

PIEZOELECTRIC PROPERTIES OF CEMENT PIEZOELECTRIC COMPOSITES CONTAINING NANO-QUARTZ POWDERS

HUANG HSING PAN¹, WEI-REN LIN¹, and KUAN HUANG²

¹Dept of Civil Engineering, National Kaohsiung University of Science and Technology Kaohsiung, Taiwan ²Dept of Civil Engineering, National Cheng Kung University, Tainan, Taiwan

In order to increase piezoelectric properties of 0-3 type cement piezoelectric composites (piezoelectric cement) developed for structural health monitoring, nano-quartz powders, as the replacement of cement matrix, were added into PZT/cement composites. The piezoelectric cement consists of 50% PZT and 50% cement by volume. Two gradations of PZT inclusions, single-grading and medium-grading, were chosen to fabricate the piezoelectric cement. Nano-quartz powders of 1% to 6% were added to form nanoquartz piezoelectric cement. Experimental results indicate that nano-quartz powders can reduce the porosity of piezoelectric cement. The single-grading piezoelectric cement (PSQ) with 4% nano-quartz powders and the medium-grading one (PMQ) with 2% have the lowest porosity. The maximum values on both piezoelectric strain factor d_{33} and relative dielectric constant ε_r always occur at the minimum porosity of nano-quartz piezoelectric cement. Both the PSQ and the PMQ have the optimum $d_{33}=104$ pC/N. For the PSQ, 4% nano-quartz powders provide a 22% enhancement on thickness electromechanical coupling coefficient K_t . However, the effect of nano-quartz powders displays as less effective to the K_t of the PMQ due to non-uniform distribution of PZT particles. Nano-quartz piezoelectric cement has higher piezoelectric properties able to monitor and detect concrete structural health.

Keywords: Optical microscope, PZT gradation, Structural health monitoring, Sensor, Porosity, Impedance.

1 INTRODUCTION

Cement-based piezoelectric composites have been developed as sensors and actuators for monitoring and detecting the health of concrete structures for 20 years (Li *et al.* 2001, Shen *et al.* 2006, Chaipanich and Jaitanong 2008, Pan *et al.* 2016a). Among them, lead zirconate titanate (PZT) ceramics are often chosen as the inclusion in cement matrix. The 0-3 type of PZT/cement piezoelectric composite, with randomly distributed PZT inclusions, can reduce the differences in acoustic impedance and volume compatibility between PZT and cement (Li *et al.* 2002, Dong and Li 2005). However, the piezoelectricity of the 0-3 type piezoelectric composite is essentially smaller due to the different alignment and orientation of PZT in the cement matrix compared to the 1-3 type and 2-2 type composites at the same volume fraction of PZT inclusions.

Adding adequate additions and admixtures in the 0-3 type cement piezoelectric composite is one of methods to improve their piezoelectric properties, particularly piezoelectric the strain factor d_{33} and relative dielectric constant ε_r . For example, the 0-3 type cement piezoelectric composites



adding suitable amounts of silica fume (Chaipanich 2007), carbon (Jaitanong *et al.* 2008), carbon black (Huang *et al.* 2009), carbon nanotube (Gong *et al.* 2011), and kaolin (Pan *et al.* 2015) can effectively increase d_{33} and ε_r . To be a sensor, the 0-3 type cement piezoelectric composite with higher d_{33} values always has higher sensitivity for structural health monitoring (SHM), especially for d_{33} greater than 70 pC/N. In order to promote the piezoelectricity of the 0-3 type cement piezoelectric composite are still in progress.

For evaluating the piezoelectric properties, some factors such as d_{33} , ε_r , and thickness electromechanical coupling coefficient K_t are often used to show the quality of piezoelectric properties. Quartz powders innately own higher dielectric constant advantageous to increasing d_{33} and ε_r of piezoelectric materials. In this study, quartz particles with nano dimension as the replacement of cement matrix in piezoelectric cement are considered to enhance its piezoelectric property. Two piezoelectric cements, with PZT gradations, single-grading, and medium-grading, are investigated.

2 EXPERIMENTS

The 0-3 type PZT/cement composite consists of Type I Portland cement and PZT particles, both with equal volume. This 0-3 type PZT/cement composite with 50% PZT is called piezoelectric cement after the polarization was applied. The PZT ceramic belongs to Ka type with a specific gravity of 7.9, $d_{33} = 470$ pC/N, $\varepsilon_r = 2100$, $K_t = 0.72$ and a dielectric loss *D* of 1.5, provided by commercial company (Eleceram Technology). The PZT ceramic was initially a flat disk and then pulverized into particles. PZT particles were uniformly distributed in the cement matrix to form a 0-3 type composite. This type of piezoelectric cement is denoted as PP material.

Two PZT gradations were used in the PP. The single-grading (75–150 μ m) PP material is denoted as PSQ material, and the medium-grading (75–600 μ m) one as PMQ material. The quartz particles are spherical with a size of 75–225 nm and called nano-quartz powders. Cement matrix was replaced by nano-quartz powders with six volumes of 1%, 2%, 3%, 4%, 5%, and 6% by volume. For example, PSQ2 and PMQ5 mean the PP material with single-grading PZT and 2% nano-quartz powders, and with medium-grading PZT and 5% nano-quartz powders, respectively.

To make the specimens, first mix fresh cement, PZT particles, and nano-quartz powder without adding water and then put them in a solar planetary mill and spin for 5 minutes to ensure that the raw materials are fully dispersed. The mixture was put into a cylindrical steel mode with a diameter of 15 mm. Then, an 80 MPa compression was applied to the mixture in the mold for 5 min to get circular specimens. Finally, the specimens were cured at 90 °C and 100% relative humidity for 24 hours to make sure that the specimen has enough strength. After curing the specimens, polish them to a thickness of 2 mm.

In the process of making electrodes on the specimen surface, the pretreatment temperature technology of heating the specimen to 140° C (Pan *et al.* 2016b) was applied to promote the piezoelectricity. The specimens were subjected to a 1.5 kV/mm electric field at 150°C for 40 minutes to make them produce piezoelectric properties. Experimental data was measured under controlled conditions of 23°C and 50% relative humidity. The measurement time is between 1 hour (day 0) and 90 days after the polarization was completed. Each experimental value here is taken from the average of three specimens.

3 RESULTS AND DISCUSSION

3.1 Porosity

After curing the specimens for 24 hours, observe specimens with an optical microscope (OM) with a magnification of 350 times. The OM photos were analyzed for pore images, and the porosity of



the specimens of PSQ and PMQ was calculated using PIA software. The results are shown in Figure 1. For the PSQ, the porosity decreases from 2.56% to 2.29% as the content of nano-quartz increases from 0% to 6%. This means that adding nano-quartz powders can reduce the porosity of are single-grading piezoelectric cement. In addition, all porosities of the PMQ are greater than those of the PSQ. This is because the medium-grading piezoelectric cement (PMQ) have three groups of PZT particle size leading to more voids within particles, compared with the size of cement and nano-quartz.

For the PMQ, the values of the porosity exhibit in Figure 1 with 3.57%, 3.14%, 3.03, 3.36%, 3.97, 3.21% and 3.31% for the replacement of nano-quartz is from 0% to 6%. The porosity of PMQ first decreases to the minimum value at 2% nano-quartz, and then increases to the maximum one at 4%. From the OM images shown in Figure 2, the PZT particles showed non-uniform distribution in the PMQ containing 3%–4% nano-quartz powders, resulting in higher porosity of PMQ. The PMQ with 2% nano-quartz powders seems better choice to reduce the porosity.



Figure 1. The relation of the porosity and nano-quartz powders for PSQ and PMQ.



Figure 2. The OM image of PMQ with (a) 2% and (b) 4% nano-quartz powders.



3.2 Relative Dielectric Constant

The values of ε_r of piezoelectric cement depends on material age, and it will approach a stable value after 60 days. Figure 3 is the comparison between the PSQ and the PMQ related to the relation of the relative dielectric constant and the content of nano-quartz powders at 90 days. The PSQ4 with 4% nano-quartz and the PMQ2 with 2% one have the highest ε_r values—545 and 440 respectively. The PSQ4 and the PMQ2 have the optimal ε_r values with corresponding to the minimum porosity of piezoelectric cement shown in Figure 1. This implies that piezoelectric cement might need lower porosity for the target of pursuing higher dielectric constant.

In addition, the ε_r values of PMQ with 3%–6% nano-quartz are lower than that with 0% nanoquartz added. After the inspection of OM images, one finds the distribution of PZT particles is the influence factor on ε_r because the PZT dispersion within the PMQ with 3%–6% nano-quartz are not uniform.



Figure 3. Comparisons of relative dielectric constant ε_r between the PSQ and the PMQ.

3.3 Piezoelectric Strain Factor

The d_{33} value of piezoelectric cement is also age-dependent, and the relation of the d_{33} and nanoquartz content for the PSQ and the PMQ at 90 days shown in Figure 4. The d_{33} development with increasing nano-quartz content is similar to the ε_r in which the PSQ at 4% nano-quartz and the PMQ at 2% have the highest d_{33} values. Both the PSQ and the PMQ have the same maximal d_{33} = 104 pC/N, providing the capability of structural health monitoring.

3.4 Thickness Electromechanical Coupling Coefficient

Electromechanical coupling coefficient is a measure of the conversion efficiency between electrical and mechanical energy in piezoelectric materials. Cement-based piezoelectric composites with higher electromechanical coupling coefficient provides higher energy harvesting in concrete structures. It is known that the K_t of piezoelectric cement is age-independent (Pan *et al.* 2013). Figure 5 shows the relation of the K_t and nano-quartz content in nano-quartz piezoelectric cement. Adding 1%–4% nano-quartz powders in the PSQ can enhance the K_t value from 13.7% to 16.7%, almost a 22% increment. However, the effect of nano-quartz powders in the PMQ is less effective on K_t .





Figure 4. Comparisons of piezoelectric strain factor d_{33} between the PSQ and the PMQ.



Figure 5. Comparisons of K_t between the PSQ and the PMQ.

Comparing the results from Figures 3, Figure 4 and Figure 5, without nano-quartz powders (0%), the piezoelectric properties of PMQ are all higher than those of PSQ. This is because the particle size of PZT in PMQ is greater than in PSQ. Larger PZT particles in piezoelectric cement always provide higher piezoelectric properties. As the amount of nano-quartz powders increases in the PMQ, the piezoelectric properties do not have apparent increments but decreases, except for 2% of nano-quartz. The dispersion for PZT particle and nano-quartz powders is one of dominant factors on the piezoelectric properties of the PMQ.

4 CONCLUSIONS

Two piezoelectric cements with single-grading and medium-grading PZT, respectively, were investigated by adding nano-quartz powders from 1% to 6%. The following conclusions are drawn.

(1) Adding nano-quartz powders can properly reduce the porosity of piezoelectric cement. An optimum amount of nano-quartz powders to have minimum porosity is 4% for single-grading piezoelectric cement and 2% for medium-grading one.



- (2) Both ε_r and d_{33} on piezoelectric cement have the optimal values at minimum porosity. For the PSQ, $d_{33} = 104$ pC/N if 4% nano-quartz powders were added, a 30% increment by comparing with no nano-quartz powders added.
- (3) Adding 4% nano-quartz powders in single-grading piezoelectric cement provides a 22% enhancement on thickness electromechanical coupling coefficient.
- (4) Except for 2% content, the effect of nano-quartz powders to medium-grading piezoelectric cement exhibits less effective on the piezoelectric properties, such as ε_r , d_{33} and K_t , because PZT particles show less uniform dispersion.
- (5) Piezoelectric cement with suitable amounts of nano-quartz powders has d_{33} >100 pC/N, providing the capability of structural health monitoring.
- (6) To increase the piezoelectric properties of nano-quartz piezoelectric cement, suitable dispersions between PZT gradation and nano-quartz powders are needed.

Acknowledgments

The author would like to thank the support by the Ministry of Science and Technology of Taiwan (MOST 108-2221-E-992-008-MY3).

References

- Chaipanich A. Dielectric and Piezoelectric Properties of PZT-Silica Fume Cement Composites, Current Applied Physics, 7, 532-536, 2007.
- Chaipanich, A., and Jaitanong, N. N., *Effect of Poling Time On Piezoelectric Properties of 0-3 PZT-Portland Cement Composites*, Ferroelectrics Letters Section, 35, 73–78, 2008.
- Dong, B., and Li. Z. J., Cement-Based Piezoelectric Ceramic Smart Composites, Composites Science and Technology, 65, 1363–1371, 2005.
- Gong, H., Zhang, Y., Quan, J., and Che, S., *Preparation and Properties of Cement Based Piezoelectric Composites Modified by CNTs*, Current Applied Physics, 11, 653–656, 2011.
- Huang, S., Li, X., Liu, F., Chang, L., Xu, D., and Cheng, X., *Effect of Carbon Black On Properties of 0-3 Piezoelectric Ceramic/Cement Composites*, Current Applied Physics, 9, 1191–1194, 2009.
- Jaitanong, N., Wongjinda, K., Tammakun, P., Rujijanagul, G., and Chaipanich, A., Effect of Carbon Addition On Dielectric Properties of 0-3 PZT-Portland Cement Composite, Advanced Materials Research, 55-57, 377–380, 2008.
- Li, Z. J., Zhang, D., and Wu, K. R., Cement Matrix 2-2 Piezoelectric Composite-Part 1 Sensory Effect, Materials and Structures, 34, 506-512, 2001.
- Li, Z. J., Zhang, D., and Wu, K. R., *Cement-Based 0-3 Piezoelectric Composites*, Journal of the American Ceramic Society, 85, 305–313, 2002.
- Pan, H. H., Lin, D. H., and Yeh, R. H., Influence of Pozzolanic Materials On 0-3 Cement-Based Piezoelectric Composites, in New Development Structure Engineering and Construction, Yazdani, S., and Singh, A. (eds), 929–934, Research Publishing Services, 2013.
- Pan, H. H., Yang, R. H., and Cheng, T. C., High Piezoelectric Properties of Cement Piezoelectric Composites Containing Kaolin, Proceedings of SPIE, 943, 94370R, 2015.
- Pan, H. H., Lin, D. H., and Yang, R. H., *High Piezoelectric and Dielectric Properties of 0-3 PZT/Cement Composites by Temperature Treatment*, Cement and Concrete Composites, 72, 1–8, 2016a.
- Pan, H. H., Wang, C. K., and Cheng, Y. C., Curing Time and Heating Conditions for Piezoelectric Properties of Cement-Based Composites Containing PZT, Construction and Building Materials, 129, 140–147, 2016b.
- Shen, B., Yang, X., and Li, Z. J., A Cement-Based Piezoelectric Sensor for Civil Engineering Structure, Materials and Structures, 39, 37–42, 2006.

