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Citation	Proceedings of the Thirteenth East Asia-Pacific Conference on Structural Engineering and Construction (EASEC-13), September 11-13, 2013, Sapporo, Japan, C-5-1., C-5-1		
Issue Date	2013-09-11		
Doc URL	http://hdl.handle.net/2115/54294		
Туре	proceedings		
Note	The Thirteenth East Asia-Pacific Conference on Structural Engineering and Construction (EASEC-13), September 11- 13, 2013, Sapporo, Japan.		
File Information	easec13-C-5-1.pdf		



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## AGE EFFECT ON PIEZOELECTRIC PROPERTIES OF CEMENT-BASED PIEZOELECTRIC COMPOSITES CONTAINING SLAG

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## ABSTRACT

In this study, piezoelectric properties of 0-3 type cement-based piezoelectric composite affected by blast furnace slag and material ages are investigated, where the piezoelectric inclusion is lead zirconate titanate (PZT) and the binder consists of cement and slag. 10% to 50% cements by volume are replaced by slag in the composites. Experimental results show that dielectric loss of cement-based piezoelectric composites should be less than 0.759, a threshold to overcome to ensure the success of polarization. 50% slag replacement in cement is a maximum to obtain piezoelectric properties. Adding slag into cement could diminish the resistivity of cement piezoelectric properties become steady at the 60<sup>th</sup> day after the polarization. Curing age is not an important factor to electric and piezoelectric properties. A 20 % replacement of cement by slag is the optimum quantity to improve d<sub>33</sub> and g<sub>33</sub>.

Keywords: Cement, piezoelectric composites, slag, curing, polarization.

## 1. INTRODUCTION

Cement-based piezoelectric composites were investigated to apply to health monitoring of structures more than one decade (Li et al. 2002, Wen and Chung 2002, Sun et al. 2004, Dong and Li 2005, Chaipanich 2007, Jaitanong et al. 2008, Li et al. 2009). For the applications of sensors and actuators in civil engineering structure, 0-3 cement-based piezoelectric composites developed to overcome the matching problem in concrete structures that conventional piezoelectric ceramics or polymers do not contact synchronously with concrete. 0-3 cement-based piezoelectric composites consist of cement binder as the matrix and PZT particles (piezoelectric ceramics) as the inclusion.

Most literatures for 0-3 cement piezoelectric composites discussed the piezoelectric and dielectric properties affected by volume fraction and particle size of piezoelectric ceramics (Li et al. 2005, Huang et al. 2004, Chaipanich 2007), poling time (Chaipanich and Jaitanong 2008), poling temperature (Dong et al. 2007), poling field (Huang et al. 2006), curing time (Wang et al. 2012), the thickness and the forming of specimens (Dong and Li 2005, Cheng et al. 2005, Huang et al. 2007,

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Pan and Chen 2011). To produce high piezoelectric strain factor ( $d_{33}$ ), adding carbon black into 0-3 cement-based composites was reported (Gong et al. 2009), and adding 80 vol.% nano-PZT powder can obtain  $d_{33}$ =53.7pC/N (Li et al. 2009).

Although pozzolanic materials, such as silica fume, slag and fly ash, are commonly used as the replacement of cement in concrete to save the cost and increase the strength and durability, the dielectric and piezoelectric properties of cement-based piezoelectric composites containing pozzolanic materials are seldom studied until PZT-silica fume cement composites was reported (Chaipanich 2007). Besides, mechanical properties of cement-based piezoelectric composites are aged-dependent, but some piezoelectric properties are not. Thereby, we investigate piezoelectric properties of PZT-slag cement composites, and discuss age effect including curing time and material age after the polarization. Cement-based piezoelectric composite we consider here was made by 50% PZT and 50% cement-based binder, and also partial cements were replaced by blast furnace slag from 10% to 50% by volume in the binder.

## 2. MATERIALS AND EXPERIMENTS

The specimen is a 0-3 type composite, where PZT ceramics (inclusion) are randomly oriented distribution in cement-based binders (matrix). PZT particle with a diameter of 75~150 $\mu$ m includes a specific gravity of 7.9, d<sub>33</sub> of 470pC/N, piezoelectric voltage factor (g<sub>33</sub>) of 24mV-m/N, and relative dielectric factor ( $\epsilon_r$ ) of 2100. Cement is type I Portland cement with the fineness of 349m<sup>2</sup>/kg and specific gravity of 3.16. Blast furnace slag was produced by CHC Resources Corporation (Taiwan) with a fineness of 572m<sup>2</sup>/kg and specific gravity of 2.87.

Mixture proportion is shown in Table 1, where PP is a cement-based piezoelectric composite with 50% PZT and 50% cement by volume, no PZT existing in PP is called as PC (100% cement). Meanwhile, PP containing partial slag replacement is referred as SL group, for example, SL10 represents 10% cement by volume replaced by slag, and SL50 is a 50% replacement of slag.

Material	PZT	cement	slag
PC	0	3160	0
PP	3950	1580	0
SL10	3950	1422	144
SL20	3950	1264	288
SL30	3950	1106	432
SL40	3950	948	576
SL50	3950	790	720

Table 1: Mixture proportion of Cement-based piezoelectric composites (kg/m<sup>3</sup>)

To manufacture the specimen of the composite, a combination of PZT powders and the binder was mixed by solar-planetary mill without adding water. Then, mixed materials were placed in a cylindrical steel mold of 15mm in diameter, and applied to a compressive stress of 80MPa by MTS machine for 5 minutes to form a disk-like shape. The specimens were cured in 90°C and relative humidity of 100% for 7, 28 and 56 days (curing days) respectively before polarizations. After

curing, specimens sequentially were polished to 2mm in thickness, coated with silver paint as an electrode, and baked for 30 minutes at 150°C in oven. Then, three voltages (polarizing voltage), 0.5, 1.0 and 1.5kV/mm, were subjected to the specimen in a 150°C silicone oil bath for 30 minutes (polarizing time). During the polarization, polarizing voltage increment should be applied with caution to prevent from current breakdown of the specimen. After the polarization is completed and successful, piezoelectric properties containing piezoelectric strain factor, piezoelectric voltage factor, and relative dielectric factor and resistivity ( $\rho$ ) depending on the age were measured and discussed here.

#### 3. RESULTS AND DISCUSSION

Experimental results show the age-dependent piezoelectric properties when polarizing voltage is applied at 7, 28 and 56 curing days, respectively. Each experimental value as follows is an average of three specimens at least, and also was measured at 24°C and 50% humidity condition.

#### 3.1. Prior to polarization

Before applying electric voltages to polarize the specimen, we measured electric properties of the composite including resistivity, capacitance (C) and dielectric loss (D), by using Impedance Phase Analyzer. Figure 1 shows the resistivity of seven materials, where the resistivity of PC (100% cement) is the highest, PP is the second, and adding slag into cement (SL group) decreases the resistivity except SL50 at 7 curing days. As the curing time increases from 7 to 56 days, the resistivity for PC, PP and SL group (except SL50) also rises. Obviously, excessive slag adding to SL50 has less pozzolanic reaction at the age of 7 days causing a higher resistivity, after that, the resistivity reduces dramatically, compared with the others SL-group materials. In Figure 2, capacitance for PC, PP and SL group gradually reduces with increasing curing time except for SL50. Adding slag to PP from 0 to 50% has a gain of capacitance, but this is not the case for SL50 at 7 curing days.



Figure 1: Resistivity and curing days.

Figure 2: Capacitance and curing days.

As one knows, cement-based composites do not belong to piezoelectric materials due to the instinct of high dielectric loss, but cement containing PZT (PP) does (Li et al. 2002, Dong and Li 2005, Chaipanich and Jaitanong 2008, Pan and Chen 2011). Dielectric loss for PC, PP and SL group is tested without applying voltages, and the results are shown in Figure 3. From this figure, dielectric loss for PC at 7, 28 and 56 curing days is 0.759, 0.851 and 0.765, respectively. A threshold of dielectric loss less than 0.759 is found to trigger the piezoelectricity of cement-based piezoelectric composites. SL-group materials except for SL50 at curing time of 28 and 56 days are easy to be polarized, depicted in Figure 3.



Figure 3: Dielectric loss and curing days.

#### 3.2. Piezoelectric properties

To create piezoelectric property of cement-based composites, polarizing voltage applied to the materials containing piezoelectric ceramics (for example PZT) is required. After the polarization, specimens were placed in the air to measure piezoelectric properties until 60 days. Piezoelectric strain factor was directly measured by  $d_{33}$  Piezometer, and the other electric properties were captured by Impedance Phase Analyzer. Piezoelectric voltage factor and relative dielectric factor were calculated from (Huang et al. 2007)

$$g_{33} = \frac{d_{33}}{\varepsilon_r \times \varepsilon_0} \tag{1}$$

$$\varepsilon_r = \frac{Ct}{A\varepsilon_0} \tag{2}$$

where vacuum dielectric constant  $\varepsilon_0 = 8.854 \text{pF/m}$ , Capacitance C was measured at 1.0kHz, *t* and A are specimen's thickness and electrode area in turn.

To discuss the age effect (after polarization) of piezoelectric properties, some results at 7 and 56 curing days and 1.0kV/mm are shown in Figures 4 ~11. Figure 4 and Figure 5 display the resistivity of cement-based piezoelectric composites become stable after the polarization of 7 days

approximately, and show less difference of curing time between 7 and 56 days. The results also indicate that adding slag will reduce the resistivity. However, in Figure 6 and Figure 7, the change of relative dielectric constant at 7 curing days is intense after the polarization, and at 56 curing days become steady, especially, at the 50<sup>th</sup> day after the polarization. Similarly, d33 and g33 shown in Figures 8~11 also display a steady behavior after the polarization of 50 days.

Experimental results indicate that all SL-group materials can be successfully polarized except for SL50 subjected to polarizing voltage greater than 0.5kV/mm at 28 and 56 curing days. Now, we choose experimental data at the 60<sup>th</sup> day after the polarization to show the effect of curing time and slag contents. In Figure 12, curing time and polarizing voltage show no efficiency to affect the resistivity, but adding slag into cement piezoelectric composites can diminish  $\rho$  in evidence. For relative dielectric constant depicted in Figure 13,  $\varepsilon_r$  rises with increasing slag and polarizing voltage. Nevertheless, curing time also show does less effective to enhance  $\varepsilon_r$ .



Figure 4: Age effect for  $\rho$  at 7 curing days.



PP SL 10

SL 20 SL 30 SL 40

70



Figure 6: Age effect for  $\varepsilon_r$  at 7 curing days.



Figure 7: Age effect for  $\varepsilon_r$  at 56 curing days.



Figure 8: Age effect for d<sub>33</sub> at 7 curing days.



Figure 10: Age effect for g<sub>33</sub> at 7 curing days.



Figure 9: Age effect for d<sub>33</sub> at 56 curing days.

Figure 11: Age effect for g<sub>33</sub> at 56 curing days.



Figure 12: Slag effect for  $\rho$ .

Figure 13: Slag effect for  $\varepsilon_r$ .

Piezoelectric strain constant (d<sub>33</sub>) and piezoelectric voltage constant (g<sub>33</sub>) are important parameters for piezoelectric materials. PC materials have no d<sub>33</sub> and g<sub>33</sub>, but PP and SL-group materials have a behavior of piezoelectric properties if the composite can be polarized successfully. The results of d<sub>33</sub> and g<sub>33</sub> are shown in Figure 14 and Figure 15 respectively. In Figure 14, polarizing voltage applied to SL group from 0.5 to 1.5kV/mm can increase 25% to 50% increment of d<sub>33</sub>, for example, d<sub>33</sub> of SL10 for 0.5kV/mm and 1.5kV/mm at 7 curing days is 32.05pC/N and 46.23pC/N, respectively, with a 44% increment. Figure 15 shows g<sub>33</sub> of SL10 and SL20 at 1.5kV/mm is even higher than that of PZT (g<sub>33</sub>=24mV-m/N). Both figures indicate that 20 % replacement of cement by slag is an optimum quantity to increase piezoelectric properties. The maximum value of d<sub>33</sub> and g<sub>33</sub> occurs at 7 curing days and 1.5kV/mm in SL20, with d<sub>33</sub>=55pC/N and g<sub>33</sub>=27.9mV-m/N. Similar to  $\rho$  and  $\varepsilon_r$ , curing time does not have much influence to d<sub>33</sub> and g<sub>33</sub>.



Figure 14: Slag effect for d<sub>33</sub>.

Figure 15: Slag effect for g<sub>33</sub>.

#### 4. CONCLUSIONS

Cement-based piezoelectric composites were drily pressed by 80MPa and cured at 7, 28 and 56 days, after that, three polarizing voltages of 0.5, 1.0 and 1.5kV/mm, respectively, were applied to polarize the specimen. To obtain piezoelectric property, dielectric loss of cement-based piezoelectric composites had better be less than 0.759 in order to be polarized easily. This leads to a maximum of 50% cement by volume replaced by slag if polarizing voltages are greater than 0.5kV/mm. Electric and piezoelectric properties of cement-based piezoelectric composites become steady after the polarization of the  $60^{th}$  day. Curing time is not an important factor to electric and piezoelectric properties. A 20% replacement of cement by slag is the optimum quantity to increase d<sub>33</sub> and g<sub>33</sub>.

#### 5. ACKNOWLEDGMENTS

This study was financially supported by the Taiwan National Science Council under NSC 101-2625-M-011-001.

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