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Nanocarbon-enhanced cement composites for self-sensing and monitoring in transport infrastructure

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A R T I C L E I N F O

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A B S T R A C T

Nanocarbon materials, such as carbon nanotubes (CNT), carbon nanofiber (CNTF), and graphene, have been extensively utilized for the development of electrical conductive cement composites (ECCC) due to their exceptional electrical conductivity. This review focuses on the current state of research on nanocarbon materials-engineered ECCC in the context of self-sensing applications, namely, self-sensing cement composites (SSCC), with a particular emphasis on the progress made in the last decade (2014–2024). Initially, the primary methods for preparing nanocarbon materials-engineered ECCC, including conductive cement-based ECCC and conductive aggregate-based ECCC, are comprehensively reviewed and compared. Subsequently, this review illustrates the electrical signal measurement and conductive theory of nanocarbon materials-engineered ECCC. Furthermore, the impact of nanocarbon materials on the performance of cement composites, encompassing microstructures, workability, mechanical, electrical behavior, and self-sensing properties, is thoroughly discussed. The review also presents case studies on the practical applications of nanocarbon materials-engineered SSCC. Finally, this review discusses the knowledge gaps and remaining challenges for future research. This review contributes to a deeper understanding of the preparation principles behind nanocarbon-engineered SSCC, providing insights for optimizing the design of high-performance SSCC, and holding the potential to drive the practical applications of nanocarbon-engineered SSCC in transportation infrastructures.

1. Introduction

Cement concrete is the preferred structural material for infrastructures such as highways, bridges, buildings, tunnels, and dams [1–3]. Its global consumption is second only to water, with an annual usage of 30 billion tons at the end of 2020 [4,5]. However, concrete structures undergo long-term degradation and damage due to the combined effects of external and environmental loads during their service life, sometimes leading to catastrophic accidents [6–8]. Therefore, continuous monitoring, real-time assessment, and early warning of the "health status" of concrete structures during their service life are of significant importance [9,10]. Detecting

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and addressing issues such as deterioration and defects at an early stage can help extend the lifespan of structures and ensure the safety of users [11,12]. Inspired by the human neural system's response to instructions, the process of continuously monitoring internal changes in concrete structures and assessing their "health status" in real-time is known as structural health monitoring (SHM) [13,14]. Additionally, monitoring and recording traffic information based on the stress-electric signal relationship is crucial for improving road efficiency and ensuring safe operations [15,16]. Although sensing technologies such as strain gauges, fiber optics, gratings, and piezoelectric ceramics have reached relative maturity and have been successfully applied in SHM and traffic information detection in infrastructure over the past few decades [17,18], their practical applications are limited by high costs, unstable performance in service life, and material properties that are incompatible with the parent concrete structure.

As an emerging technology, self-sensing cementitious composites (SSCC) possess the ability to perceive changes in the surrounding environment [18,19]. Also known as self-diagnostic conductive cementitious composites (ECCC) [20,21], these composites are typically prepared by directly incorporating metallic or carbon-based conductive fillers into the cement mixture along with aggregates [22–24]. The addition of conductive fillers at a critical concentration and achieving their uniform distribution within the composite material forms a conductive network, enabling the composite material to exhibit electrical conductivity [25,26]. Deformation of the composite material caused by external loads leads to changes in the conductive network, thereby altering its conductive performance (usually manifested as resistance) [27,28]. Based on this principle, ECCC can monitor stress (force) and strain (deformation) in concrete structures under loading [29–31]. Compared to metal-based fillers, carbon-based fillers do not induce issues such as rusting and corrosion and have higher conductivity [26,28,32]. Furthermore, the concept of using short carbon fibers (CF) to prepare ECCC was proposed in the 1990s and has made some development and application [9,33]. With the rapid advancement of nanotechnology, graphene, carbon nanotubes (CNTs), carbon nanofibers (CNFs), and other nanocarbon materials, have greatly contributed to the development of ECCC [20,34]. These nanocarbon materials offer superior mechanical and electrical properties, and their low dosage has the potential to reduce costs [35]. Moreover, nanocarbon materials are abundant in supply and have been industrialized, providing valuable opportunities for the development and practical application of ECCC [35,36]. Compared to traditional embedded sensors, ECCC has cementitious properties, ensuring compatibility with the parent concrete and similar service life [37,38]. Additionally, the development of ECCC offers advantages such as tunability and scalability, with immense potential to meet the requirements of different working conditions and application scenarios [9,11]. Therefore, ECCC is poised to replace traditional embedded sensors, enhancing the reliability, durability, and safety of concrete structures [39–41]. It is considered a trend in the development of SHM and traffic information detection technologies, providing promising prospects for the advancement of smart buildings and intelligent transportation.

However, nanocarbon materials possess an extremely large specific surface area [42–44], which makes their uniform dispersion within the cementitious matrix highly challenging [45–47]. In particular, the development of ECCC requires the incorporation of nanocarbon materials exceeding the percolation threshold, which undoubtedly increases the difficulty of dispersing nanocarbon materials in the cementitious matrix [39,40]. This poses challenges in establishing effective conductive pathways and inevitably results in significant degradation in the properties of the ECCC, while also increasing the manufacturing cost of ECCC [15,16]. Moreover, conventional processes used to develop conductive ECCC face the dilemma of conductivity-mechanical performance trade-offs [11,48]. Additionally, issues such as limited electrical and sensing performance, unstable electrical signals, and high costs have not been resolved [49,50].

Based on the above analysis, the primary methodologies for the preparation of nanocarbon materials-enhanced ECCC, specifically those incorporating conductive cement and conductive aggregates, are initially comprehensively reviewed and contrasted. Subsequently, this study elucidates and discusses the principles of electrical signal measurement and conductivity theory within the context of nanocarbon materials-enhanced ECCC. Additionally, the influence of integrating nanocarbon materials on the comprehensive performance of cement composites, including hydration processes, microstructural characteristics, workability, mechanical properties, electrical behavior, and self-sensing capabilities, is thoroughly examined. The review further presents illustrative case studies highlighting the practical applications of nanocarbon materials-enhanced SSCC. Ultimately, this review delves into the current knowledge gaps and ongoing challenges facing future research endeavors. By providing an effective and economically viable strategy for the advancement of high-performance SSCC, this review aims to foster and broaden the practical deployment of SSCC in the realms of SHM and traffic information detection.

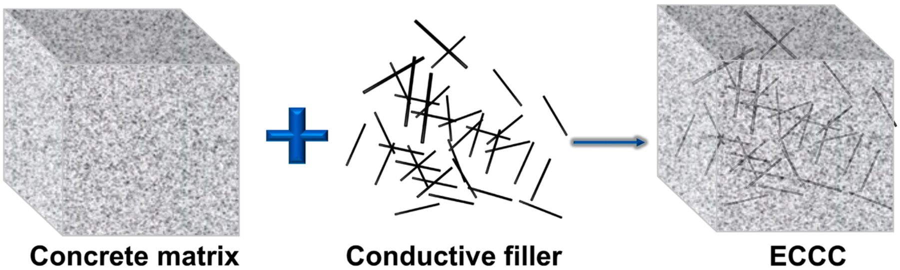

Fig. 1. Structure of traditional conductive cement-based ECCC.

2. Preparation of nanocarbon materials-engineered ECCC

2.1. Conductive cement-based ECCC

As depicted in Fig. 1, ECCC is a multi-phase and multi-component composite, which can be regarded as a composition of the matrix and dispersed conductive fillers within the matrix [29,51,52]. The conductive fillers are distributed within the cementitious composite matrix, forming a conductive network structure that facilitates electron transfer. On the other hand, the matrix is composed of cement and aggregates, providing structural support [24,53].

As shown in Fig. 2**a**, the conductive fillers (e.g., graphene, CNTs, and CNFs) are first dispersed in water to prepare a dispersion, which is then combined with cement and aggregates to develop ECCC (referred to as cement-based conductive ECCC) [54–56], the main properties of nanocarbon materials used for preparing ECCC are presented in Table 1. The key to developing high-performance ECCC lies in achieving the uniform and stable dispersion of nanocarbon materials within the matrix [13,57,58]. This section summarizes and discusses the research progress on the dispersion of graphene, CNT, and CNF in both aqueous solutions and cementitious matrices based on existing literature [59,60]. Due to the high surface energy of nanocarbon materials, their dispersion within the cementitious matrix poses significant challenges [61,62]. Typically, the nanocarbon materials are first pre-dispersed in solution and then combined with cement and aggregates, or they are directly blended with cement particles in dry powder form and subsequently mixed with water and aggregates (as shown in Fig. 2**b**) to prepare the cement-based conductive ECCC [63].

High-quality nanocarbon material dispersions are typically prepared using physical treatments, chemical treatments, and chemical-physical methods [65,66,68].

For instance, ultrasonic treatment leverages its cavitation effect to enhance the dispersion quality of nanocarbon materials in aqueous solutions, as reported in studies [53,69]. By optimizing the choice of ultrasonic power and duration, the dispersion effect can be maximized [23,24]. For a specific nanocarbon material, prolonged ultrasonic power and time typically result in a saturation state of dispersion quality [28,32]. Excessive ultrasonic treatment has minimal positive impact on dispersion quality and can lead to unnecessary energy consumption, potentially damaging the end-caps of CNTs, thereby reducing their enhancement efficiency. Therefore, careful selection of ultrasonic processing parameters, including power and time, is crucial to improve the dispersion quality of nanocarbon materials in aqueous solutions, as highlighted in studies [70,71].

Surface modification treatment can enhance the dispersion quality of nanocarbon materials [72–74]. For example, Dong et al. [75] found that pre-treating 1 wt% (based on the mass of cement) graphene with 15 % polycarboxylate superplasticizer (SP) could uniformly disperse the graphene in an aqueous solution and maintain its stability for up to 6 hours (see Fig. 3**a**). Similarly, high-quality nanocarbon dispersion can be prepared through surface treatment with surfactants, as shown in Fig. 3**b**. However, dispersants can reduce the interaction between nanocarbon materials and the cement matrix, and the chemical functionalization process is

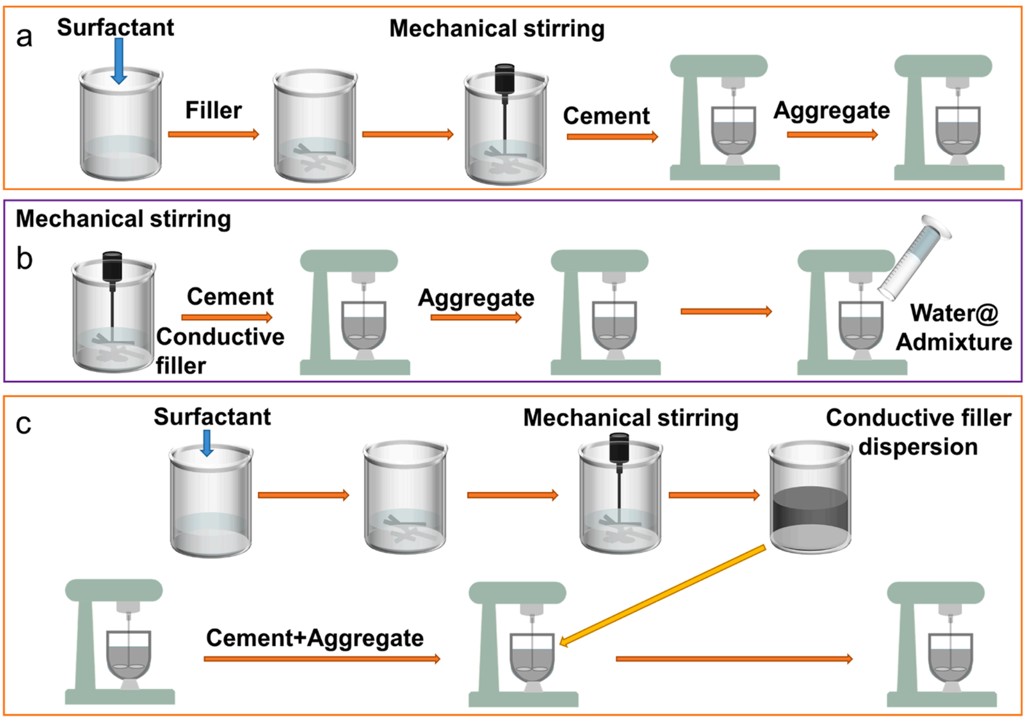

Fig. 2. The dispersion of conductive fillers in water and the preparation of cement composites: (a) surfactant is firstly mixed with conductive filler; (b) using mechanical force to disperse conductive filler; and (c) adding conductive fillers through a post-addition method.

Table 1
The main properties of nanocarbon materials used for preparing ECCC [9,14,49,64–67].

Component	Graphene	CNTs	CNFs
Diameter/thickness (nm)	~1	0.4–2.0 (SWCNTs) 1.0–100 (MWCNTs)	0.5–100
Aspect ratio	600–600,000	1000–10,000	100–1000
Specific surface area (m^2/g)	700–1500	20–1315	100–1000
Elastic modulus (GPa)	> 1100	> 1000	6–200
Tensile strength (GPa)	~125	50–200	400–600

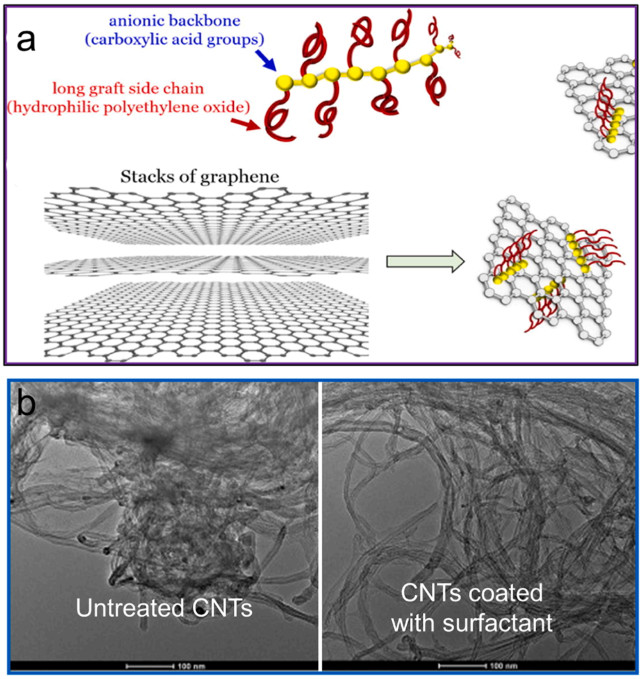

Fig. 3. Dispersion of conductive fillers in solution with the assistance of surfactant: (a) the dispersion of graphene and (b) CNTs with the help of SP [19].

cumbersome, costly, and impractical for practical application [76,77]. It's important to note that achieving high-quality dispersion of nanocarbon materials in an aqueous solution does not necessarily translate to uniform dispersion within the cementitious matrix. Furthermore, the confidentiality of the chemical structure and formulation of most commercially available surfactants (SPs) complicates the optimization of nanocarbon dispersion strategies. Additionally, as demonstrated by Birenboim et al. [78], surfactants can hinder hydration and lead to the entrapment of bubbles in cement slurry. Therefore, the benefits and drawbacks of using surfactants for nanocarbon dispersion must be carefully weighed.

Nanocarbon materials can also be used as additives, mixed with cement powder, and then blended with water and other components to prepare ECCC [79,80]. However, due to the challenging dispersion of nanocarbon materials, the mixing of dry nanocarbon powder with cement powder is extremely uneven. Recent studies have found that cementitious materials, such as fly ash and silica fume, can help improve the dispersion quality of nanocarbon materials in the cementitious matrix [72,80]. As presented in Fig. 4**a** and **b**, the use of silica fume particles can effectively disperse CNTs and graphene inside a composite [32]. However, it should be noted that the cost of this process is the severe agglomeration of volcanic ash materials, which affects the localized chemical reactions and microstructure formation around the mineral admixture particles. In addition, the pre-mixing time is another key factor affecting the dispersion quality of nanocarbon materials in the cementitious matrix. The use of high-energy ball milling equipment can improve the dispersion quality of nanocarbon materials by increasing the shear force. For instance, Ghosh et al. [81] proposed the use of a planetary ball mill to mix graphene with cement particles using 30 g of zirconium oxide balls, ultimately obtaining a uniform graphene-cement mixture. The ball milling process provides an effective solution for dispersing nanocarbon materials in the cementitious matrix and demonstrates great potential for preparing uniformly dispersed nanocarbon-modified cement-based composites. However, applying ball milling can result in the fineness of the cement, increasing the reactivity of the cement particles, and the energy consumption of this process is relatively high. Therefore, the choice of dispersion process for nanocarbon-modified cement composites should be carefully selected and systematically evaluated for its impact on the performance of ECCC.

Overall, despite extensive research aiming to improve the dispersion quality of nanocarbon materials in aqueous solutions through

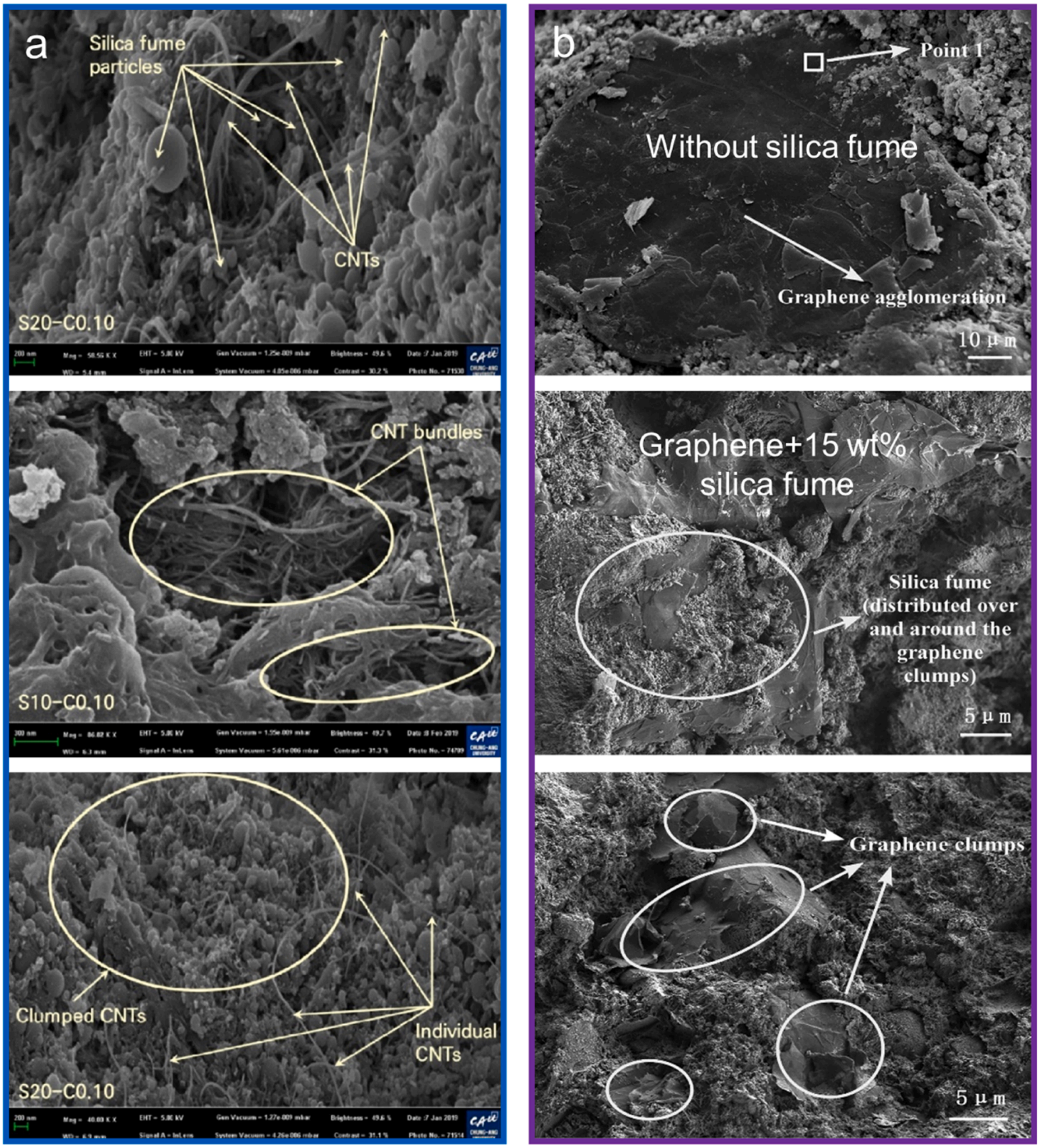

Fig. 4. SEM of the dispersion of conductive fillers assisted by silica fume within cement composites: (a) SEM images of CNT-silica fume [82] and graphene-silica fume [83].

techniques such as ultrasonic dispersion, surfactant treatment, and various combined methods [44,80,84,85], it is regrettable that even with the preparation of high-quality nanocarbon dispersion liquids, agglomeration still tends to occur when mixed with cement. Therefore, the high-quality dispersion of nanocarbon materials in aqueous solutions does not guarantee the same level in the composite. This undoubtedly significantly diminishes the enhancement efficiency of nanocarbon materials. As such, it is imperative to propose an effective solution to address the agglomeration challenge of nanocarbon materials in the cementitious matrix.

2.2. Conductive aggregate-based ECCC

Considering the challenges associated with directly introducing nanocarbon materials into the cementitious matrix, researchers have recently explored the strategy of using nanocarbon-coated aggregates to prepare conductive aggregates [86]. For instance, Gupta et al. [53] have utilized CNT-latex ink spray to prepare a conductive aggregate by coating the original aggregates' surface. Ma et al.

[87] prepared another kind of conductive aggregate by impregnating modified gelatin and carbon black into porous ceramics.

However, the complex synthesis process and high cost associated with the development of conductive aggregates using nanocarbon coatings hinder their practical application. Additionally, there is currently limited research on the development and characterization of nanocarbon-coated conductive aggregates. Therefore, new attempts are required to further explore and study nanocarbon-coated conductive aggregates, aiming to improve their efficiency by altering the utilization of nanocarbon materials and promoting their practical engineering applications.

Considering that cement only constitutes 10–15 % of the total volume of concrete, while aggregates account for a substantial 60–80 % [8,88,89], coating the surfaces of aggregates with nanocarbon materials as conductive media and incorporating the ITZ into the conductive pathways holds the potential to significantly enhance the electrical conductivity and sensing performance of ECCC. Furthermore, the lower specific surface area of aggregates could potentially reduce the amount (and cost) of nanocarbon materials required and avoid issues such as interference with hydration caused by directly adding nanocarbon materials to the cement.

3. Electrical signal measurement of nanocarbon materials-engineered ECCC

The self-sensing characteristics of ECCC arise from the changes in the internal conductive network of the composite material under external forces [18]. Thus, the volumetric resistivity/resistivity index of the composite material can be used to reflect and characterize the self-sensing ability of ECCC [90,91]. Additionally, researchers have found that other parameters such as reactance, impedance, and capacitance can also characterize the sensing behavior of ECCC [9,33,88]. Given that volumetric resistivity and resistivity index are the two most commonly used indicators to measure the sensing behavior of ECCC, this section focuses on their measurement methods.

3.1. Electrode arrangement

The electrodes serve as a bridge between the ECCC and the measurement equipment [10,61]. The material properties, fixation method, and the position of the electrodes directly influence the measurement results and accuracy of the ECCC electrical signals [11,89,92]. Therefore, this section primarily discusses the impact of these parameters on the measurement of ECCC electrical signals. The electrodes for ECCC are typically arranged in a two-probe or four-probe configuration (Fig. 5**a** and **b**).

The electrode materials used for ECCC essentially possess two characteristics [38,94]: low resistance and stable conductivity. The electrode materials reported in current literature mainly include [95,96]: perforated or non-perforated metal sheets, metal foils, metal grids, copper rings, metal/carbon rods, copper strips/wires, and conductive paints (such as silver, copper, and carbon black paints). Among these, copper and stainless steel are the most commonly used electrode. The electrodes are typically attached, embedded, adhered, or clamped to the ECCC, with attached and embedded configurations being the most widely used. Research results have shown that electrode fixation and arrangement can be achieved through six commonly used schemes [97,98]. Both electrode patches and wires are attached to the outer surface of the ECCC, and neither scheme affects the strength. However, attached electrodes are prone to detachment from the surface in practical applications. To address this issue, embedded grids, perforated plates, and ring electrodes have been developed. The advantages and disadvantages of ECCC electrode materials and their arrangement methods are summarized in Table 2, wherein the embedded grid electrode enhances the interface bonding between the electrode and the matrix.

3.2. Resistance measurement methods

Corresponding to the electrode layout schemes, the resistance measurement methods of ECCC include the two-probe and the four-probe methods [35,102,103]. The former is simpler in operation, while the latter can eliminate the contact resistance between the electrode and the ECCC, which has been confirmed in the electrical signal testing results of ECCC containing different conductive fillers [60,104]. The main conduction mechanism is ion conduction, and therefore, changes in the resistance of the composite material caused by external factors (such as stress and strain) may be disturbed [105,106]. Therefore, in practical use, it is desirable to eliminate

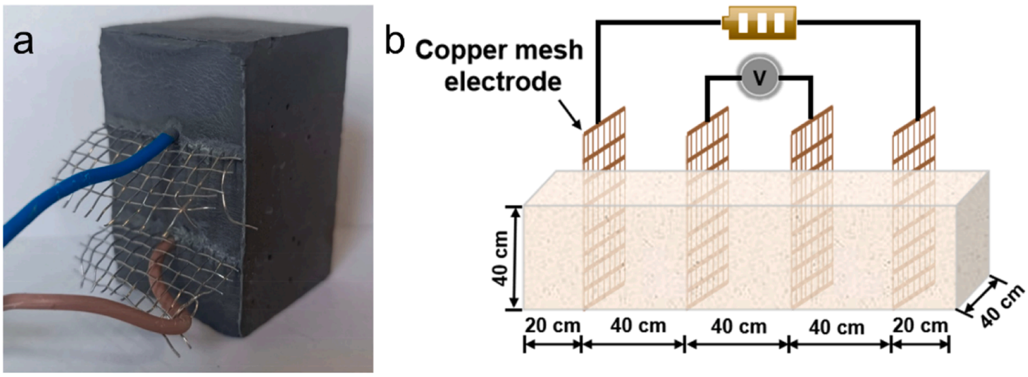

Fig. 5. Fixing style of electrodes in ECCC: (a) two-probe and (b) four-probe method [93].

Table 2
Advantages and disadvantages of electrode materials and their arrangement [37,99–101].

Electrode	Material	Advantages and disadvantages	
		Advantages	Disadvantages
Surface-mounted	Copper plate	The electrode has good contact with the sample	-
	Conductive silver paste	The electrode exhibits excellent contact and conductivity with the sample	High-cost
Embedded	Copper mesh	The electrode establishes a solid contact with the sample, minimizing errors	The insertion of the electrode is challenging
	Stainless steel	It exhibits high sensitivity toward different material systems	-
	Metal plate	-	Negatively affects the strength

the polarization phenomenon of ECCC as much as possible. Specific approaches include [107,108]: 1) applying DC voltage to the ECCC until the resistance reaches a stable state before applying load [109,110]; 2) drying the ECCC to minimize the interference of ion conduction; and 3) using alternating current with equal positive and negative peaks. The polarization effects can still be observed when using alternating current, the negative impact can be negligible by increasing the frequency [40,111].

3.3. Factors affecting sensing signal acquisition

In addition to the types and distribution of conductive additives, electrode selection and arrangement, and sensing signal acquisition, other factors that affect ECCC sensing signal testing include the specimen's curing age, temperature, and humidity [112,113]. With increasing temperature, the electron movement in the conductive fillers intensifies, leading to an increase in charge carriers and thus enhancing the sensing signal of ECCC in the working environment. Generally, the optimal operating temperature for ECCC is around 50℃. ECCC exhibits ion-electron conduction interaction, and humidity mainly affects the proportion of ion conduction in the ECCC system. To ensure measurement stability and accuracy, the contribution of ion conduction should be minimized.

4. Conductive theory of nanocarbon materials-engineered ECCC

4.1. Conductive theory

Understanding the conduction mechanism of ECCC is essential for regulating its electrical and sensing performance. Typically, the types of electrical conduction in ECCC mainly include [37,100]: electronic conduction, hole (tunnel/field emission) conduction, and ion conduction [114,115]. The first two conduction modes primarily depend on the dosage and distribution of conductive fillers, while ion conduction is mainly contributed by the pore solution inside the cementitious composites.

The introduced conductive fillers come into direct contact with each other, thereby forming efficient conductive pathways within the cementitious composite material system. When the distance between conductive fillers is sufficiently close (typically less than 10 nm), even without direct contact between the fillers, the wave nature of charged particles leads to quantum effects. This results in tunneling conduction, forming conductive pathways through potential barriers. Field emission is a special manifestation of tunneling conduction induced by localized strong electric fields [37,101]. For instance, traditional conductive fillers are unable to generate strong electric fields for field emission at low voltages. However, specific nanocarbon materials, such as CNTs, can induce localized strong electric fields at their tips. After hydration, the cementitious composite material forms a solid-liquid-gas three-phase coexistence system. Ions (cations: Ca^{2+}, Mg^{2+}, Al^{3+}, Fe^{2+}, K^{+}, Na^{+}, etc.; anions: OH^{-} and SO_4^{2-}) formed during cement hydration dissolve in the originally filled solution in the voids/pores, enabling ion conduction through interconnected capillaries [38,100]. The ion contribution to electrical conductivity is related to the concentration and migration of ions in the pore solution. Conversely, in a completely dry state, the cementitious composite material behaves as an insulator. The true conduction mechanism of ECCC is highly complex, and generally, the aforementioned conduction modes are interrelated and synergistically contribute to the overall conductivity.

In a dry state, cementitious composites behave as quasi-insulators, with resistivity typically ranging from $10^{\times 6}$ to $10^{\times 9}$ Ω·cm at room temperature [37,101]. Introducing conductive fillers can significantly reduce the resistivity by several orders of magnitude [29,115]. The conductivity of ECCC can be described by Eq. 1:

$$\delta = \delta_{f}\left(\frac{\varphi - \varphi_{c}}{1 - \varphi_{c}}\right)^{\gamma} \tag{1}$$

where Φ represents the percolation threshold, Φc denotes the concentration of fillers, δ represents the conductivity of the fillers. The structure of the conductive additives determines their distribution function of adjacent distances, which in turn determines the percolation behavior. For instance, the distance d_s among fillers can be obtained using Eq. 2:

$$d_{s} = \frac{a}{2}\left(\frac{4\pi}{3\varnothing}\right)^{\frac{1}{3}} \tag{2}$$

where a represents the diameter of the conductive fillers. For fibrous conductive fillers, the distance d_f between them can be obtained using Eq. 3:

$$d_f = \frac{a}{2}\left(\frac{\pi L}{\varnothing}\right)^{\frac{1}{3}} \tag{3}$$

where L represents the length of the fibrous conductive fillers. Generally, fibrous conductive fillers with a high aspect ratio (a=1) have better overlapping capabilities compared to spherical fillers. This allows them to significantly enhance the electrical conductivity of ECCC at lower concentration levels.

As shown in Fig. 6**a**, the variation of resistivity in ECCC with adding conductive fillers can be divided into three stages [37,100]: 1) Insulating stage (Zone A): ECCC exhibits relatively high resistivity, primarily contributed by ion conduction. 2) Percolation stage (Zone B): there is a sharp decrease in resistivity. 3) Conductive stage (Zone C): ECCC demonstrates stable and lower resistivity, with conductive pathways primarily contributed by contact conduction. In Zone A, the concentration of fillers is far low, resulting in large distances between them. Increasing the concentration of fillers, some fillers start to agglomerate and overlap, leading to a critical point, where the resistivity drops significantly by several orders of magnitude. At this point, the composite material transitions from an insulating state to a semiconductor or conductor (Fig. 6**b** and **c**). Consequently, the resistivity remains relatively stable at a lower value. However, ECCC exhibits lower sensitivity. Overall, selecting an appropriate concentration of conductive filler is crucial for fabricating high-performance ECCC.

4.2. *Self-sensing mechanism*

The self-sensing behavior of ECCC arises from the changes in its electrical properties, particularly resistivity, under the influence of external forces [29,51,116]. As shown in Fig. 7**a**, when ECCC is subjected to external loads, its resistivity undergoes various changes [11,29,115]: 1) The redistribution of conductive fillers due to external forces alters their contact status. 2) Changes in the spacing between conductive fillers affect tunneling effects. 3) Local deformations of the cementitious composite material caused by external forces lead to changes in the intrinsic resistivity of the conductive fillers. 4) The stretching/compression behavior of the conductive fillers during tension/compression alters the contact resistivity between the conductive fillers and the composite. 5) Considering the ionic conduction of the cementitious matrix, the fibrous/lamellar conductive phase can be viewed as capacitor plates. The changes in the distance between the capacitor plates and the relative permittivity under external forces result in capacitance variations (see Fig. 7**b**). In fact, these factors collectively influence the sensing performance of ECCC.

In a specific region of the aforementioned conductivity curve, only one or a few factors play a dominant role. As shown in Fig. 7**a**, it is difficult to establish a conductive pathway, and the main factor affecting the resistivity change is capacitance variation. At this stage, ECCC either lacks sensing performance or exhibits poor sensing capabilities. At the beginning of Zone B, changes in capacitance, intrinsic resistivity of the conductive phase, and bonding between the conductive phase and the matrix dominate. Near the percolation threshold, the main influencing factors are changes in tunneling distance between the conductive phases, contact variation between the conductive phases, bonding between the conductive phase and the cementitious matrix, and variation in the intrinsic resistivity of the conductive phase. Towards the end of Zone B, contact variations between the conductive phases, changes in tunneling distance,

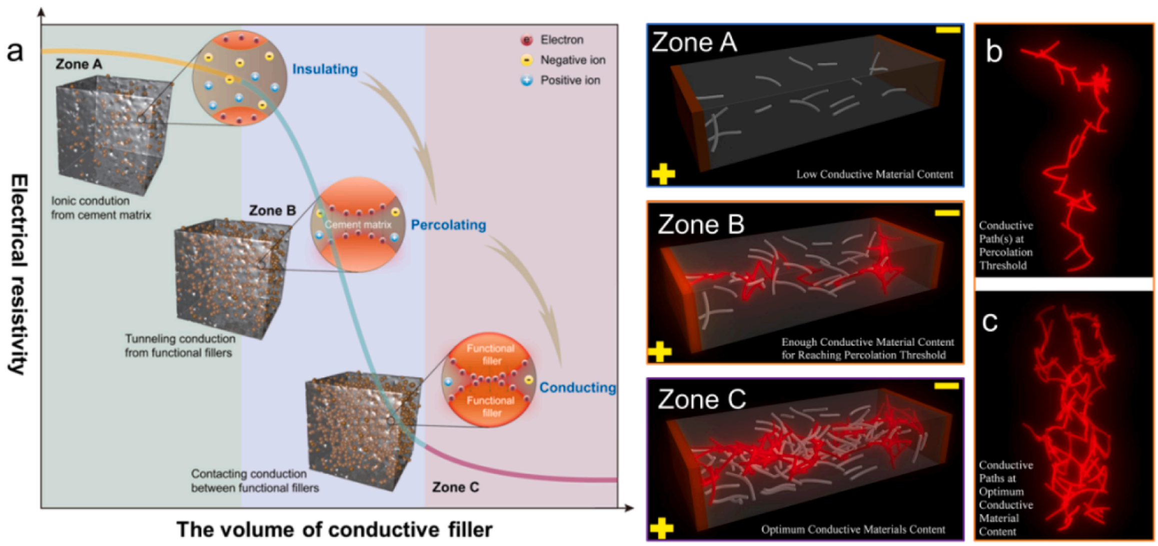

Fig. 6. Illustration of the percolation threshold of fillers in SSCC: (a) relationship between resistivity and concentration of conductive fillers: Insulating stage (Zone A); (b) transition stage (Zone B); and Optimal concentration of conductive fillers stage (Zone C) [115]; (b) conductive pathways in the percolation threshold; and (c) Conductive pathways in the optimal concentration range of conductive fillers [64].

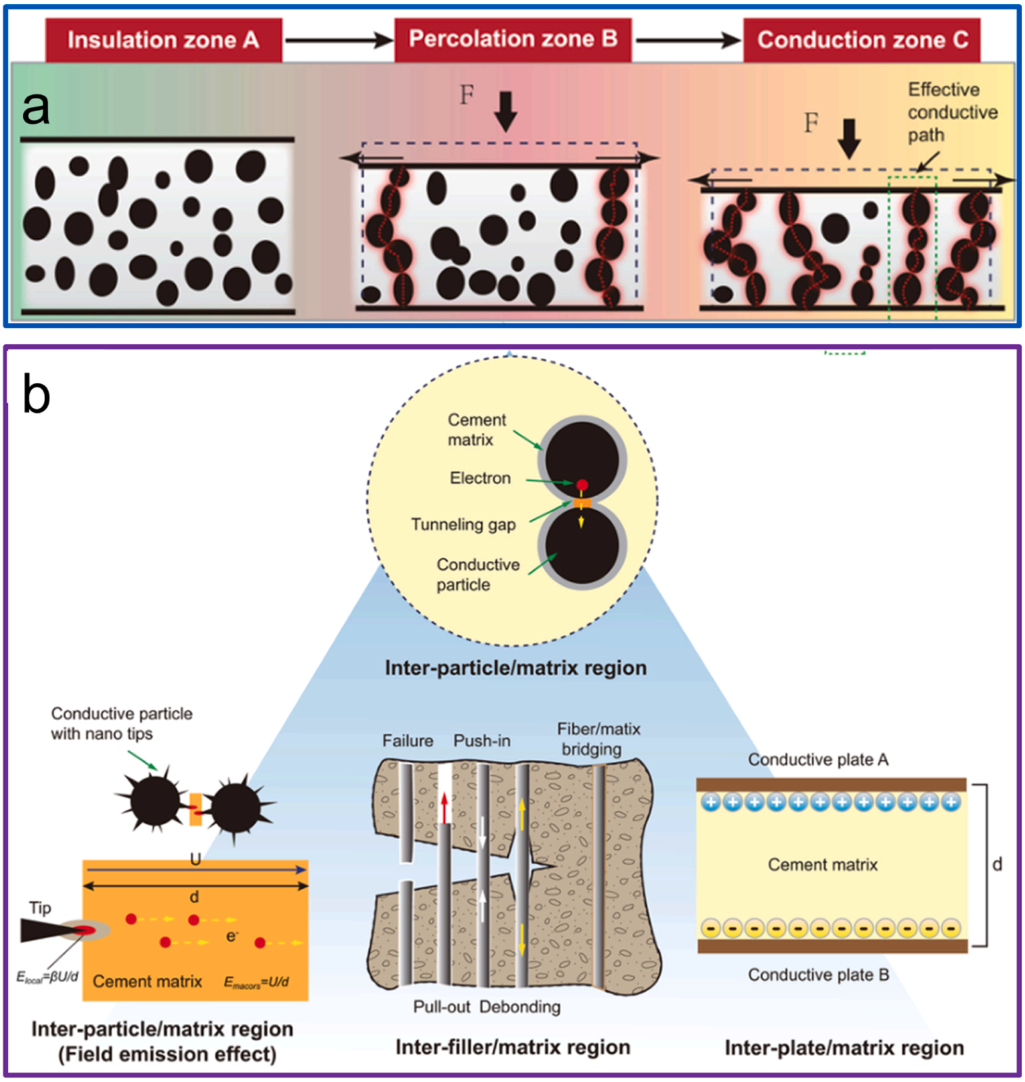

Fig. 7. Sensing mechanism of ECCC [115]: (a) conductive pathways due to deformation, and (b) capacitance variation between the bonding, tunneling resistance, and conductive fillers caused by deformation.

and variations in the intrinsic resistivity of the conductive phase play a dominant role, resulting in excellent sensing performance of ECCC during this stage. In Zone C, variations in contact between the conductive phases and changes in intrinsic resistivity become the primary factors. The entire conductive network stabilizes under external forces, resulting in ECCC exhibiting more stable sensing performance and lower sensitivity [5,9]. Generally, in self-sensing monitoring, a low resistivity helps improve the signal-to-noise ratio but makes it difficult to achieve high sensitivity. Near the percolation threshold, a balance between an appropriate concentration of the conductive phase, high sensing sensitivity, and low resistivity can be achieved. Temperature influences can be categorized into three aspects [11,115]. The relationship between tunneling current and temperature T can be represented by Eq. 4:

$$J = \frac{C_1}{\mathrm{d}^2}\mathrm{e}^{\left(-\mathrm{C}_0\mathrm{d}-\frac{F_a}{kT}\right)} \tag{4}$$

where V represents the tunneling voltage, k is the Boltzmann constant, and E_a is the activation energy. As the temperature increases, the thermal deformation caused by the tunneling gap enlarges, leading to tunneling conduction. This, in turn, hurts the resistivity change of the ECCC specimen. However, since thermal deformation can be neglected, the dominant effect is the tunneling conduction induced by thermal fluctuations. As a result, the resistivity of ECCC exhibits a decreasing trend with increasing temperature.

Based on the principle of Seebeck effect, ECCC can be used to monitor temperature changes in large-volume concrete structures [37,108]. The influence of humidity on the resistivity of ECCC can be mainly attributed to three factors [108,115]: 1) Changes in capillary pore structure, where the loss or infiltration of moisture may cause the contraction or expansion of capillary pores. 2)

Variations in ionic conduction, as water provides a pathway for ion transport, particularly prominent in Zone A. 3) The presence of water affects the contact resistance between the conductive fillers-matrix.

5. Interaction between nanocarbon materials and the cement matrix

Upon contact with water, cement immediately undergoes hydration reactions, which include cement particle dissolution, ion diffusion, and precipitation of hydration products in the solid-liquid coexistence system [117]. Among these, Calcium-Silicate-Hydrate (C-S-H) accounts for 70 % of the total hydration products, and it consists of basic units at the nanoscale [118–121]. Based on this, nanocarbon materials have the potential to regulate and optimize the composition and microstructure of hydration products from a nanoscopic perspective. Therefore, it is of significant importance to deeply understand the influence of nanocarbon materials on cement hydration and their mechanisms in the ECCC system.

Previous research has found that the introduction of nanocarbon materials can significantly promote cement hydration, however, some research results contradict this finding [122–124]. For instance, Makar et al. [125] observed through SEM that some CNTs exhibited a pulled-out morphology in the samples after 24 hours of hydration, while some CNTs were attached to the surface of C-S-H. Li et al. [126] suggested that the influence of CNTs on the cement hydration process is mainly related to their dosage. When the CNT dosage is 0.1 wt%, there is virtually no nucleation effect (see Fig. 8**a**). In fact, the agglomerates of CNTs tend to capture surrounding water molecules, resulting in a decrease in cumulative heat release. The surfactants commonly used to prepare CNT dispersion liquids can also have an impact on cement hydration. Therefore, the impact of CNTs on hydration is not strongly correlated with their dispersion quality but is primarily influenced by their total available specific surface area. Krystek et al. [127] optimized the microstructure of the cement matrix using 0.05 wt% graphene (see Fig. 8**b**). However, some researchers believe that surface-inert nanocarbon materials such as CNTs and graphene cannot significantly promote cement hydration and may even inhibit the hydration process. For instance, Tafesse et al. [128] concluded from non-evaporable water test results that pure CNTs cannot activate or delay hydration; they merely act as nanofillers in the system. Li et al. [126] demonstrated that due to their poor crystallinity and surface curvature, CNTs cannot promote the nucleation and growth of C-S-H (see Fig. 8**a**). However, when inert nanocarbon materials are functionalized with oxygen-containing groups on their surfaces, they acquire the ability to promote cement hydration. For example, Sobolkina et al. [129] used nitric acid pre-treatment on nanocarbon materials, which accelerated the early-stage hydration of C_3S (see Fig. 8**c**). This is mainly because the oxygen-containing groups grafted on the surface of nanocarbon materials can not only bind

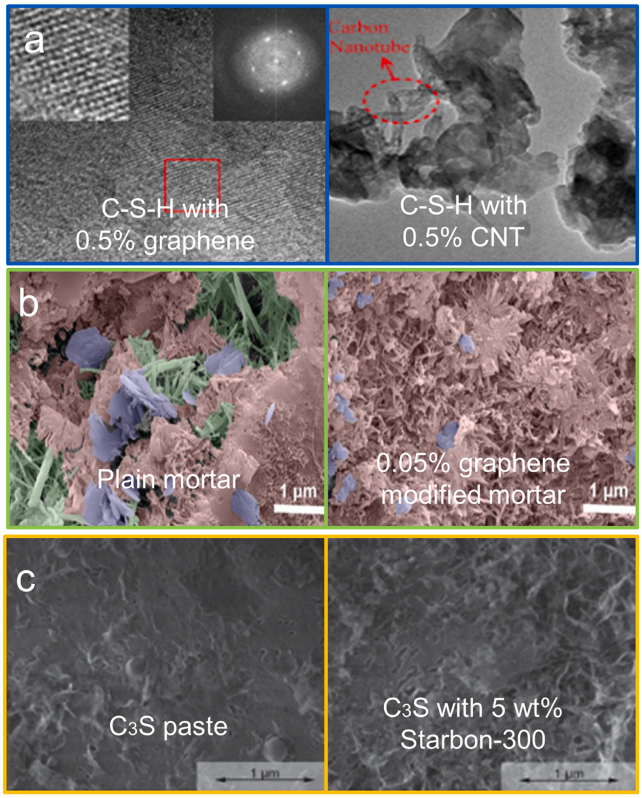

Fig. 8. Effect of nanocarbon materials (CNMs) on hydration and microstructure of composites: (a) TEM analysis of CNM-modified C-S-H [130]; (b) SEM analysis of ordinary and 0.05 % graphene-modified mortar [127]; and (c) SEM analysis of ordinary and 5 wt% Starbon-300 modified C_3S [131].

with Ca^{2+} in the aqueous phase but also increase their interaction with C_3S.

The nanoscale dimensions of nanocarbon materials, with their high specific surface areas, do not guarantee the ability to promote cement hydration. The chemical affinity of their surfaces directly determines whether they possess nucleation effects. Chen et al. [132] investigated the influence of CNTs on the structure of cement hydration products using nanoindentation and found that the introduction of CNTs can transform low-density C-S-H into high-density C-S-H, particularly when the CNT dosage is in the range of 0.1–0.5 wt%, resulting in a 200 % increase in the content of high-density C-S-H. The introduction of nanocarbon materials can also occupy the internal space of the cement matrix, exerting a filling effect and further densifying the microstructure of the matrix. In general, nanocarbon materials contribute to reducing the porosity of the cement matrix, but the final distribution of pores depends on the dosage, dispersion, and morphology of the nanocarbon materials. Additionally, Lin et al. [133] proposed that graphene with wrinkled surfaces can bridge microcracks and fill pores in cement-based composites, while needle-like to rod-like ettringite is connected at the edges of graphene, and C-S-H acts as a glue coating on the surface of graphene, ultimately developing into an interconnected system of graphene-ettringite-C-S-H, leading to densification of the microstructure of composites.

Overall, there is significant inconsistency regarding whether nanocarbon materials possess nucleation effects. From a microscopic perspective, the interfaces in ECCC primarily include the conductive filler-cement matrix interface, conductive filler-conductive filler interface, and conductive filler-aggregate interface. Due to the ultra-large specific surface area of nanocarbon materials, the interfaces have a significantly increased area and a variety of types, directly affecting the electrical contact form and interface bonding quality, thereby determining the electrical performance of ECCCs. Based on this, the regulation and optimization of the interface bonding quality between the aforementioned nanocarbon conductive materials and the cement matrix have the potential to enhance the performance of ECCC.

6. Performance of nanocarbon materials-engineered ECCC

6.1. Workability

The performance of composite significantly affects their mixing, transportation, and pouring quality, and it is of great importance for ensuring and optimizing subsequent mechanical and durability properties [59,134]. This section aims to summarize and discuss existing research findings on the performance of nanocarbon-modified cement-based composites, to understand and comprehend the influence of introducing nanocarbon materials on the performance of cement-based composites.

Generally, adding nanocarbon materials to the cement matrix leads to a significant increase in the viscosity of the mixture (reducing workability) [135,136]. Additionally, the introduced nanocarbon materials are typically unable to achieve uniform dispersion in the cement matrix and often exist in the form of agglomerates. These agglomerated nanocarbon materials trap a significant amount of free water, hindering the lubrication of cement particles. Moreover, the relatively high aspect ratio of most nanocarbon materials leads to a severe decrease in the flowability of the mixture [23,24].

Surfactants can adsorb onto the surface of carbon materials and are considered an ideal choice for mitigating the adverse effects of nanocarbon materials on the flowability of the mixture [65]. For instance, Li et al. [87] found that pre-treating graphene with 0.75 wt % of a surfactant significantly reduced the yield stress and plastic viscosity of the graphene-modified slurry (W/C=0.24) by 80 % and 70 %, respectively. This is mainly because the main chains of the surfactant, which are adsorbed on the graphene surface, extend into the pore solution, while the anions act as adsorption groups through electrostatic interaction with charged cement particles. Therefore, the repulsive force provided by the side chains of the surfactant can simultaneously disperse both the cement particles and graphene nanosheets. Additionally, some research suggests that certain nanocarbon materials, such as graphene nanosheets, have a lubricating effect, which improves the flowability of the mixture. For example, Chougan et al. [88] found that the rheological parameters of the graphene-modified cement slurry groups were superior to those of the corresponding pure cement slurry groups. This may be

Table 3
Effect of nano carbon materials on mechanical strengths of composites.

Type	Nanocarbon materials	Dosage (%)	w/b	Improving compressive strength enhances efficiency (%)	Improving flexural strength enhances efficiency (%)	Reference
Paste	CNT	0.1–1.0	0.20	−3.5–16.8	-	[137]
Paste	CNT	0.1	0.35	27	16.9	[138]
Paste	O-CNT	0.2	0.40	37.1	24.3	[139]
UHPC	CNT	~2	0.28	−4.9–5.5	-	[140]
UHPC	CNT	~0.3	0.20	15.4	-	[30]
Paste	CNF	0.1–1.0	0.20	−28.6–1.8	-	[137]
Paste	CNF	0.1	0.35	0.01	20.96	[141]
Mortar	CNF	0.1	0.485	6	28	[69]
Concrete	CNF	0.1	0.51	8	-	[69]
UHPC	Graphene	0.25–0.5	0.30	29.2–95.7	8.4–39.1	[142]
Paste	GNP	0.02	0.45	39	38	[143]
Mortar	GNP	0.01	0.18	6	< 5	[144]
Mortar	GNP	0.01	0.18	14	< 5	[144]
Concrete	GNP	0.5	0.50	−13–17	6	[137]
UHPC	GNP	0.3	0.20	15.4	-	[30]

attributed to the hydrophobic characteristics of graphene surfaces and the slip effect of the layered structure itself.

Up to date, the research and understanding of the fresh properties of nanocarbon-modified cement-based materials are currently limited, which hinders their widespread acceptance and practical application. Therefore, it is highly necessary to gain a deeper understanding and uncover the influence of nanocarbon materials on the fresh properties of the mixture. Further experimental studies are still required to ensure the effective application of nanocarbon materials in practical applications.

6.2. Mechanical properties

The mechanical performance of cement composites is commonly regarded as a crucial indicator for their practical application, directly affecting the serviceability and service life of concrete structures, among other factors. As shown in Table 3, there remains some controversy regarding the influence of directly incorporating CNTs and graphene into the cement matrix on the mechanical properties of cement-based materials. Some results demonstrate that nanocarbon materials have an enhancing effect, while others indicate that they have little to no strengthening effect, or even a slight decrease in the mechanical performance of cement-based composites. Such variations in results and perceptions are primarily caused by factors such as the type of nanocarbon material added, the dosage, dispersion quality, morphology, and surface chemical characteristics.

Furthermore, Ramezani et al. [145] revealed that CNTs with an average diameter of 20–32.5 nm and length of 10–20 μm exhibited the most effective improvement in mechanical properties. Additionally, Tamimi et al. [128] demonstrated that CNT-COOH was significantly superior to CNT-OH in improving the mechanical strength of the composite. Furthermore, Gholampour et al. [146] conducted a comparative study on the influence of graphene with different levels of reduction on the strength of cement-based composites. Konsta-Gdoutos et al. [147], through nanoindentation studies, discovered that the introduction of 0.5 wt% CNTs tended to generate high-density C-S-H within the system and decrease the total porosity of the matrix (see Fig. 9**a**). Nanocarbon materials, due to their exceptional aspect ratio, can exhibit bridging effects and suppress the initiation and propagation of microcracks (see Fig. 9**b**).

In conclusion, the impact of introducing nanocarbon materials such as CNTs and graphene on the mechanical properties of cement composites depends primarily on factors such as their surface chemical affinity (whether chemically treated), morphology, dosage, and dispersion quality. To ensure the efficient enhancement of the performance of cement-based composites by nanocarbon fillers, it is crucial to ensure that the introduced nanocarbon materials are uniformly and stably dispersed within the system.

6.3. Electrical performance

Generally, the electrical resistivity of cement-based composite materials decreases with an increase in the dosage of conductive fillers [10,105,106]. By studying the correlation between the resistivity of cement-based composites and the dosage of conductive fillers, the optimal dosage of the fillers (i.e., percolation threshold) can be determined. However, as mentioned earlier, the significant

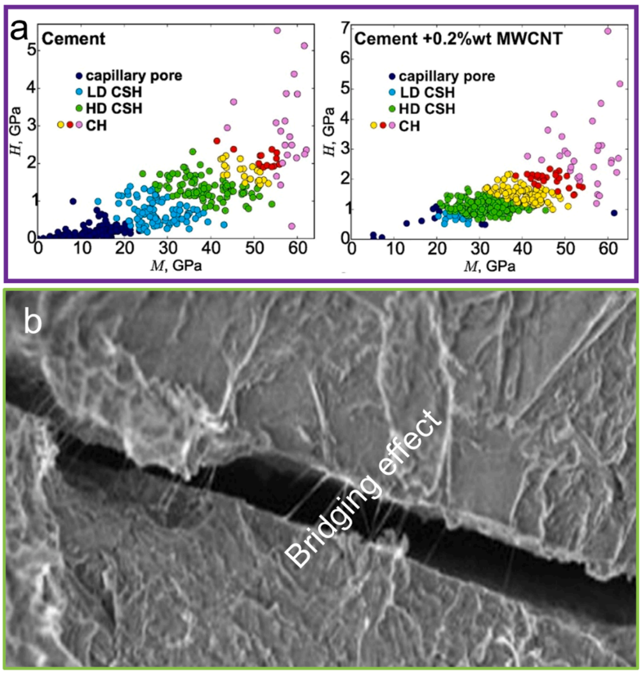

Fig. 9. The roles of nanocarbon materials in SSCC: (a) influence of CNTs on the indentation hardness (H) and modulus (M) of cementitious matrix [132], and (b) bridging effect of CNTs [19].

variations in resistivity are attributed to the differences in the preparation methods of cement-based composites, indicating that the type and dosage of conductive fillers are the main influencing factors but not the sole determinants of resistivity changes.

For one-dimensional nanocarbon materials like CNTs, their incorporation can significantly decrease the electrical resistivity of cement-based composite materials, and the dosage of CNTs generally exhibits a linear relationship with the resistivity [18,107]. Extensive research has shown that the percolation threshold of CNTs is typically around 0.1–1.0 wt%, which primarily depends on the mix proportion of the cement-based materials and the physical parameters of the CNTs, such as specific surface area and aspect ratio [10,105,108]. It is worth noting that the introduction of CNTs often leads to an initial increase followed by a decrease in the compressive strength of cement-based composites. The initial increase is mainly attributed to the ability of CNTs to inhibit the initiation and propagation of microcracks, optimizing the pore structure of the matrix. However, due to their large surface area, CNTs tend to adsorb a considerable amount of free water, resulting in poor workability of the mixture and ultimately leading to a decrease in mechanical performance. Additionally, the formation of a weak CNT-cement interface can occur with inert CNTs, and excessive incorporation of CNTs can create internal defects within the cement-based composites, further contributing to the deterioration of mechanical properties.

Due to its high conductivity, stable resistivity, and sensitivity, two-dimensional graphene nanosheets have emerged as a potential choice for the fabrication of ECCC. Studies have shown that the percolation threshold of graphene is around 2 vol%, at which point the introduced graphene can form a stable conductive pathway within the cement-based material [10,105]. However, the poor dispersibility of graphene and the weak interface bonding between graphene and the cement matrix pose significant technological challenges in the development of high-performance ECCC.

6.4. Self-sensing property

The self-sensing performance of ECCC refers to the ability to change the internal conductive network structure of the material under external forces/deformation, resulting in a variation in resistivity. This variation can be used to assess the deformation of concrete structures and identify microcracks [109–111]. Parameters such as sensitivity, repeatability, hysteresis, and signal-to-noise ratio are used to characterize the self-sensing performance of ECCCs. Among these parameters, sensitivity and repeatability are the most important ones. Sensitivity can be characterized by indicators such as the maximum fractional change in resistivity (FCR), the stress sensitivity coefficient, and the strain sensitivity coefficient (Gauge Factor, GF) [11,18,112]. Repeatability can be evaluated by curve coincidence/maximal difference during cyclic loading and testing. It should be noted that different loading conditions may result in different sensitivity and repeatability evaluation parameters for ECCCs, indicating that ECCCs exhibit varying self-sensing performance under different loading conditions. [13] illustrates the variation in the self-sensing performance of ECCCs under monotonic

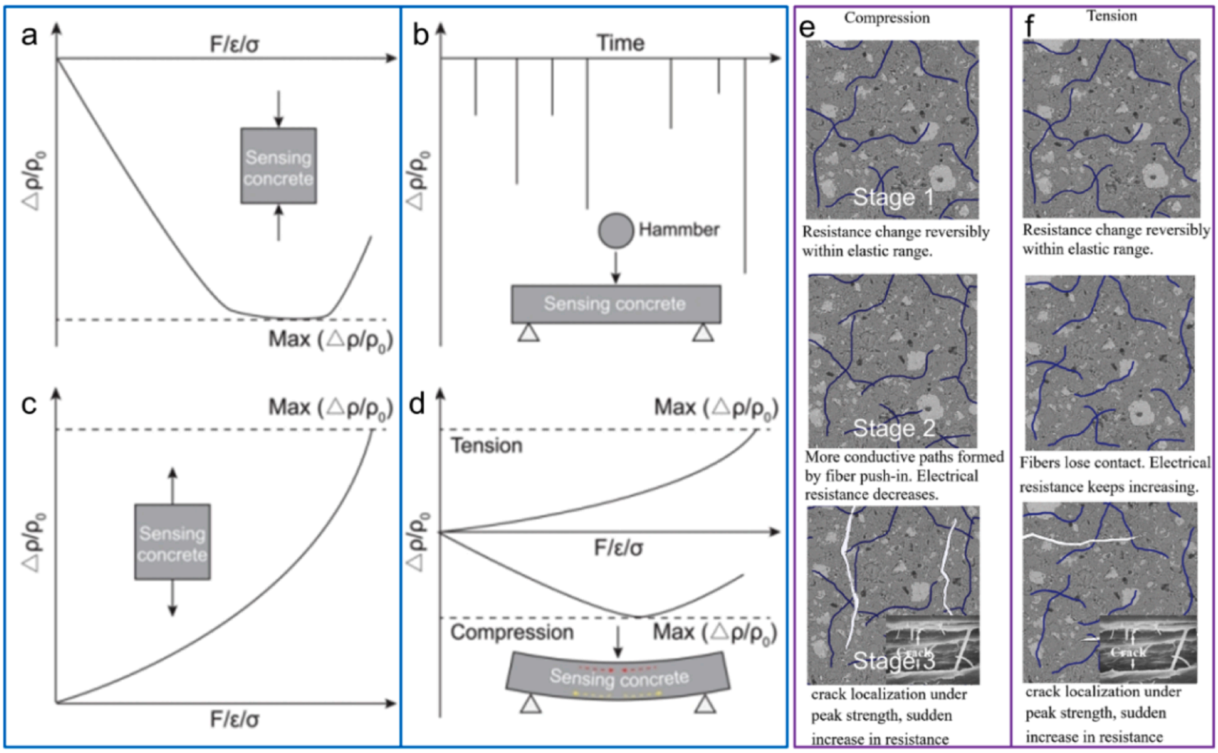

Fig. 10. Typical sensing behavior of ECCC samples under monotonic loading including [13]: (a) compression, (b) impact, (c) tension, (d) bending, (e) compression, and (f) tension crack evolution schematics.

loading conditions, including compression, impact, tension, and bending states.

During uniaxial compression, the FCR value exhibits a decreasing trend followed by a sudden increase as the pressure increases (see Fig. 10**a**). These stages correspond to compaction, the generation of new cracks, and crack propagation, respectively. Applying an appropriate force can reduce the distance between conductive fillers, thereby enhancing the self-sensing performance of ECCCs. As the pressure continues to increase, new cracks form within the ECCCs, leading to the disruption and reconstruction of conductive pathways. With further pressure increase, the conductive network eventually fractures, indicating macroscopic damage to the concrete (see Fig. 10**e**).

Under impact loading, the FCR suddenly decreases and then returns to zero (see Fig. 10**b**). The magnitude of the FCR depends on the amplitude of the impact load. High-amplitude impacts or repeated impacts result in internal damage to the concrete, preventing the FCR from returning to zero.

Under monotonic tensile strain, the FCR shows a linear increase as the tensile stress increases (see Fig. 10c). This is mainly attributed to the increasing distance between conductive fillers under tensile forces, corresponding to the gradual formation and expansion of microcracks (see Fig. 10**f**). After reaching the ultimate tensile strain, the FCR undergoes rapid changes due to the formation and accumulation of concrete damage. This implies that the sensing behavior of ECCCs is not only a function of tensile strain but is also influenced by material cracking behavior. Therefore, the crack propagation mechanism and estimation of crack information (such as length and width) can be inferred based on the electrical signals of ECCCs.

Under bending, the self-sensing behavior of ECCCs exhibits opposite trends when both tension and compression sides are present. However, when only the tension or compression side is present, the sensing behavior of ECCCs and the resistance results on both sides are consistent (see Fig. 10**d**). Under the same deflection, the sensitivity of the tension side of ECCCs is typically much greater than that of the compression side. This is mainly because ECCCs have higher compressive strength compared to their tensile strength.

Some studies have also reported the sensing behavior of ECCCs under dynamic loading to evaluate their repeatability and sensitivity [5,113]. Regardless of the type of load applied, under cyclic loading conditions (with amplitudes less than about 30 % of the ultimate strength of ECCCs), the FCR undergoes reversible changes, indicating that the ECCCs are in an elastic deformation state. When the loading amplitude exceeds the range of elastic deformation, irreversible changes in FCR occur in each cycle. Additionally, after cyclic loading, the initial resistivity of ECCCs also undergoes certain changes, mainly due to the formation of microcracks within the ECCCs and subsequent reconstruction of the conductive network. As the number of loading cycles increases, the irreversibility of FCR increases due to the occurrence of permanent deformation and damage [11,24,113]. The sensing performance of ECCCs is also dependent on the dynamic loading rate, which requires further experimental verification.

Overall, the formation and distribution of the conductive network in ECCCs, which determine their electrical and self-sensing performance, are influenced by the conductive fillers. Enhancing the electrical and sensing performance of ECCCs can be achieved by adjusting the characteristics of the conductive fillers (such as type, concentration, geometric shape, and surface treatment), optimizing their distribution (by improving dispersion and dispersion stability), and improving the interface bonding between the conductive fillers and the cementitious matrix. Furthermore, the conventional approach of developing cement-based conductive ECCCs often compromises their operational performance, mechanical properties, and durability. Therefore, there is an urgent need to develop high-performance ECCCs that meet the requirements of working performance, mechanical properties, and durability, to overcome the current technological bottlenecks and challenges in research and promote the practical engineering applications of ECCCs in SHM and road information monitoring fields.

7. Case studies and applications of nanocarbon materials-engineered SSCC

Based on the collected electrical signals, SSCC can reflect information such as stress, strain, cracks, and damage within cementitious composites. Therefore, SSCCs can be applied in transportation information detection [18,107,114,115]. In terms of engineering applications in transportation information detection: Integrating SSCCs into road surfaces or bridge pavement layers enables the monitoring and recording of traffic information, such as traffic volume, vehicle speed, and dynamic weighing, based on the stress-electrical signal relationship. This enhances the efficiency of road usage and ensures the safety and intelligent operation of roads.

Table 4 and Fig. 11 summarize the practical engineering cases of using SSCCs for transportation information monitoring. For instance, Han et al. [123] developed a self-sensing concrete pavement system based on CNTs for road traffic detection (see Fig. 11**a**). Prefabricated and cast-in-place CNT-cement-based resistive sensors were integrated into a controlled road test section. The results showed that the pavement system accurately detected the passage of different vehicles under different speeds and test environments. This demonstrates that the developed self-sensing pavement system achieves real-time traffic flow detection with high detection rates

Table 4
Nanocarbon materials-engineered SSCC for traffic detection.

Application form	Testing method	Monitoring parameters	Reference
SSCC roller compactor	Rotating car tires with laboratory roller wheels	Dynamic weighing	[151]
Integrated SSCC strip components in the pavement.	Testing the response of SSCCs using a testing machine	Vehicle speed monitoring	[152]
		Dynamic weighing	[78]
		Traffic volume/loading	[153]
	Conducting tests using a five-axis semi-trailer and a truck	Traffic volume	[29]
Integrated SSCC array in the pavement	Conducting road tests outdoors using a car	Traffic volume	[148]

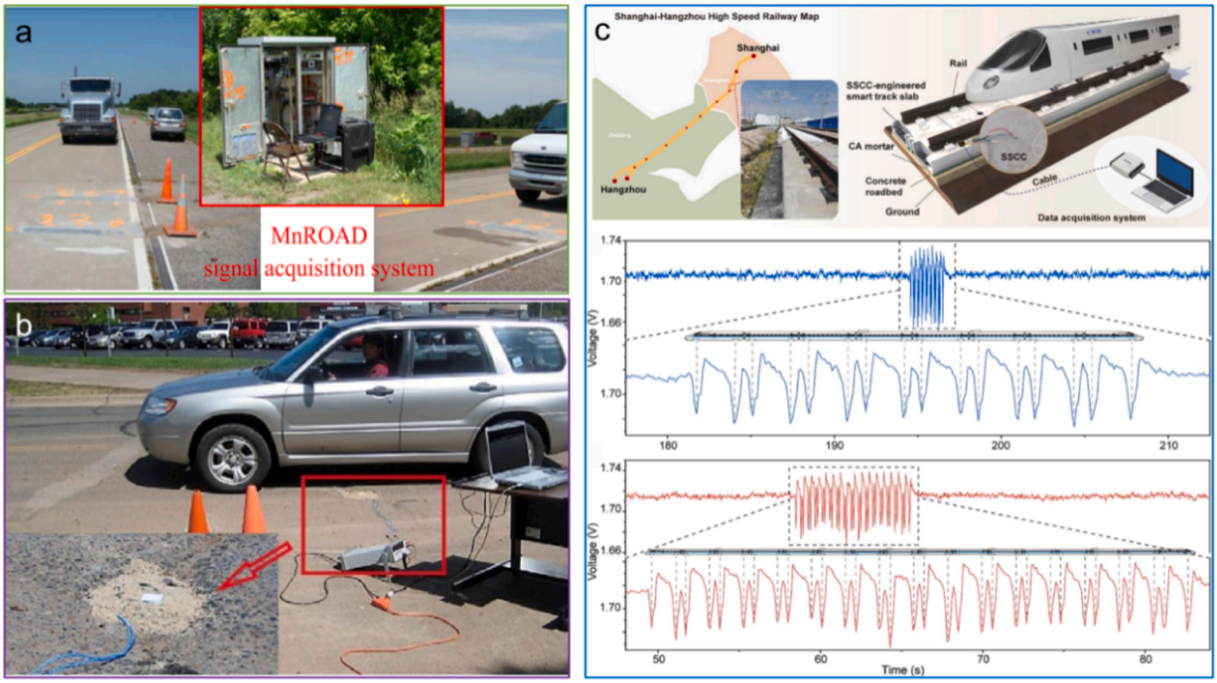

Fig. 11. Practical applications of SSCC in the field of information detection for concrete infrastructures, such as concrete pavements and bridges: (a) utilization of CNT-based SSCCs for road information testing [148]; (b) Vehicle load testing of CNT-modified SSCCs embedded in concrete pavement [149]; and (c) SSCC track panels for high-speed railway monitoring [150].

and low error rates. Additionally, Han et al. [124] developed another CNT-cement-based resistive sensor for traffic monitoring. This sensor exhibited sensitive and stable responses to repetitive loadings, indicating its significant potential in traffic monitoring applications such as traffic volume detection, dynamic vehicle weighing, and vehicle speed detection (see Fig. 11**b**). Recently, Ding et al. [125] developed a novel cement-based resistive sensor (see Fig. 11**c**) by growing CNTs in situ on the cement surface and fabricating them into resistive concrete. The sensor demonstrated a high maximum stress sensitivity of 2.87 %/MPa and a sensitivity coefficient of 748, exhibiting good repeatability and stability. The developed self-sensing concrete intelligent track slab successfully performed axle and speed detection, opening up a new field for establishing high-performance and multifunctional self-sensing concrete intelligent components/structures. This can be applied for long-term, widespread, and cost-effective monitoring of high-speed railway infrastructure.

In summary, prefabricated or cast-in-place engineered SSCC are predominantly incorporated into road surfaces as strip elements. The variations in sensing signals induced by polarization and environmental factors can be effectively filtered during the post-processing phase of the measured signals, thereby ensuring that the detection accuracy of the system remains unaffected by these extraneous influences. The integrated SSCC system boasts several advantages, including exceptional detection accuracy, robust anti-interference capabilities, ease of installation and maintenance, as well as a prolonged service life.

Currently, CNT is predominantly utilized as the conductive phase in practical engineering applications of engineered SSCC, owing to their exceptional conductivity properties and ease of incorporation, either by direct mixing into cement or coating onto aggregate surfaces. However, there is a scarcity of engineering instances involving SSCC prepared from CNT-coated aggregates. Further research is essential to explore and elucidate the optimal design and conductivity mechanisms of SSCC modified with nanocarbon materials, thereby facilitating their broader adoption in practical engineering applications.

8. Research challenges and limitations of nanocarbon materials-engineered SSCC applications

Nanocarbon materials typically possess a large specific surface area and van der Waals forces, which make their dispersion in solutions and cementitious matrices highly challenging [56,61]. High-quality nanocarbon material dispersions can be prepared using techniques such as mechanical stirring, ultrasonic dispersion, and surfactants. However, it is important to note that good dispersion of nanocarbon materials in aqueous solutions does not necessarily guarantee the same level of high-quality and stable dispersion in cementitious matrices [63,115]. Furthermore, the introduction of nanocarbon materials can significantly compromise the workability of the mixture, which is unfavorable for transportation and casting processes. Moreover, agglomerated nanocarbon materials can adversely affect the mechanical, durability, and electrical properties of cementitious composites. Therefore, it is necessary to explore more convenient and cost-effective processes to achieve good dispersion of nanocarbon materials in cementitious composites, thereby fully harnessing their enhancement efficiency in terms of mechanical, electrical, and sensing performance.

The development strategy for cement-based conductive ECCCs involves directly adding conductive fillers to the cement, followed by mixing with aggregates. Therefore, the electrical and sensing performance of ECCCs is primarily influenced by factors such as the type and dosage of fillers, dispersion quality, distribution, and preparation processes [9,37]. To achieve high conductivity and self-sensing performance in ECCCs, a significant amount of nanocarbon materials (beyond the percolation threshold) is typically required. However, this poses significant challenges in terms of nanocarbon material dispersion, leading to a substantial decrease in the working and mechanical properties of the composite material. The technical bottleneck of achieving a balance between strength and electrical performance, along with the high cost, has limited the development and practical engineering application of ECCCs. Therefore, it is crucial to change the method of incorporating nanocarbon materials and develop a low-cost ECCC that overcomes the conflicting nature of its conductivity and mechanical properties, to expand the practical engineering application of ECCCs.

In traditional processes for developing cement-based conductive ECCCs, the ITZ is located at the edge of the conductive pathway, which limits the sensitivity to the initial microcracks in the ITZ and results in very limited electrical and sensing capabilities. Additionally, the composite material is subjected to environmental factors (such as temperature/humidity and corrosive media) during service, which can interfere with the electrical signals and affect the electrical and sensing performance of ECCCs [37,100]. In the cement hydration environment, the presence of numerous ions as conductive media, along with the electron transport in the conductive phase, collectively form the conductive framework of the ECCC. Fluctuations in ion concentration in the pore solution led to variations in the electrical and sensing performance of ECCCs. Furthermore, the significant difference in particle size between nanocarbon materials and other components of concrete has limited the development and research of ECCCs primarily to the realm of paste and mortar, with very few studies focused on the application of conductive concrete in engineering structures. Therefore, it is crucial to change the method of incorporating nanocarbon fillers, reduce their dosage, and optimize the design and process flow to significantly enhance the self-sensing performance of ECCCs, while achieving low-cost, efficient, and widespread development for practical engineering applications.

9. Summaries and recommendations

This comprehensive review has focused on the current state of research on nanocarbon materials-engineered ECCC, specifically within the context of self-sensing applications, namely, self-sensing cement composites (SSCC). Initially, a thorough review and comparison of the primary methods for preparing nanocarbon materials-engineered SSCC have been presented. Additionally, this work has provided an illustration and discussion of the electrical signal measurement and conductive theory associated with nanocarbon materials-engineered SSCC. Furthermore, the impact of incorporating nanocarbon materials into cement composites has been extensively discussed, covering various aspects such as hydration and microstructures, workability, mechanical and electrical behavior, as well as self-sensing properties. Moreover, case studies highlighting practical applications of nanocarbon materials-engineered SSCC have been presented. The following findings can be made:

(1) The dispersion of nanocarbon materials within ECCC is crucial. While preparing a high-quality nanocarbon materials dispersion does not guarantee that these nanomaterials will disperse evenly and stably within the ECCC system, the cumbersome and costly pre-processing procedures are not conducive to the practical applications of traditional processes. In contrast, coating the nanocarbon materials onto the surface of aggregates can avoid the challenges associated with nanocarbon material agglomeration. Additionally, the smaller specific surface area of aggregates allows for a reduction in the amount of nanocarbon materials required.

(2) While the two-probe method is simpler to operate, the four-probe method offers the advantage of eliminating contact resistance between the electrode and the ECCC specimen. This advantage has been confirmed through electrical signal testing results of ECCC containing various conductive fillers. In addition to considerations regarding the types and distribution of conductive fillers, electrode selection, and arrangement, and the acquisition of sensing signals, other factors that influence the testing of ECCC sensing signals include the curing age, temperature, and humidity of the specimen. Although polarization effects can still be observed when using alternating current, their negative impact can be minimized by increasing the frequency or reducing the amplitude of the AC voltage.

(3) The introduced nanocarbon materials can establish direct contact with each other, thereby creating efficient pathways for electrical conduction within the ECCC system. When the distance between nanocarbon materials is sufficiently close (typically less than 10 nm), even without direct contact, the wave-like behavior of charged particles gives rise to quantum effects. This phenomenon, known as tunneling conduction, allows for the formation of conductive pathways through potential barriers. Field emission, on the other hand, is a special manifestation of tunneling conduction that occurs due to localized strong electric fields. Traditional nanocarbon materials, for example, are unable to generate strong electric fields for field emission at low voltages. However, specific nanocarbon materials like CNT can induce localized strong electric fields at their tips.

(4) The preparation of SSCC with high sensitivity and reliable repeatability holds significant importance for their practical applications in monitoring pavement information. The electrical and self-sensing performance of SSCC is heavily influenced by the formation and distribution of the conductive network, which is in turn affected by the conductive fillers. To enhance the electrical and sensing performance of SSCC, it is necessary to adjust the characteristics of the conductive fillers (such as type, concentration, geometric shape, and surface treatment), optimize their distribution (by improving dispersion and dispersion stability), and enhance the interface bonding between the conductive fillers and the cementitious matrix.

(5) The conventional approach to developing cement-based conductive SSCC often compromises their operational performance, mechanical properties, and durability. Therefore, there is an urgent need to develop high-performance SSCC that meet the

requirements of workability, mechanical properties, and durability. This will help overcome the current technological bottlenecks and challenges in research and promote the practical applications of SSCC in the fields of SHM and traffic information monitoring. In the future, it is crucial to focus on researching the relationship between ITZ crack propagation and the self-sensing performance of SSCC and establish a corresponding correlation.

CRediT authorship contribution statement

Xinran Chen: Writing – review & editing. **Shumei Zhou:** Writing – review & editing. **Yun Wang:** Writing – review & editing. **Jing Zhong:** Writing – review & editing. **Dong Lu:** Writing – review & editing, Methodology. **Jian Yuan:** Writing – original draft, Data curation, Conceptualization. **Suhui Yu:** Writing – original draft, Funding acquisition, Formal analysis.

Declaration of Competing 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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Data Availability

Data will be made available on request.

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