial loading because its surfaces were not regular. The initial values of the voltage corresponding to the initial loading show a noticeable difference between the R and E specimens. In general, the E specimens have more initial voltage at the start of the load application process, which can be attributed to better piezoelectric properties. It is also apparent from the results that the E specimens show enhanced piezoelectrical response when the loading changed between the minimum and the maximum values. It was noticed during the experiment that when the faces were switched the sign of the voltage reading also switches from positive to negative for the E specimens. The mentioned observation can be attributed to specific polarization of the material because of the current application during the curing process. In addition, Figure  shows that the absolute value of the voltage reading at the starting instant of load applying is higher than the peak values of the voltage during the loading process. This can be attributed to realignment of the ions of the nonevaporable moisture inside the pores of the dried cement paste (Sun et al., 2004). After this realignment, the effect of the ions of the nonevaporable moisture diminish, and the piezoelectrical behavior of the specimens starts to be more proportional to the applied load.

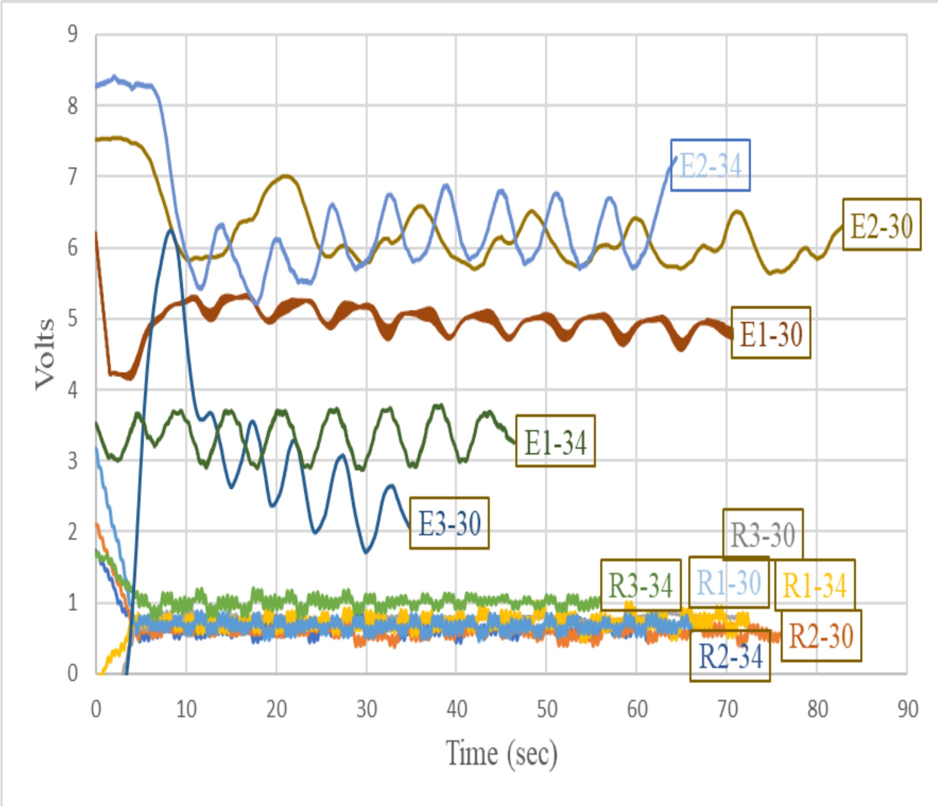


**Figure (3) Piezoelectric voltage due to fluctuating applied load (colored)**

It is worth mentioning that during the application of the load on the E specimens, whenever the load kept at certain level, the piezoelectric voltage measured some value. In other words, there was a sustained charge on the specimens when there is constant applied load on it. On the other hand, this behavior was not observed on the nonpolarized specimens. In addition, when the load was applied on any other faces than the polarized ones on the E specimens, there was minimal piezoelectricity produced from them. When comparing the piezoelectric voltages produced by the E specimens, a noticeable discrepancy can be noticed. This discrepancy can be attributed to the heterogeneous nature of the cement paste and the distribution of the ions inside the cured specimens. Hence, it is apparent that more investigation of the nano structure of the specimens is of a great help to understand the effect of the morphology on the piezoelectrical properties. Moreover, the effect of other factors should also be investigated, such as: curing temperature, and curing voltage, to increase the repeatability of the results.

## Conclusion and recommendations:

This study investigates the effect of the w/c on the electrical conductivity and the piezoelectrical properties of cement hydrates cured under static electrical potential. The experiment included preparing reference specimens to be compared with the polarized specimens. The following conclusions can be drawn from the results of this specific experiment:

1. Curing the cement hydrates while applying static electrical potential polarizes its ions in the direction of the passing current.
2. There is no evidence on any correlation between the electrical conductivity and the piezoelectric property of the polarized specimens and the w/c of the original mix.
3. Polarizing the cement paste specimens using 10 V DC decreased its bulk resistivity by 67% when compared to the nonpolarized specimens.
4. The polarized specimens exhibit well-defined piezoelectric behavior under external applied loading. In contrast, the nonpolarized specimens exhibit very low piezoelectric properties.
5. The polarizing of the specimens is direction-dependent, and the sign of the measured piezoelectric voltage switches when the specimen inverted.

Polarizing the cement hydrates using a static electrical potential showed signs of enhancement of the electrical conductivity of the hydrates, as well as the piezoelectric properties. However, this technique still needs additional investigation to determine the effect of different factors on the polarizing effect, such as: temperature, and voltage. In addition, this technique could be more effective if combined with electrically conductive fillers, such as: metallic fibers, and carbon nanotubes. More investigation is also needed to assess the efficiency of this technique on the concrete since the aggregate in the concrete could disrupt the polarization paths of the ions in the cement paste.

**Declaration of Competing Interest:**

The authors declare that there is no conflict of interest regarding the publication of this paper

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