

Research Article

Effect of Carbon Nanotube Size on Electrical Properties of Cement Mortar under Different Temperatures and Water Content

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The effects of temperature and water content on the electrical conductivity of cement mortar with different sizes of carbon nanotubes were studied, and the effect of the size of carbon nanotubes on the electrical conductivity of cement mortar was revealed. The results show that the small diameter carbon nanotubes have the best enhancement effect on the electrical conductivity of cement mortar. The electrical conductivity of cement mortar with different diameters of carbon nanotubes is positively correlated with water content, and with the decrease of the diameter of carbon nanotubes in the sample, the effect of water content on the electrical conductivity of cement mortar becomes smaller and smaller. With the increase of temperature, the electrical conductivity of cement mortar containing carbon nanotubes of different diameters increases in varying degrees, but the increase of samples with small diameter carbon nanotubes is the smallest, indicating that the electrical conductivity of cement mortars with small diameter carbon nanotubes is the smallest, indicating that the electrical conductivity of cement mortar containing carbon nanotubes.

1. Introduction

During the service life of concrete structures, cracks, deformation, and other structural defects will occur, and the structural defects will further expand under continuous load, which will lead to major safety problems. If the real-time monitoring and crack diagnosis of the building structure can be carried out, and the remedial measures for structural damage can be taken in time, the probability of accidents can be greatly reduced. Therefore, much attention has been paid to the health monitoring of concrete structures [1-4]. Currently, the commonly used method for structural health monitoring is to set up sensors on the surface or inside the structure. However, common sensors suffer from limitations such as high construction cost, short lifespan, and easy to destroy the structural integrals. Therefore, it is important to develop a sensor with low cost, stable long-term performance, and good compatibility with concrete.

Intrinsic self-sensing concrete (ISSC) can be used as an ideal sensing material because of its good compatibility and high sensitivity [3, 5, 6]. The principle of ISSC is to add conductive fillers to cement-based materials to achieve self-sensing ability; these conductive fillers are generally carbon fibers (CFs) [7], carbon nanotube (CNTs) [8, 9], carbon nanofibers (CNFs) [8], multi-layer graphene (MLG) [10], and carbon black (CB) [11]. Compared with other conductive fillers, CNTs have higher specific stiffness and specific strength, as well as good electrical conductivity and corrosion resistance. These excellent properties make CNTs widely used in composites.

Carbon nanotube reinforced composites (CNTRC) have excellent functional properties, such as mechanical properties [12–15], electrical properties [16–19], and high-temperature properties [20–22]. Based on these properties, many scholars have investigated the electrical conductivity of carbon nanotube cement-based materials. Yoo et al. [23] found that MWCNTs had a better enhancement effect on the electrical conductivity of cement paste than graphite nanofibers (GNF) and graphene (G), and the fluidity of the composite had an effect on the resistivity of the material. Lee et al. [24] found that the simultaneous addition of dispersed MWCNTs solution and MWCNTs base film to cement composites was able to better enhance the heating properties of the cement paste and obtain the lowest resistance. Cerro-Prada et al. [25] studied the effect of four different mass fractions of MWCNTs on the strength and electrical properties of cement mortars. It was found that the resistivity of cement mortars decreased by 10% at both 0.015 wt% and 0.01 wt% dosing. These studies show that carbon nanotube composites can be used to make sensors for the health monitoring of concrete structures.

In practical applications, concrete structures usually serve in extremely complex dynamic environments, and the changes of ambient temperature and water content are bound to cause changes in the electrical conductivity of carbon nanotube cement-based materials. Some researchers have also paid attention to this problem and carried out research work. Song and Choi [26] studied the effect of internal water content on the initial resistivity and piezoresistive response of carbon nanotube cement-based composites, and found that the resistivity and piezoresistive sensitivity of carbon nanotube cement-based materials increased significantly with the decrease of water content. Han et al. [27] found that there is a nonlinear relationship between piezoresistive sensitivity and moisture content of carbon nanotube cement composites, that is, with the increase of moisture content, the piezoresistive sensitivity of carbon nanotube cement composites increases at first and then decreases. Similarly, the resistance of carbon composites varies with temperature. On the one hand, the increase in temperature leads to an increase in the probability of electron transition and an increase in electrical conductivity [28, 29]. On the other hand, the increase in temperature causes the material to produce expansion strain, which increases the potential barrier of electron transition, and the expansion strain will increase the interface resistance between fiber and matrix and decrease the conductivity. Therefore, in order to use carbon nanotube cement-based composites as a sensor for health monitoring of concrete structures in the real environment, the electrical conductivity of carbon nanotube cement-based composites under different water content and temperature must be studied. However, only single-size CNTs are used in the existing research, and the effect of size effect on the electrical conductivity of the composites remains to be further studied.

In this paper, cement mortar specimens containing four kinds of multi-walled carbon nanotubes with different sizes were prepared. When the moisture content was 0%, 1.62%, 3.16%, 4.87%, and 5.34%, respectively, and the test temperature was 12°C, 20°C, 30°C, 40°C, and 50°C, respectively, the change law of electrical conductivity of multi-walled carbon nanotubes cement mortar (MWCNTs/CM) specimens was studied by the four-electrode method. The effect of different sizes of MWCNTs on the electrical conductivity of MWCNTs/CM was revealed.

TABLE 1: Physical properties of the MWCNTs.

Specification	Diameter	Length	Purity	Ash%	Surface area
L-1020	10-20 nm	5-15 µm	>97%	<3%	$100 \sim 160 \text{ m}^2/\text{g}$
L-2040	20-40 nm	5-15 µm	>97%	<3%	$80 \sim 140 \text{ m}^2/\text{g}$
L-4060	40-60 nm	5-15 µm	>97%	<3%	$40 \sim 70 \text{ m}^2/\text{g}$
S-1020	10-20 nm	$<2\mu m$	>97%	<3%	$100 \sim 160 \text{ m}^2/\text{g}$

2. Experimental Program

2.1. Raw Materials. The MWCNTs are all produced by Shenzhen Nano Port Company, and their physical properties are shown in Table 1; the parameters of MWCNTs in the table are provided by the manufacturer. The cementitious material was 42.5R ordinary Portland cement. Its chemical composition and mechanical properties are shown in Table 2. The sand was natural river sand (sieved 2.3 mm). Polyvinylpyrrolidone (PVP) was selected as the dispersant. The performance parameters of this dispersant are listed in Table 3. Stainless steel mesh was used as the electrode to prevent the electrode from rusting during the maintenance of the specimen.

2.2. Dispersion of MWCNTs. MWCNTs will be entangled in the natural state, and they need to be dispersed in order to function in cement mortar. At present, there are many known dispersion methods, and each method has its advantages and disadvantages. Only one method cannot effectively disperse MWCNTs and maintain a stable dispersion state; so in this study, a combination of physical and chemical methods was used to disperse the MWCNTs to prepare MWCNTs suspensions. Reference [15] has been confirmed through experiments that this dispersion method can achieve a good dispersion effect of MWCNTs in cementbased materials. In this study, a combination of physical and chemical dispersion was used to disperse the MWCNTs to prepare MWCNTs suspensions. The specific actions are as follows (see Figure 1): MWCNTs and PVP were mixed in water at a mass ratio of 1:2 and stirred magnetically for 10 mins, then the mixed solution was dispersed in an ultrasonic disperser for 60 mins.

2.3. Specimen Preparation. The cement mortar specimens were divided into different groups according to the diameter of MWCNTs. The specimens without MWCNTs were used as the control group (marked as group N). The MWCNTs/ CM specimens with diameters of 10~20 nm, 20~40 nm, and 40~60 nm and length of $5~15\,\mu$ m were marked as group M15, M30, and M50, respectively. The MWCNTs/ CM specimens with a diameter of 10~20 nm and a length of $<2\,\mu$ m were marked as group S15. The content of MWCNTs in each group of specimens was 0.1 wt% by cement mass of MWCNTs. The mix ratio and specimen grouping are shown in Table 4.

The cube specimens of 70 mm were prepared by mixing the MWCNTs suspension with cement and sand. Before vibrating the specimen, four electrodes were embedded into

MgO (%)	SO ₃ (%)	Loss (%)	Cl ⁻ (%)	Surface area (m²/kg)	Fineness (%)	Setting time (min)		Compressive strength (MPa)	
						Initial setting time	Final setting time	3d	28d
≤5.0	≤3.5	≤3.5	≤0.06	300	≤8.0	≥45	≤600	≥16.0	≥42.5

TABLE 2: Properties of ordinary Portland cement.

TABLE 3: Properties of surfactant.

Property	Value
pH (5%, 25°C)	3.0-5.0
Ethanol dissolution test	Qualified
Ash of sulfate	≤0.1%
Water content	≤5.0%



FIGURE 1: Specimen preparation process.

the paste according to the spacing of 14 mm, as shown in Figure 1(c).

2.4. Testing Process

2.4.1. Measurement. In this study, the electrical conductivity of MWCNTs/CM specimen was measured by voltammetry. During the measurement, the contact surface between the electrode sheet and the cement-based material will produce additional contact resistance, which will lead to a large error in the test results. Previous studies have shown that the fourelectrode method (see Figure 2) could eliminate this error to a great extent [30]. The voltage is set to a stable DC voltage of 24 V for this test, and the specimen is electrified in advance and then measured after the current is stable to eliminate the influence of polarization effect [31]. The formula for measuring electrical conductivity by fourelectrode method is as follows:

$$\sigma = \frac{IL}{US},\tag{1}$$

where σ is the electrical conductivity, *U* is the voltmeter reading (V), *I* is the ammeter reading (A), *S* is the contact area between the electrode and the specimen, which is 0.0049 m² in this experiment, and *L* is the distance between

the middle two electrodes, which is 0.014 m in this experiment.

2.4.2. Water Content Dependence Test. After 28 days of curing, the specimens were soaked in water at room temperature, and the surface moisture was dried after soaking. The water content was measured by an electronic scale with an accuracy of 0.01 g, and its electrical conductivity was measured by four-electrode method. With the increase of water content, the solution concentration in mortar pores increases, which will lead to the enhancement of ion migration ability, and the polarization effect will be more obvious. In order to ensure the accuracy of the test results, the specimen was electrified for 10 mins before the measurement, and the conductivity was measured when the polarization current no longer decreased.

2.4.3. Temperature Dependence Test. The measurement experiments of electrical conductivity at different temperatures were carried out. Firstly, the specimens of each group were dried in the indoor natural state to avoid the effect of water content, and the surface temperature was measured at room temperature. Secondly, put the specimen into the oven and set the temperature at 20°C, 30°C, 40°C, and 50°C for not less than two hours each time. Finally, when the temperature of the specimen reaches the temperature set in advance and does not change for a period of time, the electrical conductivity is tested immediately. In order to ensure the accuracy of the test data, the infrared thermometer was used to measure the surface temperature of the specimen while measuring the electrical conductivity.

2.4.4. Pore Distribution Characteristics. Pore structure is an important part of cement-based materials. The addition of MWCNTs can improve and refine the pore size of the composites, reduce the porosity of the composites, and reduce the effects of liquid conductivity and water on field emission. In this paper, Auto Pore Iv 9510 high performance automatic mercury porosimeter is used to test the indexes of pore structure in the specimen, including porosity and pore size distribution.

3. Results and Discussion

3.1. MWCNTs Size Dependence of Electrical Conductivity

3.1.1. The Effect of MWCNTs Diameter on Electrical Conductivity. The electrical conductivity of MWCNTs/CM under dry condition is measured. The experimental results show that the diameter of MWCNTs has a great influence on the electrical conductivity of MWCNTs/CM (see Figure 3). When the content of MWCNTs is 0.1 wt%, the

Group	Specification of the MWCNTs	Cement (g)	Sand (g)	Water (g)	MWCNTs (g)	PVP (g)
N	_	600	1800	300	0.6	1.2
M15	L-1020	600	1800	300	0.6	1.2
M30	L-2040	600	1800	300	0.6	1.2
M50	L-4060	600	1800	300	0.6	1.2
S15	S-1020	600	1800	300	0.6	1.2

TABLE 4: Mix design of cement mortar.



FIGURE 2: Testing method of electrical conductivity.



FIGURE 3: Relationship between electrical conductivity of MWCNTs/CM and MWCNTs diameter.

larger the diameter of MWCNTs is, the smaller the electrical conductivity of MWCNTs/CM is. Compared with the cement mortar without MWCNTs, the electrical conductivity of the cement mortar with MWCNTs of diameter 10-20 nm, 20-40 nm, and 40-60 nm increased by 130.6%, 106.5%, and 80.2%, respectively. It shows that under this content, the small diameter MWCNTs can enhance the electrical conductivity of cement mortar better. This phenomenon can be explained by the repulsive volume. In general, polymer chains such as MWCNTs have repulsive volume [32, 33]. If the repulsive volumes of the two polymer chains overlap, another similar polymer chain will not be allowed between them. Previous studies have shown that the size of the repulsive volume decreases with the increase of the length-diameter ratio of the MWCNTs [32]. Therefore, the MWCNTs with large length-diameter ratio produce smaller

TABLE 5: Electrical conductivity and growth rate of cement mortar with different lengths of MWCNTs.

MWCNTs length (µm)	Electrical conductivity σ (S/m)	Growth rate of electrical conductivity (%)
5-15	0.001070	130.6
<2	0.000644	38.8

repulsive volume in the cement mortar matrix, so that more MWCNTs can be distributed in all parts of the cement mortar matrix, so it is easier to lap together to form a conductive path, and then form a conductive network. It can also be understood that when the length-diameter ratio of the MWCNTs is too small, it is more difficult to form conductive networks, just as spherical or flaky conductive fillers are not as good as thin tubular conductive fillers.

3.1.2. The Effect of MWCNTs Length on Electrical Conductivity. The electrical conductivity of the MWCNTs/ CM is affected not only by the diameter of the MWCNTs, but also by the length of the MWCNTs. Table 5 shows the variation of electrical conductivity of the MWCNTs/CM with the length of MWCNTs. As can be seen from the table, compared with the cement mortar without MWCNTs, the electrical conductivity of the cement mortar with MWCNTs of length 5-15 μ m and <2 μ m increased by 130.6% and 38.8%, respectively. It shows that the longer length of MWCNTs can enhance the electrical conductivity of cement mortar better. When the diameter of MWCNTs is the same, although the dispersion effect of MWCNTs with small length in cement-based materials is better, at the same content, it is difficult for them to overlap with each other because of their small length. It does not fully play the role of the conductive path provided by the extremely high length-diameter ratio of MWCNTs for electron transmission. On the other hand, the longer MWCNTs are more

Geofluids



FIGURE 4: (a) Electrical conductivity of MWCNTs/CM under different water contents at 12° C. (b) The growth rate of electrical conductivity when the water content of the specimen is 5.34%.

TABLE 6: Pore structure characteristic parameters of CM and MWCNTs/G	CM.
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Group	Porosity (%)	Average aperture (nm)	Most probable aperture (nm)	Critical aperture (nm)	Total specific surface area (m ² /g)
Ν	16.66	38.91	77.20	130.95	8.228
M15	13.55	34.69	73.49	95.51	7.142
M30	13.72	33.85	74.14	95.45	7.475
M50	14.09	32.39	76.02	120.77	7.917

likely to overlap each other, forming a complete conductive path, which is more conducive to electron transport.

3.2. Moisture Dependence of Electrical Conductivity

3.2.1. The Effect of MWCNTs Diameter. Figure 4 shows the electrical conductivity curve of CM and MWCNTs/CM in the process of water content change at 12°C (the measured temperature on the surface of the specimen during the test). Table 6 shows the fitting results of the relationship between electrical conductivity and water content. It can be seen that the effect of water content change on the electrical conductivity of CM is significantly higher than that of MWCNTs/ CM. In the process of moisture absorption, when the water content of CM increases to 5.34%, the electrical conductivity increases from 0.00452 S/m to 0.00796 S/m, with a growth rate of 76.10%. When the water content of MWCNTs/CM increased to 5.34%, the electrical conductivity of the group with the largest increase in electrical conductivity increased from 0.00832 S/m to 0.00994 S/m, with a growth rate of only 19.47%. The test results show that the electrical conductivity of CM is more sensitive to the water content of the specimen, which is mainly attributed to the different conductive mechanism of CM and MWCNTs/CM. For CM, its conductivity comes from the ionic conductivity of pore solution [34, 35]. The conductivity of MWCNTs/CM is not only related to the ionic conductivity of pore solution, but also to the content and distribution of MWCNTs. The effect of MWCNTs content on the electrical conductivity of cement mortar under different water content has been studied by scholars [26, 36, 37], so we will not repeat it here. The main work of this paper is to explore the effect of MWCNTs size on the electrical conductivity of cement mortar under different water content.

It can be seen from Figure 4, with the increase of water content, the electrical conductivity of M30 and M50 increased by 11.34% and 19.47%, respectively, while the electrical conductivity of M10 tended to be stable after a slight increase in the initial stage, with a growth rate of only 0.934%. As can be seen from Figure 4(a), the functional relationship between the electrical conductivity of cement mortar containing MWCNTs of different diameters and the water content of the specimen accords with Eq. (2).

$$\sigma = Ae^{-W/B} + C, \tag{2}$$

where σ is the electrical conductivity of MWCNTs/CM and W is the water contents at 12°C. The results show that with the decrease of the diameter of MWCNTs in the specimen, the effect of water content on the electrical conductivity of MWCNTs/CM becomes smaller and smaller.

The reasons for the above results are as follows: the analysis of Section 3.1.1 shows that the number of conductive paths formed by large diameter MWCNTs in the specimen is less. When the water content of the specimen increases,



FIGURE 5: Differential curve of pore size distribution of mortar specimen.

the conductive ions are more easily transported through the pore solution to improve the electrical conductivity of cement mortar, which makes the electrical conductivity of cement mortar with large diameter MWCNTs increase more. However, the MWCNTs with small diameter are easier to overlap with each other to form a conductive path. At this time, the electrical conductivity of the specimen mainly depends on the transmission of electrons between MWCNTs, although the change of water content has a positive effect on the migration of conductive ions in the pore solution, but the effect is much smaller than the direct transfer of electrons between MWCNTs, so the change range of electrical conductivity of cement mortar with small diameter MWCNTs is not obvious.

Another important reason is that the transport of solution in cement mortar is affected by pore structure [38]. Figure 5 shows the differential curve of pore size distribution of cement mortar specimens with MWCNTs of different diameters measured by mercury porosimetry, and Table 6 shows the corresponding pore parameters. As can be seen from Figure 5 and Table 6, with the increase of the diameter of MWCNTs, the porosity of MWCNTs/CM increases from 13.55% to 14.09%, and the most probable pore aperture of MWCNTs/CM increases from 73.49 nm to 76.02 nm, that is, the increase of MWCNTs diameter leads to an increase in the number of pores in cement mortar, in which the increment of larger pores is more obvious. As cement mortar is mixed, water will take up a part of the space in the mortar, and as the water is consumed or evaporated, this part of the space eventually forms capillary pores. Therefore, with the increase of the diameter of MWCNTs, the capillary porosity of mortar will also increase, that is, the material becomes more loose. When the internal water content of the specimen increases, the connectivity of pores in the specimen is enhanced, and the ability of conductive ions to transport through the solution is enhanced, and finally the electrical conductivity of cement mortar with large diameter MWCNTs is greatly improved.



FIGURE 6: Variation of electrical conductivity of MWCNTs/CM with water content and MWCNTs length.

3.2.2. The Effect of MWCNTs Length. Figure 6 shows the change of electrical conductivity of MWCNTs/CM with moisture content and MWCNTs length. The enhancement of the electrical conductivity of cement mortar by short lengths of MWCNTs is significantly less than that of long lengths of MWCNTs at the same dosing and moisture content. The main reason for this is that longer lengths of MWCNTs are more likely to lap each other to form a complete conductive pathway.

As the moisture content increases, the electrical conductivity of the specimen containing S-1020 increases from 0.000631 S/m to 0.000797 S/m, with a growth rate of 26.3%. However, the electrical conductivity of the specimen containing L-1020 only increases from 0.01073 S/m to 0.010816 S/m, with an increase rate of 0.8%. This is because the short-length MWCNTs are not easy to overlap with each other and cannot form a complete conductive path at the same content, which leads to the resistance of the composites is greatly affected by the migration of conductive ions in the pore fluid. Therefore, the effect of moisture content on the electrical conductivity of the composites is more significant.

3.3. Temperature Dependence of Conductivity

3.3.1. The Effect of MWCNTs Diameter. Figure 7 demonstrates the effect of the diameter of the MWCNTs on the electrical conductivity of the cement mortar at different temperatures. The electrical conductivity of all three groups of specimens increased to varying degrees with increasing temperature, for example, when the temperature was increased from 12° C to 50° C, the electrical conductivity of cement mortars mixed with 10-20 nm, 20-40 nm, and 40-60 nm MWCNTs increased by 18.69%, 31.30%, and 29.81%, respectively. It can be seen that the electrical conductivity of MWCNTs/CM is less affected by temperature when the diameter of the MWCNTs is small. Further analysis shows that with the increase of temperature, the change of electrical



FIGURE 7: (a) Variation of electrical conductivity of MWCNTs/CM with temperature. (b) The growth rate of electrical conductivity at 50°C.

conductivity mainly goes through two stages. In the first stage (12°C-30°C), the electrical conductivity of each group increases rapidly with the increase of temperature, and the growth rate is less affected by the diameter of MWCNTs. In the second stage (30°C-50°C), the growth rate of conductivity slowed down for all groups of specimens as the temperature increased, and the smaller the diameter of the MWCNTs, the greater the slowdown in the growth rate of the conductivity of the MWCNTs/CM. This may be due to (1) the cement mortar matrix will expand with the increase of temperature, and the MWCNTs with small diameter are more likely to be broken under the action of the expansion force of the mortar matrix, which affects the formed conductive network and restricts the improvement of electrical conductivity. (2) With the increase of temperature, the electrons in the composites can absorb more energy, which makes it easier for electrons to transition and current transfer between MWCNTs. However, the cement mortar with small diameter MWCNTs mainly relies on the overlap of MWCNTs to transfer current, so the increase of electrical conductivity caused by the increase of tunneling effect is relatively small. The number of overlapping MWCNTs in cement mortar with large diameter MWCNTs is less, so the electrical conductivity of MWCNTs/CM is greatly increased with the increase of tunneling effect.

3.3.2. The Effect of MWCNTs Length. At the same temperature and diameter, the longer the length of MWCNTs is, the better the electrical conductivity of cement mortar is (see Figure 8). With the increase of temperature, the electrical conductivity of the specimen with small length increased from 0.000661 S/m to 0.000916 S/m, and the growth rate reached 45.16%. The electrical conductivity of the specimen with longer length only increases from 0.0107 S/m to 0.0127 S/m, with an increase rate of 18.69%. The test results show that the shorter the length of MWCNTs in cement mortar, the greater the electrical conductivity is affected by temperature. The main reason for the above results is that the short-length MWCNTs are not easy to overlap each



FIGURE 8: Variation of conductivity of MWCNTs/CM with temperature and MWCNTs length.

other to form a conductive network. In this case, the resistance of the composite is mainly determined by the resistance caused by the electron transfer between MWCNTs and the resistance produced by the pores of cement mortar. According to the previous analysis, it is known that temperature has a great influence on the electrical conductivity of MWCNTs/CM which does not form a conductive network. Therefore, the effect of short-length MWCNTs on the electrical conductivity of cement mortar is more affected by temperature.

4. Conclusion

In this study, the effects of MWCNTs size, water content, and temperature on the electrical conductivity of MWCNTs/CM were investigated. Through this study, the following conclusions are drawn.

- (1) When the content of carbon nanotubes is 0.1 wt%, the smaller the diameter of carbon nanotubes is, the more obvious the electrical conductivity of cement mortar is. The cement mortar with carbon nanotube diameter of 10-20 nm has the highest electrical conductivity, which is 112% and 128.3% of that of carbon nanotube cement mortar with diameter of 20-40 nm and 40-60 nm, respectively
- (2) The increase of water content can increase the electrical conductivity of MWCNTs/CM in different degrees. When the diameter of carbon nanotube is 10-20 nm, 20-40 nm, and 40-60 nm, the electrical conductivity of carbon nanotube cement mortar increases by 1.20%, 8.19%, and 18.15%, respectively, with the increase of moisture content. The results show that the water content has less effect on the electrical conductivity and higher stability after adding small diameter carbon nanotubes into cement mortar. The electrical conductivity of cement mortar containing large diameter carbon nanotubes is greatly affected by water content
- (3) With the increase of temperature, the electrical conductivity of cement mortar containing carbon nanotubes of different diameters increases in varying degrees, but the increase of samples with small diameter carbon nanotubes is the smallest, indicating that the electrical conductivity of cement mortars with small diameter carbon nanotubes is less affected by temperature

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of 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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