IRON-DECORATED GRAPHENE NANOPLATELETS PRODUCED BY ATMOSPHERIC-PRESSURE FERROCENE PYROLYSIS AS NANO-ADDITIVES FOR CEMENT MORTARS

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Abstract: *This study examines iron-based graphene nanoplatelets (Fe-GNPs), synthesized through a modified ferrocene pyrolysis route, as hybrid nano-additives for cement mortars. The nanomaterial was characterized by scanning electron microscopy, X-ray diffraction, Brunauer–Emmett–Teller surface area analysis, and thermogravimetric analysis, confirming a graphitic carbon network decorated with crystalline α-Fe nanoparticles. Mortars containing 0.001, 0.01, 0.05, and 0.1 wt.% Fe-GNPs, with and without surfactant-assisted ultrasonication, were tested in flexure and compression after 7 and 28 days of curing. The results showed a clear concentration-dependent response, with 0.01 wt.% yielding the highest strength gains. At 28 days, this dosage increased flexural strength by 14.5% without surfactant and by 21.2% with surfactant, while compressive strength rose by 17.8% and 22.5%, respectively, relative to the control mortar. Higher dosages reduced the reinforcing effect, mainly because agglomeration limited efficient stress transfer and matrix densification. The findings indicate that Fe-GNPs synthesized by a continuous atmospheric-pressure route are promising hybrid nano-additives for improving the mechanical performance of cement mortars, provided that dosage and dispersion are carefully controlled.*

Keywords: *iron-based graphene nanoplatelets, cement mortars, nano-reinforcement, dispersion, flexural strength, compressive strength.*

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Introduction

Cement mortars are among the most widely used construction materials due to their cost-effectiveness and relatively high compressive strength. Despite these advantages, conventional cement mortars exhibit comparatively low tensile and flexural strength and limited fracture toughness, making them susceptible to crack initiation and propagation under tensile stresses, shrinkage, and environmental loading. These microcracks can significantly compromise long-term durability by facilitating the ingress of water, chlorides, and other aggressive agents that accelerate deterioration processes such as corrosion and chemical attack [1].

To overcome these limitations, significant research efforts have focused on reinforcing cementitious materials by incorporating micro- and nanoscale additives. Traditional reinforcement approaches include steel fibers, polymer fibers, and other macro-scale fillers that improve crack resistance and energy absorption [2-4]. More recently, nanomaterials such as carbon nanotubes (CNTs), graphene oxide, graphene-based nanostructures, nanosilica, and metal or metal-oxide nanoparticles have attracted considerable attention due to their exceptionally high surface area, mechanical strength, and ability to interact with the cement hydration products [5-7]. These nanomaterials can act as nucleation sites for hydration, promote the formation of additional calcium silicate hydrate (C–S–H) phases, and refine the pore structure of the cement matrix. Each

class of nanomaterial contributes differently to mechanical enhancement, depending on its morphology, optimal concentration, and interaction with the cement hydration process.

In particular, CNTs and other carbon-based nanomaterials offer unique advantages due to their high aspect ratio and extremely high mechanical strength [8]. When properly dispersed within the cement matrix, they can enhance load transfer, bridge microcracks, and improve both mechanical and functional properties of cement-based composites. Consequently, integrating nanostructured reinforcements into cement mortars represents a promising strategy for developing next-generation, high-performance, multifunctional construction materials. In mortar matrices, the addition of CNTs has been shown to increase compressive strength by up to 35% and flexural strength by 28% compared to control mixes at optimal dosages (ranging from 0.02 to 0.5 wt.% of cement) under standard curing conditions [9]. Field-specific investigations also demonstrate peak enhancements in both compressive and flexural strengths at around 0.3 wt.% CNTs, with up to ~32–36% improvements after 28 days of curing [10].

Graphene nanoplatelets (GNPs), thin, plate-like stacks of graphene sheets with thicknesses of a few nanometers, provide a two-dimensional reinforcement morphology that can be even more effective under certain conditions. GNPs improve the mechanical properties of cementitious composites primarily through nanofiller effects, enhanced hydration, and crack bridging, thereby reducing porosity and creating a more compact microstructure [9,11].

Experimental studies have demonstrated that incorporating GNPs can significantly influence the mechanical performance of cementitious materials. For example, when GNPs are added at an optimal concentration of ~0.04 wt.% relative to cement, flexural strength has been reported to increase by up to 12.8%. Compressive strength increases by up to 33.9% after 28 days of curing compared with plain mortar [5]. In study [11], the addition of surface-modified GNPs at ~0.05 wt.% resulted in flexural strength increases of ~15-24% and compressive strength increases of ~3-8% in cement paste specimens. These findings highlight the strong dependence of reinforcement efficiency on nanoplatelet dispersion, surface modification, and concentration within the cement matrix.

The degree of strength enhancement depends strongly on the dispersion of GNPs, their concentration in the matrix, and their interactions with other components of the mortar. Excessively high concentrations of GNPs can lead to agglomeration, diminishing reinforcement efficiency, and potentially reducing the resulting strength gains. Effective dispersion methods, such as ultrasonication in the presence of surfactants or polymers (e.g., PVP), have been shown to significantly improve the uniformity of nanoplatelet distribution, thereby enhancing the mechanical performance of the composites [12,13].

In the present study, iron-based graphene nanoplatelets (Fe-GNPs) synthesized by a modified ferrocene pyrolysis route were evaluated as nano-reinforcements for cement mortars. Unlike conventional graphene-based additives, the present material combines graphitic nanoplatelets with iron nanoparticles generated via a single synthesis route, making it relevant from both functional and processing perspectives. The study focuses on two questions: first, whether Fe-GNPs can improve flexural and compressive performance at very low dosages; and second, how strongly the resulting behavior depends on dispersion quality. Special attention is therefore given to the combined influence of Fe-GNPs dosage and surfactant-assisted ultrasonication, as well as to the structural characterization of the synthesized nanomaterial. The aim is not only to identify an optimum dosage window, but also to clarify the practical conditions under which this hybrid additive can act efficiently in a cementitious matrix.

Materials and Methods

Raw materials

Ordinary Portland cement (OPC, CEM I 52.5 N) conforming to GOST 31108-2020 and supplied by Araratcement Factory (Yerevan, Armenia) was used as the binder. Its chemical composition and key physical

properties, determined in accordance with GOST EN 196-1-2002 to GOST EN 196-3-2002, are summarized in Table 1. These parameters govern hydration kinetics, setting behavior, and the mechanical response of the reference mortar against which the effect of Fe-GNPs incorporation is evaluated.

Table 1. Chemical composition and key physical properties of the cement used in this study

Characteristics	Units		Results Obtained
Al_2O_3	Content wt. %		3.11
SiO_2			24.4
Fe_2O_3			1.1
CaO			56.9
MgO			4.8
SO_3			2.7
Free CaO			1.2
Insol. Resid.			1.79
Loss of Ignition			4.0
Specific gravity	g/cm^3		3.1
Blaine's fineness	m^2/kg		430
Standard consistency	%		29
Setting time	min	Initial	50
		Final	300
Flexural strength	MPa	7 days	4.4
		28 days	4.8
Compressive strength	MPa	7 days	44.4
		28 days	51.1

DISPERBYK-199, a commercial wetting and dispersing agent, was used to improve nanoparticle dispersion in the aqueous phase before mixing. According to the manufacturer, the recommended dosage is 0.2–1.0 wt.% of the total binder content. In the present study, the dispersant amount was adjusted in proportion to the Fe-GNPs content to promote nanoparticle wetting and reduce agglomeration during ultrasonication. This step was introduced because uniform dispersion is essential for effective reinforcement, whereas poorly dispersed nanomaterials may behave as defects rather than as strengthening inclusions.

Natural river sand was used as the fine aggregate. Its main physical properties are listed in Table 2. Before mixing, the sand was washed to remove impurities and oven-dried at (105 ± 5)°C for 24 h to constant mass. These precautions were taken to reduce unintended variations in moisture content and effective water-to-cement ratio. The use of well-characterized, clean, and dry fine aggregate was necessary to ensure that the observed changes in mechanical performance could be interpreted primarily in relation to Fe-GNPs incorporation rather than variability in aggregate condition.

The selected raw materials provided a stable basis for mortar production. They ensured that the measured changes in mechanical performance could be attributed primarily to the incorporation of Fe-GNPs rather than to uncontrolled variation in the constituent materials.

Table 2. Key physical properties of the fine aggregate (natural river sand) used in mortar preparation are presented as average values

Characteristics		Units	Results Obtained
Specific Gravity		g/cm^3	2.54
Bulk density in the loose state		kg/m^3	1650
Bulk density in compact state		kg/m^3	1790
Sieve Residue			
		Partial	Total
Sieve size (mm)	2.5	13.80	13.80
	1.25	21.05	34.85
	0.63	28.14	60.99
	0.315	19.94	89.13
	0.16	10.58	99.71
Particle content of < 0.16 mm		0.29	
Fineness modulus		-	2.98

Synthesis of Fe–GNPs

Iron-based graphene nanoplatelets were synthesized by a modified ferrocene pyrolysis method using a two-zone horizontal tube furnace. Ferrocene served as the organometallic precursor, while nitrogen-carrying benzene vapor transported the precursor and provided an additional carbon source. The first heating zone was maintained at 180 °C to promote ferrocene sublimation, whereas the second zone was held at 1000 °C to induce thermal decomposition and formation of iron–carbon nanostructures. Compared with sealed high-pressure ferrocene pyrolysis, this atmospheric-pressure configuration offers better process continuity and greater practical relevance for scale-up. The presence of iron species formed during decomposition promotes catalytic graphitization, leading predominantly to graphene nanoplatelets together with a minor fraction of other carbon nanostructures (Fig. 1).

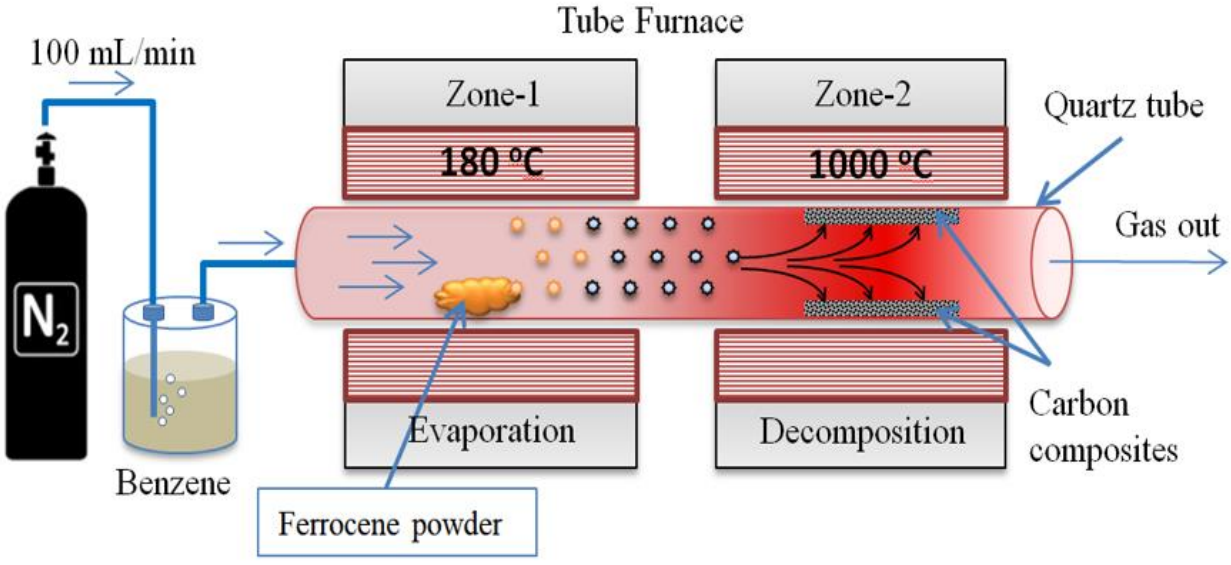


Fig. 1. *Schematic illustration of the ferrocene decomposition process and production of Fe–GNPs*

The first zone of the furnace is maintained at 180 °C to sublimate ferrocene, while the second zone reaches 1000 °C to induce thermal decomposition and formation of iron–carbon nanocomposites. Nitrogen flow

enriched with benzene transports the ferrocene vapors between the zones, promoting the growth of graphene nanoplatelets. This setup enables continuous operation, controlled particle morphology, and scalable production of Fe–GNPs.

Mixing and Specimen Preparation

Cement mortars incorporating Fe-GNPs were prepared by a controlled multi-step procedure designed to improve nanoparticle dispersion and mixture reproducibility. First, cement and sand were dry-mixed for 3 min to obtain a homogeneous powder blend. In parallel, the mixing water was ultrasonicated for 40 min using a Hielscher UP400St digital sonicator (400 W, 24 kHz) to facilitate nanoparticle deagglomeration and dispersion. For Fe-GNPs-containing mixtures, the nanomaterial was gradually introduced into the water during sonication. In mixtures containing DISPERBYK-199, the dispersant was added to the same aqueous phase before mixing with the dry constituents. The liquid phase was then introduced gradually into the cement–sand blend, followed by controlled mechanical mixing. This sequence was selected to improve nanoparticle distribution and to limit the persistence of coarse agglomerates in the fresh mortar. The detailed mixing proportions of all mortar compositions are presented in Table 3.

Table 3. Mix proportions of cement mortars with varying Fe-GNPs content, with and without dispersant

Mix design	Sand (g)	Cement (g)	W/C	Fe-GNPs (wt.% of cement)	Dispersant (Disperbuk-199 wt.% of cement)
Without DISPERBYK-199					
C0	2200	880	0.47	0	0
C1	2200	880	0.47	0.001	0
C2	2200	880	0.47	0.01	0
C3	2200	880	0.47	0.05	0
C4	2200	880	0.47	0.10	0
With DISPERBYK-199					
C0	2200	880	0.47	0	0
C1	2200	880	0.47	0.001	0.001
C2	2200	880	0.47	0.01	0.01
C3	2200	880	0.47	0.05	0.05
C4	2200	880	0.47	0.10	0.10

For each composition, six prismatic specimens (40×40×160 mm) were cast for mechanical testing at 7 and 28 days. The mortar was placed in two layers and compacted on a vibrating table (C278, Matest, Brugherio, Italy) to reduce entrapped air and improve packing density. After 24 h, the specimens were demolded and cured at (20 ± 2) °C and 98% relative humidity. This procedure was adopted to maintain consistent curing conditions across all mixtures and to ensure that differences in mechanical response reflected material design rather than curing variability (Fig. 2).

The diagram illustrates the sequential steps, including ultrasonic dispersion of nanoparticles, incorporation of dispersant, dry mixing of cement and sand, gradual addition of the liquid phase, and layerwise casting with compaction. This procedure ensures uniform nanoparticle distribution, improved workability, and reproducible mechanical performance of the resulting mortars.

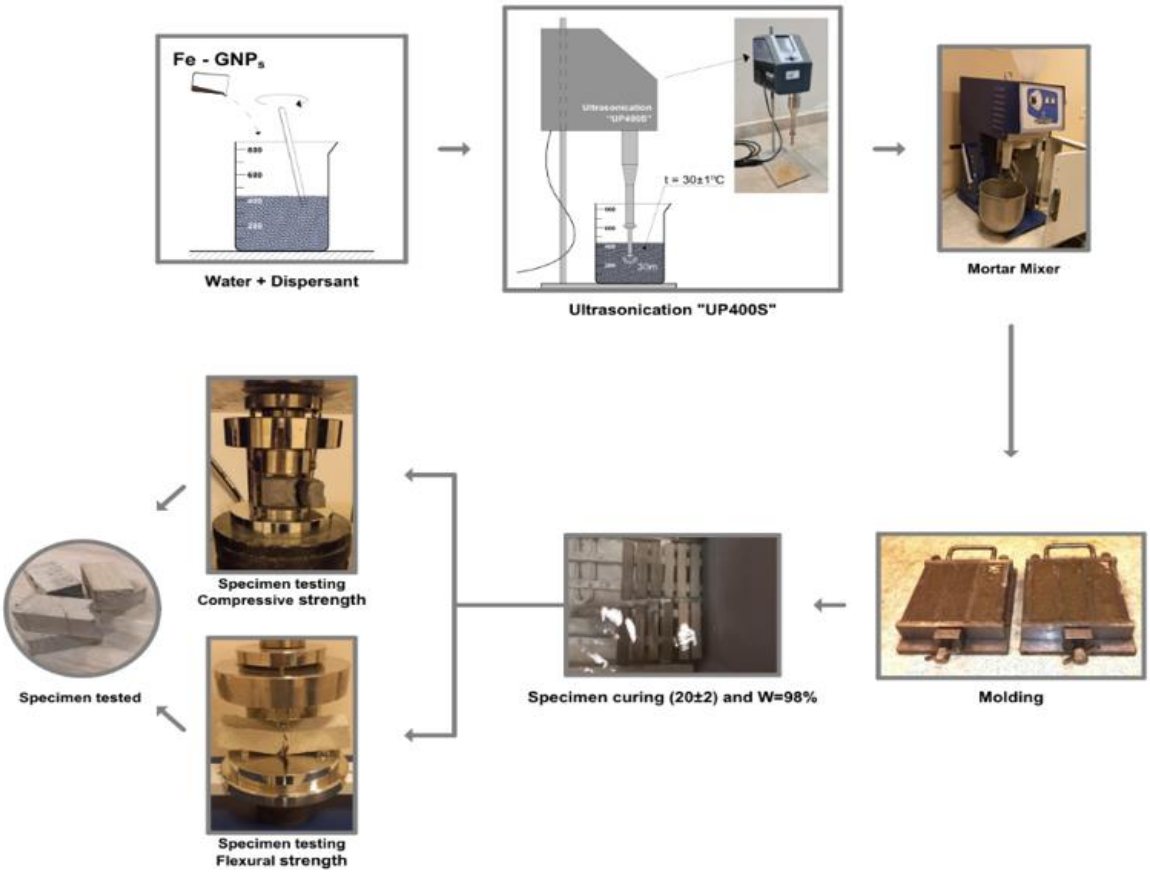


Fig. 2. *Schematic representation of the experimental procedure for Fe-GNPs-modified mortar mixtures*

Characterization Methods

Microstructural and Phase Analysis (SEM, XRD, TGA)

The synthesized Fe-GNPs were characterized for morphology, phase composition, and thermal behavior using scanning electron microscopy (SEM), X-ray diffraction (XRD), and thermogravimetric analysis (TGA/DSC). SEM observations were carried out on a LEO 1530 microscope operated at 30 kV with a field-emission gun, enabling direct visualization of graphene nanoplatelet morphology and the spatial distribution of iron nanoparticles. Phase analysis was performed using an X'PERT PRO diffractometer with Cu Kα radiation (λ = 1.5418 Å), enabling identification of the graphitic carbon and α-Fe phases present in the hybrid material.

Thermogravimetric analyses were performed on an SDT Q600 thermal analyzer to assess thermal stability and oxidation behavior. Approximately 11.4 mg of sample was heated from room temperature to 1000 °C at 20 °C min^{-1} under flowing air. The combined mass-loss and heat-flow data were used to distinguish carbon fractions of different structural order and to evaluate the contribution of the iron-containing phase during oxidation.

The specific surface area of the graphene-based nanomaterial was determined by nitrogen adsorption using a Micromeritics ASAP-2020M analyzer. The isotherms were interpreted using the Brunauer–Emmett–Teller method. Because surface area strongly affects interactions with hydration products and the nanomaterial's ability to act as a nucleation substrate, this parameter is relevant to interpreting subsequent mortar results.

Mechanical testing (Flexural and Compressive Strength)

The mechanical performance of the mortars was evaluated by flexural and compressive strength testing. Flexural strength was determined on 40×40×160 mm prismatic specimens using a Unitronic 50 kN machine

under three-point bending in accordance with EN 196-1-2002, clause 9.2. Compressive strength was measured on 40×40×40 mm specimens using a Matest 2000 kN automatic testing machine in accordance with EN 196-1-2002, clause 9.3. Six specimens were tested for each composition, and average values were used for comparison. This testing program was selected to capture the influence of Fe-GNPs incorporation on both crack-sensitive flexural behavior and bulk load-bearing capacity.

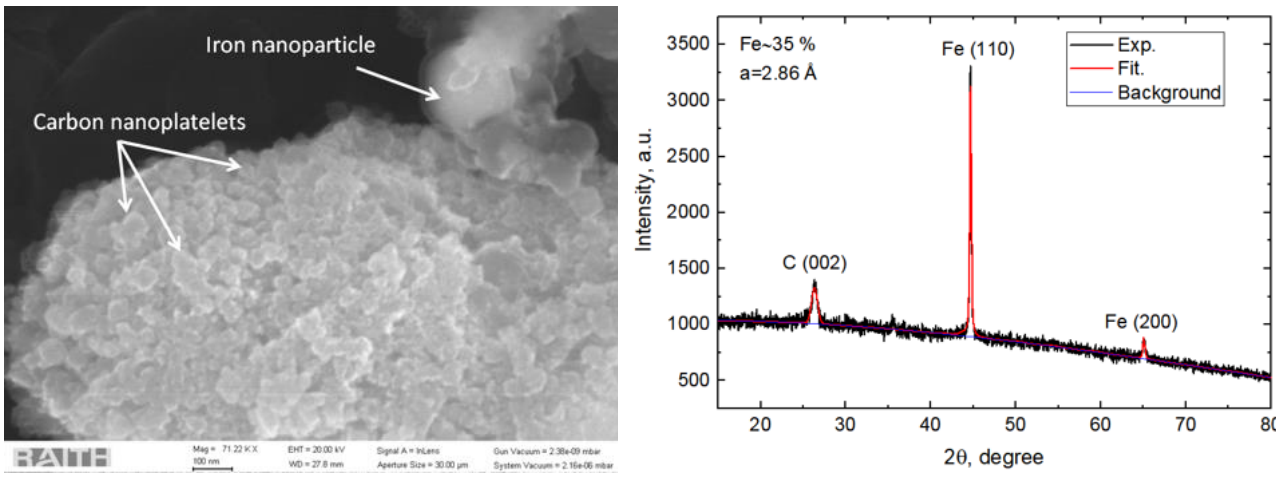
Results and Discussion

Structural Characterization of Fe-GNPs

Figure 3a shows the morphology of the synthesized Fe-GNPs. The SEM image reveals thin graphene nanoplatelets together with iron nanoparticles distributed across the carbonaceous matrix. In addition to platelet-like graphitic structures, a minor fraction of tubular carbon species is also visible, indicating that the pyrolysis process generated a mixed carbon nanostructure rather than a strictly single-morphology product. Such a hybrid architecture is relevant because platelet-like carbon structures can contribute to pore filling and crack deflection, whereas the iron particles may influence both graphitization during synthesis and interfacial behavior after incorporation into cementitious systems.

Figure 3b presents the XRD pattern of the synthesized nanocomposite. Rietveld refinement indicates the presence of two main phases: graphitic carbon and α-Fe. The reflection at $2\theta \approx 26°$ corresponds to the (002) planes of graphitic carbon, while the peaks near 45° and 65° are assigned to body-centered cubic α-Fe. The carbon reflection is relatively broad, which is consistent with limited stacking order and the presence of few-layer graphitic structures rather than bulk graphite. The sharpness of the iron peaks indicates that the metallic phase is comparatively well crystallized.

Quantitative phase analysis estimated the iron content of the nanocomposite to be approximately 35 wt.%, confirming the formation of a hybrid carbon–metal system. This result is important because the present additive is not a simple graphene powder modified after synthesis, but a material in which graphitic carbon and iron are generated together under the same pyrolytic conditions. That hybrid character is likely to influence both the powder's dispersion behavior and its interaction with the cementitious matrix.



a. **b.**

Fig. 3. **a.** *SEM micrograph*, **b.** *X-ray diffraction (XRD) pattern of Fe–GNPs*

The SEM and XRD results together confirm that the synthesized product is a hybrid nanocomposite composed predominantly of graphitic nanoplatelets and crystalline α-Fe nanoparticles, with a minor fraction of carbon nanotubes. This mixed nanostructure is potentially advantageous for cement reinforcement because it provides multiple interfaces for nucleation, stress transfer, and crack interaction.

Thermogravimetric analysis is widely used to assess the structural quality of graphene-based materials because the oxidation temperature depends strongly on graphitic order [14]. More ordered carbon structures generally oxidize at higher temperatures than defective or amorphous fractions. Accordingly, the TGA/DSC response of Fe-GNPs provides indirect information about the coexistence of carbon species with different structural organization in the synthesized material.

Figure 4 shows the TGA/DSC response of Fe-GNPs in air. The curves reveal a multi-step oxidation profile, indicating the coexistence of carbon species with varying degrees of structural order and iron-containing phases. Rather than a single oxidation event, the thermal behavior reflects the heterogeneous nature of the synthesized nanocomposite [14].

The first oxidation stage, observed at approximately 300–350 °C, is associated with the combustion of amorphous and highly defective carbon. A smaller mass change in the range of 400–500 °C, accompanied by an exothermic feature, can be linked to iron-containing regions that are less effectively protected by graphitic carbon. This behavior suggests that the metallic phase is not uniformly encapsulated throughout the sample, consistent with the morphology observed by SEM.

The dominant mass loss, about 52%, occurs between 500 and 700 °C and is accompanied by a sharp exothermic peak. This stage is assigned to the oxidation of the more ordered graphitic carbon fraction, including graphene nanoplatelets and related graphitized structures. Its higher oxidation temperature, relative to the defective carbon fraction, is consistent with stronger structural order and greater thermal stability.

Above approximately 780 °C, only a weak thermal feature is observed, with negligible additional mass loss. The final residue, around 52 wt.%, consists predominantly of iron oxides and is consistent with the iron content estimated from XRD. Overall, the thermal analysis supports the interpretation that the Fe-GNPs material is a heterogeneous hybrid system comprising graphitic carbon, more defective carbon domains, and iron-containing particles.

This behavior can be further explained by considering the mechanism of nanomaterial formation and subsequent interaction with the cement matrix. During synthesis, iron nanoparticles act as catalytic centers that promote the decomposition of ferrocene and the graphitization of carbon species, thereby forming graphene nanosheets and small carbon nanotubes [15,16]. Thus, the presence of α-Fe is directly related to the mechanism of hybrid nanostructure formation.

Second, after incorporation into the cement matrix, iron nanoparticles are embedded within the graphitic carbon network. These iron-containing particles may enhance interfacial interactions between the nanomaterial and hydration products and may serve as additional nucleation sites for the formation of calcium silicate hydrate (C–S–H), thereby contributing to matrix densification [16].

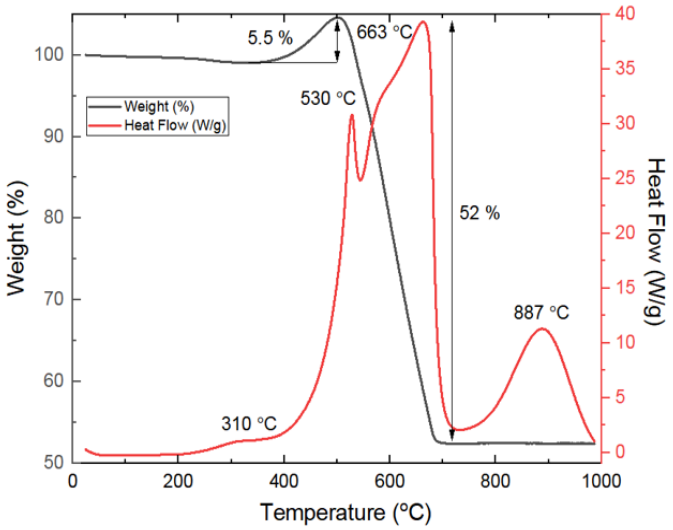


Fig. 4. *TGA and DSC curves of Fe-GNPs*

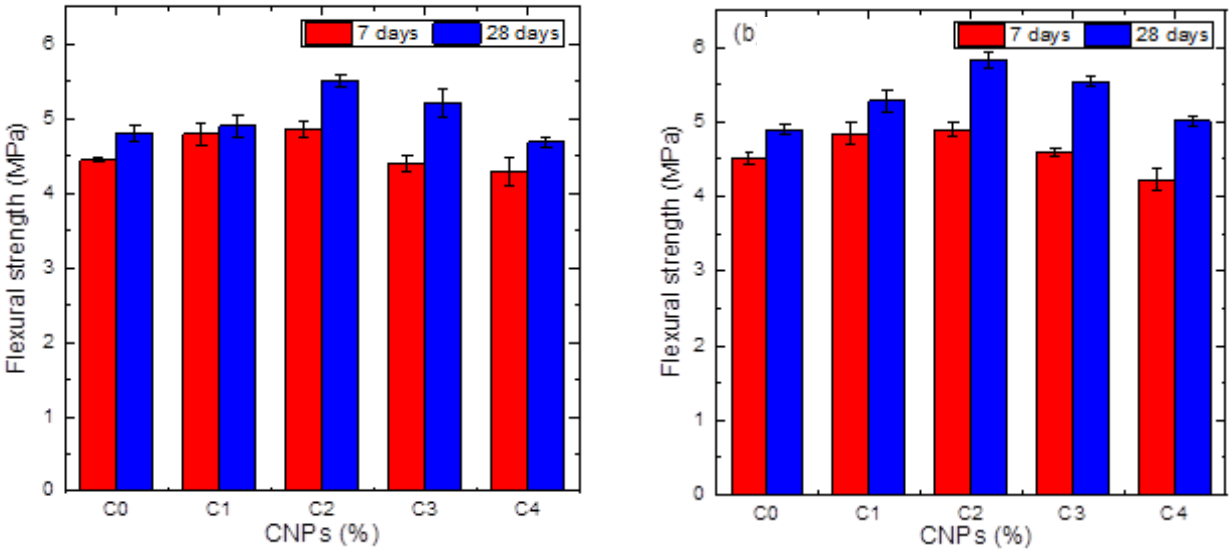
Flexural and Compressive Strength

The synthesized Fe-GNPs were incorporated into cement mortars as nano-additives to evaluate their influence on mechanical performance. Mortar compositions containing 0.001, 0.01, 0.05, and 0.1 wt.% Fe-GNPs relative to cement were prepared to identify a dosage range within which reinforcement

could be achieved without severe agglomeration. Flexural and compressive strengths were determined after 7 and 28 days of curing, and the role of surfactant-assisted dispersion was examined in parallel.

In addition to the Fe-GNPs dosage, the effect of the dispersing agent DISPERBYK-199 was also examined. Before mixing with cement, the nanocomposites were ultrasonically dispersed in water for 40 min to improve distribution within the fresh mortar. In the surfactant-containing mixtures, the dispersant was introduced at this stage to stabilize the suspension and reduce re-agglomeration. This comparative design makes it possible to distinguish between the effect of the nanomaterial itself and the effect of improved dispersion quality.

Figures 5 and 6 summarize the flexural and compressive strength results for mortars containing different Fe-GNPs dosages, with and without surfactant. The designations C0, C1, C2, C3, and C4 correspond to Fe-GNPs contents of 0, 0.001, 0.01, 0.05, and 0.1 wt.% of cement, respectively. The reference composition C0 serves as the baseline against which the effect of Fe-GNPs incorporation is assessed.



a. **b.**

Fig. 5. *Flexural strength of cement mortars containing different wt.% concentrations of graphene nanoplatelets (GNPs).* **a.** *Mixtures without surfactant,* **b.** *Mixtures with the addition of surfactant*

The results are presented for specimens cured for 7 and 28 days, illustrating the influence of nanoplatelet concentration and dispersion conditions on the flexural performance of the mortar composites. The comparison highlights the role of surfactant-assisted dispersion in improving the effectiveness of GNPs reinforcement.

The experimental results show that both flexural and compressive strengths increased with curing age for all mixtures, as expected from continued cement hydration. The incorporation of Fe-GNPs further improved mechanical performance, but the magnitude of the improvement depended strongly on dosage and dispersion condition. Among the investigated mixtures, composition C2 containing 0.01 wt.% Fe-GNPs produced the highest strength values at both curing ages. After 28 days, the flexural strength of C2 increased by 14.5% without surfactant and by 21.2% with surfactant relative to the control mortar. Under the same conditions, compressive strength increased by 17.8% without surfactant and by 22.5% with surfactant. These results indicate that a low Fe-GNPs dosage can be effective when the nanomaterial is sufficiently dispersed within the cementitious matrix.

When the Fe-GNPs dosage exceeded the optimum level, both flexural and compressive strengths decreased. This behavior is attributed mainly to nanoparticle agglomeration, which reduces the effective surface area available for interaction with hydration products and creates locally weak regions in the matrix. The results therefore underline that the reinforcing effect is governed not only by the presence of the nanomaterial but also by the balance between dosage and dispersion quality.

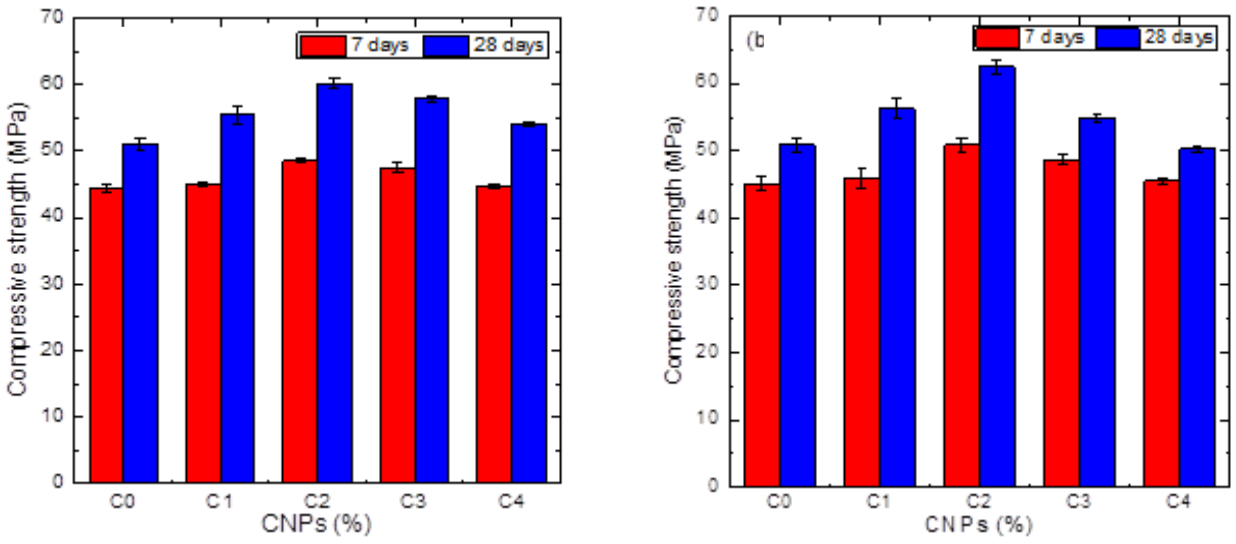


Fig. 6. *Compressive strength of cement mortars containing different wt.% concentrations of graphene nanoplatelets (GNPs).* **a.** *Mixtures without surfactant*, **b**. *Mixtures with the addition of surfactant*

The results correspond to specimens cured for 7 and 28 days and illustrate the influence of nanoplatelet concentration and dispersion conditions on the compressive performance of the mortar composites. The comparison highlights the role of surfactant-assisted dispersion in improving nanoparticle distribution and enhancing the load-bearing capacity of the cement matrix.

The strength enhancement observed in the optimized Fe-GNPs mixtures can be explained by several complementary mechanisms. First, the platelet-like carbon phase may act as a nanofiller, contributing to matrix densification and reducing fine voids [15-17]. Second, graphitic nanostructures can hinder the propagation of microcracks by promoting crack deflection and local bridging. Third, the high surface area of the Fe-GNPs system may facilitate the nucleation of hydration products, particularly in the vicinity of the dispersed nanomaterial [16,18]. These mechanisms should be understood as mutually reinforcing rather than mutually exclusive, and their relative contribution is expected to depend strongly on the quality of dispersion achieved in the fresh mixture [19,20].

The use of DISPERBYK-199 substantially improved nanoparticle dispersion and, consequently, the mechanical performance of the mortars. Better dispersion reduces the likelihood of coarse agglomerates acting as stress concentrators and allows a larger fraction of the Fe-GNPs surface to participate in matrix modification. The fact that the surfactant-bearing mixtures outperformed their unsurfacted counterparts at the same Fe-GNPs dosage supports the interpretation that dispersion quality is a decisive factor in the effectiveness of this nano-additive system [19,21].

At dosages above 0.01 wt.%, the reduction in strength indicates that the negative effect of agglomeration outweighs the potential benefits of additional nanomaterial [22-24]. This concentration-sensitive behavior is consistent with the broader literature on graphene-based cementitious composites, where reinforcement efficiency typically decreases beyond an optimal dosage window.

The structural characterization results help explain the mechanical response of the Fe-GNPs-modified mortars. The combined SEM, XRD, BET, and TGA/DSC analyses confirmed that the synthesized additive is a hybrid carbon–metal nanomaterial with graphitic nanoplatelets, crystalline α-Fe, and a high specific surface area. These features are compatible with the observed strengthening effect, particularly at low dosage and under improved dispersion conditions. At the same time, the present results suggest that structural quality alone is insufficient. Without adequate distribution within the cementitious phase, the potential benefit of the nanomaterial cannot be fully translated into mechanical gain.

Overall, the results demonstrate that Fe-GNPs can serve as effective nano-reinforcements for cement mortars when used at low dosage and with adequate dispersion. The combination of measurable strength enhancement, low optimum content, and an atmospheric-pressure synthesis route makes the system technically promising. However, further work on matrix-level microstructure and durability would be useful for broader practical validation.

Fe-GNPs: Scalable Synthesis for Cementitious Applications

In addition to mechanical performance, the practical relevance of the synthesis route is an important consideration for potential application in cement-based materials. From this perspective, the Fe-GNPs used in the present study were obtained via a modified ferrocene pyrolysis process at atmospheric pressure in a continuous two-zone furnace, which differs from more restrictive, closed, high-pressure routes [25].

Compared with sealed high-pressure solid-phase pyrolysis, the present synthesis route offers two practical advantages: continuous operation and a less demanding pressure regime. These features are relevant because they simplify process control and improve the prospects for scaling the synthesis beyond laboratory batch production. At the same time, the present study does not attempt a full techno-economic analysis; therefore, the practical significance of the route should be interpreted as indicative rather than definitive [19,26].

From a processing standpoint, the proposed route is attractive because it combines continuous operation with moderate process complexity. In addition, the optimal Fe-GNPs dosage identified in this study is low, which limits the amount of nanomaterial needed to achieve measurable mechanical improvement. This point is important in cementitious systems, where even technically effective nano-additives may become impractical at high required dosages [26].

The optimum incorporation level identified in this study, 0.01 wt.% of cement (C2 mix in Table 3), corresponds to approximately 0.1 kg of Fe-GNPs per ton of cement. In the present work, all mortar mixtures were prepared at a laboratory scale using 880 g of cement per batch. This confirms that only a small amount of additive is required to achieve a measurable strengthening effect under the investigated conditions.

Energy demand is also an important consideration. Atmospheric-pressure continuous systems avoid the use of reinforced high-pressure equipment and reduce the repeated heating–cooling cycles characteristic of strictly batch-based closed processes. For this reason, the proposed method may be more attractive for further scale-up studies than synthesis routes that rely on more restrictive pressure conditions [27,28].

Taken together, the low optimum dosage, the continuous atmospheric-pressure synthesis route, and the demonstrated mechanical benefits suggest that Fe-GNPs warrant further consideration as hybrid nano-additives for cementitious materials. However, any claim regarding large-scale implementation should remain provisional until supported by dedicated analyses of production, durability, and cost.

Comparative Assessment of Nano-Additives

A comparative evaluation of Fe-GNPs with other widely investigated nano-additives, particularly multi-walled carbon nanotubes (CNTs) and graphene oxide (GO), is useful for positioning the present results within the broader field of cement nanomodification. Such a comparison must be interpreted cautiously because performance reported in the literature depends strongly on matrix composition, water-to-cement ratio, dispersion protocol, curing conditions, and specimen geometry.

CNTs are well known for their crack-bridging ability and for their potential to improve compressive, flexural, and tensile performance [2,24,29]. However, their effectiveness strongly depends on dispersion, and relatively high dosages are often associated with loss of workability and agglomeration. These limitations do not diminish their importance, but they do highlight the practical challenge of consistently translating intrinsic nanomaterial properties into composite-level gains.

Graphene oxide is highly reactive due to its oxygen-containing functional groups and can enhance hydration kinetics and matrix refinement. At the same time, GO performance is sensitive to the synthesis route, degree

of oxidation, and dispersion protocol. In some systems, its strong chemical activity is beneficial; in others, it complicates reproducibility or control of the fresh state [30,31].

In the present study, the Fe-GNPs system achieved a compressive strength increase of about 22% at a low optimum dosage of 0.01 wt.% when surfactant-assisted dispersion was used. On this basis, Fe-GNPs may be viewed as a technically promising compromise between mechanical benefit, dosage efficiency, and synthesis practicality. The comparison does not suggest that Fe-GNPs are universally superior to CNTs or GO; rather, it indicates that they represent a competitive hybrid alternative worthy of further investigation [32-34].

The comparison in Table 4 should therefore be considered indicative rather than absolute. Its purpose is to position the present Fe-GNPs system within the current landscape of carbon-based nano-additives for cementitious composites, not to establish a definitive ranking across studies conducted under different experimental conditions.

Table 4. Comparative analysis of nano-additives used in cementitious composites, highlighting mechanical performance improvements, production complexity, and relative cost considerations

Nano-Additive	Typical Optimum Dosage (wt.%)	Compressive Strength Increase (%)	Production Complexity	Relative Cost Level	Key References
CNTs	0.2–0.3	30–35	High (CVD + purification)	High	[2,29]
GO	0.03–0.05	15–25	Moderate–High (chemical oxidation)	Moderate	[30,32]
Fe–GNPs (this work)	0.01	~22	Moderate (continuous pyrolysis, atmospheric pressure)	Low–Moderate	Present study

As shown in Table 3, CNTs- and GO-based systems can yield substantial performance gains, but the present Fe-GNPs system is notable for combining a low optimum dosage, meaningful mechanical improvement, and a synthesis route that is potentially compatible with continuous production. This combination is one of the main practical strengths of the present study [30,34].

Although the comparative assessment is encouraging, direct comparisons among nano-additive systems remain difficult because published studies vary in matrix design, curing conditions, dispersion strategies, and testing methodologies. Accordingly, broader validation of the Fe-GNPs concept should include durability, rheological behavior, and matrix-scale microstructural analysis in future work.

Conclusion

Iron-based graphene nanoplatelets (Fe-GNPs) were successfully synthesized by a modified ferrocene pyrolysis route operating under continuous atmospheric-pressure conditions. Structural characterization confirmed the formation of a hybrid graphitic carbon–iron nanomaterial composed of graphene-based nanoplatelets, crystalline α-Fe, and a minor fraction of additional carbon nanostructures.

When incorporated into cement mortars, Fe-GNPs produced a clear reinforcing effect that depended strongly on dosage and dispersion quality. Among the concentrations evaluated in this study, 0.01 wt.% of cement was identified as the optimal Fe-GNPs dosage, yielding the highest flexural and compressive strengths. The use of surfactant-assisted ultrasonication further increased the measured gains relative to the control mortar.

The observed improvement in mechanical performance is consistent with a combination of matrix densification, crack-growth restraint, and enhanced interaction between the nanomaterial and hydration products. At higher dosages, the reinforcing effect declined, indicating that agglomeration and dispersion limitations outweighed the potential benefits of additional Fe-GNPs content.

From a practical perspective, the study suggests that Fe-GNPs synthesized by an atmospheric-pressure route deserve further consideration as hybrid nano-additives for cementitious materials. Their low optimum dosage and measurable strengthening effect are encouraging, but broader validation should include hardened-matrix microstructure, durability-related properties, and process-scale assessment before making strong application claims.

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Conflict of Interest

The authors declare no conflicts of interest.

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