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Influence of Polycarboxylate Ether Type and Dosage on Electrokinetic and Hydrodynamic Descriptors of Graphene Oxide Suspensions in an Alkaline Model Medium

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Abstract

Polycarboxylate ether (PCE) superplasticizers are widely used in cementitious systems. They are also used to disperse solid particles in aqueous suspensions, including cement particles and nano-additives such as graphene oxide (GO). Their performance depends on both polymer formulation and dosage. This study compared four commercial PCEs in a NaOH-adjusted alkaline model medium. The electrokinetic and hydrodynamic behavior of GO suspensions was evaluated over GO:PCE mass ratios from 1:0 to 1:6. Zeta potential (ZP) and dynamic light scattering (DLS) were used as comparative descriptors. The control GO suspension showed a mean ZP of −58 mV, a Z-average hydrodynamic size of 531 nm, and a polydispersity index (PDI) of 0.37. After PCE addition, the PDI decreased to 0.26–0.30, indicating lower DLS-derived apparent heterogeneity relative to the control suspension. The response depended on PCE formulation. ACE 300 and MC-PowerFlow 2695 gave the most negative ZP values, reaching −76 mV and −65 mV, respectively, at a GO:PCE ratio of 1:5. At 1:6, the magnitude decreased relative to the peak values. By contrast, the ViscoCrete series showed a more moderate ZP response, from −53 to −58 mV, while the smallest apparent sizes were obtained with the ViscoCrete series, reaching 477 nm. These results show that GO dispersion in this alkaline model medium cannot be inferred from a single descriptor or from applying a single dosage rule across different PCE formulations. Joint interpretation of electrokinetic and hydrodynamic measurements is therefore needed for comparative screening.

Keywords

graphene oxide, polycarboxylate ether, zeta potential, dynamic light scattering, alkaline model medium

1 Introduction

Graphene oxide (GO) has drawn sustained interest in cement-based materials because its oxygen-containing surface groups and two-dimensional structure make its behavior strongly dependent on suspension chemistry and dispersion quality [1–3]. Reported effects of GO on cementitious systems remain inconsistent. Similar nominal dosages have produced different outcomes across studies. These differences have been linked not only to dosage, but also to exfoliation state, agglomeration history, preparation procedure, and the chemistry of the liquid phase into which GO is introduced [4–10]. Dispersion control should therefore be treated as a major experimental variable rather than as a minor preparatory step [6, 8–10]. When stabilization is inadequate, attractive interactions between sheets can promote re-agglomeration. This can reduce accessible surface area and alter interactions with dissolved species in the surrounding medium. As a result, suspensions prepared at the same nominal GO concentration may still behave differently if they differ in agglomerate size distribution, degree of separation, or interfacial stabilization [10–15].

Among the available dispersion strategies, polycarboxylate ether (PCE) superplasticizers are especially relevant. They are already widely used in cementitious systems and have also been investigated as GO dispersants [11–15]. In cementitious materials, PCE superplasticizers act as high-range water-reducing admixtures. Their plasticizing effect is expressed macroscopically as improved flowability at a given water content, or as lower water demand

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at a given workability. This effect mainly arises from the dispersion of flocculated solid particles through polymer adsorption and the development of electrostatic and steric repulsive forces [16–19]. In the present study, no fresh cement paste rheology was measured. Therefore, the role of PCE is considered as a dispersion-related action in alkaline GO suspensions, where polymer adsorption or association and steric effects may modify GO–GO interactions, zeta potential (ZP), and apparent hydrodynamic size.

PCEs are commonly described as comb-like polymers. They consist of an anionic carboxylate-bearing backbone and non-ionic polyether side chains [16–19]. The carboxylate groups can contribute to adsorption and electrostatic effects, while the solvated polyether side chains provide steric separation between particles or sheets [15–19]. In GO-containing suspensions, these architecture-related features may affect GO–GO interactions, ZP, and apparent hydrodynamic size through electrokinetic and polymer-related interfacial effects [15]. The exact molecular architecture, including molar mass, side-chain length, and grafting density, varies among commercial products [16–19]. For the commercial products investigated here, detailed molecular architecture is not disclosed in the publicly available technical datasheets.

Their action is commonly discussed in terms of combined electrostatic and steric contributions. However, the relative importance of these effects depends on both formulation and medium chemistry [15–20]. Preparation and measurement conditions also require control. Ultrasonication has been reported to affect the morphology, structure, dispersion state, and performance of GO or GO-containing cementitious systems [21–24]. In cementitious suspensions, zeta-potential measurements have also been used to evaluate surface-charge behavior and cement–superplasticizer interactions [25, 26].

Differences in composition and molecular design can affect adsorption behavior, solution conformation, and dispersion efficiency [16–19]. A PCE that performs well as a cement superplasticizer is therefore not necessarily the most effective dispersant for GO suspensions. Previous studies comparing different water-reducing agents or PCE-based systems have also reported different electrokinetic and particle-size responses under otherwise similar conditions [11–15].

Suspension-medium chemistry is itself a controlling variable. Behavior observed in distilled or near-neutral water cannot be assumed to represent behavior in highly alkaline solutions relevant to cementitious systems [10–13, 20, 27]. Under alkaline conditions, both GO and PCE respond to changes in pH and ionic composition. The ionization state of GO surface groups can change, and dissolved polymers may also alter their interaction behavior in solution [11–13, 16–18]. Consequently, the same GO:PCE combination can show different ZP, apparent hydrodynamic size, or size distribution depending on the chemistry of the suspension medium [10–13, 20]. For cement-related applications, screening in a chemically relevant medium is therefore more informative than screening in pure water alone [10, 20].

In this study, a NaOH-adjusted alkaline medium was used as a controlled intermediate system. It captures the effect of high pH under controlled conditions while avoiding the additional complexity of a true cement pore solution. In cement pore solution, dissolved calcium and other ions may introduce specific adsorption, complexation, and ion-bridging effects [10, 12, 13, 20]. Although this simplified medium does not reproduce the full chemistry of cement pore solution, it provides a practical basis for controlled comparative screening of GO:PCE interactions before moving to more complex cementitious environments [10, 12, 13, 20].

Preparation history also affects the apparent dispersion state. Ultrasonication is widely used to disperse GO because cavitation and shear can reduce agglomeration. However, sonication conditions can influence the material state. Mode, duration, and energy input can affect the final suspension state, and excessive treatment may alter GO structure or defect state [21–24, 28]. Comparisons among dispersants are therefore meaningful only when the mechanical preparation protocol is kept consistent across formulations [21–24].

Descriptor choice requires similar caution. ZP provides an electrokinetic indicator of interfacial behavior. The Z-average hydrodynamic size and polydispersity index (PDI), obtained by dynamic light scattering (DLS), provide complementary information on apparent agglomerate size and distribution width [10, 25, 26, 29]. No single parameter is sufficient to describe GO dispersion fully. For two-dimensional materials such as GO, DLS does not measure the true lateral sheet size. Instead, it reflects an apparent hydrodynamic response affected by agglomeration state, orientation, and the solvated interfacial layer [10, 29]. Likewise, ZP alone does not fully describe dispersion behavior in polymer-containing systems, where polymer-related interfacial effects may remain important even when the measured ZP is not maximal [16, 25, 26]. Combined evaluation of electrokinetic and hydrodynamic descriptors is therefore more informative than reliance on a single metric.

Despite the growing literature on GO dispersion and GO:PCE systems, only a limited number of studies directly compare several commercial PCE superplasticizers under one common alkaline protocol while evaluating both electrokinetic and DLS-derived descriptors [11, 12, 14, 15]. Much of the available literature focuses either on cement performance outcomes or on individual dispersant systems. This makes side-by-side comparison across commercial products more difficult [1, 2, 4, 5, 7]. Such comparison remains necessary because a formulation with a more negative ZP does not necessarily produce the smallest apparent hydrodynamic size or the narrowest size distribution [11, 14–16]. The dosage window judged favorable may therefore depend on the descriptor considered and on how a given formulation affects interfacial and hydrodynamic behavior under alkaline conditions [11, 14–16].

Against this background, the present study examines the dosage-dependent effects of four commercial PCE superplasticizers on the electrokinetic and hydrodynamic behavior of GO suspensions in a NaOH-adjusted alkaline model medium prepared at target pH 12. GO mass ratios from 1:0 to 1:6 were evaluated using a standardized alkalization and sonication protocol. ZP, Z-average hydrodynamic size, and PDI were used to compare how different commercial PCE formulations affect the apparent dispersion state of GO under controlled alkaline conditions. The aim was not to reproduce the full chemistry of cement pore solution, but to establish a controlled comparative screening framework for later application in more complex cementitious environments.

2 Materials and methods

2.1 Materials

Suspensions of graphene oxide (GO) were prepared using four commercial polycarboxylate-based superplasticizers: MasterGlenium ACE 300, ViscoCrete-4269, ViscoCrete-6711, and MC-PowerFlow 2695 (Fig. 1). These products are commercial high-range water-reducing/plasticizing admixtures based on polycarboxylate polymer technology. In this study, they were evaluated as dispersing polymers for GO suspensions. The products were used as received from the manufacturers and were dosed relative to the dry GO mass at GO:PCE mass ratios from 1:1 to 1:6.

The GO used in this study was obtained as a commercial aqueous stock dispersion from William Blythe Ltd. (UK), with a nominal stock concentration of 1 wt.% (10 mg/mL) [30]. This concentration refers only to the as-received stock dispersion. Aliquots of the stock dispersion were subsequently diluted during suspension preparation to obtain the GO content used for DLS and ZP measurements.

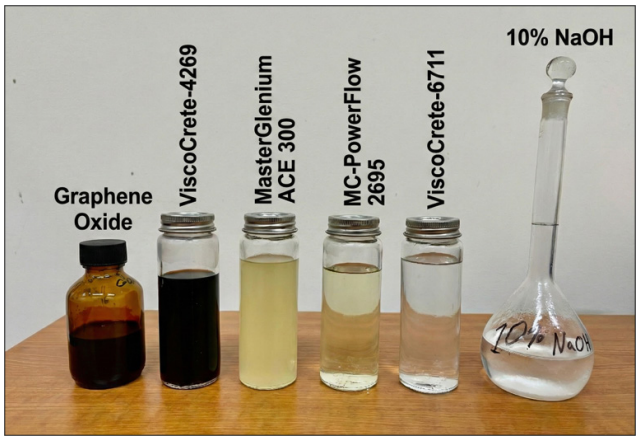


Fig. 1 Graphene oxide (GO) dispersion and the investigated PCE superplasticizer samples

Additional supplier characterization was used to describe the chemistry and morphology of the GO dispersion. AFM and SEM observations indicated that the GO sheets had lateral dimensions commonly above 5 µm, while AFM analysis gave a sheet depth below 2 nm, consistent with predominantly single- to few-layer GO. Raman spectroscopy indicated oxidized GO, with an ID/IG ratio of approximately 1. FTIR-ATR analysis identified oxygen-containing functional groups, including O–H, C=O carboxyl/carbonyl, and C–O-related groups, together with C=C bonding. These groups are relevant to the electrokinetic response because their ionization state under alkaline conditions can contribute to the negative surface charge of GO. The supplier data identify the main functional groups qualitatively; however, quantitative functional-group fractions were not provided. The corresponding supplier-provided GO characterization data are presented in Fig. S1–S5 and Table S1, available in the Supplement.

Essential physicochemical characteristics of GO and datasheet properties of the investigated superplasticizers are summarized in Tables 1 to 3.

Table 1 Physical properties of GO reported by the supplier

Material	Appearance	Concentration	Sheet size	pH
Graphene oxide (GO)	Orange-brown aqueous dispersion	1 wt.% (10 mg/mL)	>5 µm	> 2

Table 2 Composition of GO reported by the supplier

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

Table 3 Datasheet properties of the investigated PCE superplasticizers

PCE (SP) name	MasterGlenium ACE 300	MC-PowerFlow 2695	ViscoCrete-4269	ViscoCrete-6711
Manufacturer	Master Builders	MC-Bauchemie	Sika	Sika
Appearance	Yellowish opalescent liquid	Yellowish-brown liquid	Dark brown liquid	Whitish-yellow liquid
Density (20–21 °C)	1.035–1.075 kg/dm^3	~1.06 kg/dm^3	1.08 ± 0.02 kg/dm^3	1.04 ± 0.02 kg/dm^3
pH (20 °C)	Not stated in the datasheet	Not stated in the datasheet	4.5 ± 1	4.1 ± 1 at 20 °C
Chloride content	≤ 0.1% by mass	< 0.1% (max)	≤ 0.10%	≤ 0.10%
Alkali content (Na_2O eq.)	≤ 5.0% (Na_2O equivalent)	< 2.0% (max)	Not stated in the datasheet	Not stated in the datasheet

2.2 Methods

2.2.1 Preparation of GO:PCE suspensions

Graphene oxide (GO) suspensions were prepared as pre-dispersed GO–polycarboxylate ether (PCE) systems. The formulation matrix is given in Table 4.

The PCE was first diluted with distilled water, and GO was then added. A 10 wt.% sodium hydroxide (NaOH) solution was added gradually to achieve the target pH of 12. The pH was checked using colorimetric pH indicator paper, comparing the color with the supplied color scale. The suspension was first homogenized by magnetic stirring at 40 °C for 10 min (Fig. 2). Probe ultrasonication was then performed using a Hielscher UP100H device (100 W, 30 kHz) at full amplitude in pulsed mode with a 1–2 s pulse interval for 30 min. The pulsed mode was selected to reduce continuous heat accumulation during sonication. After sonication, the suspension was magnetically stirred again at 40 °C for 10 min. This post-sonication stirring step was used to restore bulk homogeneity before zeta-potential and dynamic light scattering (DLS) measurements. No post-sonication solid-content correction was applied; therefore, the results are interpreted comparatively across formulations prepared under the same pulsed-sonication protocol.

Table 4 Mixture matrix used for GO:PCE suspension preparation

Material	Mass ratio	Amount (g)
Equivalent dry GO mass, mGO	1 ×	0.025
GO stock dispersion (1 wt.%)	–	2.5
Distilled water	Balance	q.s. to 50.0
Superplasticizer	1–6 × mGO	0.025–0.15
NaOH solution (10 wt.%)	Adjusted to pH 12	~0.35

Notes: mGO = 0.01 × m (stock dispersion); therefore 2.5 g of 1 wt.% GO stock corresponds to 0.025 g dry GO equivalent.

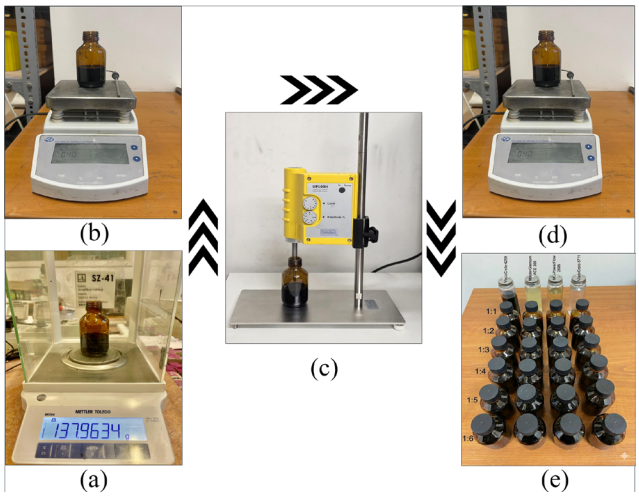


Fig. 2 Preparation sequence for PCE-stabilized GO suspensions: (a) material dosing; (b) initial magnetic stirring at 40 °C; (c) probe ultrasonication; (d) post-sonication magnetic stirring at 40 °C; (e) prepared GO:PCE suspensions

2.2.2 Zeta potential (ZP) and dynamic light scattering (DLS) characterization of GO:PCE suspensions

The electrokinetic and hydrodynamic behavior of the GO suspensions was assessed using zeta potential (ZP) and dynamic light scattering (DLS). Measurements were performed on freshly prepared suspensions. ZP was used as an electrokinetic descriptor, whereas DLS provided the Z-average hydrodynamic diameter and polydispersity index (PDI) as comparative indicators of the apparent dispersion state [10, 11, 15].

Measurements were performed at 25 °C using a Malvern Zetasizer Nano ZS instrument. DLS measurements were carried out using disposable 40 µL micro-cuvettes, while ZP measurements were performed using DTS1070 folded capillary cells. The DLS autocorrelation function was evaluated using the cumulants method to obtain the Z-average hydrodynamic diameter and PDI. ZP was calculated from electrophoretic mobility using the Smoluchowski model. A comparable Malvern Zetasizer-based ZP measurement procedure has also been reported for alkaline geopolymer-related construction-material suspensions, where ZP was used to evaluate surface-charge-related behavior during material stabilization [31].

For each formulation, one prepared suspension was analyzed, and three instrument runs were performed for both DLS and ZP measurements. The reported mean ± SD values, therefore, reflect within-instrument repeatability

for a single prepared suspension rather than independent preparation-to-preparation reproducibility. Possible fluorescence of GO was not separately quantified. However, the Zetasizer Nano ZS uses a 633 nm He–Ne laser, a wavelength at which GO fluorescence interference is comparatively limited. All suspensions were processed identically. Therefore, the DLS-derived size and PDI values are interpreted as comparative apparent descriptors rather than as absolute measures of the GO sheet-size distribution.

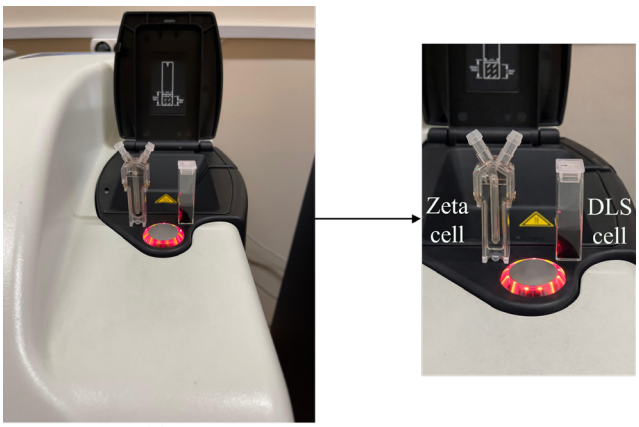
In this study, the NaOH-adjusted medium served as a simplified model system for comparative screening. Accordingly, the measurements reflect the behavior of GO:PCE suspensions in an alkaline aqueous medium rather than in a complete cement pore solution containing multivalent ions. The analyses were conducted at the Microstructural Characterization Laboratory Unit, Department of Construction Materials and Technologies, BME (Fig. 3).

3 Results and discussion

3.1 Zeta potential (ZP)

The ZP measurements obtained in the NaOH-adjusted model medium at target pH 12 showed that all graphene oxide (GO) suspensions carried a negative electrokinetic charge across the GO:PCE mass ratios investigated (Fig. 4).

The control GO (GOC) suspension (1:0), in which no PCE was added and the medium was adjusted only with NaOH solution, gave a mean ZP of −58 ± 4.95 mV under the tested alkaline conditions. The addition of polycarboxylate ether (PCE) superplasticizers altered this response in a formulation-dependent manner. ACE 300 and MC-PowerFlow 2695 increased the magnitude of the negative ZP, reaching peak values of −76 ± 3.19 mV and −65 ± 3.06 mV, respectively, at a GO:PCE ratio of 1:5.



(a) (b)

Fig. 3 Malvern Zetasizer Nano ZS measurement setup: (a) instrument; (b) cells containing GO suspensions for ZP and particle-size measurements

Because both GO and PCE are expected to carry negative charge under alkaline conditions, the more negative ZP values are not interpreted as simple opposite-charge adsorption [14, 15]. Instead, they are interpreted as changes in polymer–GO interfacial behavior. The supplier FTIR-ATR data described in Section 2.1 indicate oxygen-containing GO groups, including O–H, C=O carboxyl/carbonyl, and C–O-related groups, which may provide sites for surface association with PCE molecules. In the general comb-like PCE architecture discussed above, carboxylate-bearing backbone groups may contribute to the electrokinetic response, while solvated polyether side chains can contribute to steric separation between GO sheets or agglomerates [16–19].

When the dosage was increased to a GO:PCE ratio of 1:6, the absolute ZP magnitude decreased for both ACE 300 and MC-PowerFlow 2695. This non-monotonic response is interpreted cautiously as an overdosing-type effect. It may be associated with excess dissolved polymer, charge screening, changes in the effective shear plane, or reduced additional benefit after partial saturation of available interaction sites [11, 12, 15].

By contrast, the ViscoCrete series, VC-4269 and VC-6711, remained within a relatively narrow ZP range across the tested dosages, approximately −53 to −58 mV. This limited variation suggests that the electrokinetic response of these suspensions was less sensitive to dosage than that of ACE 300 and MC-PowerFlow 2695. In polymer-containing systems, however, ZP alone may not fully describe dispersion behavior, because adsorbed or extended polymer layers can shift the effective shear plane and reduce the apparent charge measured during electrophoretic analysis [16, 18].

3.2 Apparent hydrodynamic size of GO:PCE suspensions

The dynamic light scattering (DLS) results, expressed as Z-average hydrodynamic diameter, showed that both PCE type and GO:PCE mass ratio influenced the apparent dispersion state of the GO suspensions (Fig. 5). Because DLS is based on the Stokes–Einstein relation and reflects an equivalent hydrodynamic response rather than the true geometry of a two-dimensional nanosheet, the reported Z-average values should be interpreted as comparative indicators of apparent aggregate size rather than as the actual lateral size of GO sheets [10, 29].

The control GO suspension (1:0) showed an apparent mean size of 531 ± 2.15 nm and the highest polydispersity index (PDI = 0.37). After PCE addition, most formulations showed PDI values of 0.26–0.30 (Table 5), lower than

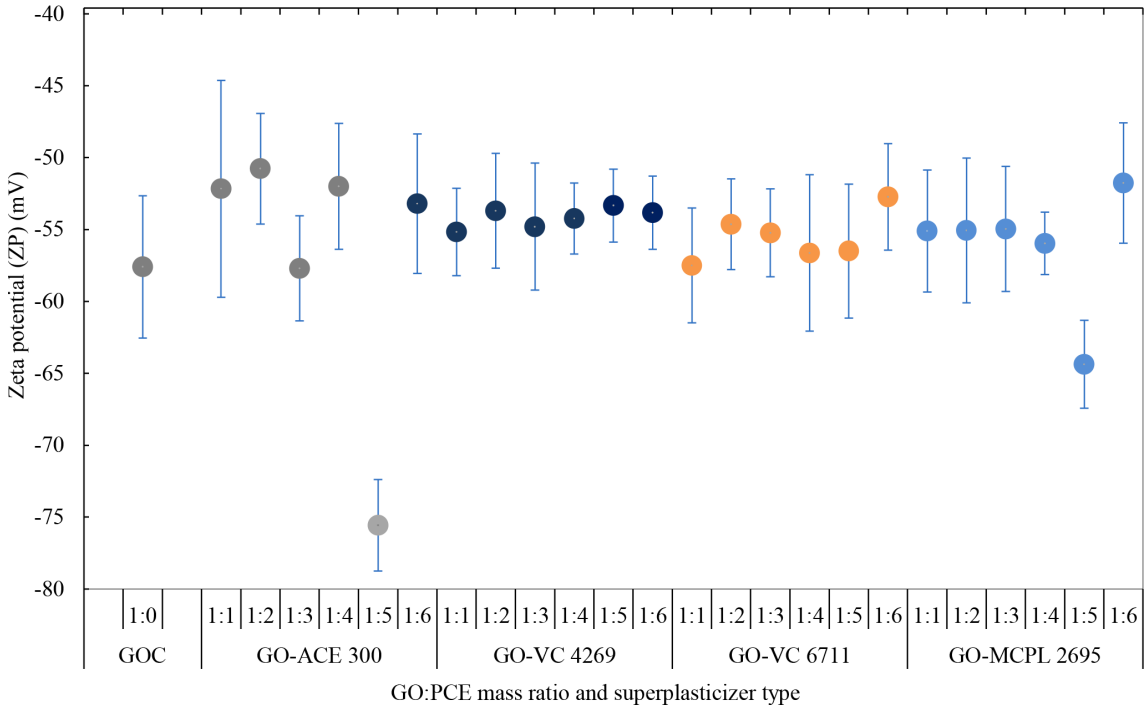


Fig. 4 Zeta potential (ZP) of GO:PCE suspensions as a function of GO:PCE mass ratio (1:0–1:6). Values are reported as mean ± SD from three instrument runs on the same prepared suspension

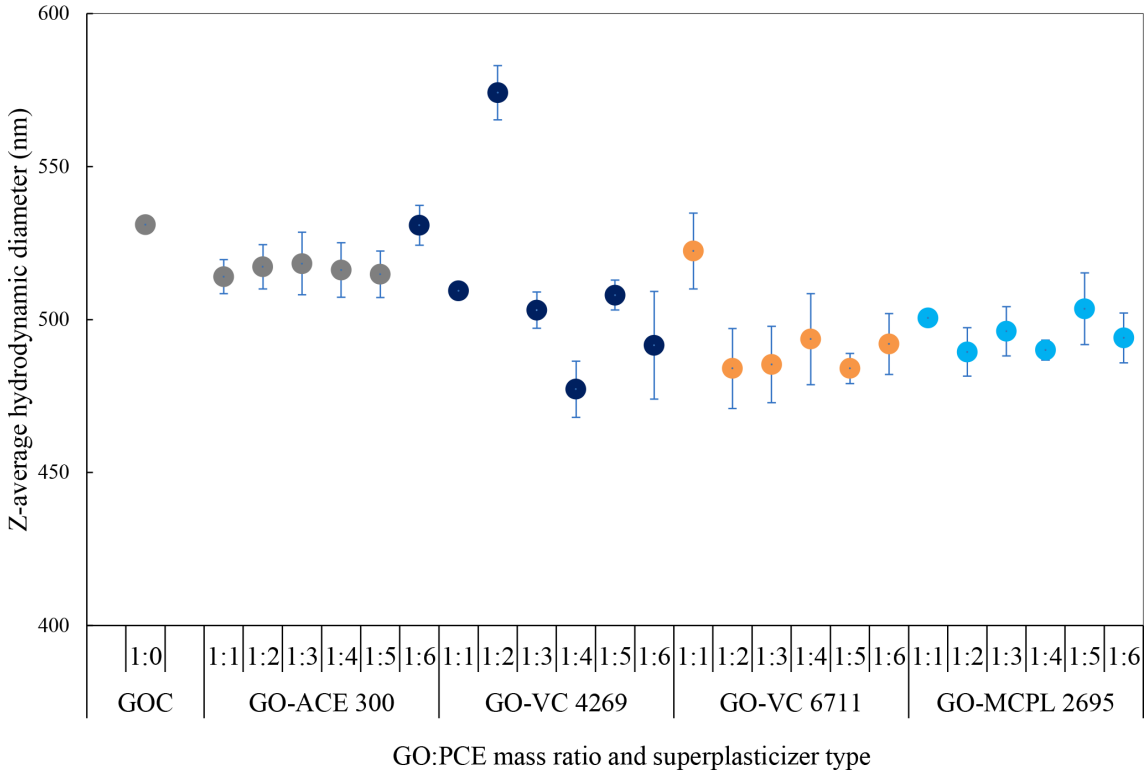


Fig. 5 Apparent particle size of GO:PCE suspensions, expressed as Z-average hydrodynamic diameter, as a function of GO:PCE mass ratio (1:0–1:6). Values are reported as mean ± SD from three instrument runs on the same prepared suspension

the control value. In this study, PDI was used as a DLS-derived comparative descriptor of apparent distribution width. These values indicate lower apparent heterogeneity than in the control suspension, but they should not be interpreted as evidence of a uniform or monodisperse distribution of GO sheets.

Table 5 Summary of electrokinetic and DLS parameters of GO:PCE suspensions at different GO:PCE mass ratios

PCE type	GO:PCE ratio	Mean ZP (mV)	Zeta deviation (mV)	Conductivity (mS/cm)	Mean size, Z-average (d, nm)	PDI	Zeta result quality	DLS result quality
GOC	1:0	−58	9.91	3.81	531	0.37	Good	Good
ACE 300	1:1	−53	10.3	2.46	514	0.27	Good	Good
	1:2	−51	11.0	3.22	517	0.28	Good	Good
	1:3	−58	12.8	3.61	518	0.28	Good	Good
	1:4	−52	11.2	3.00	516	0.30	Good	Good
	1:5	−76	13.3	2.83	515	0.29	Good	Good
	1:6	−54	10.6	3.42	531	0.29	Good	Good
VC-4269	1:1	−56	11	3.10	509	0.29	Good	Good
	1:2	−54	12	3.76	574	0.27	Good	Good
	1:3	−55	11.7	3.92	503	0.29	Good	Good
	1:4	−55	11.4	3.57	477	0.30	Good	Good
	1:5	−54	11.6	3.57	508	0.27	Good	Good
	1:6	−54	10.1	3.15	492	0.29	Good	Good
VC-6711	1:1	−58	11.5	3.62	522	0.27	Good	Good
	1:2	−55	9.53	3.06	484	0.27	Good	Good
	1:3	−56	10.7	3.50	485	0.30	Good	Good
	1:4	−57	12.5	3.93	494	0.28	Good	Good
	1:5	−57	10.1	3.66	484	0.27	Good	Good
	1:6	−53	9.7	3.26	492	0.26	Good	Good
MCPL-2695	1:1	−55	10.6	2.91	500	0.27	Good	Good
	1:2	−55	10.6	3.4	489	0.29	Good	Good
	1:3	−55	10.3	3.46	496	0.26	Good	Good
	1:4	−56	11.5	3.83	490	0.29	Good	Good
	1:5	−65	11.6	2.81	503	0.27	Good	Good
	1:6	−52	11.5	3.05	494	0.28	Good	Good

Representative DLS intensity-weighted hydrodynamic size distributions for the control GO suspension and selected GO:PCE suspensions are presented in Fig. S6, available in the Supplement. This distinction is important because DLS reports an equivalent hydrodynamic response rather than the true lateral size distribution of two-dimensional GO sheets [11–15].

Among the investigated admixtures, VC-6711 produced consistently low apparent Z-average values, around 484 nm at GO:PCE ratios of 1:2 and 1:5. The single lowest value in the dataset, 477 nm, was recorded for VC-4269 at 1:4. However, VC-4269 also showed a higher Z-average value at 1:2. This indicates that its hydrodynamic response was less consistent across the tested dosage range than that of VC-6711. The decrease in apparent size occurred even though the corresponding ZP values in Section 3.1 remained within a moderate range. Within this dataset, the reduction in apparent size therefore cannot be explained by ZP magnitude alone.

In polymer-containing systems, a smaller apparent aggregate size does not necessarily correspond to a more negative ZP, because polymer-related interfacial effects can influence the hydrodynamic response [16–19]. MC-PowerFlow 2695, by contrast, showed a relatively narrow range of apparent sizes, from 489 to 503 nm, across the tested dosages. This limited variation indicates that the measured apparent size changed less across the examined dosage window than for the other investigated products. Rather than assigning this behavior to a specific unmeasured mechanism, it is more appropriate to interpret it as a comparatively consistent hydrodynamic response within the tested alkaline dosage window.

3.3 Joint interpretation of ZP and particle size

Fig. 6 compares the absolute ZP magnitude with the apparent Z-average hydrodynamic size of the GO:PCE suspensions. No single monotonic relationship was observed. Instead, the data formed formulation-dependent clusters. This indicates

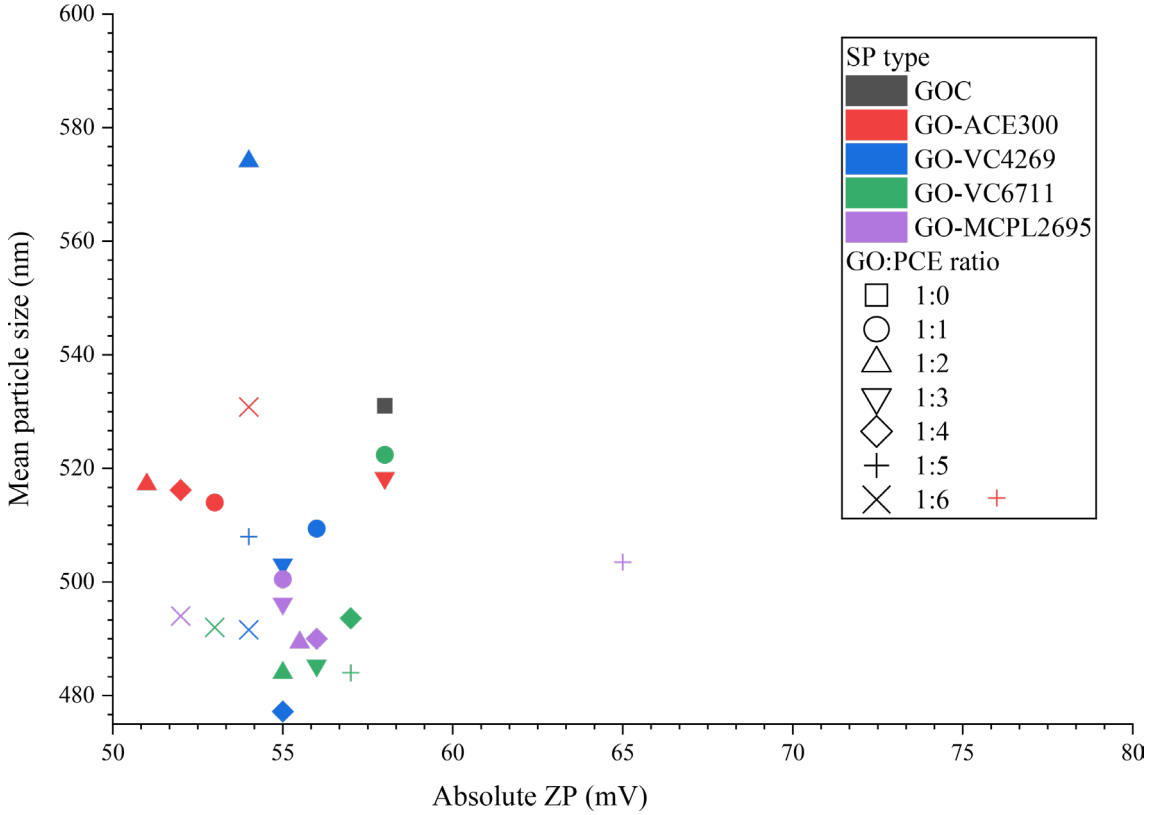


Fig. 6 Relationship between absolute ZP magnitude and apparent particle size, expressed as Z-average hydrodynamic diameter, for GO:PCE suspensions at different GO:PCE mass ratios

that ZP and apparent hydrodynamic size did not respond in the same way to changes in PCE type and dosage.

The absence of a single tendency in Fig. 6 is consistent with the fact that commercial PCEs may differ in molecular architecture, including backbone charge density, side-chain length, grafting density, molecular mass distribution, and auxiliary formulation components. These parameters can affect adsorption or association with GO, the position of the electrokinetic shear plane, and the thickness of the solvated interfacial layer. However, the observed product-dependent clustering is interpreted as a formulation-dependent response rather than being assigned to a specific molecular parameter.

This comparison shows that ZP alone is not sufficient to characterize the apparent dispersion state across different superplasticizer formulations. For example, the ACE 300 suspension at a GO:PCE ratio of 1:5 showed a high absolute ZP magnitude while retaining a moderate apparent size of about 515 nm. This indicates that the two descriptors did not rank the formulation in the same way. By contrast, the ViscoCrete series, particularly VC-6711, fell within a lower apparent size range despite only moderate absolute ZP values. This pattern suggests that the hydrodynamic behavior of these suspensions cannot be interpreted from ZP alone and may reflect additional polymer-related interfacial effects [16–19].

MC-PowerFlow 2695 showed intermediate behavior. It combined a relatively narrow apparent size range with a moderate increase in the magnitude of negative ZP. Overall, these results indicate that comparative screening of GO:PCE systems should rely on multiple descriptors. If only ZP is considered, descriptor-based differences between formulations may be overlooked. If only DLS-derived apparent size is considered, electrokinetic differences between formulations may be missed [10, 15, 29].

The NaOH-adjusted medium used here provided a useful basis for comparative screening of GO:PCE suspensions. However, it does not reproduce the full chemistry of a cement pore solution. In such media, dissolved multivalent ions, particularly Ca^{2+}, can alter adsorption behavior, compress the electrical double layer, and promote additional interparticle interactions [10, 12, 13, 20]. For cementitious applications, this distinction is relevant because PCE-modified GO systems have also been reported to influence hydration characteristics and mechanical behavior of cement [32]. Related studies have likewise shown that the microstructure of polycarboxylate superplasticizers can influence graphene-based dispersion behavior [33]. Selection of a suitable GO:PCE

formulation should therefore consider dosage, product formulation, and the chemical environment in which the suspension will ultimately be used [11–15].

Data plotting for Figs. 4 and 5 was performed using Microsoft Excel for Microsoft 365 [34], while Fig. 6 was prepared using OriginPro 2025b [35].

4 Conclusions

This study investigated the dosage-dependent effects of four commercial PCE superplasticizers on the electrokinetic and hydrodynamic behavior of graphene oxide (GO) suspensions in an alkaline model medium:

- Under the tested conditions, no single universal optimum could be defined for GO dispersion. Instead, the observed behavior depended on the combined influence of PCE type, dosage, and the selected evaluation parameter. Following PCE addition, the PDI decreased from 0.37 to 0.26–0.30. This indicates lower DLS-derived apparent heterogeneity relative to the control suspension, but not a uniform or monodisperse distribution of GO sheets.
- The electrokinetic response varied in a formulation-dependent manner. ACE 300 and MC-PowerFlow 2695 produced the most negative ZP values, reaching maximum magnitude at a GO:PCE ratio of 1:5, followed by a slight decrease at the higher dosage. In contrast, the ViscoCrete series showed a more moderate ZP response while producing the smallest apparent hydrodynamic sizes. VC-6711 gave the most consistent low-size response within the tested dosage window.
- The results demonstrate that electrokinetic and hydrodynamic descriptors reflect different aspects of dispersion behavior. A more negative ZP did not necessarily correspond to a smaller apparent particle size. Similarly, reduced apparent size alone did not identify the governing stabilization mechanism. The observations are consistent with contributions from electrokinetic effects and polymer-related interfacial or steric effects, but the specific molecular origin cannot be assigned from ZP and DLS alone.
- The NaOH-adjusted model medium provided a controlled basis for comparative analysis; however, it does not reproduce the full chemical complexity of cement pore solution. Therefore, the applicability of the results to real cementitious systems should be interpreted with caution.
- From an engineering perspective, selecting appropriate GO:PCE combinations requires consideration of polymer formulation, dosage, and chemical environment together. The results support a multi-parameter approach for evaluating GO dispersion behavior in cementitious applications.

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References

[1] 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, 15(3), 216, 2025. https://doi.org/10.3390/nano15030216

[2] 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", Construction and Building Materials, 241, 117939, 2020. https://doi.org/10.1016/j.conbuildmat.2019.117939

[3] Ghazizadeh, S., Duffour, P., Skipper, N. T., Bai, Y. "Understanding the behaviour of graphene oxide in Portland cement paste", Cement and Concrete Research, 111, pp. 169–182, 2018. https://doi.org/10.1016/j.cemconres.2018.05.016

[4] Kedir, A., Gamachu, M., Alex, A. G. "Cement-Based Graphene Oxide Composites: A Review on Their Mechanical and Microstructure Properties", Journal of Nanomaterials, 2023(1), 6741000, 2023. https://doi.org/10.1155/2023/6741000

[5] Mohammed, A., Al-Saadi, N. T. K., Sanjayan, J. "Inclusion of graphene oxide in cementitious composites: state-of-the-art review", Australian Journal of Civil Engineering, 16(2), pp. 81–95, 2018. https://doi.org/10.1080/14488353.2018.1450699

[6] Prudente, I. N. R., Santos, H. C. d., Fonseca, J. L., Oliveira, J. F. R., Shibutani, A. B., Gimenez, I. d. F., Santos, E. A. d., Barreto, L. S. "Highlights on the Effects of Graphene-Family Materials Dispersion on Hydration and Mechanical Properties of Cement-Based Materials", ACS Omega, 10(46), pp. 55161–55184, 2025. https://doi.org/10.1021/acsomega.5c07530

[7] Bagheri, A., Negahban, E., Asad, A., Abbasi, H. A., Raza, S. M. "Graphene oxide-incorporated cementitious composites: a thorough investigation", Materials Advances, 3(24), pp. 9040–9051, 2022. https://doi.org/10.1039/D2MA00169A

[8] Fonseka, I., Mohotti, D., Wijesooriya, K., Lee, C.-K., Mendis, P. "Influence of graphene oxide properties, superplasticiser type, and dispersion technique on mechanical performance of graphene oxide-added concrete", Construction and Building Materials, 428, 136415, 2024. https://doi.org/10.1016/j.conbuildmat.2024.136415

[9] Saravanan, S., Huang, L., Babaahmadi, A. "Effect of graphene oxide dispersion on hydration and carbonation performance of low-clinker cement", Construction and Building Materials, 508, 144950, 2026. https://doi.org/10.1016/j.conbuildmat.2025.144950

[10] Lu, Z., Hou, D., Hanif, A., Hao, W., Li, Z., Sun, G. "Comparative evaluation on the dispersion and stability of graphene oxide in water and cement pore solution by incorporating silica fume", Cement and Concrete Composites, 94, pp. 33–42, 2018. https://doi.org/10.1016/j.cemconcomp.2018.08.011

[11] Zhao, L., Guo, X., Liu, Y., Ge, C., Chen, Z., Guo, L., Shu, X., Liu, J. "Investigation of dispersion behavior of GO modified by different water reducing agents in cement pore solution", Carbon, 127, pp. 255–269, 2018. https://doi.org/10.1016/j.carbon.2017.11.016

[12] Zhou, H., Li, D. "The dispersion and influence of graphene-oxide-modified polycarboxylate superplasticiser in alkaline cement solution", Advances in Cement Research, 36(12), pp. 587–598, 2024. https://doi.org/10.1680/jadcr.23.00217

[13] Liu, B., Wang, L., Pan, G., Li, D. "Dispersion of graphene oxide modified polycarboxylate superplasticizer in cement alkali solution for improving cement composites", Journal of Building Engineering, 57, 104860, 2022. https://doi.org/10.1016/j.jobe.2022.104860

[14] Anwar, A., Liu, X., Zhang, L. "To what extent can polycarboxylate superplasticizers enhance the dispersion characteristics of modified graphene oxide in cement? Insights from ultrasonication and high-shear mixing methods", Cement and Concrete Research, 195, 107881, 2025. https://doi.org/10.1016/j.cemconres.2025.107881

[15] Qin, W., Guodong, Q., Dafu, Z., Yue, W., Haiyu, Z. "Influence of the molecular structure of a polycarboxylate superplasticiser on the dispersion of graphene oxide in cement pore solutions and cement-based composites", Construction and Building Materials, 272, 121969, 2021. https://doi.org/10.1016/j.conbuildmat.2020.121969

[16] Javadi, A., Jamil, T., Abouzari-Lotf, E., Soucek, M. D., Heinz, H. "Working Mechanisms and Design Principles of Comb-like Polycarboxylate Ether Superplasticizers in Cement Hydration: Quantitative Insights for a Series of Well-Defined Copolymers", ACS Sustainable Chemistry & Engineering, 9(25), pp. 8354–8371, 2021. https://doi.org/10.1021/acssuschemeng.0c08566

[17] Ma, R., Wang, Y., Li, H., Bai, Y. "Progress in the polycarboxylate superplasticizer and their structure-activity relationship – A review", Materials Today Communications, 35, 105838, 2023. https://doi.org/10.1016/j.mtcomm.2023.105838

[18] Sha, S., Wang, M., Shi, C., Xiao, Y. "Influence of the structures of polycarboxylate superplasticizer on its performance in cement-based materials-A review", Construction and Building Materials, 233, 117257, 2020. https://doi.org/10.1016/j.conbuildmat.2019.117257

[19] Marchon, D., Sulser, U., Eberhardt, A., Flatt, R. J. "Molecular design of comb-shaped polycarboxylate dispersants for environmentally friendly concrete", Soft Matter, 9(45), pp. 10719–10728, 2013. https://doi.org/10.1039/C3SM51030A

[20] Amini, K., Ghasemi, A., Soleimani Amiri, S., Mirvalad, S., Habibnejad Korayem, A. "The synergic effects of metakaolin and polycarboxylate-ether on dispersion of graphene oxide in cementitious environments and macro-level properties of graphene oxide modified cement composites", Construction and Building Materials, 270, 121462, 2021. https://doi.org/10.1016/j.conbuildmat.2020.121462

[21] Mellado, C., Figueroa, T., Baez, R., Meléndrez, M., Fernández, K. "Effects of probe and bath ultrasonic treatments on graphene oxide structure", Materials Today Chemistry, 13, pp. 1–7, 2019. https://doi.org/10.1016/j.mtchem.2019.04.006

[22] Le, G. T. T., Chanlek, N., Manyam, J., Opaprakasit, P., Grisdanurak, N., Sreearunothai, P. "Insight into the ultrasonication of graphene oxide with strong changes in its properties and performance for adsorption applications", Chemical Engineering Journal, 373, pp. 1212–1222, 2019. https://doi.org/10.1016/j.cej.2019.05.108

[23] Xiong, G., Ren, Y., Wang, C., Zhang, Z., Zhou, S., Kuang, C., Zhao, Y., Guo, B., Hong, S. "Effect of power ultrasound assisted mixing on graphene oxide in cement paste: Dispersion, microstructure and mechanical properties", Journal of Building Engineering, 69, 106321, 2023. https://doi.org/10.1016/j.jobe.2023.106321

[24] Ríos, J. D., Leiva, C., Martinez de la Concha, A., Ariza, M. P., Cifuentes, H. "Influence of Graphene Oxide Concentration and Ultrasonication Energy on Fracture Behavior of Nano-Reinforced Cement Pastes", Crystals, 14(8), 707, 2024. https://doi.org/10.3390/cryst14080707

[25] Plank, J., Hirsch, C. "Impact of zeta potential of early cement hydration phases on superplasticizer adsorption", Cement and Concrete Research, 37(4), pp. 537–542, 2007. https://doi.org/10.1016/j.cemconres.2007.01.007

[26] Srinivasan, S., Barbhuiya, S. A., Charan, D., Pandey, S. P. "Characterising cement–superplasticiser interaction using zeta potential measurements", Construction and Building Materials, 24(12), pp. 2517–2521, 2010.
https://doi.org/10.1016/j.conbuildmat.2010.06.005

[27] Lu, Z., Hanif, A., Ning, C., Shao, H., Yin, R., Li, Z. "Steric stabilization of graphene oxide in alkaline cementitious solutions: Mechanical enhancement of cement composite", Materials & Design, 127, pp. 154–161, 2017.
https://doi.org/10.1016/j.matdes.2017.04.083

[28] Coleman, B. R., Knight, T., Gies, V., Jakubek, Z. J., Zou, S. "Manipulation and Quantification of Graphene Oxide Flake Size: Photoluminescence and Cytotoxicity", ACS Applied Materials & Interfaces, 9(34), pp. 28911–28921, 2017.
https://doi.org/10.1021/acsami.7b08585

[29] Bhattacharjee, S. "DLS and zeta potential - What they are and what they are not?", Journal of Controlled Release, 235, pp. 337–351, 2016.
https://doi.org/10.1016/j.jconrel.2016.06.017

[30] Sayed, A., Mazrouaa, A. M., Mohamed, M. G., Abdel-Raouf, M. E.-S. "Green synthesis of chitosan/erythritol/graphene oxide composites for simultaneous removal of some toxic species from simulated solution", Environmental Science and Pollution Research, 30(10), pp. 25903–25919, 2023.
https://doi.org/10.1007/s11356-022-23951-4

[31] Khalaf, A. A., Kopecskó, K. "Utilizing residual red mud by geopolymerization process – From an expansive soil to a rock-like material", Cleaner Engineering and Technology, 14, 100647, 2023.
https://doi.org/10.1016/j.clet.2023.100647

[32] Li, Q., He, C., Zhou, H., Xie, Z., Li, D. "Effects of polycarboxylate superplasticizer-modified graphene oxide on hydration characteristics and mechanical behavior of cement", Construction and Building Materials, 272, 121904, 2021.
https://doi.org/10.1016/j.conbuildmat.2020.121904

[33] Wang, Q., Zhan, D., Qi, G., Wang, Y., Zheng, H. "Impact of the microstructure of polycarboxylate superplasticizers on the dispersion of graphene", New Carbon Materials, 35(5), pp. 547–558, 2020.
https://doi.org/10.1016/S1872-5805(20)60508-X

[34] Microsoft Corporation "Microsoft Excel for Microsoft 365", [computer program] Available at: https://www.microsoft.com/en-us/microsoft-365/excel [Accessed: 25 May 2026]

[35] OriginLab Corporation "OriginPro, (2025b)", [computer program] Available at: https://www.originlab.com/ [Accessed: 25 May 2026]