

Effect of a Pre-dispersed GO:PCE Suspension on Compressive Strength and Density of Cement Paste

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Abstract

This study examines whether a fixed preparation route using a pre-dispersed graphene oxide (GO) and polycarboxylate ether (PCE) suspension (GO:PCE suspension) produces a dosage-dependent compressive-strength response in cement pastes prepared with CEM I 42.5 N-SR0 and CEM II/B-LL 42.5 N, and whether apparent bulk density helps explain the resulting strength ranking. Graphene oxide (GO) was supplied as a 1 wt% aqueous dispersion (10 mg/mL) and combined with a polycarboxylate ether (PCE) at a fixed GO:PCE ratio of 1:5 by dry GO mass. After probe sonication, the prepared GO:PCE suspension was diluted with the mixing water to prepare pastes containing 0, 0.01, 0.03, and 0.05% GO by mass of cement. Compressive strength was measured at 7 and 28 days, and apparent bulk density was determined at 28 days. Both binders showed a non-linear response, with the 0.03% GO mixture giving the highest 28-day compressive strength, corresponding to increases of 23% for CEM I and 19% for CEM II/B-LL relative to the corresponding controls. Increasing the GO dosage to 0.05% did not provide a proportional additional benefit. Apparent bulk density showed only partial correspondence with strength, indicating that density alone could not explain the observed strength ranking.

Keywords

cement paste, compressive strength, graphene oxide (GO), polycarboxylate ether (PCE), Portland-limestone cement (PLC)

1 Introduction

Cement production remains a major source of process-related CO₂ emissions, which continues to drive the development of lower-clinker binders and more efficient use of cementitious constituents [1]. Under CEN EN 197-1:2011, Portland-limestone cements (PLC), including CEM II/A-LL and CEM II/B-LL, are classified as common cements, and their use is increasingly discussed in the context of reducing clinker content and lowering the embodied CO₂ of cement-based materials [1, 2]. However, the behavior of PLCs should not be interpreted as a simple diluted extension of CEM I. Limestone affects not only particle packing but also hydration chemistry. It can modify the hydrate assemblage, stabilize carboaluminate-bearing phases, and alter early hydration kinetics relative to plain Portland cement [3]. Subsequent studies on Portland-limestone systems have further shown that the magnitude of these effects depends on limestone content, clinker type, fineness, grinding, and the processing route used to prepare the binder [4–8].

Accordingly, performance trends established for admixtures or nano-additives in CEM I should not be transferred directly to CEM II/B-LL without explicit validation for the specific binder chemistry.

Graphene oxide (GO) has been studied in cementitious materials because of its sheet-like morphology, high specific surface area, and oxygen-containing functional groups, which can influence particle interactions, early hydration, and the evolution of the hardened microstructure [9, 10]. These characteristics make GO a plausible nanoscale modifier for cement paste, but the reported literature remains inconsistent. Improvements in strength and microstructure vary widely across studies, and the variability is large enough that GO should not be discussed as a universally beneficial additive. Instead, the response depends primarily on dosage, dispersion state, and the chemical environment into which GO is introduced [9, 10]. This dependence is especially evident in

cement paste. Several studies report that low GO dosages, typically in the range of a few hundredths of a percent by binder mass, can improve compressive strength when the nanosheets remain well dispersed [11, 12]. However, increasing the nominal GO dosage does not necessarily produce greater benefit. Beyond an optimal range, re-agglomeration may become more likely and fresh-state behavior may become less stable [12, 13]. Differences in dispersion state may therefore contribute to the divergent responses reported across nominally similar GO-modified cement paste studies [10]. A central technical difficulty is that GO is not intrinsically stable under the physicochemical conditions of fresh cement paste. The pore solution generated during hydration is strongly alkaline and contains substantial concentrations of dissolved alkalis and calcium ions [14]. Such an environment can compress electrostatic repulsion, alter surface interactions, and promote the re-agglomeration of GO sheets. This problem has been observed directly in alite paste, where GO tends to aggregate unless dispersion is maintained [15]. Complementary work on alkaline cement pore solution has further shown that Ca^{2+} plays a major role in destabilization and can intensify GO aggregation through ion-mediated interactions [16]. A related study comparing different water-reducing agents in cement pore solution found that GO modified with a polycarboxylate superplasticizer dispersed more effectively than GO treated with lignosulfonate- or naphthalene-based dispersants under the same alkaline conditions [17]. These findings indicate that dispersion should be treated as a controlled experimental variable rather than as a routine preparation step. For this reason, pre-dispersion using polycarboxylate ether (PCE) superplasticizers has become a practical and scientifically relevant strategy [18–20].

PCE modification of GO has been reported to improve stability in alkaline cementitious solutions through steric stabilization [18]. More recent studies show that the effectiveness of this approach depends on the GO:PCE ratio, the characteristics of the superplasticizer, and the applied mixing procedure [19, 20]. In cement paste, these factors can affect agglomerate size and repeatability and may, in turn, influence hydration behavior and mechanical performance. Therefore, the use of a GO:PCE suspension in the present study is not a generic admixture choice; it is a deliberate step intended to reduce uncontrolled agglomeration and improve the interpretability of GO-related effects. Binder chemistry adds a further level of complexity, since most mechanistic and performance-oriented GO studies have been performed in ordinary Portland cement systems [9–13, 15–20].

By contrast, direct comparative data remain limited for CEN EN 197-1:2011 [2] CEM I and CEM II/B-LL cement pastes prepared through the same pre-dispersed GO:PCE suspension route, with a fixed GO:PCE ratio and systematically varied dry-GO-equivalent dosage. The specific remaining gap is the lack of a dosage-resolved comparison showing whether one controlled GO:PCE suspension preparation route produces a comparable hardened-state response in these two binder chemistries under their respective reference-consistency conditions.

This gap matters because CEM II/B-LL differs from CEM I not only in clinker content but also in phase evolution, aluminate–carbonate interactions, and early-age chemical conditions [3–8]. These differences can influence both the stability of the prepared GO:PCE suspension and the hardening pathway into which it is introduced. Although recent work on ternary limestone clay cement paste indicates that GO can also improve the performance of lower-clinker, limestone-bearing systems, that binder is not equivalent to CEN EN 197-1:2011 [2] Portland-limestone cement and cannot be used as a direct substitute for CEM II/B-LL evidence [21]. A second, more limited objective of the present study is to examine whether apparent bulk density helps interpret the compressive-strength trend across mixtures. In this context, apparent bulk density is treated only as a practical indicator of external compactness. It cannot resolve pore-size distribution, pore connectivity, or local defects, and therefore cannot serve as a direct proxy for compressive strength [22].

Against this background, the present study addresses this gap by comparing cement pastes incorporating a pre-dispersed GO:PCE suspension in two binder systems: a sulfate-resistant CEM I 42.5 N-SR0 cement and a limestone-containing CEM II/B-LL 42.5 N cement. GO was introduced through a pre-dispersed GO:PCE suspension to reduce uncontrolled agglomeration and improve repeatability of the preparation route. Compressive strength was evaluated at 7 and 28 days, and apparent bulk density was measured at 28 days. The central research question was whether the same GO:PCE suspension preparation route produces a dosage-dependent strength response in both binder chemistries and whether density trends help interpret the corresponding strength ranking.

The comparison is therefore intended as a controlled performance assessment under a fixed preparation route rather than as a universal mechanistic model of GO action in cement paste.

2 Materials and methods

2.1 Materials

Two commercial cements were used as binders:

1. a sulfate-resistant Portland cement, CEM I 42.5 N-SR0, produced by Danucem at the Rohožník plant in Slovakia, and
2. a Portland-limestone cement, CEM II/B-LL 42.5 N, manufactured by Duna-Dráva Cement at the Vác plant in Hungary.

According to the manufacturer's product information, the CEM II/B-LL cement contained approximately 21% limestone and 5% fly ash. Graphene oxide (GO) was supplied by William Blythe Ltd. (UK) as an aqueous dispersion with a nominal concentration of 1 wt% (10 mg/mL). GO was used at dosages of 0, 0.01, 0.03, and 0.05% by mass of cement. A polycarboxylate ether (PCE) superplasticizer (MasterGlenium ACE 300) was used to prepare a pre-dispersed GO:PCE suspension prior to mixing. The fixed GO:PCE ratio was 1:5, expressed as dry GO mass per unit of as-received liquid PCE product mass. The physicochemical characteristics of GO, as reported by the supplier, are summarized in Tables 1 and 2. GO was introduced into the cement paste mixtures as a pre-dispersed GO:PCE suspension to reduce uncontrolled agglomeration and improve repeatability of the preparation route.

2.2 Methods

2.2.1 Preparation of (GO:PCE) suspension

To reduce the risk of GO agglomeration upon exposure to alkaline, Ca^{2+} -bearing cement pore solution, GO was introduced as a pre-dispersed GO:PCE suspension. The GO:PCE suspension preparation workflow is shown in Fig. 1. The required amount of PCE (MasterGlenium ACE 300) was added to the as-received GO dispersion (step 1), and the mixture was homogenized by magnetic stirring for 10 min at 40 °C (step 2). The mixture was then subjected to probe sonication using a finger-type sonicator (Hielscher UP100H, 100 W, 30 kHz) at 100% amplitude in pulsed mode (1 s on/1 s off) for 30 min to limit excessive heating during dispersion (step 3).

Table 1 Physical Properties of GO (Values reported by the supplier)

Material	Appearance	Concentration	Sheet size	pH
Graphene Oxide (GO)	orange-brown aqueous dispersion	1 wt% (10 mg/mL)	$>5 \mu\text{m}$	> 2

Table 2 Elemental Analysis of GO (Values reported by the supplier)

Material	Oxygen	Carbon	Nitrogen	Sulfur	Trace metals
GO	$> 30\%$	60–70%	$< 1\%$	$< 2\%$	$< 0.1\%$

After sonication, the GO:PCE suspension was magnetically stirred for an additional 10 min at 40 °C and used immediately for paste preparation (step 4). Before cement addition, the prepared GO:PCE suspension was diluted into the mixing water to improve its distribution in the paste mixture (step 5). The same GO:PCE suspension preparation route was used across all GO dosages to maintain a constant preparation method. This control is important because PCE-based treatment improves GO stability in cement-like alkaline media but does not completely prevent calcium-driven re-agglomeration [18, 23]. In addition, dispersion quality remains sensitive to superplasticizer characteristics and mixing procedures [19, 20, 23–25].

Therefore, the present work interprets dosage effects only within the fixed GO:PCE ratio and sonication protocol used here, rather than treating nominal GO dosage as an independent variable.

2.2.2 GO-modified cement paste preparation

Eight cement paste mixtures were prepared using two cement types and four graphene oxide (GO) dosage levels, as summarized in Table 3. The GO-free reference pastes were designated R0 for CEM I and B0 for CEM II/B-LL. The GO-modified pastes were designated RG1, RG3, RG5 and BG1, BG3, BG5, corresponding to GO dosages of 0.01%, 0.03%, and 0.05% by cement mass, respectively. Mixing was carried out using a Hobart-type mixer following a procedure adapted from the mixing regime specified in CEN EN 196-1:2016 [26]. The water-to-cement ratio (w/c) for each cement series was determined separately by Vicat standard-consistency testing of the corresponding GO-free reference mixture, R0 or B0, according to CEN EN 196-3:2016 [27]. The w/c ratio required to achieve standard consistency depends on the cement's composition and fineness. The resulting ratios were 0.332 for the R series and 0.315 for the B series, and these values were retained unchanged across all mixtures within each cement series.

For the GO-modified mixtures, the 1 wt% aqueous GO dispersion was combined with polycarboxylate ether (PCE; MasterGlenium ACE 300) to prepare the GO:PCE suspension. The reported GO dosages represent dry GO equivalent mass relative to cement mass. The fixed GO:PCE ratio of 1:5 refers to the dry GO mass contained in the GO dispersion relative to the mass of PCE as supplied. PCE was introduced exclusively through the prepared GO:PCE suspension, and no additional PCE was added during paste mixing. The prepared GO:PCE suspension was diluted into the mixing water and stirred for 10 min before cement addition. For the GO-free reference

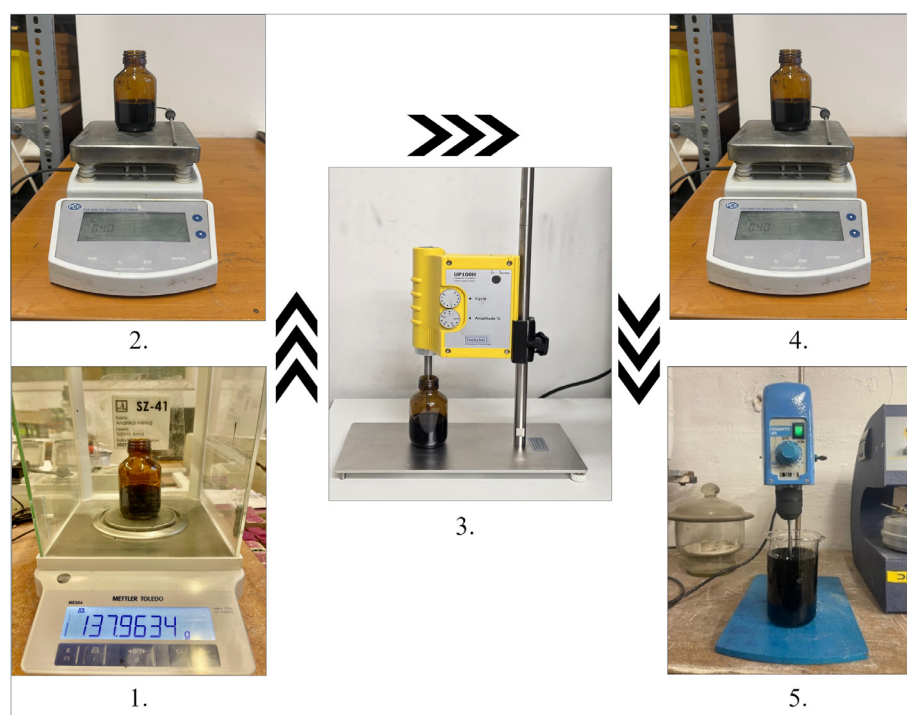


Fig. 1 GO:PCE suspension preparation workflow: 1. weighing/dosing; 2. initial magnetic stirring after combining GO dispersion and PCE; 3. probe sonication; 4. re-stirring after sonication; 5. dilution of the prepared GO:PCE suspension into the mixing water before paste preparation

Table 3 Mixture matrix and specimen identifiers

Series (Cement type; w/c)	GO 0%	GO 0.01%	GO 0.03%	GO 0.05%
R (CEM I; 0.332)	R0	RG1	RG3	RG5
B (CEM II/B-LL; 0.315)	B0	BG1	BG3	BG5

mixtures, R0 and B0, PCE (MasterGlenium ACE 300) was added directly to the mixing water. The PCE dosage in the GO-free reference mixtures was not matched to the PCE amount contained in the corresponding GO:PCE suspensions, and was adjusted using a flow-table procedure adapted from MSZ EN 12350-5:2009 [28] to obtain the selected semi-plastic spread range of approximately 18–20 cm. The flow-table readings were used to guide mixture preparation and a practical pre-casting consistency-control step to maintain comparable handling conditions within each cement series. Accordingly, the present comparison evaluates the performance response of cement pastes incorporating a pre-dispersed GO:PCE suspension at different GO dosages, rather than the isolated contribution of GO at a constant PCE dosage. The cement was then added gradually to the liquid phase. Wet mixing was performed at low speed for 2 min, followed by a brief pause to scrape the bowl, and then at high speed for an additional 2 min. After a short rest period, the fresh paste was cast into molds and compacted on a vibrating table to reduce entrapped air. The specimens were cured at 20 ± 2 °C

for 24 h. After demolding, the specimens were stored in sealed containers lined with wet towels. The containers were kept in a climate-controlled room at 20 ± 2 °C and 65% relative humidity until testing at 7 and 28 days.

2.2.3 Compressive strength examination

The compressive strength of the cement pastes was determined at 7 and 28 days. Each mixture was prepared as a single paste batch, from which separate sets of $30 \times 30 \times 30$ mm cube specimens were cast and tested destructively at each age, with five cubes tested per age. Compressive-strength results were analyzed separately for each cement series and test age. Within each R-series or B-series age set, overall differences among the four mixtures were assessed using a Welch one-way analysis of variance (ANOVA), which accommodates unequal within-group variances. Games-Howell post-hoc comparisons were subsequently used to compare each GO-modified mixture with its corresponding control mixture (R0 or B0). Statistical significance was considered at $p < 0.05$. Five cube-strength values were tested at each age for each mixture. As each mixture was prepared from a single paste batch, the analysis describes specimen-level variation among the tested cubes from that batch. Compression testing was performed using a hydraulic compression testing machine at a loading rate of 2400 ± 200 N/s, as shown in Fig. 2. This loading regime was applied consistently to all specimens.

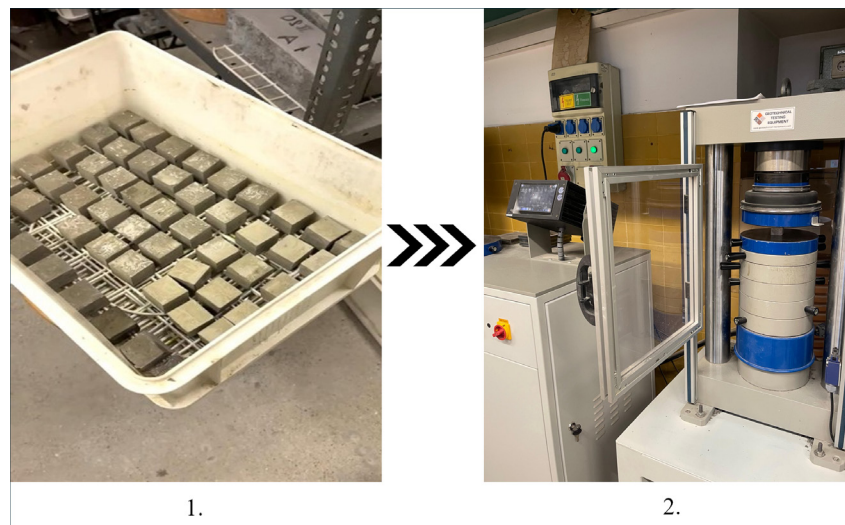


Fig. 2 Compressive strength testing process: 1. cement paste cubes ($30 \times 30 \times 30$ mm); 2. hydraulic compression testing machine

Apparent bulk density was determined at 28 days on the same cube specimens used for compressive strength testing, immediately before compression. The specimens were removed from the sealed containers immediately before testing. Visible surface moisture was gently wiped off, after which the specimen mass and external dimensions were measured. Apparent bulk density was calculated as the specimen mass divided by its geometric external volume. Accordingly, the reported values represent an as-conditioned moist state with visible free water removed; they do not represent oven-dry density, saturated density, or a standardized surface-dry condition. Density and compressive strength were therefore compared using specimens in the same conditioning state.

3 Results and discussion

3.1 Compressive strength analysis

The incorporation of the pre-dispersed GO:PCE suspension produced a non-linear compressive-strength response in both binder series, with the highest mean response observed at a dosage of 0.03% (Fig. 3). In the R series (CEM I), the mean compressive strength increased from 80 MPa for R0 to 93 MPa for RG3 at 7 days and from 92 MPa for R0 to 113 MPa for RG3 at 28 days, corresponding to increases of 16% and 23%, respectively. Increasing the dosage to 0.05% did not provide an additional benefit; RG5 reached 102 MPa at 28 days and remained below RG3. In the B series (CEM II/B-LL), the same non-linear dosage-response pattern was observed. At 7 days, the mean compressive strength increased from 69 MPa for B0 to 79 MPa for BG3, corresponding to a 15% increase. At 28 days, it increased from 90 MPa for B0 to 107 MPa for BG3, corresponding to a

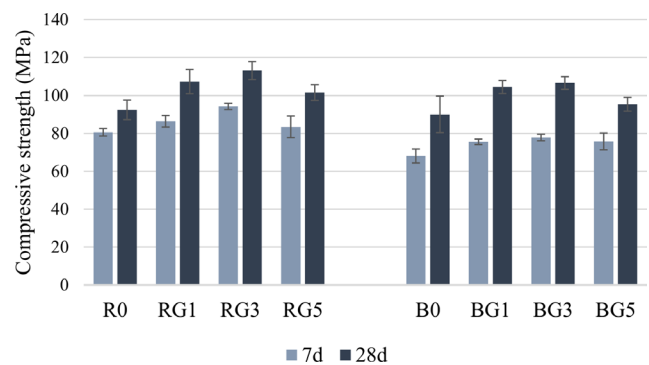


Fig. 3 Compressive strength at 7 and 28 days for CEM I (R-series) and CEM II/B-LL (B-series) pastes with GO dosages of 0.01%, 0.03%, and 0.05% by cement mass (controls: R0 and B0). Values are reported as mean $\pm$ standard deviation (SD) for five cube specimens tested at each age from one paste batch per mixture.

19% increase. Increasing the dosage to 0.05% again did not improve performance, with BG5 remaining below BG3.

Because the water-to-cement ratios of the R and B series were selected separately by applying the same Vicat standard-consistency criterion to R0 and B0, the absolute strengths of the two series are not interpreted as a direct binder-to-binder strength comparison. Instead, the effect of the GO dosage within the fixed GO:PCE suspension route is evaluated within each cement series relative to its corresponding GO-free control.

Fig. 4 presents the corresponding relative changes with respect to the appropriate controls (RG mixtures versus R0; BG mixtures versus B0). These within-series relative changes provide the appropriate basis for comparing the dosage-response pattern under the respective reference-consistency conditions and show the highest measured relative gains at the 0.03% dry-GO-equivalent dosage at both ages.

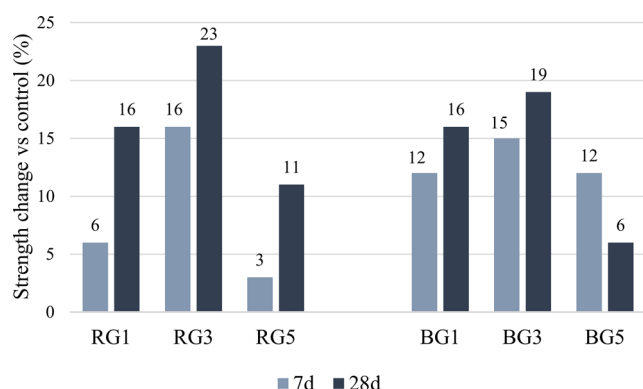


Fig. 4 Relative change in compressive strength at 7 and 28 days with respect to the corresponding controls (RG mixtures versus R0; BG mixtures versus B0), calculated from mixture mean values. Each mean was based on five cube specimens tested at each age from one paste batch per mixture.

To assess whether the observed differences in compressive strength exceeded specimen-level variability, the results were analyzed separately for each cement series and testing age using Welch's one-way ANOVA, followed by Games–Howell post hoc comparisons with the corresponding control mixture (Table 4). Overall differences among the four mixtures were indicated in the R series at 7 and 28 days and in the B series at 7 and 28 days. In the R series, RG1 and RG3 differed significantly from R0 at both ages, whereas RG5 did not differ significantly from R0 at either age. In the B series at 7 days, BG1 and BG3 differed significantly from B0, while BG5 did not. At 28 days, although the overall analysis indicated differences among the B-series mixtures, none of the

individual Games–Howell comparisons with B0 reached the $p < 0.05$ threshold; BG3 showed the closest comparison ($p = 0.053$). Overall, the statistical results are consistent with the observed non-linear dosage-dependent response, with 0.03% GO providing the highest observed mean compressive strength in both binder series.

In the literature, such improvements are often associated with improved dispersion, hydration-related effects, and microstructural refinement when GO remains effectively distributed in the cement system [9–11]. Gong et al. [11] reported substantial gains in compressive strength in Portland cement paste at a GO dosage of approximately 0.03%, providing a relevant low-dose comparison with the highest observed mean response at 0.03% in the present study (RG3 and BG3). Reduced performance at higher nominal GO dosages has also been reported in some cementitious systems [12, 13]. In alkaline, Ca^{2+} -rich environments, GO aggregation has been reported [15, 16]; however, aggregation was not measured in the present pastes. Limestone-containing binders differ from ordinary Portland cement because limestone can modify early hydration and hydrate assemblage, including stabilization of carboaluminate phases [3]. These changes may influence early-age microstructure development and, therefore, the additional benefit obtainable from nanoscale additives such as GO. In low-clinker limestone-bearing systems, recent work has shown that improved GO dispersion can promote early hydration and mechanical performance, while also affecting carbonation behavior [29]. Studies of

Table 4 Compressive-strength summary and statistical comparison of R- and B-series cement pastes at 7 and 28 days

Series	Age (d)	Mixture	Compressive strength, MPa (mean $\pm$ SD)	Change vs. corresponding control (%)	Overall Welch ANOVA p -value	Games–Howell post-hoc p -value vs. corresponding control
R	7	R0	80.57 $\pm$ 2.02	–	<0.001	–
		RG1	86.42 $\pm$ 3.09	+7.26		0.038
		RG3	94.25 $\pm$ 1.65	+16.98		<0.001
		RG5	83.45 $\pm$ 5.72	+3.57		0.725
R	28	R0	92.38 $\pm$ 5.14	–	0.001	–
		RG1	107.27 $\pm$ 6.40	+16.12		0.017
		RG3	113.16 $\pm$ 4.74	+22.49		<0.001
		RG5	101.57 $\pm$ 4.21	+9.94		0.061
B	7	B0	68.03 $\pm$ 3.68	–	0.008	–
		BG1	75.55 $\pm$ 1.39	+11.07		0.027
		BG3	77.82 $\pm$ 1.77	+14.39		0.008
		BG5	75.71 $\pm$ 4.46	+11.29		0.071
B	28	B0	89.98 $\pm$ 9.63	–	0.003	–
		BG1	104.43 $\pm$ 3.42	+16.05		0.086
		BG3	106.56 $\pm$ 3.34	+18.42		0.053
		BG5	95.23 $\pm$ 3.69	+5.83		0.685

GO modified with water-reducing agents and PCE stabilization provide related evidence that admixture choice can influence the stability of GO dispersions under cement-pore-resolution conditions [17, 18]. These literature-reported mechanisms provide context for interpreting the observed non-linear strength response.

Note: Values are mean $\pm$ standard deviation for five cube specimens tested at each age. Within each cement-series/age set, overall differences among the four mixtures were assessed using Welch one-way analysis of variance (ANOVA). Games–Howell post-hoc comparisons were subsequently performed among all mixtures within each set; only comparisons of GO-modified mixtures with their corresponding control are reported here. Bold p-values indicate statistical significance at $p < 0.05$. Each mixture was prepared as one paste batch; therefore, the analysis describes specimen-level variability among the tested cubes.

Taken together, all GO-containing mixtures showed higher observed mean compressive strengths than their corresponding controls, although the statistical support for individual control-versus-GO comparisons varied with binder series, age, and dosage. Within the tested dosage range, the highest observed strength in both binder series occurred at the 0.03% dry-GO-equivalent dosage. Similar non-monotonic responses, including optimum performance at low GO dosage, have been reported in cement paste and mortar [11–13, 30]. Review literature further indicates that the reported optimum may vary with binder chemistry, GO:PCE suspension preparation route, and test conditions [31]. The influence of dispersion state on the observed strengthening response has been examined directly in prior work [32]. Studies have also investigated hydration-related and microstructural responses of GO-modified cementitious systems [33, 34].

3.2 Compressive strength and apparent bulk density

Figs. 5 and 6 present the relationship between 28-day compressive strength and 28-day apparent bulk density, shown separately for the two binder systems: the R-series (CEM I) in Fig. 5 and the B-series (CEM II/B-LL) in Fig. 6. The mean apparent bulk density of each mixture was compared with the corresponding mean 28-day compressive strength ($n = 5$ cubes per mixture). Across the plotted mixture means, apparent bulk density ranged from 2069 to 2111 kg/m³ in the R-series and from 2030 to 2060 kg/m³ in the B-series, while mean compressive strength ranged from 92.4 to 113.2 MPa in the R-series and from 90 to 107 MPa in the B-series.

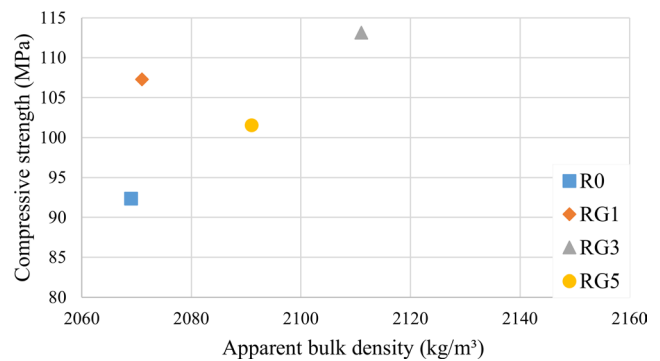


Fig. 5 Relationship between mean 28-day compressive strength and mean apparent bulk density of R-series cement pastes (R0, RG1, RG3, and RG5). Each marker represents a mixture mean based on five cube specimens measured immediately before compression

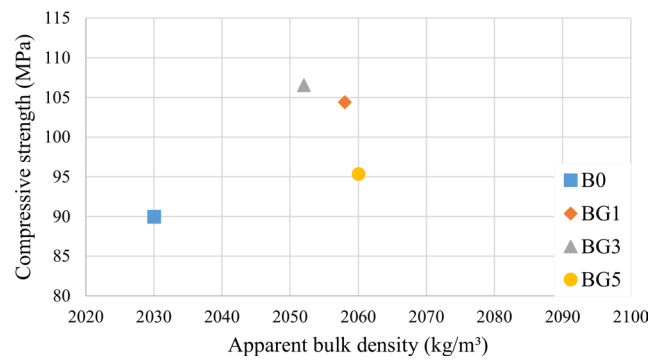


Fig. 6 Relationship between mean 28-day compressive strength and mean apparent bulk density of B-series cement pastes (B0, BG1, BG3, and BG5). Each marker represents a mixture mean based on five cube specimens measured immediately before compression

Within each binder series, higher apparent bulk density generally tended to be associated with higher compressive strength; however, the relationship was not strictly monotonic and did not uniquely determine the strength ranking. This is consistent with the broader understanding that higher external compactness often accompanies higher strength. However, apparent bulk density is an external-volume measure and, by itself, does not identify pore-size distribution, pore connectivity, or internal heterogeneity in hardened cement pastes. In the present results, mixtures with similar mean apparent bulk densities still exhibited different compressive strengths (e.g., BG1 and BG5), and the highest mean apparent bulk density did not necessarily coincide with the highest mean compressive strength within a binder series (e.g., BG5 and BG3). Accordingly, apparent bulk density is useful as a comparative compactness indicator, but it cannot by itself explain the full strength ranking of the GO-modified pastes. Apparent bulk density reflects gross external compactness, but it does not resolve whether the internal pore system is uniformly refined or whether local heterogeneity and defect-like features remain.

Complementary pore-structure methods are required to examine such features more directly [35–38].

The Supplement provides additional descriptive hardened-state characterization: MIP pore-entry distributions and normalized MIP-accessible intrusion-volume metrics are reported in Fig. S1 and Table S1, qualitative XRD patterns are reported in Fig. S2, and illustrative SEM micrographs are reported in Figs. S3–S4. Individual compressive-strength values underlying the reported mean $\pm$ SD results are provided in Table S2. An additional limitation is that only one fixed GO:PCE ratio (1:5, expressed as dry GO mass to as-received PCE product mass) and one sonication-based preparation protocol were examined. Calorimetry and direct dispersion measurements were not included in the current experimental program.

4 Conclusions

This study evaluated the effect of graphene oxide introduced through a pre-dispersed GO:PCE suspension on the compressive strength and apparent bulk density of cement pastes prepared with two binders: CEM I 42.5 N-SR0 and CEM II/B-LL 42.5 N. The results were used to assess:

- the strength response of each binder to GO dosage and
- whether apparent bulk density could explain strength differences in GO-modified pastes.

The main conclusions are as follows:

- Within the tested dosage range, the GO:PCE suspension route showed a non-linear strength response in both binder series. The 0.03% GO dosage by cement mass produced the highest measured 28-day strength, increasing compressive strength by 23% in CEM I and 19% in CEM II/B-LL. At 7 days, the same dosage increased compressive strength by 16% in CEM I and

15% in CEM II/B-LL. Although 0.03% GO gave the highest measured strength in both binders, the response peak was more clearly defined in CEM I than in CEM II/B-LL, indicating that the dosage sensitivity remained binder-dependent. Increasing GO to 0.05% by mass of cement did not provide additional benefit and resulted in lower strength than measured at 0.03%.

- Apparent bulk density, measured on the same cube specimens immediately before compressive testing, served as a practical indicator of external compactness. Within each binder series, higher apparent bulk density generally tended to coincide with higher strength; however, density did not uniquely explain the strength ranking of the GO-modified pastes.
- Future work should directly quantify dispersion stability in alkaline, Ca^{2+} -rich cementitious environments and relate it to hydration and pore-structure development using complementary measurements, rather than inferring mechanisms from compressive strength and apparent bulk density alone.

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