

Review

Graphene-Based Coating Strategies to Realize High Performance Cementitious Composites: A Perspective from Carbon-Neutrality

Shupeí Dong ¹, Mingrui Du ^{1,2,*}, Yuan Gao ³ and Xupei Yao ¹

¹ School of Water Conservancy and Transportation, Zhengzhou University, Zhengzhou 450001, China; xupeiyao@zzu.edu.cn (X.Y.)

² State Key Laboratory of Intelligent Construction and Healthy Operation and Maintenance of Deep Underground Engineering, China University of Mining and Technology, Xuzhou 221116, China

³ School of Transportation and Civil Engineering, Nantong University, Nantong 226019, China; y.gao@ntu.edu.cn

* Correspondence: dumingrui@zzu.edu.cn

Abstract

Graphene-based nanosheets (GNS), including graphene, graphene oxide (GO), reduced graphene oxide (rGO), and graphene nanoplatelets (GNPs), have attracted increasing attention for developing high-performance and sustainable cementitious composites. Compared with conventional dispersion strategies, graphene-based coating strategies enable the targeted localization of GNS at critical interfacial transition zones (ITZs), thereby maximizing their reinforcing efficiency while mitigating agglomeration issues. This review systematically summarizes recent advances in GNS coating technologies for cementitious composites, including physical adsorption, chemical assembly, electrophoretic deposition, and in situ growth. The effects of GNS coatings on interfacial engineering, mechanical performance, durability enhancement, and smart functionalities are critically discussed. Existing studies indicate that GNS coatings can improve strength, crack resistance, impermeability, and resistance to chloride ingress, freeze–thaw cycles, and other degradation processes mainly through ITZ densification and microstructure refinement. However, these benefits are strongly dependent on the coating method, substrate type, and stability of the graphene–substrate interface in calcium-rich alkaline pore solutions. In particular, physically adsorbed GO coatings may suffer from desorption or Ca²⁺-induced aggregation, chemically assembled coatings require further validation beyond laboratory-scale systems, and electrophoretic deposition is mainly applicable to electrically conductive substrates. In addition, localized conductive networks created by GNS coatings facilitate multifunctional properties such as self-sensing, electromagnetic shielding, and electrothermal performance. From a carbon-neutrality perspective, the improvements in mechanical properties and durability provide opportunities to reduce material consumption, extend service life, and lower life-cycle carbon emissions. Nevertheless, their carbon-neutral contribution should be verified through quantitative life-cycle assessment rather than inferred directly from strength or durability enhancement alone. Finally, the remaining challenges associated with large-scale implementation, long-term stability, cost-effectiveness, and field-scale validation are discussed. Particular attention is given to the fact that most existing evidence is derived from laboratory-scale specimens rather than real structural elements exposed to service environments.

Academic Editor: Qingguo Shao

Received: 3 June 2026

Revised: 3 July 2026

Accepted: 6 July 2026

Published: 9 July 2026

Copyright: © 2026 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the [Creative Commons Attribution \(CC BY\) license](https://creativecommons.org/licenses/by/4.0/).

Keywords: graphene-based nanosheets; coating strategy; cementitious composites; mechanical properties; carbon neutrality

1. Introduction

Global cement production has reached approximately 4.1 billion tonnes annually, making cement one of the most widely consumed construction materials [1]. However, the cement industry is also one of the largest industrial sources of carbon dioxide (CO₂) emissions, contributing approximately 7–8% of global CO₂ emissions [2–6]. According to the International Energy Agency (IEA), global energy-related CO₂ emissions reached 37.4 Gt in 2023, highlighting the urgent need for deep decarbonization across energy-intensive industries [3]. Achieving carbon neutrality by 2050, as advocated by the Paris Agreement, presents a particular challenge for the cement sector because cement clinker production is inherently carbon intensive [2,4–6]. During clinker manufacture, limestone and clay are heated to temperatures approaching 1450 °C, consuming approximately 3.4–3.6 GJ of thermal energy per tonne of clinker and generating substantial CO₂ emissions from both fuel combustion and limestone calcination [2,5,6]. Process emissions from calcination alone account for approximately 60% of total clinker-related CO₂ emissions, while fuel combustion contributes most of the remainder [2,5]. Consequently, reducing the carbon footprint of cement production has become one of the most critical challenges facing the construction industry.

Various strategies have been proposed to reduce CO₂ emissions during cement production [7], including improving thermal efficiency and minimizing heat loss, developing clinker-free or low-clinker binders, increasing the utilization of renewable energy and alternative fuels, employing carbon capture utilization and storage (CCUS) technologies, and partially replacing clinker with supplementary cementitious materials (SCMs) [2,4,6]. Despite considerable progress, the large-scale implementation of many of these approaches remains constrained by technological barriers, high capital costs, infrastructure requirements, and uncertainties associated with long-term engineering performance [4,6]. Therefore, complementary and immediately applicable approaches capable of reducing material consumption and extending service life are urgently required to facilitate the transition toward carbon-neutral construction.

Recently, graphene-based nanosheets (GNS), including graphene, graphene oxide (GO), reduced graphene oxide (rGO), graphene nanoplatelets (GNPs), and functionalized graphene derivatives, have attracted extensive attention in the development of high-performance cementitious composites [8–11]. Owing to their exceptional intrinsic properties, including a tensile strength of approximately 130 GPa, a Young's modulus approaching 1 TPa, and a theoretical specific surface area of approximately 2630 m² g^{−1}, GNS possess extraordinary potential for microstructural regulation and multi-scale reinforcement [8–11]. Numerous studies have demonstrated that the incorporation of trace amounts of GNS can significantly improve the compressive strength, tensile/flexural strength, fracture resistance, and toughness of cementitious composites [12–19]. Such performance enhancement may potentially support structural optimization if design conditions allow while maintaining equivalent engineering performance [20]. However, these carbon-reduction benefits should not be regarded as automatic consequences of strength improvement, because practical material savings depend on structural design codes, safety factors, serviceability requirements, constructability, and whether engineers actually optimize the mixture or member geometry. Therefore, the carbon-neutrality potential of GNS coatings should not be interpreted as a direct or proportional consequence of mechanical performance improvement. Any potential reduction in cement consumption or CO₂ emissions

is highly scenario-dependent and only becomes meaningful under specific structural re-design conditions and full life-cycle assessment frameworks, rather than being inferred from material-level strength enhancement alone [21,22]. In addition, GNS can markedly enhance durability-related properties, including impermeability, resistance to chloride ingress, freeze–thaw resistance, and carbonation resistance of cementitious composites [13–19]. Furthermore, the excellent electrical and thermal conductivity of GNS enables the development of multifunctional cementitious materials with self-sensing, self-monitoring, and thermal-management capabilities [15–19]. These advantages contribute to carbon neutrality not only through performance enhancement and material savings but also through prolonged service life and improved infrastructure management, which collectively reduce life-cycle carbon emissions.

Moreover, recent advances in nanoscience and nanotechnology have further enhanced the sustainability of GNS production. Graphene-based materials have been successfully synthesized from various waste-derived carbon resources, including waste plastics, waste tires, and other carbon-rich wastes through emerging upcycling technologies [23–25]. These developments may reduce the environmental footprint associated with graphene production and create new opportunities for integrating circular-economy concepts into the construction sector. Consequently, GNS exhibit considerable potential for supporting carbon-neutral development in both the cement industry and broader construction sector.

Several strategies have been developed to incorporate GNS into cementitious composites. Through ultrasonication, surfactant-assisted dispersion, surface functionalization, and dry-mixing techniques, GNS can be distributed throughout the cementitious matrix with reduced agglomeration [13–19]. Uniformly dispersed GNS provide abundant nucleation sites for hydration products, accelerate cement hydration, refine pore structures, bridge nanoscale cracks, and facilitate load transfer across the matrix, thereby enhancing the overall mechanical performance of cementitious materials [13–16]. However, key engineering properties, including compressive strength, tensile/flexural strength, and durability, are strongly influenced by the interfacial properties between different phases [26–28]. In cementitious composites, the ITZ between cement paste and aggregates or reinforcing fibers is generally regarded as the weakest region because of its relatively high porosity, preferential orientation of calcium hydroxide crystals, and lower concentration of calcium silicate hydrate (C–S–H) gel [26–28]. As a result, microcracks preferentially initiate and propagate within the ITZ, eventually leading to mechanical deterioration and durability degradation [26–28]. Although dispersion-based strategies can improve the overall matrix microstructure, their ability to specifically strengthen these weak interfacial regions remains limited.

To overcome this limitation, graphene-based coating strategies have emerged as a promising approach for targeted interfacial engineering. By coating GNS onto selected components, such as fibers, sand particles, aggregates, cementitious particles, and supplementary cementitious materials, graphene can be strategically localized at critical interfacial regions where damage is most likely to initiate [19,26–28]. Benefiting from their high mechanical strength, large specific surface area, two-dimensional morphology, and abundant surface functional groups [8–11], GNS coatings can accelerate hydration, densify the ITZ, fill interfacial pores, bridge microcracks, and enhance interfacial bonding through both mechanical interlocking and physicochemical interactions [19,26–28]. Consequently, the traditionally weak interfacial regions can be transformed into mechanically robust and damage-tolerant zones, leading to substantial improvements in strength, durability, and multifunctional performance.

Although several excellent reviews have comprehensively summarized the fresh properties, hardened properties, reinforcement mechanisms, durability enhancement,

and multifunctional characteristics of GNS-reinforced cementitious composites [17–19], these studies have predominantly focused on GNS dispersion strategies. A systematic review dedicated to graphene-based coating strategies remains lacking. Therefore, this review aims to summarize recent developments in graphene-based coating technologies for cementitious composites, with particular emphasis on coating methodologies, interfacial strengthening mechanisms, mechanical enhancement, durability improvement, and smart functionalities. Furthermore, the remaining scientific challenges and future opportunities associated with GNS coatings are critically discussed from a carbon-neutrality perspective. Because most available studies are still based on laboratory-scale specimens, this review emphasizes the need for field-scale validation, realistic exposure conditions, and assessment in structural concrete elements before the practical contribution of GNS coatings to sustainable infrastructure can be confirmed.

2. GNS Properties

2.1. Morphology and Chemical Structure

GNS comprise a family of two-dimensional carbon nanomaterials constructed from sp^2 -hybridized carbon atoms and typically possessing thicknesses ranging from a single atomic layer to less than 100 nm [11,29,30]. As the fundamental structural units of other carbon allotropes, including zero-dimensional fullerenes, one-dimensional carbon nanotubes, and three-dimensional graphite, GNS mainly encompass pristine graphene, graphene oxide (GO), reduced graphene oxide (rGO), and graphene nanoplatelets (GNPs). The representative morphology and chemical structures of these four types are illustrated in Figure 1. These materials differ significantly in morphology, defect density, surface chemistry, and degree of oxidation, resulting in distinct physicochemical properties and interactions with cementitious matrices [11,29,30].

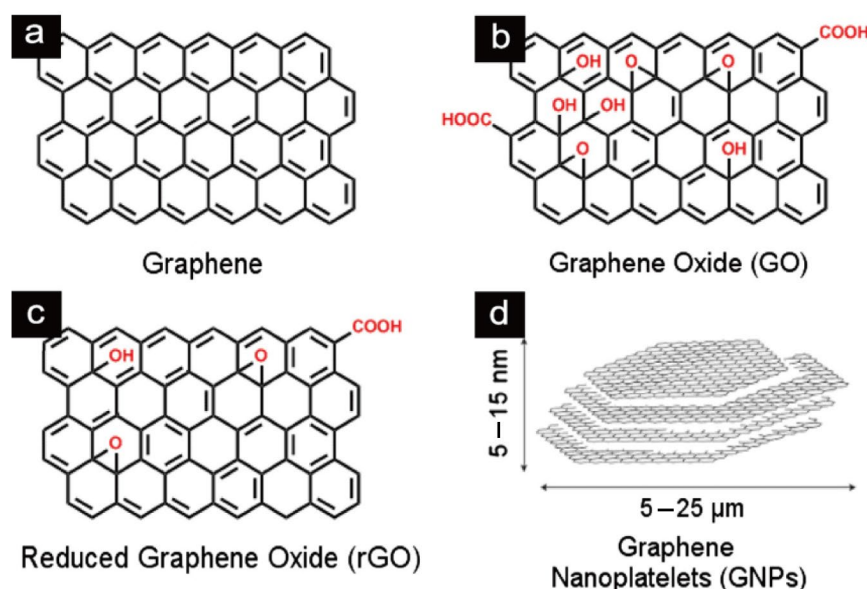


Figure 1. Morphology and chemical structure of GNS: (a) graphene; (b) graphene oxide (GO); (c) reduced graphene oxide (rGO); and (d) graphene nanoplatelets (GNPs). Adapted from Ref. [18].

Pristine graphene consists of a single layer of carbon atoms arranged in a two-dimensional hexagonal honeycomb lattice with a C–C bond length of approximately 0.142 nm and an apparent thickness of approximately 0.34 nm [8]. Owing to its fully conjugated sp^2 carbon network and nearly defect-free structure, graphene exhibits exceptional intrinsic

mechanical strength, electrical conductivity, thermal conductivity, and gas-barrier performance [8,31]. However, the absence of surface functional groups and strong van der Waals attractions between adjacent sheets render pristine graphene highly hydrophobic and prone to agglomeration, significantly limiting its dispersion in aqueous solutions and cementitious systems [31].

Graphene oxide (GO) is typically synthesized through the oxidation of graphite using modified Hummers-type methods, during which abundant oxygen-containing functional groups, including hydroxyl ($-\text{OH}$), epoxy ($-\text{C}-\text{O}-\text{C}-$), carbonyl ($\text{C}=\text{O}$), and carboxyl ($-\text{COOH}$) groups, are introduced onto both the basal planes and sheet edges [32–35]. The oxidation process partially disrupts the sp^2 -conjugated carbon network, generating sp^3 -hybridized defects, structural distortions, and characteristic nanoscale wrinkles [34–36]. Simultaneously, oxygen functionalization and water intercalation increase the interlayer spacing from approximately 0.34 nm in graphite to 0.7–1.2 nm depending on the hydration state [37,38]. Consequently, GO exhibits excellent hydrophilicity and colloidal stability in water, which facilitates its homogeneous dispersion in cementitious matrices and promotes strong interactions with hydration products.

Reduced graphene oxide (rGO) is obtained through the partial reduction in GO using chemical reducing agents, such as hydrazine hydrate and ascorbic acid, or through thermal reduction treatments [10,39,40]. During reduction, a substantial proportion of oxygen-containing functional groups is removed, partially restoring the sp^2 -conjugated structure and significantly improving electrical conductivity and mechanical properties [11,40]. Nevertheless, irreversible lattice defects generated during oxidation cannot be completely eliminated, and residual oxygen functionalities remain on the graphene surface [36]. Therefore, rGO possesses a hybrid structure intermediate between pristine graphene and GO. Importantly, these remaining functional groups contribute to aqueous dispersibility and provide active sites for interfacial bonding with cement hydration products [11].

Graphene nanoplatelets (GNPs) are multi-layer graphene structures generally consisting of 10–100 stacked graphene layers, with thicknesses below 100 nm and lateral dimensions ranging from several micrometers to hundreds of micrometers [41]. GNPs are commonly produced through graphite intercalation, thermal expansion, and subsequent mechanical exfoliation processes. Unlike GO and rGO, GNPs undergo minimal oxidation and therefore retain a highly ordered graphitic structure with relatively few surface functional groups [41,42]. As a result, GNPs exhibit outstanding electrical conductivity, thermal conductivity, and mechanical stiffness. However, their hydrophobic nature and strong inter-sheet attractions often require surfactant-assisted dispersion, ultrasonication, or surface modification before incorporation into cementitious materials [42]. Owing to their relatively low production cost, scalability, and commercial availability, GNPs have become one of the most widely investigated graphene-based nanomaterials for large-scale cementitious applications.

2.2. Physical and Mechanical Properties

The exceptional physical and mechanical properties of GNS originate from their unique two-dimensional geometry, ultrahigh aspect ratio, and robust in-plane sp^2 carbon–carbon covalent bonds [29,30]. Variations in oxidation degree, defect density, layer number, and surface chemistry result in significant differences among graphene, graphene oxide (GO), reduced graphene oxide (rGO), and graphene nanoplatelets (GNPs), leading to distinct physical and mechanical characteristics [11,41].

Physical properties. One of the most remarkable features of GNS is their extraordinarily high specific surface area (SSA). Theoretical calculations indicate that monolayer graphene possesses an SSA of approximately $2630 \text{ m}^2 \text{ g}^{-1}$, representing one of the highest

values among all known solid materials [43,44]. However, practical SSA values are generally lower because of irreversible sheet restacking and agglomeration during processing and storage. For GO, the introduction of oxygen-containing functional groups and the expansion of interlayer spacing can generate SSA values ranging from approximately 100 to 1000 m² g^{−1} depending on the synthesis route and exfoliation degree [38,45]. Reduced graphene oxide (rGO) may partially recover accessible surface area through the formation of porous structures during reduction, whereas the SSA of graphene nanoplatelets (GNPs) generally decreases with increasing layer number owing to multilayer stacking effects [40,42]. The large surface area of GNS provides abundant sites for physical adsorption and interfacial interactions, making these materials highly attractive for surface modification and composite applications.

Another important characteristic of GNS is their relatively low density, typically ranging from 1.8 to 2.2 g cm^{−3} depending on oxidation degree and structural compactness [11,40]. Furthermore, GNS possess extremely high aspect ratios owing to their nanometer-scale thickness and micrometer-scale lateral dimensions [30,44]. Such geometric characteristics provide extensive interfacial contact areas and facilitate the formation of interconnected networks within composite systems. The combination of high surface area, low density, and ultrahigh aspect ratio distinguishes GNS from conventional nanomaterials and underpins many of their unique functionalities.

Mechanical properties. Graphene is widely recognized as one of the strongest and stiffest materials ever measured. Atomic force microscopy (AFM) nanoindentation experiments conducted by Lee et al. demonstrated a Young's modulus of approximately 1.0 ± 0.1 TPa and an intrinsic tensile strength of approximately 130 ± 10 GPa for defect-free monolayer graphene [8]. In addition, graphene can sustain elastic strains approaching 20–25% before fracture [8].

The mechanical properties of GO differ substantially from those of pristine graphene because oxidation disrupts the continuous sp²-conjugated carbon network and introduces structural defects [11,44]. Nevertheless, GO still exhibits relatively high stiffness, with reported Young's modulus values commonly ranging from approximately 150 to 250 GPa depending on oxidation level, sheet size, and testing methodology [44]. Upon reduction, partial restoration of the sp² carbon framework leads to significant recovery of mechanical performance. Reported Young's modulus values for rGO generally range from approximately 200 to 700 GPa, although considerable variability exists due to differences in reduction degree and defect density [40,44].

For GNPs, the effective Young's modulus is typically reported within the range of 0.5–1.0 TPa [41,44]. Owing to their multilayer structure, the measured stiffness reflects not only the intrinsic properties of individual graphene layers but also the influence of interlayer van der Waals interactions, stacking order, and structural imperfections. Although the mechanical properties of GNPs are generally lower than the theoretical values of defect-free monolayer graphene, they remain substantially higher than those of conventional engineering materials [44]. The key physical and mechanical properties of representative GNS are summarized in Table 1.

Table 1. Key physical and mechanical properties of graphene-based nanosheets.

Property Category	Property Index	Graphene	GO	rGO	GNPs	Ref.
Physical	Specific surface area (m ² /g)	~2630	100–1000	300–1500	50–500	[38,40,42–45]
	True density (g/cm ³)	~2.25	1.8–2.1	1.9–2.2	2.0–2.25	[11,40,41]
	Aspect ratio	>10,000	1000–30,000	1000–30,000	100–5000	[44]
Mechanical	Young's modulus (GPa)	~1000	150–250	200–700	500–1000	[8,44]
	Tensile strength (GPa)	~130	strongly dependent on oxidation degree	limited data available	dependent on layer number and defects	[8,44]

2.3. Fabrication Processes

GNS can generally be produced through two major routes: top-down exfoliation and bottom-up synthesis [11,29,30]. Top-down methods generate graphene sheets by overcoming the weak van der Waals interactions between graphite layers, whereas bottom-up methods construct graphene directly from molecular carbon precursors on suitable substrates [11]. In recent years, waste-derived graphene fabrication has attracted increasing attention owing to its potential for waste valorization and sustainable production, which aligns well with the carbon-neutral development goals of the construction industry [40]. Representative fabrication routes of GNS are shown in Figure 2.

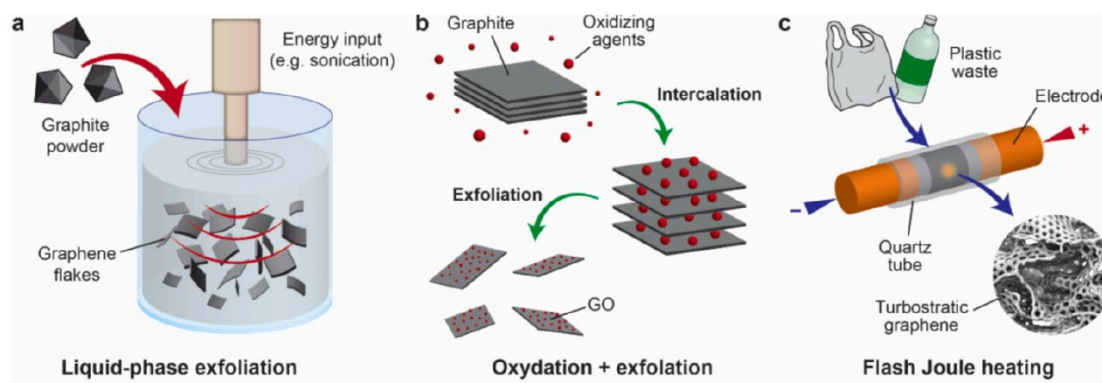


Figure 2. Fabrication routes and types of GNS: (a) liquid-phase exfoliation; (b) graphite oxidation/exfoliation; and (c) waste-derived flash joule heating method. Adapted from Ref. [19].

2.3.1. Conventional Fabrication Processes

Top-down exfoliation remains the most widely adopted route for producing graphene and its derivatives. Micromechanical exfoliation, first employed by Novoselov et al. [30], isolates graphene from highly oriented pyrolytic graphite through repeated peeling. Although this method yields graphene with exceptionally low defect density and excellent intrinsic properties, its low productivity limits large-scale applications [30,31].

Liquid-phase exfoliation (LPE) is another important top-down approach. In this process, graphite is dispersed in suitable solvents or surfactant solutions and subsequently exfoliated through ultrasonication or high-shear mixing [46,47]. Compared with micromechanical exfoliation, LPE is more scalable and avoids chemical oxidation, although the exfoliation efficiency and downstream purification remain challenging [47].

The oxidation–reduction route is currently the most common method for producing GO and rGO [11]. Graphite is first oxidized using the Hummers method or its modified variants, introducing oxygen-containing functional groups and expanding the interlayer spacing [32,33]. The resulting GO exhibits excellent aqueous dispersibility, while subsequent chemical or thermal reduction partially restores the sp^2 carbon network and produces rGO with improved conductivity and mechanical performance [38]. Despite its scalability and low cost, irreversible structural defects generated during oxidation remain a major limitation of this route [38].

Bottom-up methods construct graphene directly from carbon-containing precursors. Chemical vapor deposition (CVD) is the most widely studied technique, in which hydrocarbon gases such as methane are decomposed on catalytic metal substrates (e.g., Cu or Ni) at elevated temperatures to form large-area graphene films with high crystallinity [48,49]. Another representative method is epitaxial growth on silicon carbide (SiC), where thermal sublimation of silicon atoms leaves carbon atoms that reorganize into graphene layers [50]. Although these methods produce high-quality graphene, their high cost and complex processing restrict large-scale structural-material applications.

2.3.2. Fabrication from Waste

The conversion of waste-derived carbon resources into graphene-based materials has emerged as a promising sustainable production route. A notable breakthrough is the flash Joule heating (FJH) technique developed by Luong et al. [23], in which carbon-containing solids are subjected to a high-voltage electrical pulse, generating ultrafast heating above 2500–3000 K. Under these conditions, heteroatoms are rapidly removed and carbon atoms reorganize into turbostratic graphene structures. Various waste feedstocks, including waste plastics, waste tires, petroleum coke, coal fly ash, and biochar, have been successfully converted into graphene-like materials using this method [23,51].

Biomass wastes provide another important source of graphene-related materials. Agricultural residues such as rice husks, sugarcane bagasse, and lignocellulosic biomass can be transformed into graphitic nanosheets or graphene-like porous carbon materials through pyrolysis, activation, and graphitization processes [52,53]. These materials often possess high specific surface areas and hierarchical pore structures, making them attractive for composite and functional applications. Furthermore, the utilization of waste-derived carbon resources contributes to circular-economy strategies and enhances the sustainability of graphene production [40].

Overall, while conventional fabrication methods remain essential for producing high-quality graphene with controlled structures, waste-to-graphene technologies offer a promising pathway toward potentially lower-cost and more sustainable GNS production. Nevertheless, their actual environmental advantage over conventional graphene fabrication cannot be assumed a priori. The net carbon footprint depends on feedstock collection and pretreatment, electricity source, conversion efficiency, purification requirements, yield, and post-processing or functionalization steps. Therefore, comparative life-cycle assessment is required to quantify whether waste-derived GNS can provide a lower embodied carbon burden when used as coating materials for cementitious composites. Moreover, when waste-derived GNS are used for coating cementitious constituents, additional factors such as nanosheet quality, oxidation degree, coating uniformity, alkaline stability, suspension recyclability, drying demand, and scale-up reproducibility must be considered, because low-carbon feedstock alone does not guarantee low-impact or technically reliable coating performance.

3. Coating Strategies of GNS in Cementitious Composites

Direct incorporation of GNS into cementitious composites via conventional dispersion methods often suffers from agglomeration in the highly alkaline pore solution, non-uniform distribution, and adverse effects on workability [11,29,30]. More critically, bulk dispersion cannot selectively deliver GNS to the weakest links that govern macroscopic performance—the ITZs, including fiber/matrix and aggregate/paste interfaces [40,44]. To overcome these limitations, “coating strategies” have been developed, whereby GNS are pre-deposited onto specific constituents (e.g., fibers, sand, aggregates, clinker particles, supplementary cementitious materials) or applied as functional coatings on hardened concrete surfaces [32,46,54,55]. The principle is to target critical interfacial regions where stress concentration and deterioration originate, forming a nano-reinforced interlayer that densifies the ITZ via C-S-H nucleation, fills interfacial pores [56], and bridges microcracks, thereby transforming the ITZ from a weak zone into a damage-tolerant region [33,38,48]. GNS coating strategies can be categorized into four types based on binding mechanisms and process characteristics: physical adsorption, chemical assembly, electrophoretic deposition, and in situ growth. It should be emphasized that the feasibility of GNS coatings in cementitious composites is highly method- and substrate-dependent rather than universally applicable. The highly alkaline and calcium-rich pore solution of cement-based materials can destabilize some graphene-based suspensions, especially GO, through ionic

cross-linking and sheet aggregation [57]. Therefore, physical adsorption, although simple and cost-effective, may provide limited long-term interfacial stability under prolonged alkaline exposure [58]. Chemical assembly can improve coating stability through covalent bonding or strong interfacial coupling, but its application in real cementitious systems remains mostly restricted to laboratory-scale fibers, fine particles, or model substrates [59]. Electrophoretic deposition offers controllable and uniform coatings, but it requires electrically conductive substrates, which excludes most natural aggregates, cement clinker, and supplementary cementitious materials unless additional conductive pretreatments are introduced [60]. Accordingly, GNS coating strategies should be regarded as targeted interfacial-engineering approaches with specific applicability windows, rather than general-purpose coating solutions for all cementitious constituents.

In practical terms, physically adsorbed GO coatings are more feasible for fibers, fine particles, recycled powders, and small-batch sand treatments, but their stability may be weakened by high-shear mixing and calcium-rich alkaline pore solutions. Silane-assisted chemical assembly is more suitable for sand, recycled aggregates, and oxide-rich inorganic surfaces where surface hydroxyl groups can be functionalized, although the process involves additional pretreatment, coupling agents, washing, and drying steps. Electrophoretic deposition is technically plausible for conductive fibers or steel fibers, but it is not directly applicable to natural aggregates, cement clinker, or most SCMs without conductive pretreatment. In situ growth can provide strong interfacial integration on thermally stable substrates, especially carbon-based fibers, but its high processing temperature and energy demand restrict its use for ordinary aggregates, clinker, recycled aggregate powders, and polymer fibers. Therefore, the selection of a GNS coating route should be based on substrate conductivity, surface chemistry, thermal stability, coating uniformity, process energy, and compatibility with concrete mixing.

3.1. Physical Adsorption

Physical adsorption is the most widely adopted and operationally straightforward coating strategy, relying on non-covalent interactions such as van der Waals forces, electrostatic attraction, hydrogen bonding, and mechanical interlocking [11,44,61]. The process can be conducted under ambient conditions without altering the intrinsic structure of GNS or chemically damaging the substrate, making it simple, scalable, and cost-effective. A representative physical adsorption process for GO coating on PE fibers is shown in Figure 3.

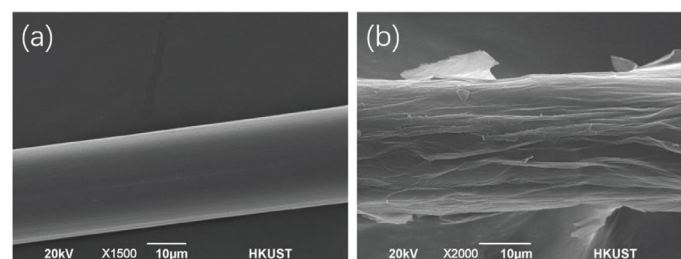
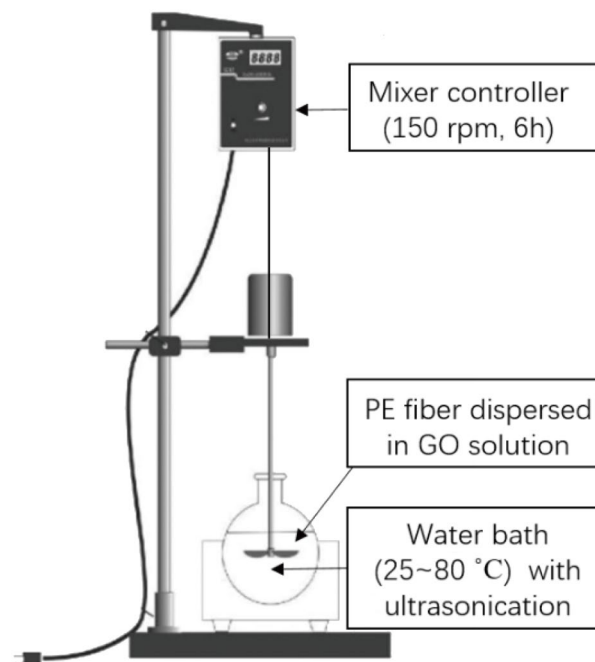


Figure 3. Physical deposition of graphene oxide on polyethylene fibers and corresponding surface morphology: schematic illustration of GO coating formation on PE fibers by simple mixing in an aqueous GO suspension; (a) SEM image of untreated PE fibers; and (b) SEM image of GO-coated PE fibers with roughened surface morphology. Adapted from Ref. [62].

For fibrous substrates, dip-coating is the most commonly employed approach. Yao et al. demonstrated that graphene oxide (GO) can be uniformly deposited onto poly(vinyl alcohol) (PVA) fibers, increasing surface roughness and hydrophilicity and thereby improving fiber–matrix interfacial bonding [61]. Similarly, Lu et al. reported that GO-coated carbon fibers exhibited enhanced surface wettability and stronger interactions with cement hydration products, leading to improved interfacial performance [63].

For particulate substrates, GNS coatings are typically achieved through liquid-phase assembly or mechanical mixing. Recent studies showed that GO-coated fly ash particles act as effective nucleation sites for cement hydration while simultaneously serving as physical spacers that reduce graphene agglomeration and promote a more uniform distribution of nanosheets within the cementitious matrix [7,64].

Although particulate coating is highly relevant for practical concrete because aggregates occupy the largest volume fraction of concrete [65], scalable processing of GNS-coated aggregates remains insufficiently developed. Most existing studies have focused on fine sand, recycled concrete aggregates, or small batches of particles under controlled laboratory conditions [66]. For commercial concrete production, however, coating methods must be compatible with large aggregate volumes, broad particle-size distributions, surface moisture variations, dust contamination, and continuous mixing processes. Key technical issues include achieving uniform GNS coverage on irregular aggregate surfaces,

preventing coating loss during storage and mixing, controlling water demand and workability, recycling or treating coating suspensions, and maintaining cost-effective throughput. Therefore, aggregate coating should be regarded as one of the most critical but least mature routes for translating GNS coating strategies from laboratory studies to ready-mixed concrete production [67].

In addition to constituent modification, physically adsorbed GNS coatings have also been explored for surface protection applications. Graphene-based coatings can refine pore structures, reduce water absorption, and suppress the transport of aggressive ions, thereby improving the durability of cementitious materials [68]. The effectiveness of physical adsorption primarily originates from the large specific surface area and unique surface chemistry of GNS. On fiber surfaces, GNS enhances mechanical interlocking and interfacial load transfer, while oxygen-containing functional groups on GO facilitate interactions with hydration products [13,69,70]. For particulate substrates, GNS provides abundant nucleation sites for C–S–H formation and helps prevent nanosheet re-agglomeration during mixing, contributing to a denser ITZ [7,14,64].

Despite these advantages, physical adsorption has inherent limitations. Since coating adhesion relies mainly on weak secondary interactions, partial desorption may occur under high-shear mixing or prolonged exposure to highly alkaline pore solutions [40]. Furthermore, calcium ions in cement pore solutions can induce GO aggregation through ionic cross-linking, reducing dispersion stability and potentially compromising the long-term effectiveness of the coating layer [57]. This behavior highlights the intrinsic instability of physically adsorbed GO coatings in calcium-rich alkaline environments, which may limit their long-term durability in structural concrete applications. These limitations have motivated the development of more robust coating approaches, such as chemical assembly and electrophoretic deposition.

3.2. Chemical Assembly

Chemical assembly refers to interfacial engineering techniques that establish irreversible covalent bonding between GNS and substrate surfaces, overcoming the weak adhesion and potential instability inherent in physical adsorption coatings [11,38]. The strategy typically involves introducing reactive functional groups onto substrate surfaces (e.g., $-\text{NH}_2$, $-\text{OH}$) that can chemically react with oxygen functional groups on GNS ($-\text{COOH}$, $-\text{OH}$, epoxy), forming covalent linkages (e.g., amide, ester, siloxane) and yielding strong interfacial integration [71]. A typical chemical assembly process for preparing GO-coated PVA fibers is illustrated in Figure 4.

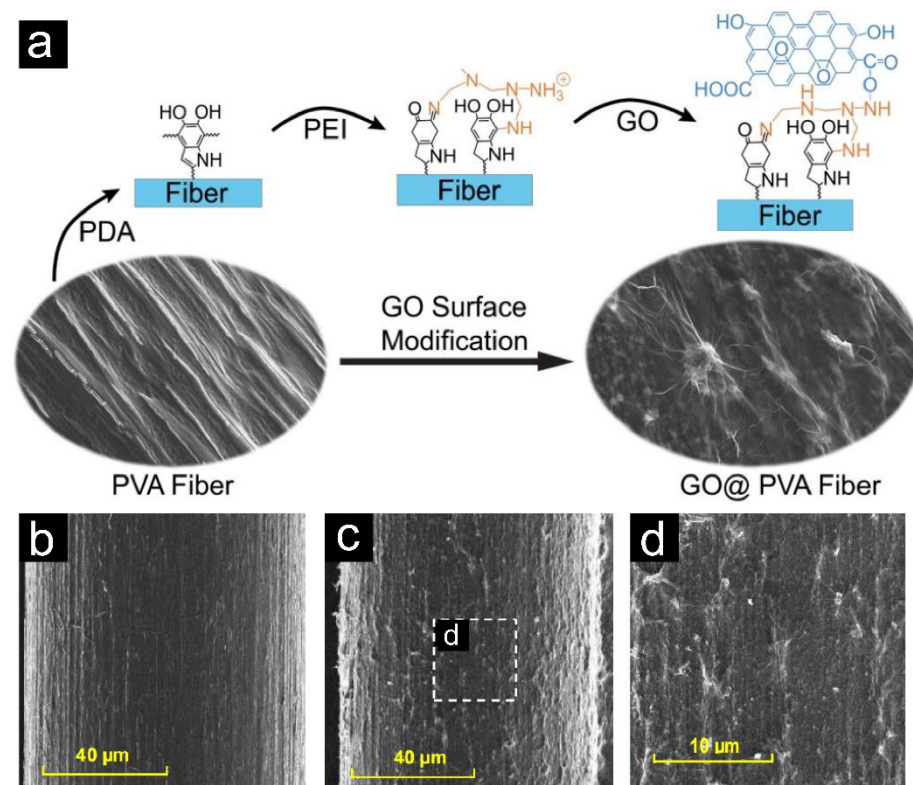


Figure 4. Chemical assembly coating approach and surface morphology of GO-coated PVA fibers: (a) schematic illustration of the fabrication process of GO@PVA fibers; (b) SEM image of pristine PVA fibers; (c,d) SEM image of GO-coated PVA fibers. Adapted from Ref. [61].

A typical chemical assembly process includes three main steps: substrate activation to generate reactive sites, grafting of coupling agents or functional molecules, and covalent anchoring of GNS. Substrate surfaces such as fibers or aggregates can be activated by chemical treatments (e.g., acid, alkali, plasma) to increase site density for subsequent functionalization [38,71]. Silane coupling agents (e.g., APTES) are commonly used to introduce terminal amino or silanol groups on substrates, which can then react covalently with GO functional groups [59]. For instance, Nguyen et al. [72] prepared GO-coated sand through amino-functionalization with 3-aminopropyltriethoxysilane followed by coupling with GO; the formation of strong interfacial bonds was supported by FTIR and XPS analyses. Functionalization with (3-aminopropyl)triethoxysilane has been shown to enable covalent bonding between GO and inorganic surfaces, enhancing interfacial microstructure and overall composite performance [73]. Although studies on exact covalent grafting between GO and typical cementitious substrates remain limited, this chemistry has been validated in related materials systems and provides a clear pathway for robust chemical bonding.

In addition to direct covalent coupling, layer-by-layer (LbL) self-assembly offers a bottom-up chemical assembly route by alternately adsorbing oppositely charged polyelectrolytes and charged GNS, constructing nanoscale multilayer films with tunable thickness and composition [74]. This technique has been widely applied in graphene-based coatings on various substrates and can be adapted for surface modification in cementitious environments, enabling precise control of interfacial architecture.

Covalently assembled GNS interfacial layers can provide multiple reinforcement benefits. First, chemically anchored GNS surfaces furnish abundant stable sites for the heterogeneous nucleation of calcium silicate hydrate (C–S–H), resulting in densification of the ITZ and improved microstructural homogeneity [75]. Second, a covalent network of GNS across the interface can bridge nanoscale microcracks and more effectively transfer stresses between phases, improving resistance to crack initiation and propagation under

load [76]. Combined with the intrinsic functional groups on GO, chemical assembly also aids the formation of stable interface phases and inhibits deleterious interfacial porosity [75,77].

Although chemical assembly is expected to provide stronger interfacial stability than physical adsorption because of covalent bonding or strong chemical coupling, such stability has not yet been fully demonstrated in real cementitious systems over service-life-relevant timescales [71]. Current evidence is still largely derived from laboratory-scale demonstrations involving fibers, fine particles, or model substrates [78]. Therefore, chemical assembly should be considered a promising but not yet mature route for long-term GNS coatings in cement-based materials. The multi-step processes involve organic coupling agents and functionalization chemicals, which increases cost and complexity. Some substrate activation treatments (e.g., strong acid etching) can degrade the mechanical properties of organic fibers.

3.3. Electrophoretic Deposition

Electrophoretic deposition (EPD) is a coating technique in which charged colloidal particles migrate and deposit onto conductive substrates under a direct-current electric field [79,80]. Compared with physical adsorption and chemical assembly, EPD enables rapid and controllable fabrication of uniform graphene-based coatings, providing high coating density, low agglomeration, and precise thickness control [11,79,81]. A typical EPD setup and the morphology of GO-coated carbon fibers are shown in Figure 5.

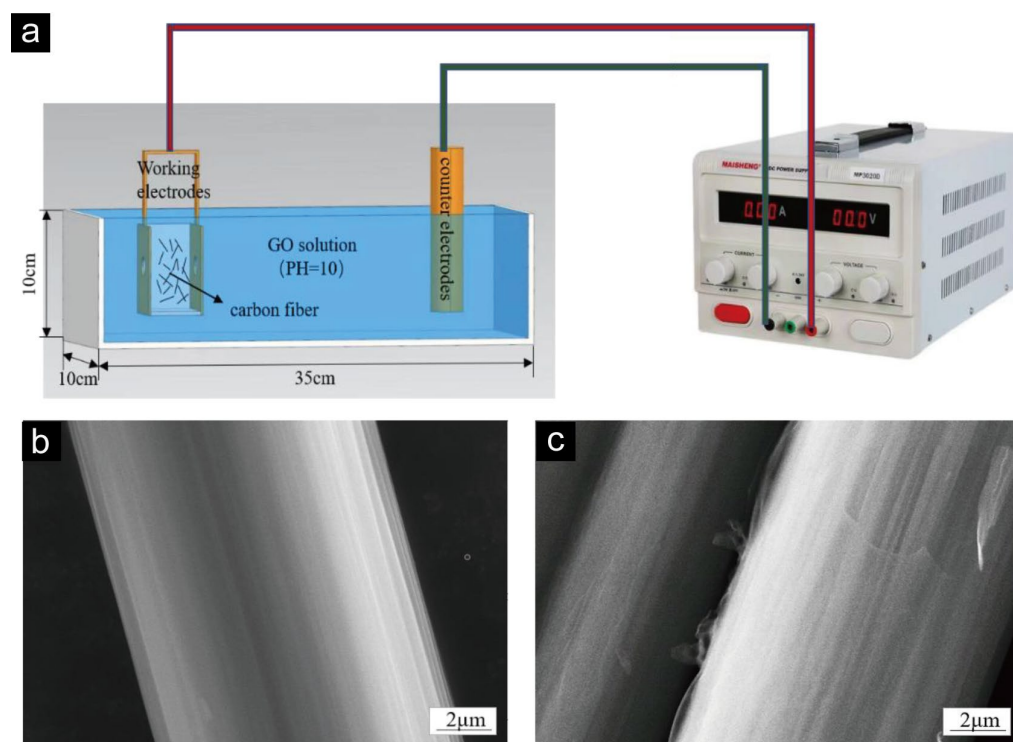


Figure 5. Electrophoretic deposition coating approach and SEM images of GO-coated carbon fibers: (a) schematic illustration of the electrophoretic deposition (EPD) process for coating graphene oxide onto carbon fibers; (b) SEM image of pristine carbon fibers; and (c) SEM image of GO-coated carbon fibers after EPD treatment. Adapted from Ref. [82].

In cementitious composites, EPD has primarily been investigated for modifying electrically conductive reinforcements such as carbon fibers and steel fibers [63,79]. Graphene oxide (GO) nanosheets dispersed in aqueous solutions carry a net negative charge due to ionization of surface carboxyl and hydroxyl groups, remaining colloidally stable [11].

When a conductive substrate serves as an electrode, the charged GO nanosheets migrate directionally under the electric field, adhere to the substrate surface, and form a continuous, compact coating without requiring surfactants or aggressive chemical reagents [79,80,83]. This preserves the intrinsic structure and functional groups of GO.

A typical EPD setup consists of a DC power supply, an electrolytic cell, and two electrodes. Deposition is usually completed within seconds to minutes, with applied voltages ranging from a few volts to tens of volts. Deposition parameters including voltage, suspension concentration, electrode spacing, and deposition time allow precise control of coating thickness from tens of nanometers to several micrometers [79,80,83].

EPD coatings have demonstrated significant enhancement in interfacial properties. In studies involving carbon fibers, GO deposited via EPD improved surface roughness and promoted interfacial adhesion with cementitious matrices, enhancing load transfer and mechanical stability [63,79,84]. Similarly, electrophoretically deposited coatings on conductive fibers can act as temporary barriers, reducing direct exposure to highly alkaline pore solutions and improving durability [63,85]. These effects collectively contribute to a denser ITZ and improved composite performance.

Despite its advantages, EPD is limited to electrically conductive substrates; natural sand, coarse aggregates, ordinary synthetic fibers, cement clinker, and most SCMs cannot be coated without conductive pretreatment [79,80,86]. Moreover, coating quality depends on suspension stability and electrical parameters, and excessive deposition may cause defects or reduce adhesion. Therefore, although EPD is highly effective for conductive reinforcements such as carbon fibers or steel fibers, it should be considered a substrate-specific coating strategy rather than a universal method for cementitious composites. Its direct application to ordinary aggregates, cement clinker, and most SCMs is restricted by their low electrical conductivity, and any extension of EPD to these constituents would require additional conductive pretreatment or hybrid processing routes.

3.4. Growth of GNS

In situ growth of GNS is a “bottom-up” coating strategy, differing from physical adsorption, chemical assembly, and electrophoretic deposition. Graphene is directly generated on the substrate from carbon precursors, forming a continuous layer with strong interfacial bonding [87–89]. This approach can maximize interface stability and stress transfer. The surface morphology of graphene nanowall-coated carbon fibers prepared by in situ growth is shown in Figure 6.

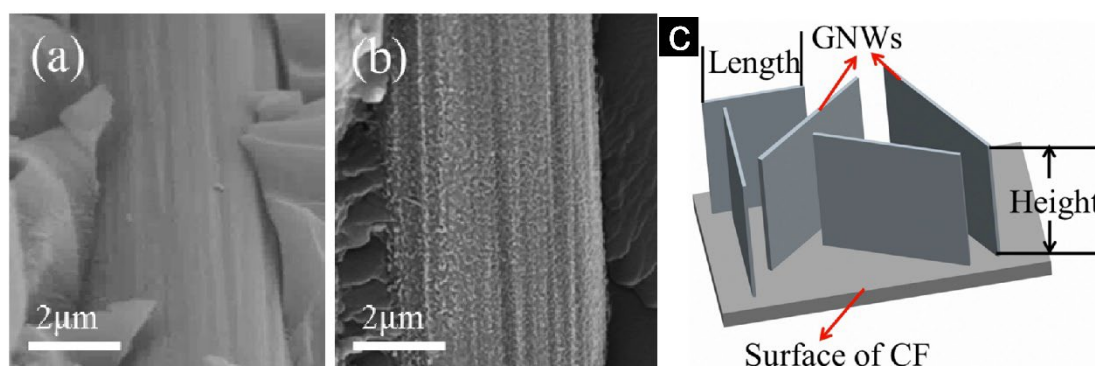


Figure 6. In situ growth coating approach and SEM images of graphene nanowall-coated carbon fibers: (a) SEM image of pristine carbon fibers; and (b) SEM image of graphene nanowall-coated carbon fibers after in situ growth. (c) simplified schematic illustration of graphene nanowalls grown on the surface of carbon fibers. Adapted from Ref. [90].

Chemical vapor deposition (CVD) is the most common method. Hydrocarbon gases such as methane decompose at high temperatures ($\sim 900\text{--}1100\text{ }^{\circ}\text{C}$) on catalytic substrates, producing graphene films on fibers [91]. Chi et al. demonstrated graphene nanowalls grown on carbon fibers, and Chen et al. successfully grew graphene on glass fiber fabrics, improving interfacial bonding and conductivity [87,92]. Zhou et al. reported in situ graphene on carbon fibers enhanced mechanical and thermal properties [93]. The exceptional mechanical properties of graphene (Young's modulus $\sim 1\text{ TPa}$, tensile strength $\sim 130\text{ GPa}$) enable these coatings to act as ultra-thin, robust interphases [88,91].

Flash Joule heating (FJH) offers rapid, energy-efficient graphene synthesis from carbon-rich wastes such as plastics or tires [23]. Although FJH mainly produces graphene powders rather than direct coatings, it represents a potential sustainable source for future applications.

Despite its advantages, in situ growth in cementitious composites is still at the laboratory stage. High temperatures of CVD limit compatibility with most cement constituents and polymer fibers, and scalable methods for aggregates or SCMs are yet to be developed [7,94]. Developing low-temperature, energy-efficient, and substrate-compatible processes remains essential for practical application [23,87,91].

4. GNS-Based Coating in Enhancing Cementitious Composites

4.1. Enhanced Mechanical Properties

To provide a descriptive comparison of representative mechanical enhancements reported for different GNS-coated cementitious systems, the improvements in compressive and tensile/flexural strengths are summarized in Figure 7. Because the original studies differ in GNS type, substrate, coating route, dosage, curing age, and testing method, the comparison is intended to be descriptive rather than statistical.

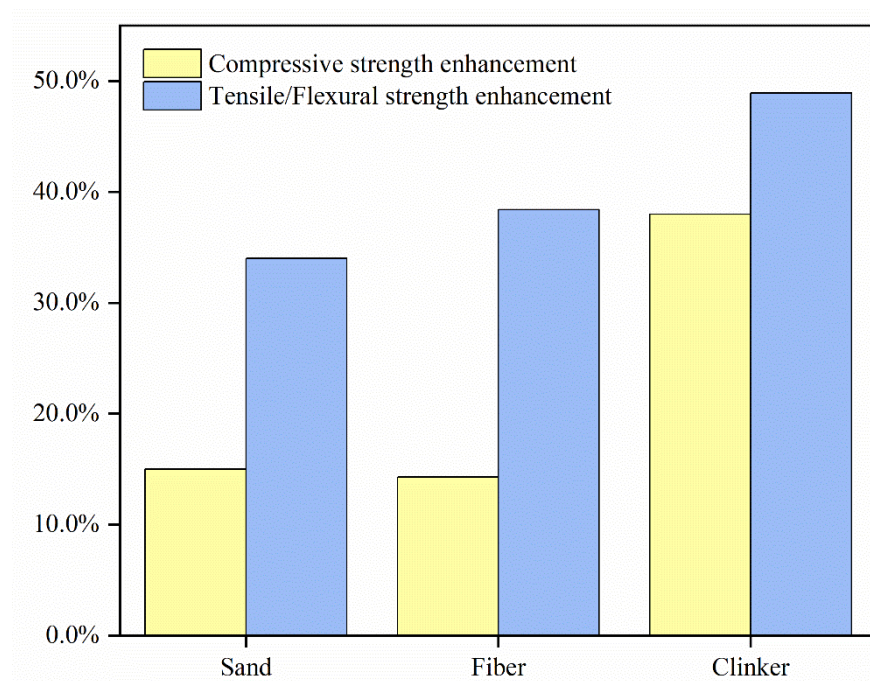


Figure 7. Comparison of the enhancement in compressive and tensile or flexural strengths achieved by GNS coating on different cement components. The second strength index refers to tensile strength or flexural strength depending on the original study. The values were extracted from representative studies on GO-coated sand, GO-coated carbon fibers prepared by electrophoretic deposition, and in situ graphene-modified clinker/cement systems. Because these studies differ in GNS type, substrate, coating route, dosage, curing age, and testing method, this figure is intended as a

descriptive comparison rather than a statistical meta-analysis. Error bars are not shown because the source studies did not report standard deviations or sample sizes in a directly comparable format. Adapted from Ref. [72,79,95].

The mechanical enhancement achieved by GNS coatings primarily originates from targeted reinforcement of ITZs, which are generally recognized as the weakest regions in cementitious composites. By localizing graphene nanosheets at critical interfaces rather than dispersing them throughout the matrix, coating strategies improve stress transfer efficiency, promote hydration product formation, and suppress crack initiation and propagation [13,76,96]. It should be emphasized that the reported mechanical enhancement values in GNS-coated cementitious systems are derived from heterogeneous experimental conditions, including variations in graphene type, coating method, substrate geometry, dosage level, curing regime, and testing age. Therefore, direct quantitative comparison between different studies should be interpreted with caution. To improve interpretability, the following discussion organizes the results based on substrate type and coating strategy, and distinguishes early-age performance from later-age performance wherever possible. The values summarized in Table 2 are intended to improve cross-study transparency rather than to provide a direct statistical comparison. The listed studies differ in GNS chemistry, coating route, substrate type, coating condition, cementitious matrix, curing regime, and testing age. Therefore, the reported enhancement ratios should be interpreted as system-specific outcomes rather than universal performance indicators of all GNS coating strategies.

Fiber–matrix interfaces play a decisive role in the mechanical performance of fiber-reinforced cementitious composites. Due to their smooth and chemically inert surfaces, conventional polymer and carbon fibers often exhibit limited interfacial bonding with cement hydrates. Coating fibers with graphene oxide (GO) significantly improves surface activity and interfacial load transfer. Yao et al. reported that the splitting tensile strength of the 35-day specimens increased by 35.6% compared with the untreated PVA fiber-reinforced cementitious composites [61]. The GO coating promoted hydration around the fiber surface and changed the failure mode from interfacial debonding to cohesive failure [61,97]. Similarly, Lu et al. demonstrated that GO-coated polyethylene (PE) fibers enhanced the first-cracking strength, ultimate tensile strength, and tensile strain capacity by 11.7%, 46.3%, and 70.4%, respectively [74]. For conductive reinforcements, Wang et al. employed electrophoretic deposition to coat carbon fibers with GO, resulting in improved interfacial bonding and flexural performance of the cementitious composites [79]. Recent studies on GO-coated steel fibers further revealed substantial increases in surface roughness and wettability, which enhanced fiber–matrix mechanical interlocking and promoted crack-bridging effectiveness [97].

The aggregate–paste ITZ is another critical weak zone due to its higher porosity and relatively loose microstructure. Coating sand or aggregates with GNS provides abundant nucleation sites for hydration products and promotes the formation of a denser ITZ [98]. Nguyen et al. reported that GO-coated sand significantly refined the interfacial transition zone (ITZ), reducing its thickness by approximately 20% and increasing ITZ hardness by about 17%. This ITZ refinement was accompanied by enhancements in 28-day compressive and tensile strengths of approximately 15% and 34%, respectively [72]. The coating also reduced water absorption by approximately 20%, indicating a more compact interfacial structure [64]. For recycled concrete aggregates (RCA), Lu et al. demonstrated that GO coating significantly improved ITZ compactness and reduced microstructural defects, resulting in enhanced mechanical performance and durability [99]. Similarly, pristine graphene-treated RCA was shown to promote hydration and improve the overall mechanical properties of recycled aggregate concrete [100]. However, the positive results obtained

from GO-coated sand or recycled aggregates should be interpreted in the context of laboratory-scale processing [66]. In real concrete production, aggregates include both fine and coarse fractions with irregular shapes, variable mineral compositions, different water absorption capacities, and non-uniform surface conditions. These factors may strongly influence GNS adsorption, coating continuity, and interfacial bonding efficiency. Moreover, because aggregates account for the majority of concrete volume, even a thin GNS coating layer would require large-scale material handling and process control. At present, systematic studies on coating coarse aggregates at industrially relevant scales, evaluating coating uniformity after mixing, and quantifying the resulting performance gain per unit processing cost remain limited [101].

Compared with fiber and aggregate coatings, studies on GNS-coated cement clinker and supplementary cementitious materials (SCMs) remain relatively limited. Existing investigations suggest that coating cementitious particles with GO can provide additional nucleation sites for calcium silicate hydrate (C–S–H) formation and accelerate early hydration [13,96,102]. The high specific surface area of GO facilitates calcium ion adsorption and promotes the growth of hydration products around particle surfaces [103]. As a result, coated particles contribute to a denser microstructure and improved early-age mechanical performance. However, systematic studies quantifying the strengthening efficiency of clinker- or SCM-based coatings are still scarce, and further research is required to establish clear structure–property relationships [102].

Beyond strength enhancement, GNS coatings improve toughness through multiple crack-resisting mechanisms. Nanosheets located at interfaces can bridge, deflect, pin, and branch propagating microcracks, thereby increasing fracture energy and delaying crack coalescence [75,76]. Molecular simulation and experimental observations have demonstrated stronger interfacial interactions between GO and hydration products, leading to improved stress transfer efficiency [75]. Consequently, GNS-coated cementitious composites exhibit enhanced ductility, crack resistance, and post-cracking load-carrying capacity compared with conventional systems. It should also be noted that most reported mechanical enhancements were obtained from paste, mortar, or small concrete specimens prepared under controlled laboratory conditions [101,104]. The transferability of these improvements to full-scale concrete members depends on coating uniformity during large-volume mixing, aggregate gradation, workability, curing conditions, reinforcement detailing, and structural loading states [105,106]. Therefore, field-scale or member-level validation is required before laboratory mechanical benefits can be translated into engineering design advantages. Although GNS coatings show promising interfacial reinforcement efficiency, their advantages should be evaluated in comparison with established cementitious-material modification strategies. Conventional approaches, such as silica fume, metakaolin, supplementary cementitious materials, optimized particle packing, and fiber reinforcement, have been widely used to improve strength, refine pore structures, and enhance durability at relatively mature cost and supply-chain levels [107]. At present, systematic comparisons between GNS coatings and these conventional strategies remain limited, particularly in terms of performance improvement per unit material cost, per unit embodied carbon, and per unit processing energy. Therefore, the reported mechanical and durability improvements of GNS-coated systems should not be interpreted as evidence of superior cost- or carbon-effectiveness unless benchmarked against established alternatives under comparable mixture designs, curing conditions, exposure environments, and functional requirements [108].

The compatibility of GNS coatings with other modern concrete technologies remains insufficiently explored. In practical mixtures, GNS-coated constituents may be combined with supplementary cementitious materials, polycarboxylate superplasticizers, low wa-

ter-to-binder-ratio designs, recycled aggregates, fiber reinforcement, self-compacting concrete, or 3D-printable cementitious systems. These combinations may generate unintended interactions, including competition between admixtures and GO for adsorption sites, changes in pore-solution chemistry and calcium availability caused by SCMs, altered rheology and water demand, coating loss under mixing shear, and modified hydration or crack-bridging behavior. Therefore, the synergy between GNS coatings and existing concrete technologies should be verified through systematic compatibility studies rather than assumed [109,110]. A compact comparison of representative GNS coating systems, including GNS type, substrate, coating route, cementitious system, and main performance improvement, is summarized in Table 2.

Table 2. Compact comparison matrix of representative GNS coating systems in cementitious composites.

GNS Type	Substrate	Coating Route	Cementitious System	Main Performance Improvement	Ref.
GO	PVA fibers	Physical adsorption	PVA fiber-reinforced cementitious composites	Splitting tensile strength increased by 35.6% compared with untreated PVA fiber-reinforced cementitious composites.	[61]
GO	PE fibers	Physical adsorption	PE fiber-reinforced strain-hardening cementitious composites	First-cracking strength, ultimate tensile strength, and tensile strain capacity increased by 11.7%, 46.3%, and 70.4%, respectively.	[74]
GO	Sand	Silane-assisted chemical assembly	Cement mortar with GO-coated sand	ITZ thickness reduced by approximately 20%; ITZ hardness increased by about 17%; 28-day compressive and tensile strengths increased by approximately 15% and 34%, respectively; water absorption decreased by approximately 20%.	[72]
GO	Carbon fibers	Electrophoretic deposition, EPD	GO-coated carbon fiber-reinforced cement-based composites	Improved interfacial bonding and flexural/mechanical performance were reported.	[79]
Graphene-based coating	Clinker or cement particles	In situ growth	Graphene-based cement material	Representative strength enhancement was reported for in situ graphene-modified clinker/cement systems.	[95]

4.2. Enhanced Durability

The durability deterioration of cementitious composites is primarily governed by the ingress of water, chloride ions, carbon dioxide, and other aggressive species through interconnected pores and ITZs. Compared with conventional bulk incorporation, GNS coating strategies localize graphene nanosheets at critical interfaces, where they can refine pore structures, densify hydration products, and suppress microcrack development. These effects have been shown to improve short-term durability-related indicators, such as water absorption and chloride transport resistance. However, whether such improvements can be maintained over service-life-relevant timescales remains insufficiently verified, particularly under prolonged exposure to calcium-rich alkaline pore solutions and coupled environmental actions [111,112]. The freeze–thaw deterioration behavior and microstructural changes of GO-modified concrete are illustrated in Figure 8.

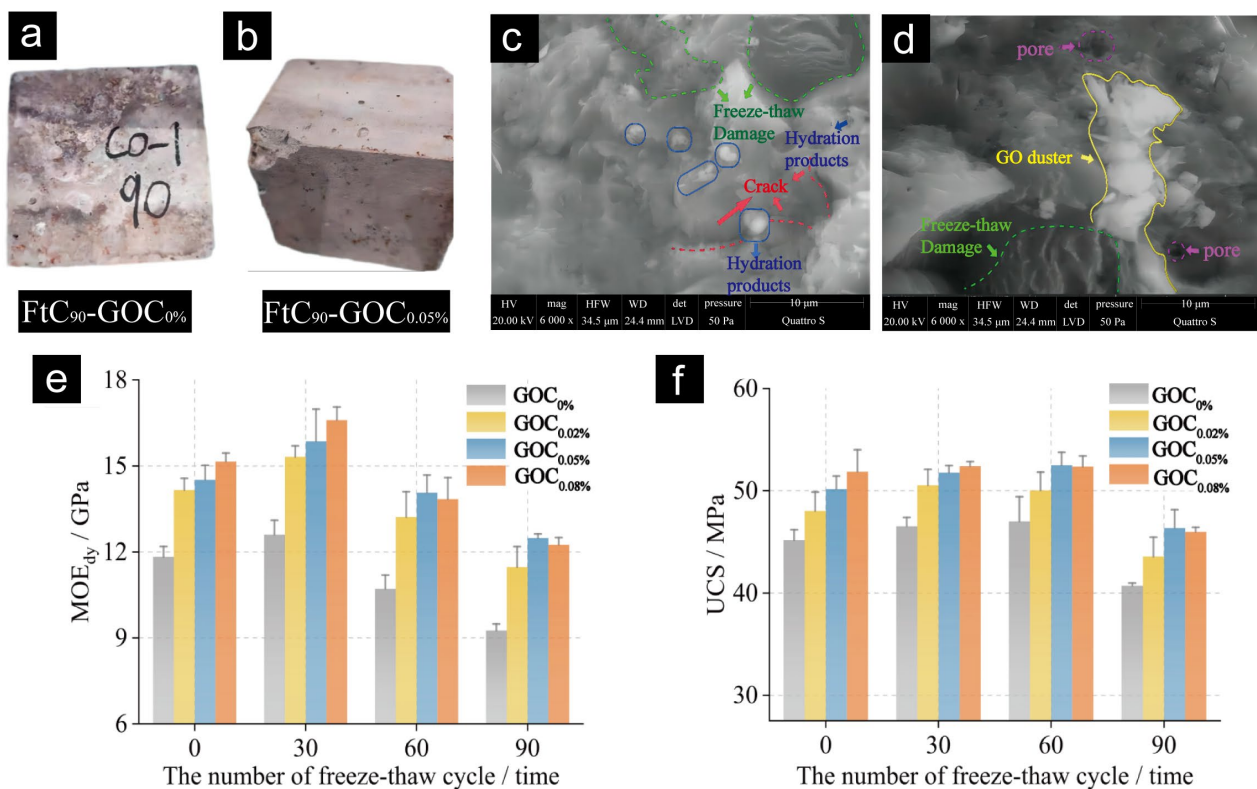


Figure 8. Microstructural and quantitative deterioration of GO-modified concrete under freeze-thaw cycling: (a,b) SEM images of unmodified concrete and GO-modified concrete after 90 freeze-thaw cycles; (c,d) surface morphology of unmodified and GO-modified concrete after freeze-thaw exposure; (e) relative dynamic elastic modulus; and (f) uniaxial compressive strength. Adapted from Ref. [113].

One of the most significant durability benefits of GNS coatings is the reduction in mass transport through the ITZ. Due to their two-dimensional morphology and high aspect ratio, graphene nanosheets create tortuous diffusion pathways that hinder the migration of water and ions [103]. Nguyen et al. reported that GO-coated sand reduced the water absorption of cement mortar by approximately 20% compared with the control specimen, accompanied by a denser ITZ and improved mechanical performance [72]. Similarly, Lu et al. demonstrated that GO-coated recycled concrete aggregates (RCA) effectively decreased water absorption and chloride-ion transport by improving the microstructural integrity of the aggregate–paste interface [114]. It should be noted that reductions in water absorption and chloride-ion transport mainly represent improved transport-related performance within the specific testing periods adopted in existing studies. These indicators do not by themselves confirm long-term coating stability or service-life-level durability [72]. Most available studies on GNS-coated cementitious composites are based on laboratory specimens with relatively short exposure durations, commonly ranging from 28 days to several months, and therefore should not be directly extrapolated to the 50–100-year service life required for structural concrete. For physically adsorbed GO coatings, prolonged exposure to aqueous alkaline environments may weaken non-covalent interactions between GNS and substrates, while Ca^{2+} in cement pore solutions may induce GO aggregation through ionic cross-linking. In addition, repeated wet–dry cycles, thermal cycles, freeze–thaw cycles, chloride exposure, carbonation, sulfate attack, and coupled mechanical–environmental actions may further affect coating retention, interfacial bonding, and transport pathways. Therefore, the durability benefits obtained from GNS coatings

should be interpreted as short-term or medium-term improvements unless their resistance to desorption, ionic cross-linking, environmental cycling, and time-dependent degradation is systematically demonstrated [115].

Recent studies further indicate that the durability effects of GNS coatings should be interpreted from the perspective of evidence convergence and remaining inconsistency. The relatively convergent evidence is that GO-containing coatings can promote hydration-product nucleation, densify the ITZ, refine interfacial pores, and reduce short-term water absorption or chloride transport. These effects have been reported in GO-coated sand, GO-treated recycled aggregates, GO-coated fly ash, and GO-coated fiber systems, suggesting that targeted interfacial engineering is a plausible mechanism for improving transport-related durability indicators. However, the evidence is still not fully unified across material classes and exposure conditions. In particular, long-term alkaline stability, drying shrinkage, freeze–thaw resistance, coupled chloride–carbonation or sulfate–freeze–thaw deterioration, and durability retention in UHPC, recycled aggregate concrete, and ordinary mortar systems may exhibit different responses. The differences among GO, rGO, and GNPs should also be considered because their oxygen-containing functional groups, colloidal stability, electrical conductivity, and bonding ability with cement hydrates are substantially different. Therefore, durability enhancement should be regarded as a system-specific outcome rather than a general property of all graphene-based coatings. Future studies should combine prolonged alkaline aging, environmental cycling, and coupled deterioration tests with microstructural characterization to clarify where the evidence is convergent and where contradictory or exposure-dependent responses remain.

GNS coatings also improve the resistance of cementitious composites to interfacial deterioration. SEM observations from several studies revealed that GO-coated fibers and aggregates promote the formation of compact hydration products around the interface, resulting in fewer microcracks and reduced porosity [61,72,99,116]. Yao et al. observed that hydration products were densely attached to GO-coated PVA fibers, indicating stronger fiber–matrix interactions and a more stable interfacial microstructure [61]. Similar interfacial densification has been reported for GO-coated carbon fibers and recycled aggregates, where graphene nanosheets served as nucleation sites for C–S–H growth and reduced the presence of large interfacial voids [63,99].

The enhanced interfacial structure further contributes to improved resistance against freeze–thaw damage and carbonation [117]. A denser ITZ reduces the amount of freezable water and restricts crack propagation induced by volumetric expansion during freezing cycles. Moreover, the reduced pore connectivity slows carbon dioxide diffusion into the cement matrix, delaying carbonation-front advancement and helping maintain the alkalinity required for steel passivation [99,100]. Although quantitative studies on coating systems remain limited, existing results generally indicate improved durability-related indicators compared with unmodified specimens. However, the magnitude and persistence of these improvements remain dependent on GNS chemistry, coating stability, substrate type, matrix composition, and exposure conditions.

Resistance to sulfate and chemical attack is also improved by the barrier effect of graphene nanosheets [118]. The compact interfacial structure generated by GNS coatings hinders the ingress of sulfate ions and other corrosive species, reducing the formation of expansive products such as ettringite and gypsum within the ITZ [13,76,99]. In addition, the chemically stable graphene framework provides an effective physical barrier that mitigates acid penetration and microstructural degradation under aggressive exposure conditions [99,100,119].

In addition to exposure duration, the chemical degradation kinetics of graphene-based coatings under realistic pore-solution chemistry remain insufficiently quantified

[104]. Potential processes such as Ca^{2+} -induced aggregation, loss or transformation of oxygen-containing functional groups, weakening of coating–substrate interactions, and changes in interfacial bonding may alter the transport-blocking and crack-bridging effects of GNS coatings over time [120]. Long-term exposure tests combined with microstructural characterization and service-life prediction models are therefore required to determine whether short-term durability improvements can persist under real service conditions [121].

Overall, the durability enhancement observed in GNS-coated cementitious composites originates from the combined effects of pore refinement, transport inhibition, interfacial densification, and crack suppression [122]. Nevertheless, current evidence mainly confirms improvements in laboratory durability indicators rather than service-life-level durability. The long-term effectiveness of GNS coatings, especially physically adsorbed coatings, still needs to be validated under prolonged alkaline aging, wet–dry cycling, freeze–thaw cycling, chloride exposure, carbonation, sulfate attack, and coupled mechanical–environmental deterioration. Therefore, GNS coatings should be regarded as promising durability-enhancement strategies whose long-term reliability remains to be established.

4.3. Smart Functionality

Beyond mechanical reinforcement and durability enhancement, GNS coating strategies provide an effective route for imparting smart functionalities to cementitious composites [123]. By concentrating graphene nanosheets on fiber or aggregate surfaces, conductive pathways can be established at relatively low nanomaterial dosages [66], enabling self-sensing, electromagnetic shielding, and electrothermal functions [63,97,124]. The conductive pathways and self-sensing mechanism of graphene-modified cementitious composites are schematically shown in Figure 9.

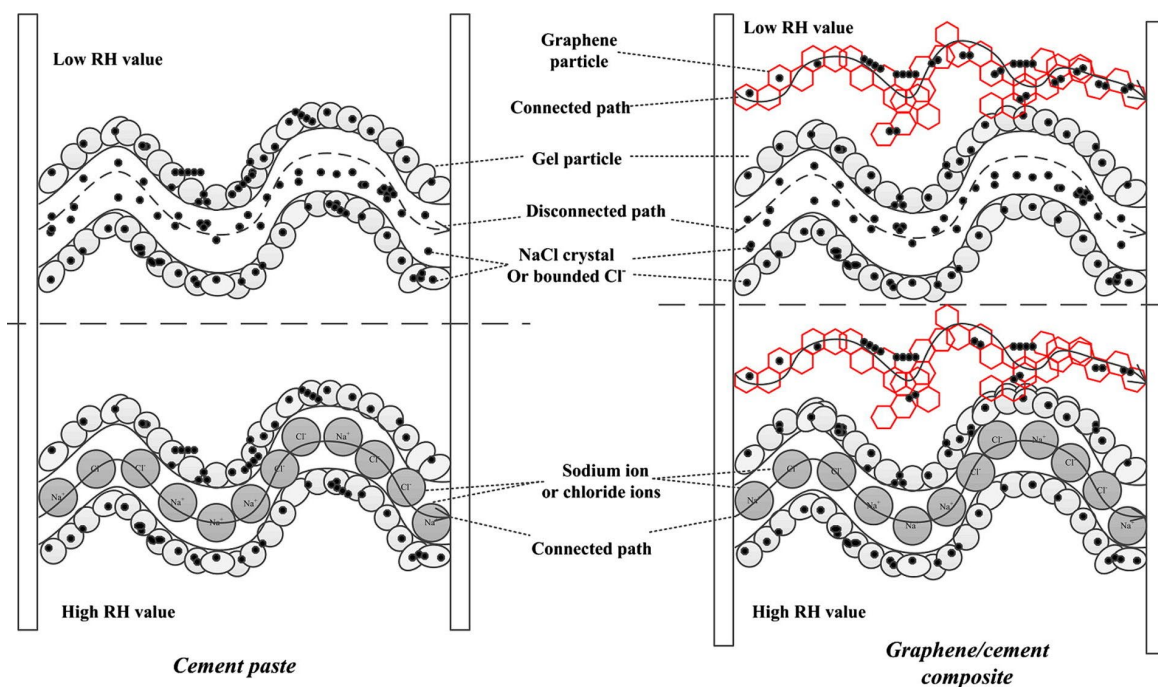


Figure 9. Conceptual schematic of conductive pathways formed by graphene-modified interfaces in cementitious composites, showing their potential roles in electrical conductivity, piezoresistive self-sensing, and structural health monitoring. Adapted from Ref. [125].

Self-sensing based on the piezoresistive effect is one of the most promising functionalities of GNS-coated cementitious composites [126,127]. For electrically conductive graphene-coated reinforcements, especially carbon-fiber-based systems, the coated fibers may act simultaneously as reinforcing elements and conductive sensors. Under external loading, the initiation and propagation of microcracks alter conductive pathways, leading to measurable changes in electrical resistance that can be correlated with stress and damage evolution [124,128–130]. Similarly, Lu et al. demonstrated that GO-electrodeposited carbon fibers formed stable conductive networks while improving interfacial performance, enabling reliable strain monitoring during mechanical loading [63,131]. These studies indicate that GNS-coated reinforcements can detect damage development before visible cracking occurs, providing opportunities for real-time structural health monitoring [132].

The conductive network generated by graphene-coated fibers and particles can substantially improve the electrical conductivity of cementitious materials [101,133]. Compared with direct dispersion, coating strategies localize conductive fillers more efficiently and reduce the amount of graphene required to establish conductive pathways [101,134,135]. Enhanced conductivity further contributes to electromagnetic interference (EMI) shielding performance. The layered structure and large specific surface area of graphene nanosheets facilitate multiple reflection, scattering, and absorption of incident electromagnetic waves [13,76,96,134]. Studies on graphene-based cement composites have reported shielding effectiveness exceeding 20 dB in the X-band frequency range, indicating attenuation of more than 99% of incident electromagnetic radiation [134,136]. Such performance is attractive for applications requiring electromagnetic protection and smart infrastructure systems.

The electrically conductive networks formed by GNS coatings also enable efficient electrothermal conversion through the Joule heating effect. When an external voltage is applied, electrical energy is converted into heat, producing a rapid and uniform temperature increase within the composite [13,76]. Experimental studies have demonstrated that graphene-based conductive cementitious materials can generate sufficient heat for snow-melting and deicing applications in pavements, bridge decks, and airport runways. Compared with conventional chemical deicers, electrothermal systems offer advantages including environmental friendliness, lower maintenance requirements, and compatibility with automated control systems [137]. The integration of self-sensing and electrothermal capabilities further highlights the potential of GNS-coated cementitious composites for next-generation intelligent infrastructure.

Overall, GNS coating strategies provide a versatile platform for transforming conventional cementitious materials into multifunctional smart materials [138]. Through the synergistic realization of sensing, conductive, shielding, and electrothermal functions, GNS-coated composites exhibit significant potential for future resilient and intelligent infrastructure applications.

The end-of-life behavior of GNS-coated cementitious composites remains largely unexplored. Graphene-coated aggregates, fibers, or cementitious particles may influence concrete crushing, separation, recycled aggregate quality, recycled fines utilization, dust generation, and disposal pathways. Depending on coating adhesion and degradation state, GNS may remain attached to recycled aggregates, be incorporated into fine recycled powders, or be partially released during demolition and mechanical processing. Therefore, future studies should assess the fate, stability, and potential environmental release of GNS coatings during recycling and disposal to ensure that their sustainability benefits are evaluated over the full life cycle.

5. Conclusions

Graphene-based coating strategies represent a targeted and highly efficient approach for enhancing the performance of cementitious composites through interfacial engineering. Unlike conventional dispersion methods, GNS coatings selectively localize graphene-based nanosheets at critical interfaces, including fiber–matrix, aggregate–paste, and recycled aggregate–paste ITZs, where mechanical deterioration and durability degradation are most likely to initiate. Based on the literature reviewed, the following conclusions can be drawn:

(1) GNS coating strategies effectively improve the mechanical performance of cementitious composites by promoting hydration, refining interfacial microstructures, enhancing interfacial bonding, and suppressing crack initiation and propagation. Reported studies indicate that GO-coated fibers can increase tensile strength by up to 35–46% and tensile strain capacity by over 70%, while GO-coated sand has been reported to reduce ITZ thickness by approximately 20%, increase ITZ hardness by about 17%, and enhance 28-day compressive and tensile strengths by approximately 15% and 34%, respectively. These improvements originate primarily from ITZ densification, enhanced load-transfer efficiency, and graphene-induced crack-bridging mechanisms.

(2) GNS coatings can improve durability-related indicators by reducing pore connectivity, improving interfacial compactness, and creating tortuous transport pathways. Existing studies have reported reductions of approximately 20% in water absorption and decreases in chloride-ion penetration. However, these results are mainly derived from laboratory tests with limited exposure durations, and they should not be directly extrapolated to 50–100-year service lives without long-term alkaline-aging and coupled-deterioration validation.

(3) Beyond structural reinforcement, GNS coating strategies provide an effective platform for multifunctional cementitious materials. By concentrating conductive nanosheets at interfaces, efficient conductive networks can be established at relatively low graphene dosages, enabling self-sensing, electromagnetic shielding, and electrothermal functionalities that support intelligent infrastructure systems and condition-based maintenance.

(4) From a carbon-neutrality perspective, GNS coating strategies may contribute to emission reduction by improving material efficiency and extending service life. However, the estimated cement savings and CO₂ reductions should be regarded as scenario-based potentials rather than verified net carbon benefits, because strength improvement does not automatically lead to structural or mixture optimization. Moreover, the embodied carbon associated with GNS synthesis, functionalization, coating deposition, drying, energy input, and substrate pretreatment must be included in future life-cycle assessments.

(5) Durability enhancement may provide additional life-cycle carbon benefits by reducing repair and replacement demand. Nevertheless, most current evidence is derived from short-term laboratory tests, and the persistence of GNS coatings under prolonged alkaline exposure, wet–dry cycling, freeze–thaw cycling, and coupled deterioration conditions remains insufficiently verified.

(6) Despite these advantages, several challenges remain before large-scale implementation becomes feasible. Current research is still dominated by laboratory-scale investigations, and key issues such as long-term coating stability in calcium-rich alkaline environments, coating uniformity, scalable aggregate processing, field-scale validation, and quantitative life-cycle assessment remain insufficiently resolved. Future studies should clarify the method-specific limitations of physical adsorption, chemical assembly, and electrophoretic deposition, and compare GNS coatings with established low-carbon strategies to evaluate their practical cost- and carbon-effectiveness. Particular attention should be paid to matching coating routes with substrate characteristics, for example, physical adsorption or chemical assembly for sand and recycled aggregates, EPD for conductive fibers or

steel fibers, and low-temperature or hybrid approaches for clinker, SCMs, and recycled aggregate powders.

Overall, graphene-based coating technologies provide a promising route for simultaneously improving mechanical performance, durability, multifunctionality, and sustainability. By transforming weak interfaces into highly efficient reinforcement zones, GNS coatings may become an important enabling technology for the next generation of low-carbon and high-performance cementitious materials.

Author Contributions: Validation, S.D.; data curation, Y.G.; writing—original draft, S.D.; writing—review and editing, X.Y.; supervision, M.D.; visualization, S.D.; project administration, M.D.; funding acquisition, M.D. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by National Natural Science Foundation of China, grant number 52508319; China Postdoctoral Science Foundation, grant number 2023TQ0299; China Postdoctoral Science Foundation, grant number 2023M743218 and Natural Science Foundation of Henan Province, grant number 252300423465.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: No new data were created or analyzed in this study.

Conflicts of Interest: The authors declare no conflicts of interest.

References

1. *Mineral Commodity Summaries 2025*; U.S. Geological Survey: Reston, VA, USA, 2025.
2. IEA. *Net Zero Roadmap: A Global Pathway to Keep the 1.5 °C Goal in Reach*; IEA: Paris, France, 2023.
3. IEA. *CO₂ Emissions in 2022*; IEA: Paris, France, 2024.
4. Cement, G. *Global Cement and Concrete Association*; GCCA: London, UK, 2021.
5. Andrew, R.M. Global CO₂ emissions from cement production. *Earth Syst. Sci. Data* **2018**, *10*, 195–217.
6. Habert, G.; Miller, S.A.; John, V.M.; Provis, J.L.; Favier, A.; Horvath, A.; Scrivener, K.L. Environmental impacts and decarbonization strategies in the cement and concrete industries. *Nat. Rev. Earth Environ.* **2020**, *1*, 559–573.
7. Cheng, Z.; Liu, Y.; Wu, J.; Guo, X.; Chen, W.; Gao, Y. Graphene oxide-coated fly ash for high performance and low-carbon cementitious composites. *J. Mater. Res. Technol.* **2023**, *25*, 6710–6724.
8. Lee, C.; Wei, X.; Kysar, J.W.; Hone, J. Measurement of the elastic properties and intrinsic strength of monolayer graphene. *Science* **2008**, *321*, 385–388.
9. Katsnelson, M.I. Graphene: Carbon in two dimensions. *Mater. Today* **2007**, *10*, 20–27.
10. Stankovich, S.; Dikin, D.A.; Piner, R.D.; Kohlhaas, K.A.; Kleinhammes, A.; Jia, Y.; Wu, Y.; Nguyen, S.T.; Ruoff, R.S. Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide. *Carbon* **2007**, *45*, 1558–1565.
11. Dreyer, D.R.; Park, S.; Bielawski, C.W.; Ruoff, R.S. The chemistry of graphene oxide. *Chem. Soc. Rev.* **2010**, *39*, 228–240.
12. Sanchez, F.; Sobolev, K. Nanotechnology in concrete—A review. *Constr. Build. Mater.* **2010**, *24*, 2060–2071.
13. Pan, Z.; He, L.; Qiu, L.; Korayem, A.H.; Li, G.; Zhu, J.W.; Collins, F.; Li, D.; Duan, W.H.; Wang, M.C. Mechanical properties and microstructure of a graphene oxide–cement composite. *Cem. Concr. Compos.* **2015**, *58*, 140–147.
14. Lv, S.; Ma, Y.; Qiu, C.; Sun, T.; Liu, J.; Zhou, Q. Effect of graphene oxide nanosheets of microstructure and mechanical properties of cement composites. *Constr. Build. Mater.* **2013**, *49*, 121–127.
15. Dimov, D.; Amit, I.; Gorrie, O.; Barnes, M.D.; Townsend, N.J.; Neves, A.I.S.; Withers, F.; Russo, S.; Craciun, M.F. Ultrahigh performance nanoengineered graphene–concrete composites for multifunctional applications. *Adv. Funct. Mater.* **2018**, *28*, 1705183.
16. Tong, T.; Fan, Z.; Liu, Q.; Wang, S.; Tan, S.; Yu, Q. Investigation of the effects of graphene and graphene oxide nanoplatelets on the micro- and macro-properties of cementitious materials. *Constr. Build. Mater.* **2016**, *106*, 102–114.
17. Xu, Y.; Zeng, J.; Chen, W.; Jin, R.; Li, B.; Pan, Z. A Holistic Review of Cement Composites Reinforced with Graphene Oxide. *Constr. Build. Mater.* **2018**, *171*, 291–302.

18. Shamsaei, E.; de Souza, F.B.; Yao, X.; Benhelal, E.; Akbari, A.; Duan, W. Graphene-based nanosheets for stronger and more durable concrete: A review. *Constr. Build. Mater.* **2018**, *183*, 642–660.
19. de Souza, F.B.; Yao, X.; Lin, J.; Naseem, Z.; Tang, Z.Q.; Hu, Y.; Gao, W.; Sagoe-Crentsil, K.; Duan, W. Effective strategies to realize high-performance graphene-reinforced cement composites. *Constr. Build. Mater.* **2022**, *324*, 126636.
20. Ge, Z.; Qin, J.; Sun, R.; Guan, Y.; Zhang, H.; Wang, Z. The Effect of the Addition of Graphene Nanoplatelets on the Selected Properties of Cementitious Composites. *Front. Built Environ.* **2021**, *7*, 673346.
21. Izhar, T.; Ahmad, S.A.; Mumtaz, N. Multiobjective design optimization of reinforced concrete beam coupled with market practice based constructability function using simulated annealing. *Asian J. Civ. Eng.* **2024**, *25*, 3901–3914.
22. Almasailam, F.; Purnell, P.; Black, L. Factors affecting the carbon footprint of reinforced concrete structures. *Mater. Struct.* **2025**, *58*, 116.
23. Luong, D.X.; Bets, K.V.; Algozeeb, W.A.; Stanford, M.G.; Kittrell, C.; Chen, W.; Salvatierra, R.V.; Ren, M.; McHugh, E.A.; Advincula, P.A.; et al. Gram-scale bottom-up flash graphene synthesis. *Nature* **2020**, *577*, 647–651.
24. Algozeeb, W.A.; Savas, P.E.; Luong, D.X.; Chen, W.; Kittrell, C.; Bhat, M.; Shahsavari, R.; Tour, J.M. Flash graphene from plastic waste. *ACS Nano* **2020**, *14*, 15595–15604.
25. Wyss, K.M.; De Kleine, R.D.; Couvreur, R.L.; Kiziltas, A.; Mielewski, D.F.; Tour, J.M. Upcycling end-of-life vehicle waste plastic into flash graphene. *Commun. Eng.* **2022**, *1*, 3.
26. Scrivener, K.L.; Crumbie, A.K.; Laugesen, P. The interfacial transition zone (ITZ) between cement paste and aggregate in concrete. *Interface Sci.* **2004**, *12*, 411–421.
27. Bentz, D.P.; Garboczi, J.; Stutzman, P.E.; Maso, J.C. *Computer Modelling of the Interfacial Zone in Concrete*; National Institute of Standards and Technology: Gaithersburg, MD, USA, 1992.
28. Zhang, P.; Wittmann, F.H.; Zhao, T.-J.; Lehmann, E.H.; Vontobel, P. Neutron radiography, a powerful method to determine time-dependent moisture distributions in concrete. *Nucl. Eng. Des.* **2011**, *241*, 4758–4766.
29. Geim, A.K.; Novoselov, K.S. The rise of graphene. *Nat. Mater.* **2007**, *6*, 183–191.
30. Novoselov, K.S.; Geim, A.K.; Morozov, S.V.; Jiang, D.; Zhang, Y.; Dubonos, S.V.; Grigorieva, I.V.; Firsov, A.A. Electric field effect in atomically thin carbon films. *Science* **2004**, *306*, 666–669.
31. Geim, A.K. Graphene: Status and prospects. *Science* **2009**, *324*, 1530–1534.
32. Hummers, W.S., Jr; Offeman, R.E. Preparation of graphitic oxide. *J. Am. Chem. Soc.* **1958**, *80*, 1339.
33. Marcano, D.C.; Kosynkin, D.V.; Berlin, J.M.; Sinitskii, A.; Sun, Z.; Slesarev, A.; Alemany, L.B.; Lu, W.; Tour, J.M. Improved synthesis of graphene oxide. *ACS Nano* **2010**, *4*, 4806–4814.
34. Lerf, A.; He, H.; Forster, M.; Klinowski, J. Structure of Graphite Oxide Revisited. *J. Phys. Chem. B* **1998**, *102*, 4477–4482.
35. Dikin, D.A.; Stankovich, S.; Zimney, E.J.; Piner, R.D.; Dommett, G.H.B.; Evmenenko, G.; Nguyen, S.T.; Ruoff, R.S. Preparation and characterization of graphene oxide paper. *Nature* **2007**, *448*, 457–460.
36. Stankovich, S.; Dikin, D.A.; Dommett, G.H.B.; Kohlhaas, K.M.; Zimney, E.J.; Stach, E.A.; Piner, R.D.; Nguyen, S.T. Graphene-based composite materials. *Nature* **2006**, *442*, 282–286.
37. Szabó, T.; Berkesi, O.; Forgó, P.; Josepovits, K.; Sanakis, Y.; Petridis, D.; Dékány, I. Evolution of surface functional groups in a series of progressively oxidized graphite oxides. *Chem. Mater.* **2006**, *18*, 2740–2749.
38. Compton, O.C.; Nguyen, S.B. Nguyen, Graphene oxide, highly reduced graphene oxide, and graphene: Versatile building blocks for carbon-based materials. *Small* **2010**, *6*, 711–723.
39. Fernández-Merino, M.J.; Guardia, L.; Paredes, J.I.; Villar-Rodil, S.; Solís-Fernández, P.; Martínez-Alonso, A.; Tascón, J.M.D. Vitamin C Is an Ideal Substitute for Hydrazine in the Reduction of Graphene Oxide Suspensions. *J. Phys. Chem. C* **2010**, *114*, 6426–6432.
40. Pei, S.; Cheng, H.M. The reduction of graphene oxide. *Carbon* **2012**, *50*, 3210–3228.
41. Bianco, A.; Cheng, H.-M.; Enoki, T.; Gogotsi, Y.; Hurt, R.H.; Koratkar, N.; Kyotani, T.; Monthieux, M.; Park, C.R.; Tascon, J.M.; et al. All in the graphene family—A recommended nomenclature for two-dimensional carbon materials. *Carbon* **2013**, *65*, 1–6.
42. Wick, P.; Louw-Gaume, A.E.; Kucki, M.; Krug, H.F.; Kostarelos, K.; Fadeel, B.; Dawson, K.A.; Salvati, A.; Vázquez, E.; Ballerini, L.; et al. Classification framework for graphene-based materials. *Angew. Chem. Int. Ed.* **2014**, *53*, 7714–7718.
43. Stoller, M.D.; Park, S.; Zhu, Y.; An, J.; Ruoff, R.S. Graphene-Based Ultracapacitors. *Nano Lett.* **2008**, *8*, 3498–3502.
44. Papageorgiou, D.G.; Kinloch, I.A.; Young, R.J. Mechanical properties of graphene and graphene-based nanocomposites. *Prog. Mater. Sci.* **2017**, *90*, 75–127.
45. Chen, D.; Feng, H.; Li, J. Graphene oxide: Preparation, functionalization, and electrochemical applications. *Chem. Rev.* **2012**, *112*, 6027–6053.

46. Hernandez, Y.; Nicolosi, V.; Lotya, M.; Blighe, F.M.; Sun, Z.; De, S.; McGovern, I.T.; Holland, B.; Byrne, M.; Gun'Ko, Y.K.; et al. High-yield production of graphene by liquid-phase exfoliation of graphite. *Nat. Nanotechnol.* **2008**, *3*, 563–568.
47. Coleman, J.N. Liquid exfoliation of defect-free graphene. *Acc. Chem. Res.* **2013**, *46*, 14–22.
48. Li, X.; Cai, W.; An, J.; Kim, S.; Nah, J.; Yang, D.; Piner, R.; Velamakanni, A.; Jung, I.; Tutuc, E.; et al. Large-area synthesis of high-quality and uniform graphene films on copper foils. *Science* **2009**, *324*, 1312–1314.
49. Reina, A.; Jia, X.; Ho, J.; Nezich, D.; Son, H.; Bulovic, V.; Dresselhaus, M.S.; Kong, J. Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition. *Nano Lett.* **2009**, *9*, 30–35.
50. Emtsev, K.V.; Bostwick, A.; Horn, K.; Jobst, J.; Kellogg, G.L.; Ley, L.; McChesney, J.L.; Ohta, T.; Reshanov, S.A.; Ruohrl, J.; et al. Towards wafer-size graphene layers by atmospheric pressure graphitization of silicon carbide. *Nat. Mater.* **2009**, *8*, 203–207.
51. Wyss, K.M.; Beckham, J.L.; Chen, W.; Luong, D.X.; Hundi, P.; Raghuraman, S.; Shahsavari, R.; Tour, J.M. Converting plastic waste pyrolysis ash into flash graphene. *Carbon* **2021**, *174*, 430–438.
52. Safian, M.T.-U.; Haron, U.S.; Ibrahim, M.N.M. A review on bio-based graphene derived from biomass wastes. *BioResources* **2020**, *15*, 9756–9785.
53. Titirici, M.-M.; White, R.J.; Falco, C.; Sevilla, M. Black perspectives for a green future: Hydrothermal carbons for environment protection and energy storage. *Energy Environ. Sci.* **2012**, *5*, 6796–6822.
54. Coleman, J.N.; Lotya, M.; O'Neill, A.; Bergin, S.D.; King, P.J.; Khan, U.; Young, K.; Gaucher, A.; De, S.; Smith, R.J.; et al. Two-dimensional nanosheets produced by liquid exfoliation of layered materials. *Science* **2011**, *331*, 568–571.
55. Zhan, P.; Xu, J.; Wang, J.; Zuo, J.; He, Z. A review of recycled aggregate concrete modified by nanosilica and graphene oxide: Materials, performances and mechanism. *J. Clean. Prod.* **2022**, *375*, 134116.
56. Lu, D.; Qu, F.; Zhao, H.; Li, N.; Han, S. A targeted approach of using graphene oxide in cement composites. *Constr. Build. Mater.* **2024**, *456*, 139339.
57. Zhao, L.; Zhu, S.; Wu, H.; Zhang, X.; Tao, Q.; Song, L.; Song, Y.; Guo, X. Deep research about the mechanisms of graphene oxide (GO) aggregation in alkaline cement pore solution. *Constr. Build. Mater.* **2020**, *247*, 118446.
58. Antolín-Rodríguez, A.; García-González, J.; Guerra-Romero, M.I.; Juan-Valdés, A. Performance improvements of recycled concrete and ceramic aggregates using graphene oxide nanocoating. *Sci. Rep.* **2026**, *16*, 12971.
59. Huang, C.-Y.; Lin, Y.-C.; Chung, J.H.Y.; Chiu, H.-Y.; Yeh, N.-L.; Chang, S.-J.; Chan, C.-H.; Shih, C.-C.; Chen, G.-Y. Enhancing Cementitious Composites with Functionalized Graphene Oxide-Based Materials: Surface Chemistry and Mechanisms. *Int. J. Mol. Sci.* **2023**, *24*, 10461.
60. Behunová, D.M.; Gallios, G.; Girman, V.; Kolev, H.; Kaňuchová, M.; Dolinská, S.; Václavíková, M. Electrophoretic Deposition of Graphene Oxide on Stainless Steel Substrate. *Nanomaterials* **2021**, *11*, 1779.
61. Yao, X.; Shamsaei, E.; Chen, S.; Zhang, Q.H.; de Souza, F.B.; Sagoe-Crentsil, K.; Duan, W. Graphene oxide-coated Poly(vinyl alcohol) fibers for enhanced fiber-reinforced cementitious composites. *Compos. Part B Eng.* **2019**, *174*, 107010.
62. Lu, Z.; Yao, J.; Leung, C.K. Using graphene oxide to strengthen the bond between PE fiber and matrix to improve the strain hardening behavior of SHCC. *Cem. Concr. Res.* **2019**, *126*, 105899.
63. Lu, Z.; Hanif, A.; Sun, G.; Liang, R.; Parthasarathy, P.; Li, Z. Highly dispersed graphene oxide electrodeposited carbon fiber reinforced cement-based materials with enhanced mechanical properties. *Cem. Concr. Compos.* **2018**, *87*, 220–228.
64. Gao, Y.; Li, G.; Chen, W.; Shi, X.; Gong, C.; Shao, Q.; Liu, Y. Graphene oxide coated fly ash for reinforcing dynamic tensile behaviours of cementitious composites. *Constr. Build. Mater.* **2024**, *411*, 134289.
65. Wang, Y.; Zheng, C.; Zhang, Y.; Su, J.; Zhang, H.; Hu, B.; Yuan, X. Effect of graphene-oxide coated sand on the properties of cement mortar. *Acta Mater. Compos. Sin.* **2025**, *42*, 3333–3344.
66. Lu, D.; Shi, X.; Wong, H.S.; Jiang, Z.; Zhong, J. Graphene coated sand for smart cement composites. *Constr. Build. Mater.* **2022**, *346*, 128313.
67. Srisindhu, J.; Divyah, N. State of Art Review on Variants of Graphene on Properties of Concrete. *Iran. J. Sci. Technol. Trans. Civ. Eng.* **2025**. <https://doi.org/10.1007/s40996-025-02037-4>.
68. Lin, Y.; Du, H. Graphene reinforced cement composites: A review. *Constr. Build. Mater.* **2020**, *265*, 120312.
69. Chuah, S.; Pan, Z.; Sanjayan, J.G.; Wang, C.M.; Duan, W.H. Nano reinforced cement and concrete composites and new perspective from graphene oxide. *Constr. Build. Mater.* **2014**, *73*, 113–124.
70. Samraj, R.; Pannem, R.M.R. A critical review on the effect of graphene oxide in fiber reinforced cementitious systems. *J. Dispers. Sci. Technol.* **2025**, *46*, 1563–1580.
71. Bagheri, A.; Negahban, E.; Asad, A.; Abbasi, H.A.; Raza, S.M. Graphene Oxide-Incorporated Cementitious Composites: A Thorough Investigation. *Mater. Adv.* **2022**, *3*, 9040–9051.

72. Nguyen, H.D.; Zhang, Q.; Sagoe-Crentsil, K.; Duan, W. Graphene oxide-coated sand for improving performance of cement composites. *Cem. Concr. Compos.* **2021**, *124*, 104279.
73. Murali, M.; Alaloul, W.S.; Mohammed, B.S.; Musarat, M.A.; Al Salaheen, M.; Al-Sabaei, A.M.; Isyaka, A. Utilizing graphene oxide in cementitious composites: A systematic review. *Case Stud. Constr. Mater.* **2022**, *17*, e01359.
74. Lu, Z.; Yu, J.; Yao, J.; Hou, D. Experimental and molecular modeling of polyethylene fiber/cement interface strengthened by graphene oxide. *Cem. Concr. Compos.* **2020**, *112*, 103676.
75. Fan, D.; Lue, L.; Yang, S. Molecular dynamics study of interfacial stress transfer in graphene-oxide cementitious composites. *Comput. Mater. Sci.* **2017**, *139*, 56–64.
76. Pu, Y.; Yang, S.; Qi, M.; Sheng, K.; Bi, J.; Fan, F.; Yuan, X. Synergistic effect of graphene oxide and hydroxylated graphene on the enhanced properties of cement composites. *RSC Adv.* **2022**, *12*, 26733–26743.
77. Zhao, L.; Guo, X.; Song, L.; Song, Y.; Dai, G.; Liu, J. An intensive review on the role of graphene oxide in cement-based materials. *Constr. Build. Mater.* **2020**, *241*, 117939.
78. Kong, X.; Qiao, W.; Ban, T.; Kang, R.; Huo, B.; Zhang, X. Experimental and molecular simulation of graphene oxide reinforced carbon fiber/cement matrix interface. *Acta Mater. Compos. Sin.* **2025**, *42*, 5216–5226.
79. Besra, L.; Liu, M. A review on fundamentals and applications of electrophoretic deposition (EPD). *Prog. Mater. Sci.* **2007**, *52*, 1–61.
80. Chen, J.; Zhao, D.; Ge, H.; Wang, J. Graphene oxide-deposited carbon fiber/cement composites for electromagnetic interference shielding application. *Constr. Build. Mater.* **2015**, *84*, 66–72.
81. Wu, D.; Zhang, F.; Liang, H.; Feng, X. Nanocomposites and macroscopic materials: Assembly of chemically modified graphene sheets. *Chem. Soc. Rev.* **2012**, *41*, 6160–6177.
82. Wang, S.; Wang, X.; He, J.; Xin, M. Mechanical behavior and microstructure of graphene oxide electrodeposited carbon fiber reinforced cement-based materials. *Crystals* **2022**, *12*, 964.
83. Corni, I.; Ryan, M.P.; Boccaccini, A.R. Electrophoretic deposition: From traditional ceramics to nanotechnology. *J. Eur. Ceram. Soc.* **2008**, *28*, 1353–1367.
84. Li, H.; Liebscher, M.; Curosu, I.; Choudhury, S.; Hempel, S.; Davoodabadi, M.; Dinh, T.T.; Yang, J.; Mechtcherine, V. Electrophoretic deposition of nano-silica onto carbon fiber surfaces for an improved bond strength with cementitious matrices. *Cem. Concr. Compos.* **2020**, *114*, 103777.
85. Li, Z.-Q.; Zhu, J.-H.; Pei, C. Enhanced interfacial bonding in cement composites through electrophoretic deposition of nano silica on carbon fibers. *Constr. Build. Mater.* **2024**, *435*, 136835.
86. Lee, W.; Lee, J.U.; Cha, H.-J.; Byun, J.-H. Partially reduced graphene oxide as a multi-functional sizing agent for carbon fiber composites by electrophoretic deposition. *RSC Adv.* **2013**, *3*, 25609–25613.
87. Chi, Y.; Chu, J.; Chen, M.; Li, C.; Mao, W.; Piao, M. Directly deposited graphene nanowalls on carbon fiber for improving the interface strength in composites. *Appl. Phys. Lett.* **2016**, *108*, 211601.
88. Novoselov, K.S.; Fal'Ko, V.I.; Colombo, L.; Gellert, P.R.; Schwab, M.G.; Kim, K. A roadmap for graphene. *Nature* **2012**, *490*, 192–200.
89. Chen, S.; Brown, L.; Levendorf, M.; Cai, W.; Ju, S.-Y.; Edgeworth, J.; Li, X.; Magnuson, C.W.; Velamakanni, A.; Piner, R.D.; et al. Oxidation resistance of graphene-coated Cu and Cu/Ni alloy. *ACS Nano* **2011**, *5*, 1321–1327.
90. Wang, X.; Li, C.; Chi, Y.; Piao, M.; Chu, J.; Zhang, H.; Li, Z.; Wei, W. Effect of Graphene Nanowall Size on the Interfacial Strength of Carbon Fiber Reinforced Composites. *Nanomaterials* **2018**, *8*, 414.
91. Saeed, M.; Alshammari, Y.; Majeed, S.A.; Al-Nasrallah, E. Chemical Vapour Deposition of Graphene—Synthesis, Characterisation, and Applications: A Review. *Molecules* **2020**, *25*, 3856.
92. Chen, Z.; Qi, Y.; Chen, X.; Zhang, Y.; Liu, Z. Direct CVD Growth of Graphene on Traditional Glass: Methods and Mechanisms. *Adv. Mater.* **2018**, *31*, e1803639.
93. Zhou, S.; Wei, W.; Li, L.; Luo, Y.; Xia, L.; Zhang, H.; Jia, Q.; Zeng, Z.; Yang, Z.; Wu, G. In situ growth of graphene on carbon fibers to enhance the mechanical and thermal conductivity of epoxy composites. *Appl. Surf. Sci.* **2024**, *680*, 161299.
94. Sai, T.Y.; Jagadeesh, P. Mechanical and microstructural properties of ultrafine slag cement mortar reinforced with graphene oxide nanosheets. *Carbon Lett.* **2023**, *33*, 1649–1660.
95. Yao, Y.; Zhang, Z.; Liu, H.; Zhuge, Y.; Zhang, D. A new in-situ growth strategy to achieve high performance graphene-based cement material. *Constr. Build. Mater.* **2022**, *335*, 127451.
96. Li, X.; Korayem, A.H.; Li, C.; Liu, Y.; He, H.; Sanjayan, J.G.; Duan, W.H. Incorporation of graphene oxide and silica fume into cement paste: A study of dispersion and compressive strength. *Constr. Build. Mater.* **2016**, *123*, 327–335.

97. Gao, Y.; Cheng, Z.; Yu, J.; Guo, X.; Liu, Y.; Chen, W. Mechanical Performance Enhancement of UHPC via ITZ Improvement Using Graphene Oxide-Coated Steel Fibers. *Cem. Concr. Compos.* **2025**, *157*, 105931.
98. Fu, W.; Li, X.; Wang, P.; Xu, T.; Zhou, J.; Liang, K.; Wang, X.; Liu, M. Improving the mechanical performance of concrete by graphene nanoplatelets (GNP) enrichment in the interfacial transition zone (ITZ). *J. Build. Eng.* **2024**, *90*, 109373.
99. Lu, D.; Wang, D.; Wang, Y.; Zhong, J. Nano-engineering the interfacial transition zone between recycled concrete aggregates and fresh paste with graphene oxide. *Constr. Build. Mater.* **2023**, *384*, 131244.
100. Gholampour, A.; Sofi, M.; Alipooramirabad, H.; Tang, Y. Performance of concrete containing pristine graphene-treated recycled concrete aggregates. *Resour. Conserv. Recycl.* **2023**, *199*, 107266.
101. Gokhale, D.G.; Kaish, A.B.A. Recent Trends in Incorporating Graphene Coated Sand in Self-Sensing Cementitious Composites. *Mater. Proc.* **2023**, *14*, 48. <https://doi.org/10.3390/IOC2023-14544>.
102. Wang, S.; Wang, J.; Yu, J.; Qian, W.; Yang, H.; Gao, Y.; Liu, Y. Recycled concrete powder surface modification via graphene oxide for high performance, low carbon and cost-effective cement paste development. *Environ. Res.* **2026**, *291*, 123584.
103. Rathnaswamy, S.; Jagadeesh, P. Enhancing the performance of recycled aggregate concrete using graphene oxide. *Front. Built Environ.* **2025**, *11*, 1740790.
104. Hu, Z.-Y.; Wan, Y.; Duan, Y.-J.; Shi, Y.-H.; Gu, C.-P.; Ma, R.; Dong, J.-J.; Cui, D. A Review of the Impact of Graphene Oxide on Cement Composites. *Nanomaterials* **2025**, *15*, 216.
105. Landović, A.; Čeh, A.; Starčev-Čurčin, A.; Šešljica, M. Small-Scale and Large-Scale Modeling of Fiber-Reinforced Concrete Girders. *Buildings* **2024**, *14*, 3812.
106. Accornero, F.; Rubino, A.; Marano, G.C.; Carpinteri, A. Smart construction of fibre-reinforced concrete structures: Size-scale effects on minimum reinforcement and plastic rotation capacity. *Smart Constr. Sustain. Cities* **2024**, *2*, 12.
107. Abushama, W.J.; Tamimi, A.K.; Tabsh, S.W.; El-Emam, M.M.; Ibrahim, A.; Ali, T.K.M. Influence of Optimum Particle Packing on the Macro and Micro Properties of Sustainable Concrete. *Sustainability* **2023**, *15*, 14331.
108. Singh, D.; Kumar, R.; Jha, P. A Comprehensive Review of Life Cycle Assessment for Supplementary Cementitious Materials in Sustainable Construction. In *Recent Advances in Sustainable Construction Technology, Geotechnics and Structural Engineering*; Springer: Singapore, 2026.
109. Luo, J.; Wang, Q.; Wang, L.; Fang, M. A Review of the Rheological Properties of 3D-Printed Concrete: Raw Materials, Printing Parameters, and Evolution Mechanisms. *Buildings* **2026**, *16*, 1264.
110. Vollpracht, A.; Lothenbach, B.; Snellings, R.; Haufe, J. The pore solution of blended cements: A review. *Mater. Struct.* **2016**, *49*, 3341–3367.
111. Antolín-Rodríguez, A.; Juan-Valdés, A.; Guerra-Romero, M.I.; Pozo, J.M.M.-D.; Krzywon, R.; Maravelaki, P.-N.; García-González, J. Mechanical Performance of Concrete with Graphene-Oxide-Treated Recycled Coarse Ceramic Aggregates: Effects on Aggregate Water Absorption and Workability. *Ceramics* **2025**, *8*, 104.
112. Baral, S.; Raghavendra, R.; Thomas, K.; Das, R. Dispersion of Graphene Oxide: Evaluating Ionic Surfactants for Nanocomposite Cement Applications. *Nanomaterials* **2026**, *16*, 632.
113. Qian, J.; Zhou, L.-Q.; Wang, X.; Yang, J.-P. Degradation of Mechanical Properties of Graphene Oxide Concrete under Sulfate Attack and Freeze–Thaw Cycle Environment. *Materials* **2023**, *16*, 6949.
114. Lu, D.; Qu, F.; Su, Y.; Cui, K. Nano-engineered the interfacial transition zone between recycled fine aggregates and paste with graphene oxide for sustainable cement composites. *Cem. Concr. Compos.* **2024**, *154*, 105762.
115. Yang, Q.; Wang, Z.; Chen, X.; Li, J. The Deterioration of Concrete Based on the Experiments Under the Combined Effects of Freeze-Thaw Cycles, Carbonation Erosion and Sulfate Corrosion. *Buildings* **2025**, *15*, 4179.
116. Yu, J.; Chen, Z.; Xu, X.; Su, X.; Liang, S.; Wang, Y.; Hong, J.; Zhang, S. Deep Learning-Assisted Analysis of GO-Reinforcing Effects on the Interfacial Transition Zone of CWRB. *Materials* **2024**, *17*, 5926.
117. Chen, Y.; Du, S. Improving the interfacial transition zone of concrete: A targeted approach utilizing graphene oxide. *Constr. Build. Mater.* **2025**, *466*, 140323.
118. Topsakal, M.; Sahin, H.; Ciraci, S. Graphene coatings: An efficient protection from oxidation. *Phys. Rev. B* **2012**, *85*, 155445.
119. Collazo, A.; Díaz, B.; Figueroa, R.; Nóvoa, X.; Pérez, C. Corrosion resistance of a water-borne resin doped with graphene derivatives applied on galvanized steel. *Prog. Org. Coatings* **2022**, *173*, 107220.
120. Zhang, H.; Gan, X.; Lu, Z.; Li, L.; Lu, L. Polycarboxylate Superplasticizer-Modified Graphene Oxide: Dispersion and Performance Enhancement in Cement Paste. *Nanomaterials* **2025**, *15*, 419.
121. Li, S.; Zhang, X.; Zhao, Y.; Jing, H.; Lyu, P.; Shan, J. Strength Degradation and Chloride Ion Diffusion Behavior of Concrete Under Simulated Marine Exposure. *J. Mar. Sci. Eng.* **2026**, *14*, 475.

122. Singh, L.; Karade, S.; Bhattacharyya, S.; Yousuf, M.; Ahalawat, S. Beneficial role of nanosilica in cement based materials—A review. *Constr. Build. Mater.* **2013**, *47*, 1069–1077.
123. Dong, S.; Li, L.; Ashour, A.; Dong, X.; Han, B. Self-assembled 0D/2D nano carbon materials engineered smart and multifunctional cement-based composites. *Constr. Build. Mater.* **2021**, *272*, 121632.
124. Han, B.; Yu, X.; Ou, J. *Self-Sensing Concrete in Smart Structures*; Butterworth-Heinemann: Oxford, UK; Elsevier: Kidlington, UK, 2014.
125. Mu, S.; Yue, J.; Wang, Y.; Feng, C. Electrical, Piezoresistive and Electromagnetic Properties of Graphene Reinforced Cement Composites: A Review. *Nanomaterials* **2021**, *11*, 3220.
126. Dong, W.; Li, W.; Zhu, X.; Sheng, D.; Shah, S.P. Multifunctional cementitious composites with integrated self-sensing and hydrophobic capacities toward smart structural health monitoring. *Cem. Concr. Compos.* **2021**, *118*, 103962.
127. Wu, T.; Sun, L.; Liu, X.; Liu, Y.; Yu, X. Piezoresistive performance of self-sensing cement-based composites filled with multi-layer graphene. *J. Sustain. Cem. Mater.* **2024**, *13*, 815–828.
128. Han, B.; Ding, S.; Yu, X. Intrinsic self-sensing concrete and structures: A review. *Measurement* **2015**, *59*, 110–128.
129. Azhari, F.; Banthia, N. Cement-based sensors with carbon fibers and carbon nanotubes for piezoresistive sensing. *Cem. Concr. Compos.* **2012**, *34*, 866–873.
130. Wen, S.; Chung, D. Electrical-resistance-based damage self-sensing in carbon fiber reinforced cement. *Carbon* **2007**, *45*, 710–716.
131. Irfan, M.S.; Ali, M.A.; Khan, K.A.; Umer, R.; Kanellopoulos, A.; Samad, Y.A. An electronic textile embedded smart cementitious composite. *Eng. Rep.* **2022**, *4*, e12468.
132. Triana-Camacho, D.A.; Quintero-Orozco, J.H.; Mejía-Ospino, E.; Castillo-López, G.; García-Macías, E. Piezoelectric composite cements: Towards the development of self-powered and self-diagnostic materials. *Cem. Concr. Compos.* **2023**, *139*, 105063.
133. Coppola, L.; Buoso, A.; Corazza, F. Electrical properties of carbon nanotubes cement composites for monitoring stress conditions in concrete structures. *Appl. Mech. Mater.* **2011**, *82*, 118–123.
134. Wen, S.; Chung, D. Electromagnetic interference shielding reaching 70 dB in steel fiber cement. *Cem. Concr. Res.* **2004**, *34*, 329–332.
135. Chen, M.; Yao, J.; Zhong, J.; Lu, D.; Ruan, W.; Xiao, H.; Sun, Y. Optimizing self-sensing performance of conductive mortar via gradation of graphene coated aggregate. *J. Build. Eng.* **2024**, *97*, 110766.
136. Barani, Z.; Kargar, F.; Godziszewski, K.; Rehman, A.; Yashchyshyn, Y.; Rumyantsev, S.; Cywiński, G.; Knap, W.; Balandin, A.A. Graphene epoxy-based composites as efficient electromagnetic absorbers in the extremely high-frequency band. *ACS Appl. Mater. Interfaces* **2020**, *12*, 28635–28644.
137. Birgin, H.B.; García-Macías, E.; D'Alessandro, A.; Ubertini, F. Self-powered weigh-in-motion system combining vibration energy harvesting and self-sensing composite pavements. *Constr. Build. Mater.* **2023**, *369*, 130538.
138. Han, B.; Sun, S.; Ding, S.; Zhang, L.; Yu, X.; Ou, J. Review of nanocarbon-engineered multifunctional cementitious composites. *Compos. Part A Appl. Sci. Manuf.* **2015**, *70*, 69–81.

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.