Carbon Letters (2024) 34:1015–1020
https://doi.org/10.1007/s42823-023-00644-7

Online ISSN 2233-4998
Print ISSN 1976-4251

ORIGINAL ARTICLE

Effects of electrodes type and design on electrical stability of conductive cement as exposed to various weathering conditions

Daeik Jang[1] · Beomjoo Yang[2] · Giljae Cho[3,4]

Received: 5 September 2023 / Revised: 25 October 2023 / Accepted: 4 November 2023 / Published online: 6 December 2023
© The Author(s), under exclusive licence to Korean Carbon Society 2023

Abstract
In the present study, the effects of electrodes type (copper, steel or CFRP) and design (plate or mesh) on electrical stability of conductive cement as exposed to various weathering conditions were investigated. To fabricate these composites, multi-walled carbon nanotube and carbon fiber were added to the cement composites by 0.6 and 0.4% by cement mass. Seven different types of electrodes were embedded to the samples, and their electrical stability was examined during the curing period. In addition, the fabricated samples were exposed to water ingress and cyclic heating conditions. Then, the compressive strength of the samples was evaluated to observe the interfacial bonding between the cement paste and electrodes. Based on the experimental results, it was found that the samples showed different electrical stability even their mix proportion was same. Thus, it can be concluded that the type and design of the electrodes are important in measuring the electrical properties of the conductive cement composites. Specifically, an improved electrical stability of electrodes is required when they are exposed to various weathering conditions.

Keywords Conductive cement · Electrodes design · Electrical stability · Weathering conditions

1 Introduction

In recent years, various conductive fillers such as carbon nanotube (CNT), carbon fiber (CF), carbon black, graphite, and graphene have received much attention to fabricate the conductive cement composites, since they can be used in versatile applications (e.g., electromagnetic wave shielding composites, structural health monitoring sensors, and electrical-heating composites) [1–4]. In the early stage of research on conductive cementitious composites, most of the researchers have attempted to achieve an improved electrical conductivity of the cement-based composite by adding many kinds of conductive fillers [5–10]. However, in recent years, many studies have focused on investigating the electrical stability and functionality changes of the conductive cementitious composites when they are exposed to various environmental conditions including extreme exposure temperatures, freeze–thaw cycles, and water absorption conditions. For examples, Dong et al. [11] reported that the piezoresistivity of cement-based sensors can be affected by the water ingress, showing that the water ingress can degrade their piezoresistivity capabilities of the cement-based sensors. Jang et al. [12, 13] examined the effect of exposure temperatures and water ingress on the piezoresistivity and self-heating capabilities of the cement-based composites, respectively. Tafesse et al. [14] investigated electrical resistivity change of cement composites with CNT and CF when they are exposed to chloride penetration. In addition, in the studies by Dong et al. [15] and Moral et al. [16], it is reported that the sensing performance of cement-based sensors with carbon black or CNT and graphite are significantly affected by temperature, water content, and humidity conditions.

Besides, the design of electrodes in conductive cementitious composites, needed to measure the changes in electrical conductivity during applications, has been regarded

✉ Beomjoo Yang
byang@cbnu.ac.kr

1 Department of Civil and Environmental Engineering, University of Pittsburgh, Pittsburgh, PA 15261, USA

2 School of Civil Engineering, Chungbuk National University, 1 Chungdae-ro, Seowon-gu, Cheongju, Chungbuk 28644, Republic of Korea

3 Khanstone P&D Team, Hyundai L&C, 37 Buganggeumho-ro, Bugang-myeon, Sejong 30074, Republic of Korea

4 Alpha Quartz, Phu Tho Lot D3-1 Trung Ha Industrial Park, Tam Nong, Vietnam

as one of the important parameters in such relevant works. Still, most of the studies have used the identical electrodes (e.g., copper and/or steel-based electrodes) to measure the electrical properties of the conductive cement; however, the electrical stability of these electrodes when they are exposed do various weathering conditions are not considered comprehensively [17–20].

Although many studies have attempted to investigate the effects of weathering conditions on the electrical functional changes of the composites, their effects on functional stabilities of various types of electrodes are rarely investigated. In this regard, this study is aimed to observe the comprehensive effects of various electrodes types on the electrical stability of conductive cementitious composites. First, the electrical property changes of the composites with different electrodes were examined during the curing period. Then, their electrical stabilities as exposed to water ingress condition and elevated temperatures, were investigated. Last, the compressive strengths of the composites embedded with various types of electrodes were measured to examine the interfacial bond between the composites and electrodes.

2 Experimental program

In the present study, one mix proportion was used to make the conductive cementitious composites as summarized in Table 1. For a binder material, commercially purchased cement was used, and silica fume (Elkem Inc). was added to the composites. Multi-walled CNT (MWCNT) supplied from the KUMHO Petrochemical Co., Ltd and CF purchased from the Ace C&TECH. Co., Ltd were used as electrically conductive fillers [21]. Based on the previous studies by the authors, the contents of silica fume and superplasticizer (FLOWMIX 3000U) were 10 and 1 wt% of cement, respectively, to enhance CNT dispersion in the cement [12, 22]. In addition, the target flow of the mixture was decided to be 120 mm which is considered as the favorable flow ability to enhance the CNT dispersion [23, 24].

Table 1 Mix proportion used in this study by cement mass

Cement	Silica fume	MWCNT	CF	Water	SP
100	10	0.6	0.4	35	1

The fabrication process of the samples are as follows. Dry materials (cement, silica fume, CNT, and CF) were put into the Hobart mixer, and they were mixed for 5 min. Meanwhile, the water and SP solution was prepared by mixing for 1 min. Then the solution was poured to the dry-mixture and the entire mixture was mixed for an additional 5 min. Then, the mixture was put into the 50 mm cubical molds. Lastly, the seven different types of electrodes with dimension of 30 mm and 70 mm in width and height, respectively, were embedded in the composites. Here, the electrodes regardless of their type were fully embedded in the samples to ensure uniformity in the evaluation criteria. Table 2 indicates the details of the used electrodes. Here, three distinct types of commercially acquired electrodes were employed: copper, stainless steel, and CFRP. Each electrode type had two variations, comprising plated and mesh designs. Among the mesh-typed electrodes, 5-mesh and 10-mesh made of copper and steel were used. The fabricated samples were cured for 28 days, followed by their exposure to various environmental conditions. The samples were exposed to water ingress conditions for 1 week, high temperature conditions at 100 °C for 1 h using the electric furnace for ten cycles, respectively. The changes of electrical resistance were observed in each weathering condition. In addition, the compressive strength of the samples was investigated using the compression machine confirming to the ASTM C109.

3 Result and discussion

3.1 Electrical resistance changes during curing period

Figure 1 shows the changes of electrical resistances for 28 days of curing period. As reported in the previous studies, the electrical resistance of CNT-embedded cementitious composites increased as the curing days increased [22, 25]. Similar results can be observed in the samples when copper electrodes were used without the silver paste (i.e., C, C5, and C10 electrodes). However, the samples having copper electrodes with silver paste showed consistent resistance over the entire curing period. Moreover, steel-mesh type and CFRP electrodes also showed stable resistance with negligible changes.

Table 2 Various type of electrodes used in this study

ID	C–S	C	C5	C10	S5	S10	CFRP
Material	Copper with silver paste	Copper	Copper	Copper	Steel	Steel	CFRP with silver paste
Type	Plate	Plate	5-Mesh	10-Mesh	5-Mesh	10-Mesh	Plate

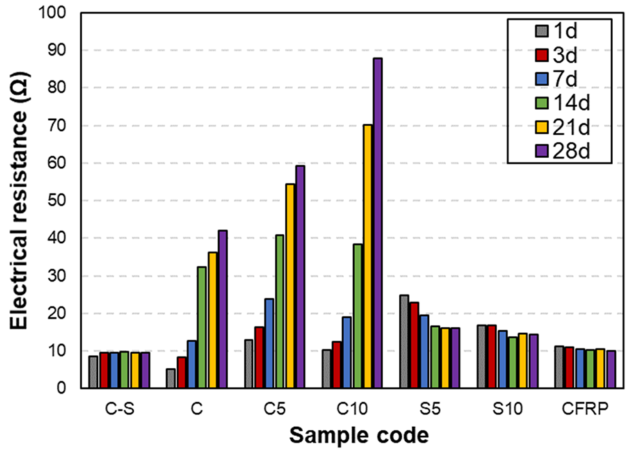


Fig. 1 Electrical resistance of the samples using various types of electrodes for 28 days

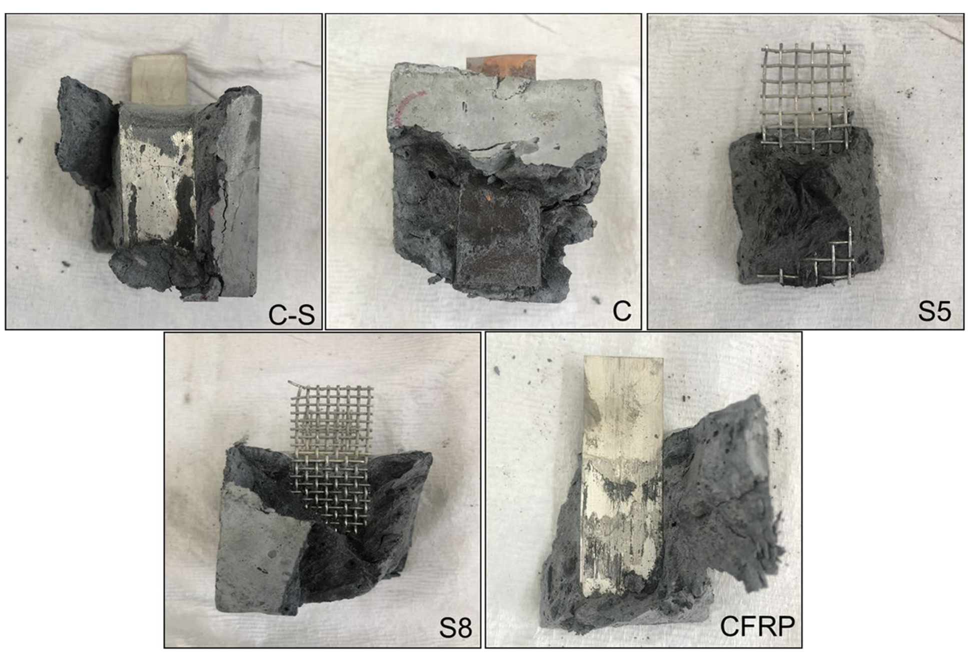
According to these results, it can be said that the copper electrodes may be affected by the moisture inside the cementitious composites and they easily corrode by reacting with the chemical ions/hydrates in the cement composites. Thus, the silver paste is necessary to be coated when copper-based electrodes are used. Alternatively, the steel and CFRP-based electrodes are recommended to be used in cementitious composites which can show the stable electrical properties. Based on the results in Fig. 1, the samples with C5 and C10 electrodes were excluded for other tests because they showed unstable electrical properties. Sample C was included in further tests for the sake of comparison with other electrodes.

After measuring the electrical resistance of the samples as seen in Fig. 1, the condition of the electrodes was visually inspected. The cross-sectional images of the cracked samples are shown in Fig. 2, to support the explained hypothesis for the results in electrical stability. It can be seen that the copper electrode without silver paste (C) was corroded in the cementitious composites due to the chemical ions formed in cement-hydration process. The silver paste coating on copper mitigated the corrosion problem (C–S), protecting the electrodes. However, it was also found that the silver paste can easily be detached from the electrodes, decreasing the electrical conductivity of the conductive cementitious composites [23].

3.2 Exposure to water ingress condition

The samples with C–S, C, S5, S10, and CFRP electrodes were used in water absorption and heating tests to observe the electrical stability as exposed to water ingress and high temperature. The water absorption test was carried out confirming to the ASTM C 1585 specifications [26]. Figure 3 indicates the electrical resistance change ratio as the water immersion time increased. It shows that the water molecules absorbed during the test can increase the electrical resistance of the composites regardless of the electrode types, which are similar with the result of a previous study [13]. However, it should be noted that the magnitude of increase in electrical resistance is dependent on the electrode type. The electrical resistance of the sample using 'C' electrodes showed much higher fraction change in resistance compared to that found in the other samples. This can be deduced from the corrosion occurred on the surface of the copper electrode due to the water molecules (discussed below). The corrosion problem can be mitigated by covering the copper electrode with

Fig. 2 Cross-sectional images of the samples using various electrodes



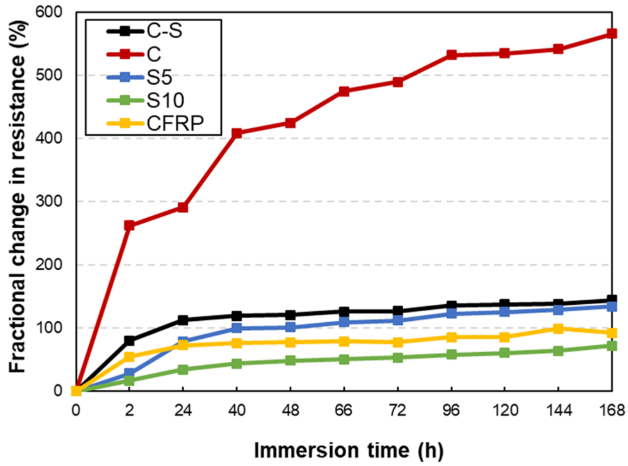


Fig. 3 Resistance change of the samples exposed to water ingress

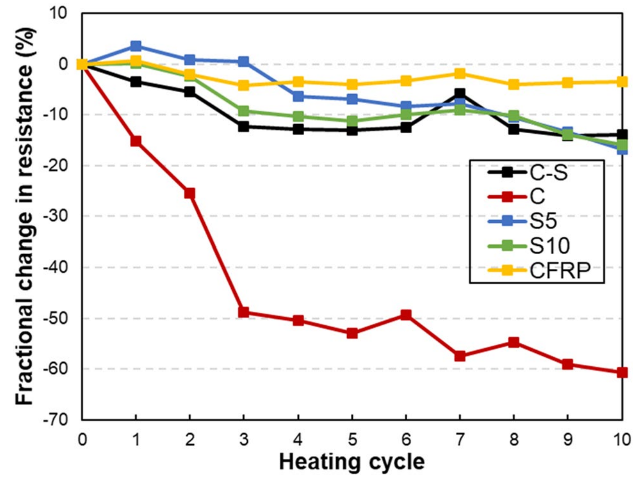


Fig. 4 Fractional change in resistance of the samples during the heating process

silver paste; thus, lower increase of electrical resistance was observed in the sample with C–S electrodes. The samples using steel-based and CFRP electrodes with high resistance to water showed relatively less change in electrical resistance compared to the copper electrodes.

In C–S samples coated with silver paste on copper, the contact resistance between the electrode and the sample is minimized. This allows the conductive network of the sample to be well-represented, resulting in negligible resistance variation over time. In other words, the C–S configuration most accurately reflects the intrinsic conductive network resistance of the sample itself. Previous studies have also frequently employed silver paste to reduce contact resistance [27, 28]. Conversely, electrodes in the copper series (i.e., C, C5, and C10) were found to be highly susceptible to the influence of moisture. It is postulated that the evaporation of moisture alters the interface between the electrode and the composite material, leading to a substantial increase in electrical resistance. This inference is corroborated by the results shown in Fig. 3, where the greatest impact of moisture is observed in the C.

In the steel series (S5, S10), electrodes exhibit a higher contact resistance, resulting in generally greater resistance values on the first day compared to samples utilizing copper (i.e., C, C5, and C10). The observed decrease in resistance over time in the steel series is hypothesized to be influenced more by disruptions in the conductive network due to hydration reactions rather than moisture. In cases where CNT and CF are jointly incorporated, the conductive network of the cementitious composite may exhibit enhanced robustness due to internal structural changes and disturbances induced by hydration reactions [29, 30]. When CF is mixed with CNT, thermal disturbances reduce overlapping regions, thereby enhancing the uniformity of the conductive network, and consequently reducing resistance. Figure 4 corroborates that in samples using steel and CFRP, thermal-induced matrix disruptions tend to decrease the rate of resistance change. Thus, the observed reduction in resistance in the steel series is interpreted as a reflection of an improved conductive network over time, compensating for the initially high resistance recorded due to contact issues. This phenomenon is less conspicuous in C–S and CFRP electrodes, which recorded lower initial resistances. In conclusion, based on the present study, samples incorporating both CNT and CF appear to exhibit relatively stable resistance changes over time when utilizing C–S and CFRP electrodes. For the copper series, the observed phenomena are thought to be influenced by the interface between the electrode and the material, whereas in the steel series, the outcomes are considered to be a reflection of both initial contact resistance and changes in the internal material structure.

3.3 Exposure to high temperature

The electrical resistances of the samples after exposure to 100 °C for ten cycles are exhibited in Fig. 4. It can be seen that all the samples showed a decrease in their electrical resistances as the heating cycles increased. This can be explained from the thermal expansion of the carbon-based conductive fillers (i.e., CNT and CF), leading to the formation of denser conductive networks as well as due the changes in the electrodes upon exposure to high temperature. As seen in the study by Jang et al. [12], the conductive cementitious composites with conductive filler higher than the percolation threshold decreased their electrical resistance as exposed to high temperatures due to the thermal expansion-induced formation of conductive networks. Thus, the observed results showed good agreement with the previous studies in terms of general trend of decrease in electrical

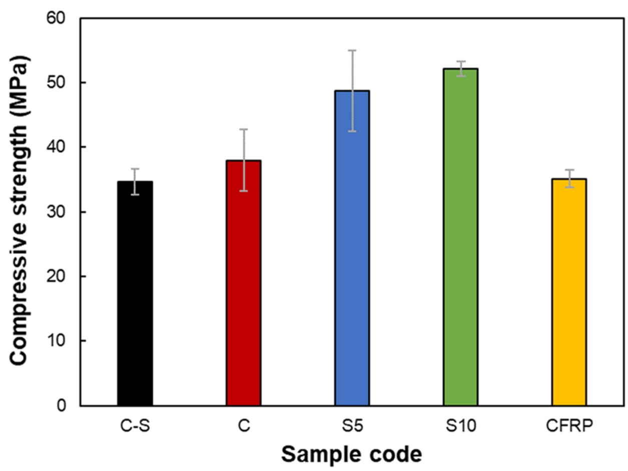


Fig. 5 Compressive strength of the samples using various types of electrodes

resistance. However, it can be found that the degree of the decrease in electrical resistance is different with the electrode types. Except the sample using 'C' electrode, comparable stable electrical resistance can be seen during the heating process. It is because that the steel and CFRP-based electrodes have high resistance to elevated temperature, maintaining the stable electrical properties. Also, the sample using copper electrode coated with silver paste showed stable value, since the silver paste can mitigate the defect occurred on the surface of copper electrode. Based on these results, it can be concluded that the type and design of the electrodes are important in measuring the electrical properties of the conductive cementitious composites. Specifically, a good electrical stability of electrodes is required when the conductive cementitious composites are exposed to various weathering conditions. Furthermore, based on the experimental findings presented in Sects. 3.1 to 3.3, the authors can now elucidate the observed phenomena that underlie the significant alterations in electrical resistivity within the cement composites following exposure to various environmental conditions, as previously reported by the authors [31–33].

3.4 Compressive strength

The compressive strengths of the samples are investigated to examine the interfacial bond between the electrodes and composites, and the results are exhibited in Fig. 5. As seen in Fig. 5, the samples embedding various types of electrodes showed different compressive strengths even the same mix proportion was used in all the samples. This phenomenon can be explained from the interfacial bond. If the electrodes are well embedded in the composites, the interfacial bond strength can increase, resulting in enhancement of compressive strength. However, the lower compressive strength can be achieved when the electrodes are not embedded well in the composites, since the gap between the electrodes and composites can be formed, lowering the compressive strength. In Fig. 5, it can be found that the compressive strengths of the samples using mesh-type electrodes are higher than those using plate-type electrodes. This can be attributed to the cement paste filling the mesh holes, thus increasing the interfacial bond and compressive strengths compared to the plate-types electrodes. A marginal increase in compressive strength of S10 sample compared to S5 samples further endorsed this fact as there were more holes to be filled with cement paste.

4 Conclusion

The present study investigated the effects of various types and designs of electrodes on the electrical stability of conductive cementitious composites as exposed to different weathering conditions. It can be found that the copper electrodes are significantly affected by the exposure to weathering conditions, and the silver paste can protect the electrodes from corrosion problems. Steel-mesh and CFRP electrodes-embedded cementitious composites showed stable electrical conductivity even after exposure to various weathering conditions. Further studies will be conducted on investigating the effects of electrodes on the functionality of composites as used in piezoresistive sensors and electrical heating. Moreover, a systematic investigation into the alterations of electrical, mechanical, and functional properties of the cementitious composites when exposed to various extreme deteriorating conditions will be undertaken.

Acknowledgements This research was supported by Chungbuk National University KNUDP program (2022).

Funding National Research Foundation of Korea, 2020R1C1C1005063, Beomjoo Yang, 2022R1A4A3029737, Beomjoo Yang, KFUPM, Civil, Hammad R. Khalid, Environmental Engineering Department, Hammad R. Khalid.

Data availability The authors confirm that the data supporting the findings of this study are available within the article.

Declarations

Conflict of interest The authors declare no competing financial interest.

References

1. Choi YC (2021) Cyclic heating and mechanical properties of CNT reinforced cement composite. Compos Struct 256:113104

2. Yoon HN, Jang D, Lee HK, Nam IW (2020) Influence of carbon fiber additions on the electromagnetic wave shielding characteristics of CNT-cement composites. Constr Build Mater 269:121238
3. Arabzadeh A, Ceylan H, Kim S, Sassani A, Gopalakrishnan K, Mina M (2018) Electrically-conductive asphalt mastic: temperature dependence and heating efficiency. Mater Des 157:303–313
4. Guo Y, Li W, Dong W, Wang K, He X, Vessalas K, Sheng D (2022) Self-sensing cement-based sensors with superhydrophobic and self-cleaning capacities after silane-based surficial treatments. Case Stud Constr Mater 17:e01311
5. Li W, Dong W, Guo Y, Wang K, Shah SP (2022) Advances in multifunctional cementitious composites with conductive carbon nanomaterials for smart infrastructure. Cem Concr Compos 128:104454
6. Abolhasani A, Pachenari A, Mohammad Razavian S, Mahdi Abolhasani M (2022) Towards new generation of electrode-free conductive cement composites utilizing nano carbon black. Constr Build Mater 323:126576
7. Dong W, Li W, Guo Y, Qu F, Wang K, Sheng D (2022) Piezoresistive performance of hydrophobic cement-based sensors under moisture and chloride-rich environments. Cem Concr Compos 126:104379
8. Li W, Qu F, Dong W, Mishra G, Shah SP (2022) A comprehensive review on self-sensing graphene/cementitious composites: a pathway toward next-generation smart concrete. Constr Build Mater 331:127284
9. Dela Vega MSDC, Vasquez MR (2019) Plasma-functionalized exfoliated multilayered graphene as cement reinforcement. Compos B Eng 160:573–585
10. Tian Z, Li Y, Zheng J, Wang S (2019) A state-of-the-art on self-sensing concrete: materials, fabrication and properties. Compos B Eng 177:107437
11. Dong W, Li W, Zhu X, Sheng D, Shah SP (2021) Multifunctional cementitious composites with integrated self-sensing and hydrophobic capacities toward smart structural health monitoring. Cem Concr Compos 118:103962
12. Jang D, Yoon HN, Seo J, Yang B (2022) Effects of exposure temperature on the piezoresistive sensing performances of MWCNT-embedded cementitious sensor. J Build Eng 47:103816
13. Jang D, Yoon HN, Seo J, Park S (2022) Enhanced electrical heating capability of CNT-embedded cementitious composites exposed to water ingress with addition of silica aerogel. Ceram Int 48:13356–13365
14. Tafesse M, Shiferaw A, Kyoung H, Cho C, Kim H (2022) Effect of chloride penetration on electrical resistivity of CNT–CF/cement composites and its application as chloride sensor for reinforced mortar. Cem Concr Compos 133:104662
15. Dong W, Li W, Lu N, Qu F, Vessalas K, Sheng D (2019) Piezoresistive behaviours of cement-based sensor with carbon black subjected to various temperature and water content. Compos B Eng 178:107488
16. del Moral B, Baeza FJ, Navarro R, Galao O, Zornoza E, Vera J, Farcas C, Garcés P (2021) Temperature and humidity influence on the strain sensing performance of hybrid carbon nanotubes and graphite cement composites. Constr Build Mater 284:122786
17. Dong W, Li W, Wang K, Han B, Sheng D, Shah SP (2020) Investigation on physicochemical and piezoresistive properties of smart MWCNT/cementitious composite exposed to elevated temperatures. Cem Concr Compos 112:103675
18. Jang D, Yoon HN, Yang B, Seo J, Farooq SZ, Lee HK (2022) Synergistic effects of CNT and CB inclusion on the piezoresistive sensing behaviors of cementitious composites blended with fly ash. Smart Struct Syst 2:351–359
19. Liu Y, Wang M, Wang W (2018) Ohmic heating curing of electrically conductive carbon nanofiber/cement-based composites to avoid frost damage under severely low temperature. Compos Part A Appl Sci Manuf 115:236–246
20. Gomis J, Galao O, Gomis V, Zornoza E, Garcés P (2015) Self-heating and deicing conductive cement. Experimental study and modeling. Constr Build Mater 75:442–449
21. Kim GM, Park SM, Ryu GU, Lee HK (2017) Electrical characteristics of hierarchical conductive pathways in cementitious composites incorporating CNT and carbon fiber. Cem Concr Compos 82:165–175
22. Jang D, Yoon HN, Seo J, Lee HK, Kim GM (2021) Effects of silica aerogel inclusion on the stability of heat generation and heat-dependent electrical characteristics of cementitious composites with CNT. Cem Concr Compos 115:103861
23. Kim GM, Nam IW, Yang B, Yoon HN, Lee HK, Park S (2019) Carbon nanotube (CNT) incorporated cementitious composites for functional construction materials: the state of the art. Compos Struct 227:111244
24. Kim GM, Naeem F, Kim HK, Lee HK (2016) Heating and heat-dependent mechanical characteristics of CNT-embedded cementitious composites. Compos Struct 136:162–170
25. Kim GM, Yang BJ, Cho KJ, Kim EM, Lee HK (2017) Influences of CNT dispersion and pore characteristics on the electrical performance of cementitious composites. Compos Struct 164:32–42
26. Jang D, Yoon HN, Farooq SZ, Lee HK, Nam IW (2021) Influence of water ingress on the electrical properties and electromechanical sensing capabilities of CNT/cement composites. J Build Eng 42:103065
27. Kim N, Jang D, Park J, Lee HK (2023) Electrical impedance-based evaluation of self-healing degree in cement composites with biological admixtures. Case Stud Constr Mater. https://doi.org/10.1016/j.cscm.2023.e02479
28. Jang D, Yoon HN, Park S, Kim S, Bae J-H, Kim N, Kim GM, Seo J (2023) Pressure-aided fabrication of CNT-incorporated composites made with fly ash or slag-blended Portland cement. Compos Struct 313:116926
29. Kim GM, Yang BJ, Yoon HN, Lee HK (2018) Synergistic effects of carbon nanotube and carbon fiber on heat generation and electrical characteristics of cementitious composites. Carbon NY 134:283–292
30. Park HM, Park C, Bang J, Lee M, Yang B (2020) Synergistic effect of MWCNT and carbon fiber hybrid fillers on electrical and mechanical properties of alkali-activated slag composites. Crystals (Basel) 10:1–10
31. Jang D, Yoon HN, Seo J, Yang B, Jang JG, Park S (2023) Effect of carbonation curing regime on electric heating performance of CNT/cement composites. J Build Eng 73:106815
32. Yoon HN, Jang D, Kil T, Lee HK (2023) Influence of various deterioration factors on the electrical properties of conductive cement paste. Constr Build Mater. https://doi.org/10.1016/j.conbuildmat.2022.130289
33. Jang D, Bang J, Jeon H (2023) Impact of silica aerogel addition on the electrical and piezo-resistive sensing stability of CNT-embedded cement-based sensors exposed to varied environments. J Build Eng 78:107700

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.