

Review

Application of Graphene-Based Nanomaterials as a Reinforcement to Concrete Pavements

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Abstract: Nanomaterials are considered to be one of the game-changing features in the modern world and nanotechnology is mostly reputed as the next-generation industrial revolution due to the extraordinary characteristics possessed by them at their very small scale. Graphene and graphene oxide are two main nanoscale materials that have seen a drastic increase in their use in cement-based composites due to exemptional enhancements in terms of strength and durability that can be imparted to compromise the inherent flaws of concrete and other cementitious composites. The main aim of this study was to investigate the effect of graphene and graphene oxide on improving the performance of cement-based composites and, particularly, of continuously reinforced concrete pavements (CRCP), which is one of the emerging trends in the transport sector due to various advantages they bring in over conventional flexible pavements and unreinforced concrete pavements. Fresh, hardened and durability properties of concrete with graphene-based nanomaterials were studied and the past experimental data were used to predict statistical interferences between different parameters attributed to concrete. According to the review, graphene-based nanomaterials seem to be promising to overcome the various CRCP distresses. Simultaneously, the possibilities and hinderances of using graphene and graphene oxide in cement-based composites as a reinforcement are discussed. Finally, the potential of using graphene in continuously reinforced concrete pavements is explored.

Keywords: graphene; graphene oxide; pavement distresses; cement composites; strength



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1. Introduction

It is well documented that concrete inherits a very low tensile strength amid its great compressive strength. However, concrete is the most common construction material used in the industry due to various other great properties instead of the low tensile strength. Rigid pavements are one such application where concrete is used effectively to give a favorable ride comfort to the drivers and obtain a significant level of durability and low maintenance [1,2]. Rigid pavements utilize concrete, while flexible pavements utilize basically asphalt for their construction and pavement design methodologies are not same. Different concrete pavements, such as jointed plain concrete pavements (JPCP), jointed reinforced concrete pavements (JRCP), fiber reinforced concrete pavements (FRCP), continuously reinforced concrete pavements (CRCP), and fiber reinforced CRCP (FR-CRCP), have taken increasing popularity over flexible pavements [1–3]. However, the present study mostly focuses on CRCP and how the performance of CRCP can be improved. Briefly, JPCP is the most common type of rigid concrete pavements used in the pavement industry. Normally, JPCP are composed of transverse joints in desirable spacing and the joints normally divide the pavement into small pavement panels. JPCP can be further categorized into undowelled and dowelled jointed pavements depending on the use of steel dowels across the joints. Using dowels across the joints improves the load transferring mechanism from between pavement panels. JRCP are mildly reinforced on the top of the slab to control transverse cracking and mostly dowels are also used in between the slab panels. In practice, plain

concrete pavements are used mostly for foot paths and roads where very low traffic is accommodated. There are enough studies where addition of fibers, such as steel [3,4], synthetic [5–8], glass [9–12] and graphene-based nanomaterials [13–28], have been used in pavements to compromise the issue in plain concrete pavements, which can be termed as fiber-reinforced concrete pavements (FRCP). Different fibers are proven to increase the flexural strength, fatigue strength and other pavement-related concrete properties in different ways and different levels of performance. The selection of fibers depends on the application of the pavements and will be addressed later in the section. However, if considerable traffic is expected in plain concrete pavements, they should be carefully designed. When the traffic increases, it is understood that the tensile stresses applied on the pavement also increase, in which plain concrete alone would not be able to withstand this after a certain level. In this case, additional rebars are used and those pavements are defined as continuously reinforced concrete pavements (CRCP) and are shown below in Figure 1a during their construction and Figure 1b after the construction.



Figure 1. (a) Continuously reinforced concrete pavements, (b) during construction and after construction [2].

Firstly, continuously reinforced concrete pavements (CRCP) are, particularly, one type of pavement of interest in the present study which has a history of 100 years and, compared to the other types of various concrete pavements, they offer greater characteristics under extreme weather conditions, high traffic loadings and life cycle cost [1,2,29]. These are reinforced with rebars to primarily provide the tensile strength to the concrete and secondary benefits, such as crack width control and fatigue performance enhancements [29]. Further, CRCP is expected to last a long while, allowing room for a possible widening due to extra traffic and several overlays due to pavement damage or aging. Moreover, fibers and additions, such as graphene-based nanomaterials, can be also included into CRCP to gain more resistance for common pavement distresses, such as spalling [30] and punchouts, while offering some other greater benefits also, such as insights of reducing slab thickness and amount of rebars. Therefore, it is necessary to investigate how these fibers and additions act in these concrete pavements and improve pavement performance indices. Even though there are various fibers [3–7,9–12] in use, the present study aims at investigating the use of graphene and graphene oxide in the above context. To start with, first, types of distresses in concrete pavements (CRCP) are discussed and the anticipated mitigation approaches are identified. Then, the nature of graphene and graphene oxide are discussed to identify the possibilities of applying graphene and graphene oxide to mitigate those distresses and other multifunctional benefits that can be imparted are discussed through previous studies.

2. Pavement Distresses and Mitigation

Various pavement distresses occur due to different mechanisms attributed to selection of suitable pavement, poor constructional practices, overloading, climatic effects, failure of layers under the top layers, etc. Some of these typical distresses are spalling, punchouts, wide longitudinal cracks, pumping and erosion, as shown in Figure 2a–d, respectively [2,31,32].

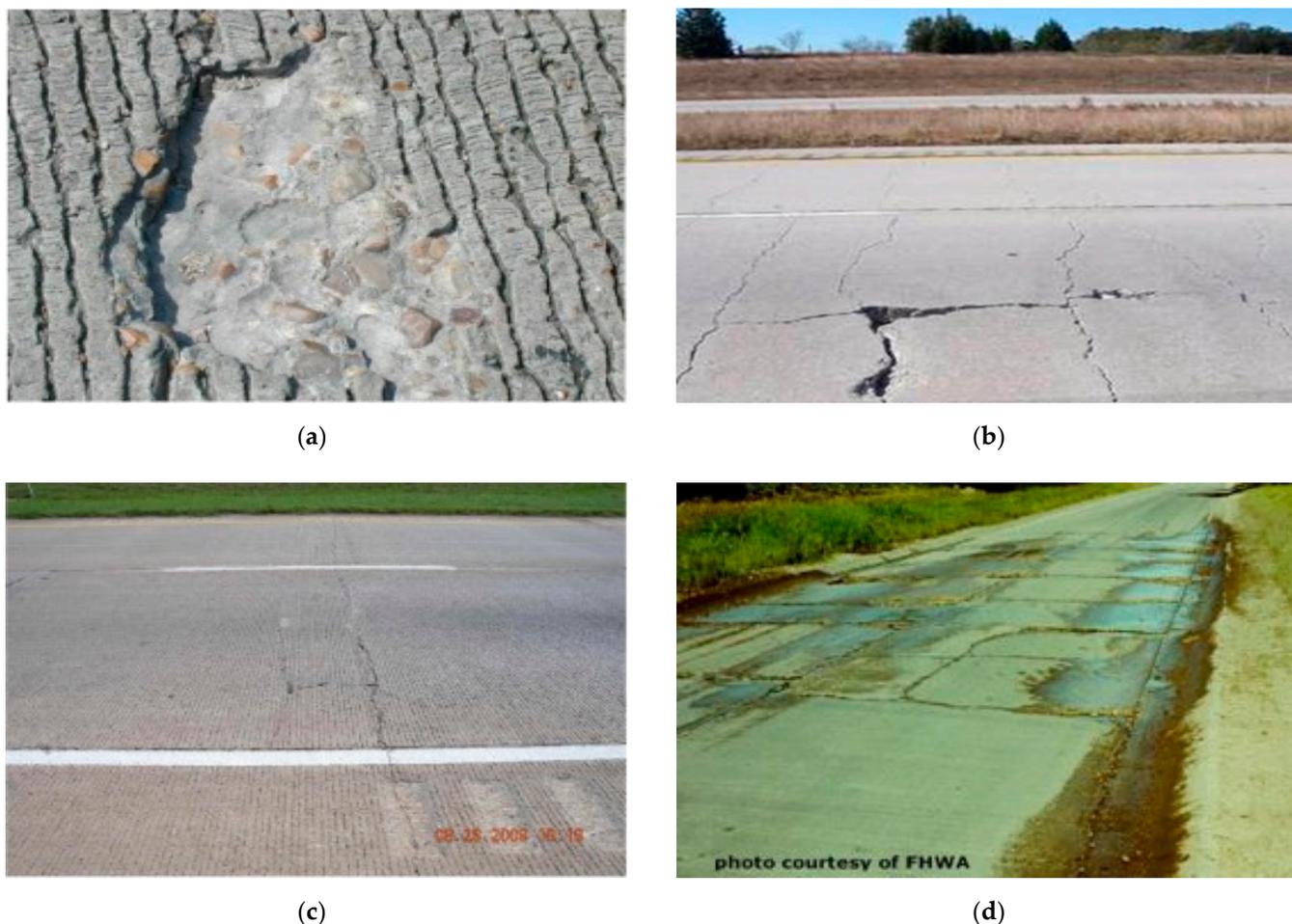


Figure 2. (a) Spalling, (b) punchouts, (c) wide longitudinal cracks, (d) pumping and erosion [2,31,32].

Among them, spalling is considered to bring in the most inimical effects to the pavements due to shear delamination of hardened concrete at the vicinity of transverse cracks. This ruinous scenario continues to grow in the longitudinal direction also with the increase in traffic and other unfavorable climatic conditions, resulting in larger spall areas [1,2,33]. Punchouts are considered to be one of the dominant parameters in the design of concrete pavements in the USA according to mechanistic-empirical (ME) design framework and the number of punchouts should be under 10 to meet satisfactory pavement requirements of CRCP [2]. These distresses create large faulty areas, such as wedges or blocks, between two transverse cracks or longitudinal cracks due to the loss of support in layers beneath [1,34]. Similar to spalling, these punchouts continue to extend with the increase in traffic and unfavorable conditions, making them harmful to the whole pavement layer from the bottom. To minimize this, reinforcement should be adequately provided while giving attention to mix design and the types of aggregates being selected to enhance the load transferring mechanism of the pavement. In terms of transverse cracks, they are allowed appear in CRCPs at desired spacings. The cause behind this is that the tensile stresses build up during the volumetric change of concrete when it hardens. These cracks continue to grow up to the edges and cause malignant effects to pavement due to intrusion of deleterious material into

the pavement. It can get worse, with additional traffic and environmental loadings causing longitudinal cracks to start as well. To mitigate the effects of transverse cracks growing bigger and undesired spacings appearing, slab effective depths, concrete curing and proper constructional practices are important [2]. Erosion of support layers, high deformations in the pavement due to heavy traffic and availability of free water in the pavement can cause bleeding of concrete where the free water tends to come on to the surface. This is termed as pumping and this can cause significant deformations in the pavement [35]. As soon as the cracks are initiated, reinforcement corrosion will start, followed by the water intrusion into the CRCP slab [1,2,5]. Reinforcement corrosion will result in reducing the tensile capacity of the steel bars, leading to various pavement distresses, as discussed above.

It is understood that performance of CRCP can greatly decrease due to the above distresses. It is of high importance to mitigate or delay these distresses to achieve high performance characteristics in CRCP. In fact, most of these distresses occur due to inherent flaws of concrete, such as low tensile and flexural capacity of concrete, which have been well identified. There are numerous studies on fiber intrusion or addition of graphene-based nanomaterials to the concrete in order to gain superior characteristics over the persisting flaws discussed above. Investigating how these methods are applicable to concrete pavements to mitigate or procrastinate these distresses getting worse by improving the flaws of concrete discussed above seems to be worthy and beneficial in many ways. Most of those studies maintain their conclusions as mixing various types of fibers and additions with desired dosages gives promising solutions in terms of flexural strength, compressive strength, tensile strength enhancements and, also fatigue and durability enhancements [3–7,9–12]. Also, few studies report the feasibility of using graphene-based nanomaterials as anticorrosive substances for steel reinforcement, which will address the reinforcement corrosion issues [34,36–38]. In the present study, in particular, how graphene and graphene oxide can be used to mitigate these issues is investigated through analyzing the past studies in context. Therefore, firstly, how graphene and graphene oxide can improve the properties of cement matrix was investigated and the conclusions were merged to the issues in CRCP, which will be discussed in the later sections.

3. Graphene-Based Nanomaterials in Cement-Based Composites

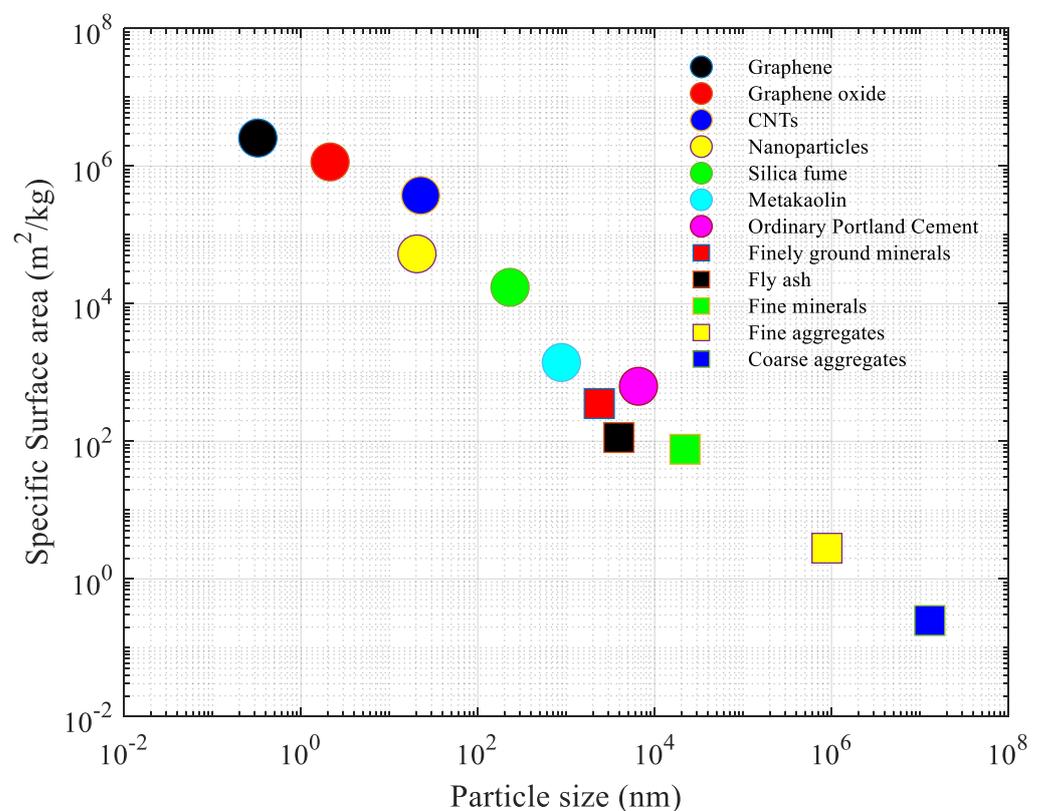
3.1. Graphene and Its Nature

Graphene, known to be thinnest material which has ever been found, consists of a one-atom-thick honeycomb two-dimensional lattice structure, and is an allotrope of carbon [20]. Graphene was first found by two of the prominent researchers, Andre Geim and Konstantin Novoselov, in 2004. Bulk graphite was exfoliated until the aforementioned 2D, one-atom-thick honeycomb lattice structure was obtained [16]. Since then, graphene has experienced a considerable growth in its use in the cement and concrete industry due to the extraordinary advantages over plain concrete [15]. Compared to other nano fibers, such as carbon nano tubes (CNT) and carbon fiber, graphene and oxide exhibit high elastic modulus, tensile strength, a greater surface area and an aspect ratio. Not only that, but they also offer a significant advantage over traditional fibers, such as synthetic, steel and glass, when mixed into cement composites. They have higher strength and aspect ratio, as mentioned before, nanoscale fiber spacings, better dispersion, improved hydration characteristics and other multifunctional benefits due to their conductive properties [15,16]. These properties are summarized in the Table 1 for more clarity.

Further, the approximate sizes of the materials that are usually employed within the concrete matrix and the sizes of graphene and graphene oxide can be compared in the following Figure 3. It is seen that graphene and graphene oxide hold a greater surface area and a smaller size, which is very beneficial in gaining a favorable pore structure in concrete matrix [14,19,27].

Table 1. Material properties of various fibers [17,18,22,27,28].

Material	Elastic Modulus (GPa)	Tensile Strength (GPa)	Elongation at Break (%)	Density (kg/m ³)	Diameter (nm)	Surface Area (m ² /g)	Aspect Ratio
Graphene	1000	130	0.8	2200	0.08	2600	6000–600,000
Graphene Oxide	23–42	0.13	0.6	1800	0.67	700–1500	1500–45,000
Carbon Nano Tubes	950	11–63	12	1330	15–40	70–400	1000–10,000
Carbon fiber	7–400	0.4–5	1.7	1770	6000–20,000	0.134	100–1000
Synthetic (Polypropylene and Nylon)	3–5	0.3–0.9	18	900	18,000–30,000	0.225	160–1000
Glass fiber	72	3.45	4.8	2540	5000–10,000	0.3	600–1500
Steel fiber	200	1.50	3.2	7800	50,000–90,000	0.02	45–80

**Figure 3.** Different particle sizes and specific surface area of materials employed within a concrete matrix [18].

There is plenty of research in which the special features of graphene or graphene oxide have been incorporated into the cement composites, such as concrete or mortar, to enhance their inherent flaws and, also, to add multifunctional benefits. Figure 4 shows graphite, which is composed with several graphene layers, graphene oxide and reduced graphene oxide. When it comes to the concrete pavements, graphene oxide, which is oxidized graphene sheet, has been widely used due to its better dispersion characteristics in the composite mix due to oxygen functionalities. Graphene oxide is chemically converted to make reduced graphene oxide, where the oxygen amount is low while having high conductivity. Reduced graphene oxide might be very helpful in smart pavements and cases where health monitoring through smart sensing is required [15].

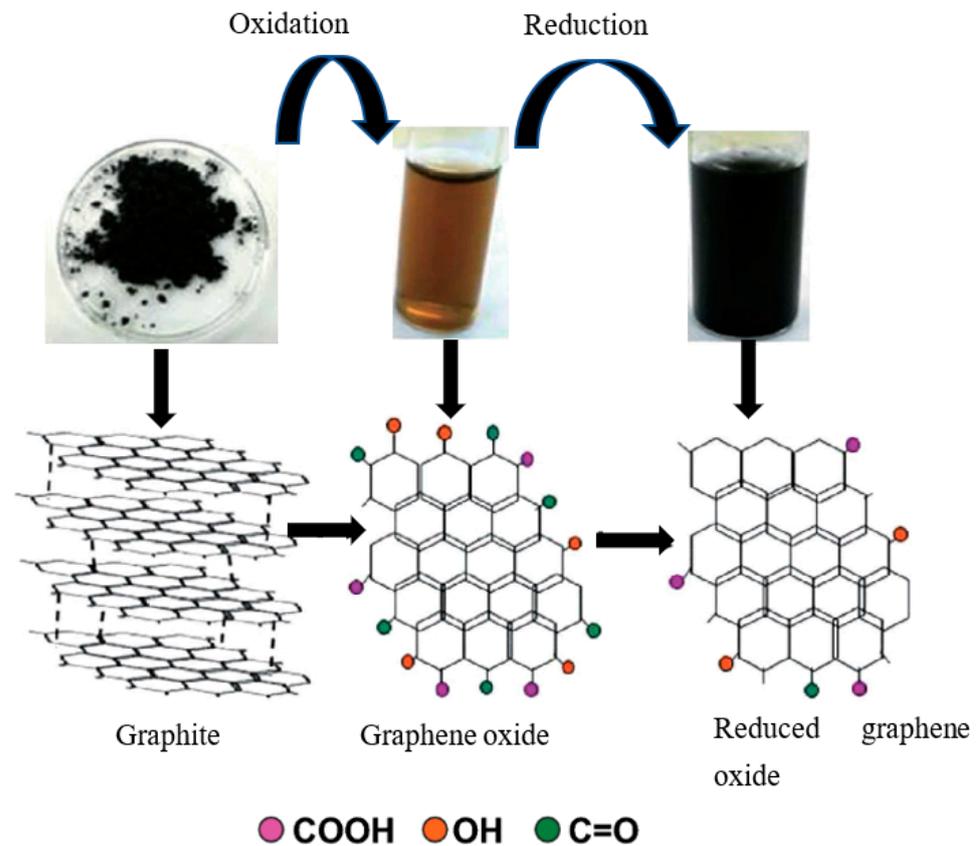


Figure 4. Graphite composed with graphene sheets, graphene oxide and reduced graphene oxide ([39]).

Moreover, some of the major potential benefits attributed to use of graphene in cement-based composites can be briefed out as: (a) reducing the amount of cement with the addition of very low dosages of graphene to reach the equivalent performance of mechanical properties [13–15,40,41], (b) increased resistance to severe environment and climatic conditions, such as freeze and thaw [23,42] (c) pseudo resistivity, which can be used in smart infrastructure [43], (d) electromagnetic interference (EMI) shielding for health- and security-related benefits [16], (d) heat sink capacity and greater thermal diffusivity for controlling cracks, (e) low cost when produced in bulk [16], and (f) sustainability achieved through low carbon dioxide equivalents [44]. One of the most important aspects that has to be considered is the dispersion of graphene or graphene oxide in the cement because poor dispersion can significantly reduce the performance of the matrix in terms of all benefits discussed above and can be worse [45]. Mainly there are three dispersion methods in use according to the past studies, such as dry dispersion, wet dispersion technique with the use of a surfactant and wet dispersion technique with a surfactant and ultrasonication. In dry dispersion, the cement and graphene are mixed in a concrete mixer. Although the method is easy, it is considered to be not effective due to the poor dispersion due to agglomeration of graphene [16]. The wet dispersion technique in water by means of mechanical stirring with a surfactant is identified to be more effective than wet dispersion, since the surfactant helps to break the Van der Waals forces between graphene layers, allowing more dispersion. However, in the third method, which is an improved version of the second method due to additional ultrasonication, the van der Waals forces break and tend to disperse graphene in aqueous solution. Surfactants generally used for this process are polycarboxylate, melamine, polyacrylic and sulfonate-based surfactants. It is important to note that overdose of surfactants and over-ultrasonication can cause serious performance losses in the concrete composite [45–47].

3.2. Scientometric Analysis of Past Studies

A comprehensive literature review was conducted on the graphene and graphene oxide in the context of cement composites and their various attributes. Graphene and graphene oxide synthesis, dispersion, mixing, pore structure of graphene-based cement composites, strength properties, durability and applications were investigated through journal articles. Web of Science (WoS) database was used to perform a bibliometric analysis and the search string used in the analysis is given below in Table 2. It should be noted that the review is performed to draw conclusions for graphene-based cement composites to use in concrete pavements. Therefore, articles which include graphene-based asphalt concrete were excluded. Articles that include strength properties were deliberately included into the search string because the concrete should satisfy strength requirements predominantly to successfully use it in the pavements. VOSviewer software was used to compare and observe various trends and occurrences of the keywords related to the above-mentioned literature. A total of 608 journal articles from 2010 to 2022 were selected from WoS and a network visualization diagram for co-occurrences was created using VoSviewer, as shown in Figure 5. A network visualization diagram includes different labels, and the size of the label (circle) represents the weight of the item. The bigger the label, the larger the number of co-occurrences the item holds. The color of an item represents different clusters. The analysis here includes 195 items and 7 different clusters with 5755 links. According to the network visualization shown below, major keywords or items appearing in the context are graphene, graphene oxide, microstructure, and strength, which indicates that those are the most researched keywords. Various other keywords are linked to those major keywords. It is important to note that, when the search string was modified by including “pavements” in the search string, it yielded only less than 10 journal articles, which indicates the significance and the potential for future research of graphene-based cement composites in concrete pavements.

Table 2. Search string used for the bibliometric analysis.

Year	Sources	Search String
2010–2021	Web of Science (All Fields)	(“Graphene”) AND (“Cement”) AND “Strength” NOT (“Asphalt”)

3.3. Effect of Graphene and Graphene Oxide on Pore Structure of Cement Composites

Graphene and GO being nanofillers offer a significant positive impact on refining the pore structure for a desirable mix. According to the experimental studies mentioned below, graphene-based nanofillers decrease the total porosity and help obtain a finer particle distribution and pore distribution. Gong et al. [15] studied that 0.03% by weight of graphene oxide has decreased porosity by 13.5%, while the degree of hydration has enhanced. The study concluded that graphene oxide enriches the pore structure to enable better hydration. Simultaneously, the mean pore diameter has drastically reduced by 36.7%, which is an excellent proof to the above. Wang et al. [46] investigated the phase analysis of graphene-included cement paste and found that hydration reaction is faster to create more hydration products in the early stage. In a similar study, Wang et al. [48] studied about the pore structure of graphene-based cement composites employing mercury intrusion porosimetry (MIP) and discovered that the median volume pore diameter and the total porosity decrease with increasing doses of graphene. However, a decrease in these parameters was observed when graphene dosage reached 0.15%, which might be due to poor dispersion of graphene in the cement matrix. Lv et al. [49] conducted several tests on pore structure incorporating graphene oxide cement composites and experienced more than 50% reduction in porosity compared to the control sample made without graphene oxide. Further, the results reveal that total pore area, median pore diameter and average pore diameter were also reduced. Du et al. [24] investigated the water sorptivity of graphene-based concrete, which is an indicator of pore structure and experienced a significant reduction in sorptivity compared

increases, as is shown in Figure 6, in which the results are based on the studies conducted by Akarsh and Bhat (2021) [14] and Chen et al. (2019) [23], Lu and Ouyang (2017) [53], Wang et al. (2016) [54], Murugan et al. (2016) [55], Jing et al. (2016) [51], and Chen et al. (2019) [23]. Figure 6 illustrates the normalized slump value calculated by dividing the slump value of a concrete mix at a certain graphene-based nanomaterials dosage by the slump value of the same concrete mix without any graphene materials dosage. Therefore, it is evident that all the normalized values lie below the region $y = 1$. A general trend line is also illustrated using a linear fit for the data to observe the decrease in the slump with increase in graphene dosage.

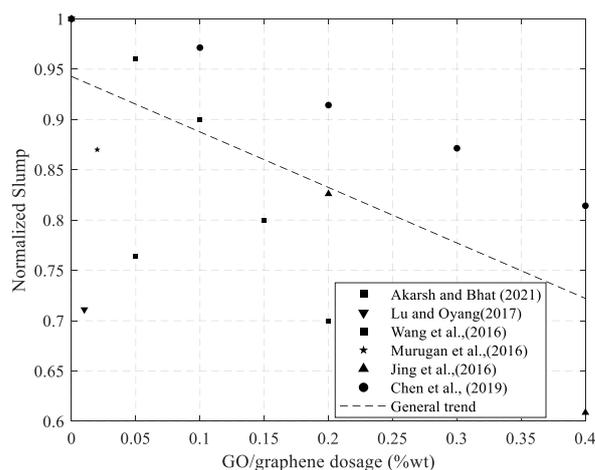


Figure 6. Normalized slump variation with GO/graphene dosage [14,23,51,53–55].

3.5. Effect on Hardened Cementitious Composites

3.5.1. Mechanical Properties; Flexural, Tensile, Compressive Strength and Fatigue Resistance

Even though the workability tends to reduce with the addition of graphene and graphene oxide, the strength gain of cement composite with a tiny percentage of those additions is remarkable. When the previous studies are investigated, this is evident and, mostly, those studies have utilized conventional strength tests, such as cube test for compression, three-point bending test for flexural strength and split tensile test of tensile strength. Gong et al. [15] concluded that, with the addition of graphene oxide in 0.03% by weight, compressive strength increased by 46% and tensile strength by 50%. Akarsh et al. [19] recently investigated the effect of concrete with graphene oxide, M sand and silica fume, where the study concludes enhanced compressive strength, flexural strength, tensile strength and fatigue resistance. The study states the importance of using silica fume in the cement matrix, as the increased dispersion of graphene oxide is due to this. The scenario was ascribed to (a) the reaction of silica fume with excess calcium hydroxide, which enables more stabilization of graphene oxide, and (b) weakening of van der Waals forces in graphene sheets to allow more dispersion.

Compared to the specimens with 0.15% by weight of graphene, the specimens of 0.15% graphene with 7% silica fume have increased the compressive, tensile and flexural strengths significantly. According to Akarsh and Bhat [14], strength of concrete greatly depends on the pore structure of the concrete at micro and nano levels. Accompanying these pores with reactive or nonreactive materials can significantly enhance strength, which happens with graphene oxide in cement due to its smaller size and larger surface area, in which the pores are filled with dense hydration products [14,19,40]. Moreover, there is much research which has investigated the effect of graphene in the cement paste, mortar and concrete. The studies are distinct to each because of the use of various dosages of superplasticizers, different ultrasonication types and different graphene dosages. However, overall, most studies have experienced a considerable strength gain with the addition of graphene or graphene oxide [41,42,46,47,53–94] and the past results are analyzed in the present paper to observe the behavioral aspects attributed to strength. Wang et al. [46] investigated

the effect of graphene in cement paste by adding 0.05% by weight of graphene with dispersants, superplasticizers, deformers and ultrasonication to reach 3–8% enhancement in compressive strength and 15–24% in flexural strength. Likewise, Liu et al. [47] examined the cement paste with addition of 0.06% by weight with the same dispersion techniques but without adding superplasticizers to reach 14.9% and 23.6% strength gains in compressive and flexural strengths, respectively, while obtaining a 15.2% enhancement in tensile strength. Further, the consequent studies by Wang in 2018 and 2019 have given the same trend results, with strength improvements up to 27.4% and 25.2% in terms of compressive and tensile strengths, respectively. The studies have employed various water-to-cement ratios as well [18,40]. Sixuan et al. [57] investigated the addition of graphene in mortar and cement paste and came up with 82% enhancement in flexural strength in the paste for 0.05% by weight of graphene, while 20% and 23% improvements were observed in compressive and flexural strengths, respectively, for 0.5% by weight fraction of graphene. Matakah et al. [58] experimented the use of graphene in concrete with the addition of 0.16% by weight with mechanical stirring and ultrasonication dispersion techniques to gain a 13.9% compressive strength improvement. In addition to that, interestingly, when the strain and toughness are considered, Gong et al. [15] concluded that the addition of graphene oxide not only increases the failure stress, but also the failure strain compared to the plain cement. Basically, it can be concluded that, with the addition of graphene oxide, the toughness of the composite increases.

3.5.2. Analysis of Mechanical Properties

When the mechanical properties of cement composites made with graphene or graphene oxide, such as compressive, flexural and tensile strengths, are considered, there seem to be correlations to estimate the effect of these additions to the strength characteristics of the concrete. Experimental results from 54 studies were taken to the analysis and the results seem scattered due to various other aspects, such as type of graphene, superplasticizer, water-to-cement ratio, aggregates, etc. To overcome this issue, all the results were normalized by the respective property corresponding to no dosage of graphene or graphene oxide. In spite of observing the effect of graphene and graphene oxide separately, the experimental data were categorized accordingly. Figure 7a–c depict the variation of strength properties with graphene oxide and Figure 8 illustrates the same for graphene. The dotted line illustrates the phenomena of normalized strength equal to 1, which, in other words, represents no change to the strength properties. The sources of data are given in Appendix A.

Here, f_c , f_f , and f_t stand for the compressive strength, flexural strength and tensile strength of graphene-oxide-based cement composites.

It is evident that GO-based cement composites reach higher normalized strength, even during very low dosages, compared to graphene-based cement composites. Also, with the existing data, it can be observed that GO enhances the strength characteristics better compared to graphene.

Here, f_c , f_f , and f_t stand for the compressive strength, flexural strength and tensile strength of graphene-based cement composites.

3.5.3. Correlation between Strength Properties

It is always better to come up with relationships and derivate functions of various parameters to quantify or measure how graphene and GO may affect cementitious composites. To determine more precise relationships, the data were analyzed for graphene and GO separately utilizing 28 day compressive, flexural and tensile strengths. Statistical linear regression analysis, which employs a linear approach to model relationships between dependent and independent variables, was performed utilizing the least squares technique. Least squares technique makes the best fit line for the data points while making the vertical difference between the original data and fitted data (error) to a minimum. The accuracy of the prediction can be understood by the variance, which is the sum of squares of the errors [95]. The present study reports better values for variance for each analysis. More-

over, a fitted line for the experimental results was used to compare the existing strength relationships provided by ACI 318 [96] and AS 3600 [97]. The sources of data are given in Appendix A.

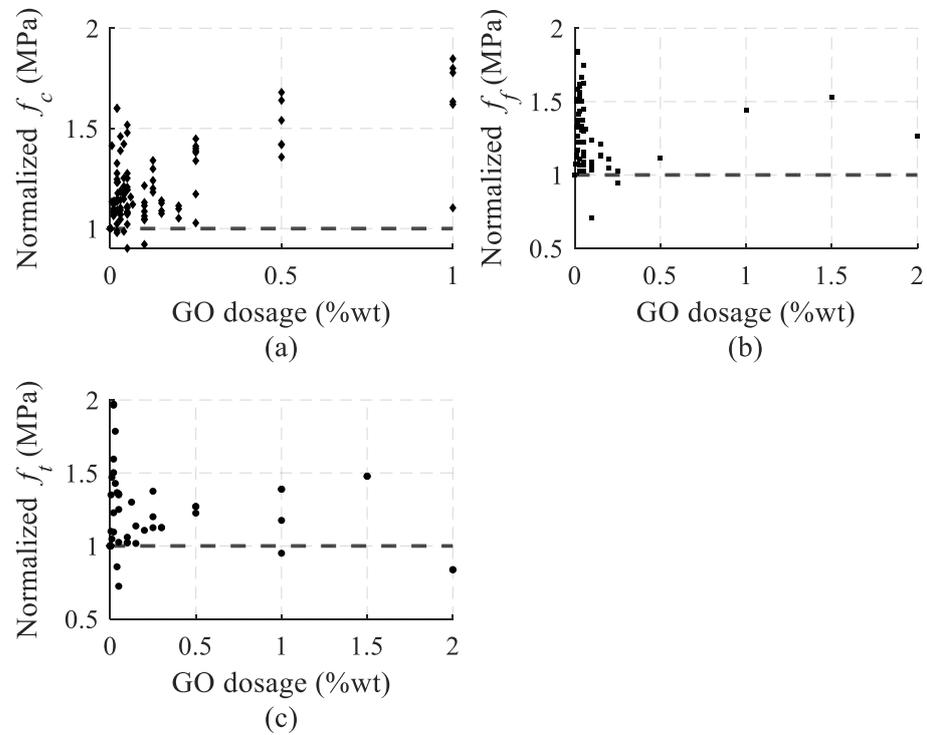


Figure 7. Normalized strengths for various graphene oxide dosages, (a) normalized compressive strength, (b) normalized flexural strength, and (c) normalized tensile strength.

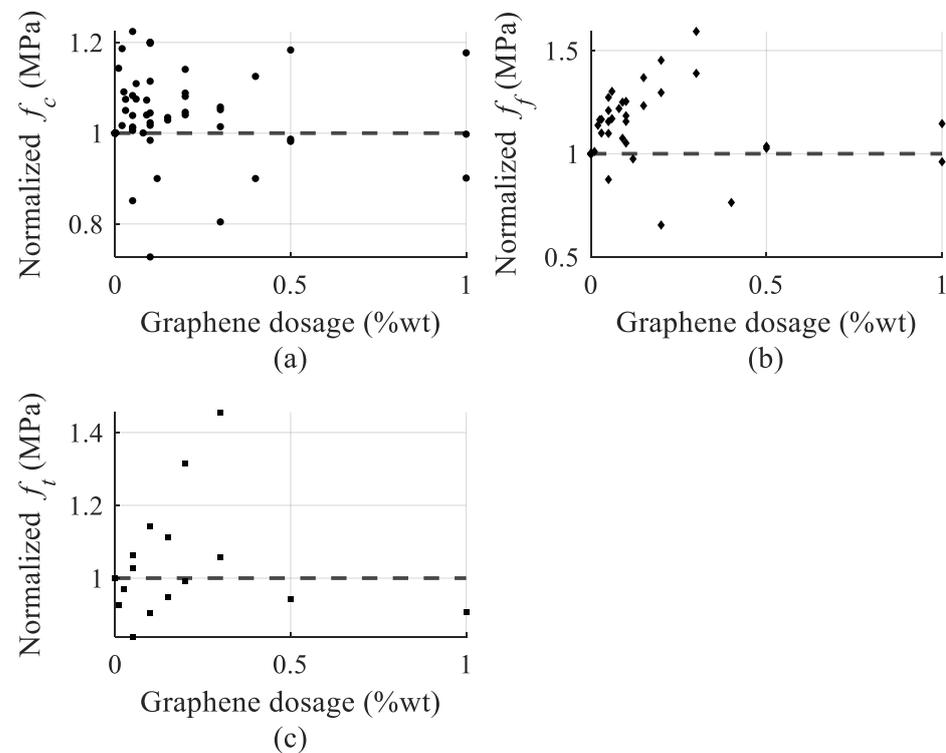


Figure 8. Normalized strengths for various graphene dosages, (a) normalized compressive strength, (b) normalized flexural strength, and (c) normalized tensile strength.

- (a) Correlation between compressive strength (f_c) and flexural strength (f_f) related to GO-based cementitious composites

ACI 318 and AS 3600 predict the relationship between flexural (f'_f) and compressive (f'_c) strengths for normal concrete, as per the following Equations (1) and (2), respectively:

$$f'_f = 0.62 \sqrt{f'_c} \quad (1)$$

$$f'_f = 0.6 \sqrt{f'_c} \quad (2)$$

The derived equation for GO-based cementitious composites from the existing experimental data is given in Equation (3) below. It should be noted that the derived equation holds an R^2 value of 0.93, which is quite better for a reasonable statistical estimation.

$$f_f = 1.19 \sqrt{f_c} \quad (3)$$

When the slope of the equations is considered, it is evident that GO-based cementitious composites give higher flexural strengths for a certain compressive strength compared to the normal concrete, which depicts the increase in flexural strength due to GO. The analysis is graphically illustrated in Figure 9a below. Figure 9a illustrates the scatter plot of experimental data, ACI 318 prediction, AS 3600 prediction and the fitted line for the experimental data. It highlights the fact that there is a considerable difference in flexural strength when GO is added to cement composites. Similarly, the same effect was observed for cementitious composites with GO mixed with other additions, such as CNT and steel fibers, and it is evident that the slope of the derived equation is higher than that without other additions, as shown in Figure 8. The derived equation for the case of GO mixed with additions is shown in Equation (4) below. The prediction is characterized by an R^2 value of 0.84 and shows a higher slope compared to that without other additions, which anticipates that the hybrid form of graphene and other additions gives improved flexural strengths.

$$f_f = 1.50 \sqrt{f_c} \quad (4)$$

- (b) Correlation between compressive strength (f_c) and tensile strength (f_t) related to GO-based cementitious composites

ACI 318 and AS 3600 predict the relationship between tensile (f'_t) and compressive strengths (f'_c) for normal concrete as per the following Equations (5) and (6), respectively.

$$f'_f = 1.50 \sqrt{f'_c} \quad (5)$$

$$f'_t = 0.4 \sqrt{f'_c} \quad (6)$$

The derived equation for GO-based cementitious composites from the existing experimental data is given in Equation (7) below. It should be noted that the derived equation holds an R^2 value of 0.87, which is, again, quite better for a reasonable statistical estimation.

$$f_t = 0.53 \sqrt{f_c} \quad (7)$$

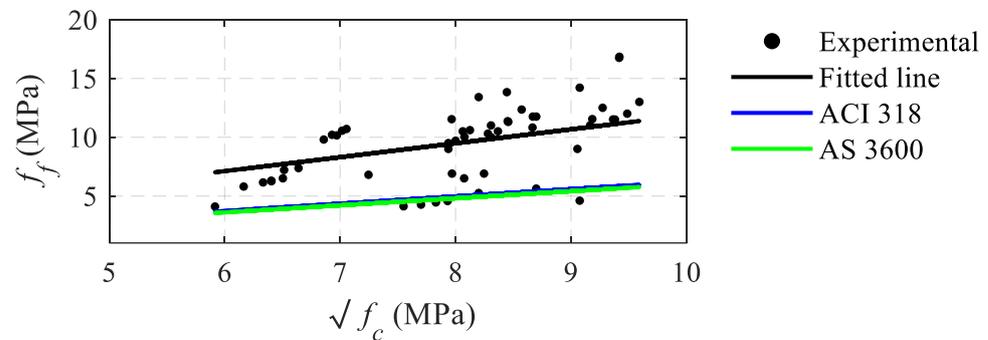
When the slope of the equations is considered, it is evident that GO-based cementitious composites give conservative strengths for a certain compressive strength, in which the slope is higher than that of AS and lower than that of ACI. The analysis is graphically illustrated in Figure 9b below.

- (c) Correlation between flexural strength and tensile strength related to GO-based cementitious composites

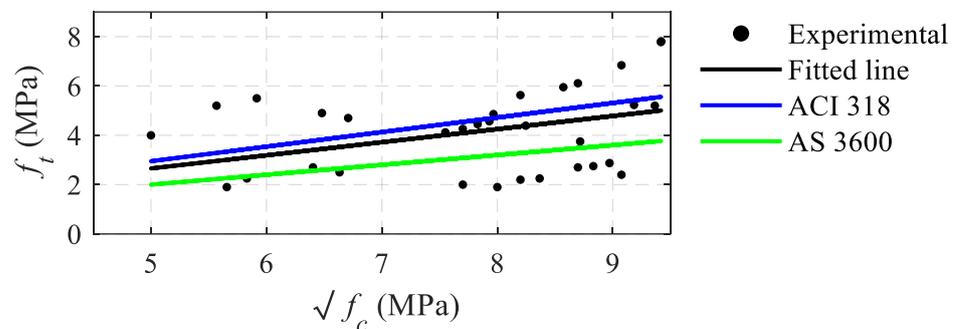
From the statistical analysis, it can be concluded that tensile strength prediction is almost a half of flexural strength for GO-based cementitious composites. The derived

equation can be interpreted as given in Equation (8) with an R^2 value of 0.95, while the relationship is graphically illustrated in Figure 9c.

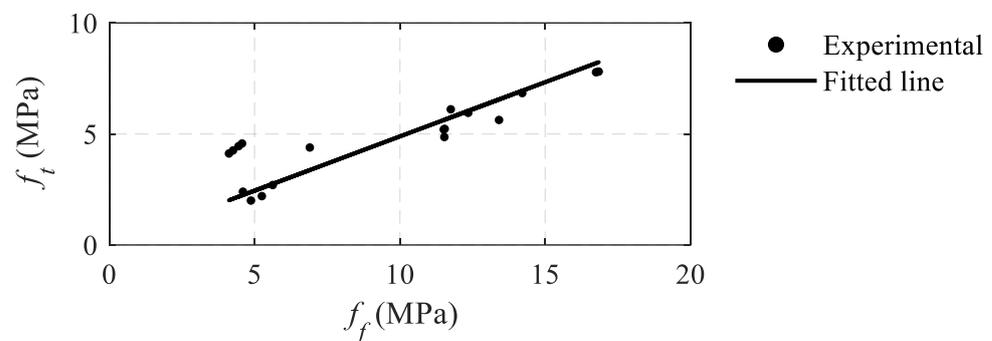
$$f_t = 0.49f_f \quad (8)$$



(a)



(b)



(c)

Figure 9. Variation of (a) f_f vs. $\sqrt{f_c}$, (b) f_t vs. $\sqrt{f_c}$, and (c) f_t vs. f_f , for GO-based cementitious composites.

(d) Correlation between compressive strength (f_c) and flexural strength (f_f) related to graphene-based cementitious composites

ACI 318 and AS 3600 predict the relationship between flexural (f'_f) and compressive (f'_c) strengths for normal concrete as per the following Equations (1) and (2), as given above.

The derived equation for graphene-based cementitious composites from the existing experimental data is given in Equation (9) below. It should be noted that the derived equation holds an R^2 value of 0.92, which is quite better for a reasonable statistical estimation.

$$f_f = 1.13 \sqrt{f_c} \quad (9)$$

When the slope of the equations is considered, it is evident that graphene-based cementitious composites give higher flexural strengths for a certain compressive strength, which depicts the increase in flexural strength due to graphene. The analysis is graphically illustrated in Figure 10a below.

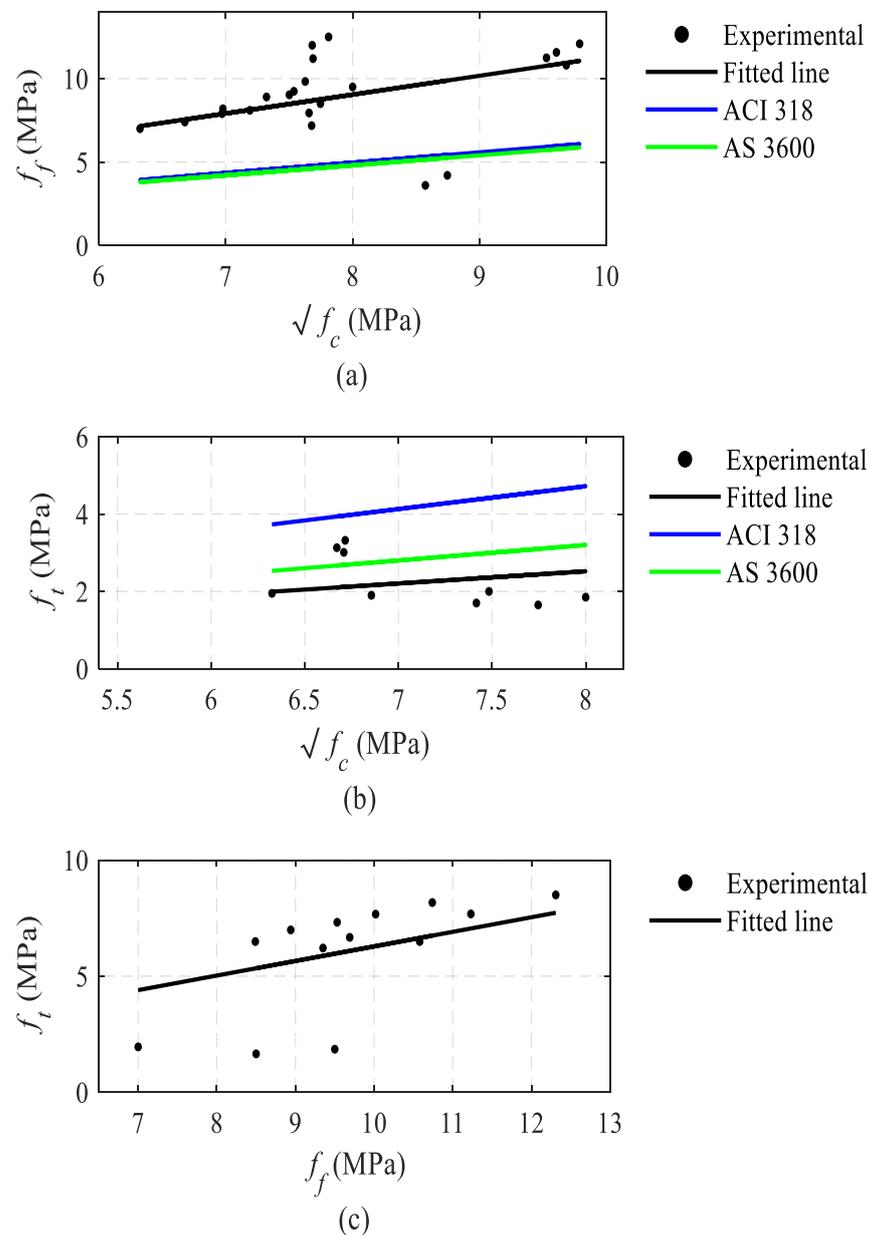


Figure 10. Variation of (a) f_f vs. $\sqrt{f_c}$, (b) f_t vs. $\sqrt{f_c}$, and (c) f_t vs. f_f , for graphene-based cementitious composites.

(e) Correlation between compressive strength (f_c) and tensile strength (f_t) related to graphene-based cementitious composites.

ACI 318 and AS 3600 predict the relationship between tensile and compressive strengths for normal concrete as per the Equations (5) and (6), as given above.

The derived equation for graphene-based cementitious composites from the existing experimental data is given in Equation (10) below. It should be noted that the derived equation holds an R^2 value of 0.96, which is, again, quite better for a reasonable statistical estimation.

$$f_t = 0.32\sqrt{f_c} \quad (10)$$

When the slope of the equations is considered, it is evident that graphene-based cementitious composites give conservative strengths for a certain compressive strength, in which the slope is higher than that of both AS and ACI. The analysis is graphically illustrated in Figure 10b below.

- (f) Correlation between flexural strength and tensile strength related to graphene-based cementitious composites

From the statistical analysis, it can be concluded that tensile strength prediction is almost two thirds of the flexural strength for graphene-based cementitious composites. The derived equation can be interpreted as given in Equation (11), with an R^2 value of 0.91, while the relationship is graphically illustrated in Figure 10c.

$$f_t = 0.63f_f \quad (11)$$

- (g) Summary of results for graphene- and graphene-oxide-based cementitious composites.

The variations discussed above can be tabulated as a summary in Table 3. In general, f_c , f_f , and f_t stand for compressive strength, flexural strength, and tensile strength of concrete with graphene- or graphene-oxide-based additions and f'_c , f'_f , and f'_t stand for compressive strength, flexural strength, and tensile strength of concrete derived in ACI and AS guidelines.

Table 3. Summary of strength relationships for GO- and graphene-based cementitious composites.

Relationship	Derived		Codes	
	GO	Graphene	ACI	AS
Flexural strength (f_f) and Compressive strength (f_c)	$f_f = 1.16\sqrt{f_c}$	$f_f = 1.13\sqrt{f_c}$	$f_f = 0.62\sqrt{f_c}$	$f_f = 0.6\sqrt{f_c}$
Tensile strength (f_t) and Compressive strength (f_c)	$f_t = 0.53\sqrt{f_c}$	$f_t = 0.32\sqrt{f_c}$	$f_t = 0.59\sqrt{f_c}$	$f_t = 0.4\sqrt{f_c}$
Tensile strength (f_t) and flexural strength (f_f)	$f_t = 0.49f_f$	$f_t = 0.63f_f$	$f_t = 0.95f_f$	$f_t = 0.67f_f$

It is seen in the summary that GO-based cementitious composites offer higher flexural and tensile strength values for a given compressive strength compared to that of graphene.

3.5.4. Estimation of Optimum Dosage Range

Even though it is difficult to come up with an ideal optimum graphene/GO dosage that should be used to obtain the best results due to various other effects that cementitious composites rely on, behavioral aspects of these cementitious composites during different dosage ranges can be roughly estimated. To do that, the maximum normalized compressive strengths from each and every study attributed to a certain dosage were selected. The strength values attributed to a certain dosage were then averaged to represent a single maximum strength for a particular dosage value. While the behavior is identified for compressive strength, it can be extended to flexural and tensile strengths with the use of the above derived relationships approximately. Figure 11a–c illustrate the variation of normalized compressive strength, flexural strength and tensile strength with graphene/GO dosage, and the variation seems quite complex. The blue line in Figure 11a–c shows the normalized averages for compressive strength, flexural strength and tensile strength, respectively, for different dosages. It should be noted that, in this analysis, graphene and graphene oxide were considered together due to similar responses. According to Figure 11, it can be again concluded that, basically, addition of graphene or graphene oxide enhances the strength characteristics because almost all the points lie above 1, which represents the normalized strength for no additions. As far as Figure 11a is concerned, it is evident that, during very low dosages up to 0.06%, the strength characteristics fluctuate, and the maximum of the fluctuation is 0.03 %wt dosage. When the dosage is increased between 0.06 and 0.4%, a slightly increasing response can be observed. When the dosage is higher than 0.4% but less than 1%, the response keeps increasing. It does not mean

that, when the graphene or graphene oxide dosage increases forever, we may experience higher strength characteristics but lower strength characteristic due to low dispersion and agglomeration effects discussed earlier. However, due to the high scatteredness of the data, an accurate prediction or an expression for the optimum dosage is hard to define from Figure 11. But it can be concluded that addition of graphene or graphene oxide brings strength improvements for certain dosage levels.

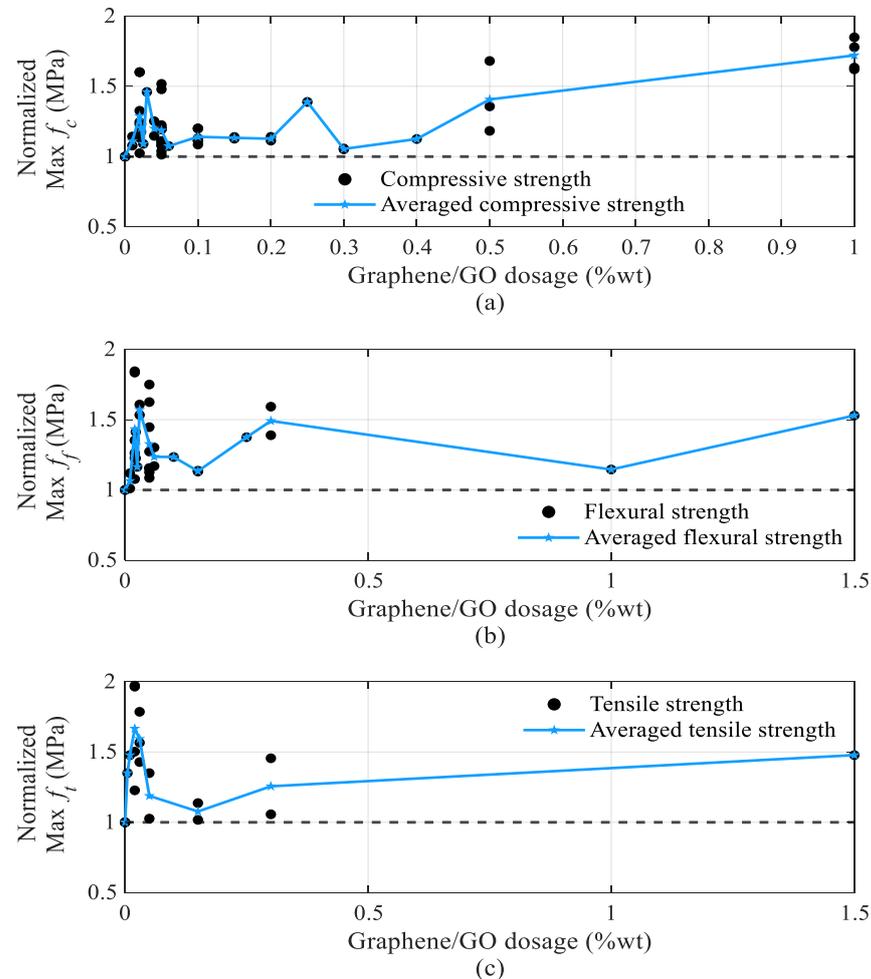


Figure 11. (a) Average normalized maximum f_c , (b) f_f and (c) f_t variation with GO/graphene dosage.

It should be noted that these fibers interact in the nano scale of the concrete, while the above discussed fibers interact in the micro scale [13]. These nano-scale fibers offer comparatively high strength, stiffness and aspect ratio. One of the dominant aspects in graphene over other traditional fibers is its smaller fiber spacing, which restrains developing cracks from the nano scale so that those cracks would not propagate up to the micro level [14,17,24]. Wang et al. [46,54,59] and Ho et al. [91] drew out the possible mechanisms for these strength enhancements after numerous studies on the context. They can be accredited to (a) the effect of filling courser pores and converting them to finer pores, (b) increased hydration due to high surface area and improved dispersion, and (c) crack bridging effect offered by graphene in the nano level, as discussed above, where the formation of micro cracks can be delayed. However, there is no proper research found in the current state of the art to support graphene or graphene oxide in CRCP but limited work has been obtained in plain concrete pavements with satisfactory results corresponding to strength and durability [13,14,92].

3.6. Durability Aspects

Durability of concrete and other cementitious composites in terms of pavements are highly important in several ways, such as cost, material sustainability, ride comfort and efficiency. Even though whatever addition enhances the strength characteristics, if the durability properties are low in performance, enhancements are invalidated and not beneficial. However, past studies have proven results for the durability enhancements in terms of graphene and GO cementitious composites. Mohammad et al. [92] investigated the pore structure enhancement due to the addition of GO and concluded that the volume of gel pores becomes higher than that of capillary pores, which gives a fine and dense microstructure which leads to improved resistance to water, chloride and acid penetration. The research also concluded that GO improves durability against chloride penetration such that penetration depth reduced in 75%, and this difference can be clearly observed in Figure 12.

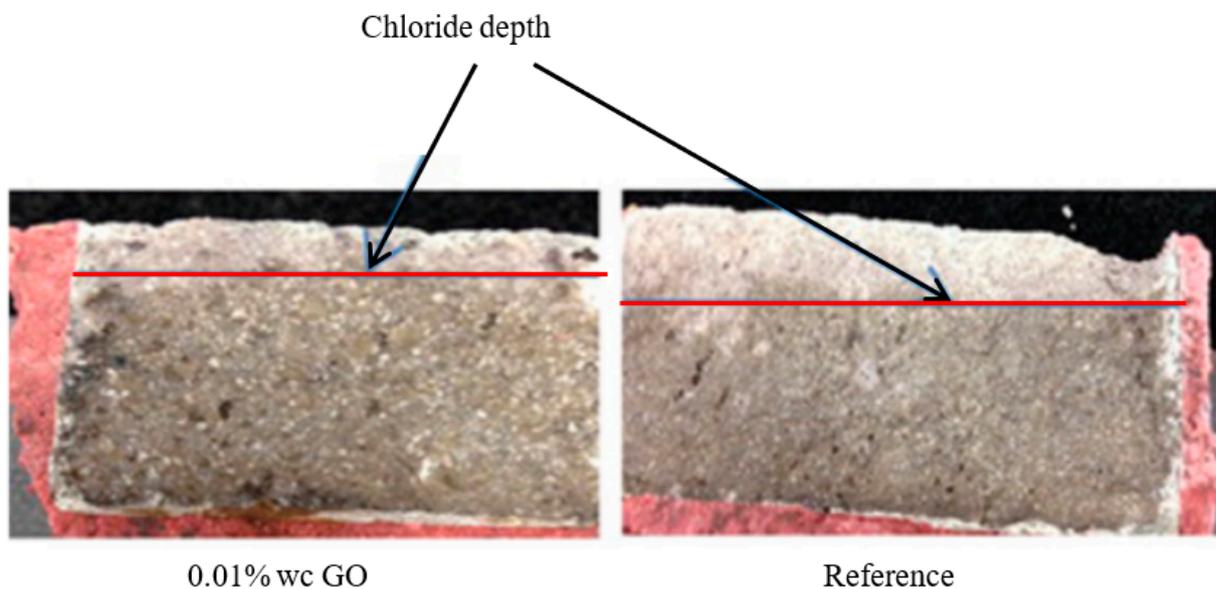


Figure 12. Reduction in chloride penetration due to GO addition. Adapted with permission from Ref. [92]. 2022, Swinburne University of Technology.

Gong et al. [15] studied that 0.03% by weight of graphene oxide has decreased porosity by 13.5%, while degree of hydration is enhanced. The study concluded that graphene oxide enriches the pore structure to enable better hydration. Simultaneously, the mean pore diameter has drastically reduced by 36.7%, which is an excellent proof to the above. Wang et al. [46] investigated the phase analysis of graphene-included cement paste and found that hydration reaction is faster to create more hydration products in the early stage. Mohammad et al. [42] investigated the freeze and thaw performance when GO is added to cement mortar and experienced a 50% reduction in weight loss when exposed to freeze and thaw. Mohammed et al. (2018) experienced 80% reduction in carbonation depth and explained that the enhancement is due to the ion interlocking on GO surface [98]. Du et al. (2016) investigated the effect of graphene on concrete and concluded 80% reduction in terms of chloride penetration due to tortuosity and pore enhancements [24]. Further, the research concluded that overdosage above 1.5% hinders the performance and drastically reduces the performance due to resulting poor pore structure, followed by the agglomeration of graphene. Akarsh et al. [19] studied acid attack and sulphate attack and fatigue performance of GO-based concrete. Results revealed that the addition of GO reduce the weight loss of specimen and decrease in the compressive strength when aging. The tested specimen, after 90 days, is shown below in Figure 13a. Even though the mechanisms behind it are not clear enough, the research concluded that the strength gain and the weight loss is due to the

damage that occurred to the hydration products, followed by the acid intrusion. In terms of the sulphate attack, the results maintained that GO-based concrete is less affected, as shown in Figure 13b. Studies on toughness and fatigue performance with the addition of graphene or graphene oxide are very low, while it can be predicted that toughness may be increased to high strains resulted in most tests. Fatigue-related durability obviously hugely attributes to pavements constructed with concrete. When the fatigue tests were performed for different stress ratios, Akarsh et al. [19] experienced no significant change in the number of cycles to failure when GO is incorporated into the concrete. The analysis was conducted utilizing S–N (stress–no. of cycles to failure) curves, which are used to characterize fatigue performance of a material. As shown in Figure 14, an almost similar number of cycles to failure can be observed for both concrete mixes. Results revealed that fatigue life does not increase significantly with the addition of graphene. Importantly, when the mix was combined with silica fume and graphene oxide, fatigue life was considerably increased.



Figure 13. (a) Resultant specimen of acid attack test and (b) sulphate attack test. Adapted with permission from Ref. [19]. 2022, Swinburne University of Technology.

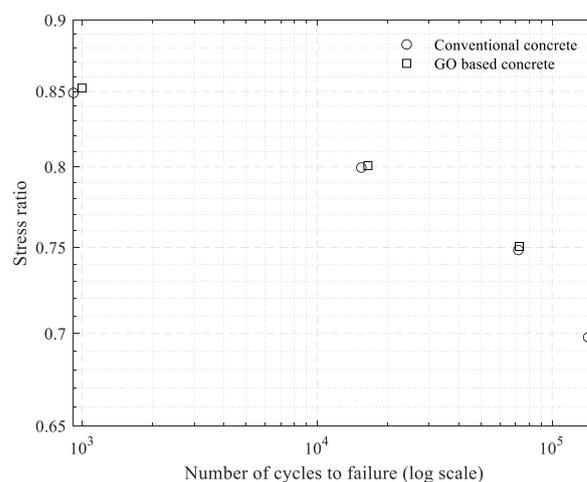


Figure 14. S–N relationship for conventional concrete and GO-based concrete. Adapted with permission from Ref. [19]. 2022, Swinburne University of Technology.

4. Application of Graphene-Based Nanofillers to CRCP

Even though research to date has used graphene-based nanofillers in cementitious composites, its usage in pavements and, particularly, in the context of CRCP is poor. However, typical issues can be anticipated, such as plastic and drying shrinkage cracking, poor steel bar performance, severe crack widths, ride quality, spalling and durability, as discussed below.

- (a) Plastic and drying shrinkage cracks: plastic and drying shrinkage cracks can appear in CRCP due to heat of hydration due to its dense mixture. Excellent heat sink capacity of graphene can significantly diffuse this heat of hydration, reducing the potential risks for plastic shrinkage cracking [99,100]. Not only that, but the improved pore structure, as discussed many times previously, imparted from graphene to concrete can significantly delay the crack initiation in the nano scale itself, which might be additionally beneficial for plastic shrinkage cracking.
- (b) Rebar performance: in terms of rebar performance, it is well documented above that intrusion of small dosages of graphene can considerably enhance the flexural strength, which rationalizes the idea that steel can have more room to take flexural loadings. Additionally, anticorrosive properties of graphene can reduce the steel corrosion, which will maintain the performance of CRCP as designed for a longer lifespan [34,36–38].
- (c) Crack width control: although transverse cracks are expected to occur due to volumetric changes in concrete when it hardens, if the widths are not controlled properly, they can cause cascade failures due to propagation [31,32,98]. Graphene can be anticipated to reduce these crack widths due to its pore bridging and uniform dispersion.
- (d) Ride comfort: basically, ride comfort is one of the prime concerns of a CRCP or any other pavement because it is one of the ultimate goals in pavement construction. Ride quality typically reduces due to various distresses discussed above. Among them, most distresses initiate due to crack propagation under high load or weather conditions [1,31,32,98]. Graphene's ability to delay crack initiation and improved resistance to tensile and compressive loads will be added advantages to keep the ride comfort as expected.
- (e) Spalling resistance: spalling is considered to be the most detrimental type of distress in CRCP, and it basically happens due to the shear delamination and debonding of matrix from the external and internal loadings. These spalls can grow bigger with increased traffic and can cause deep blow-ups. Graphene's improved bond structure is anticipated to provide resistance for these types of distress. Long-term monitoring of graphene-based pavements should be conducted [1,33].
- (f) Durability and life span: durability and life span of CRCP are basically governed by the fatigue performance of the pavement for continuous cyclic loadings occurring due to traffic. Akarsh et al. (2021) revealed that graphene oxide can considerably improve the fatigue performance of concrete, which will be very beneficial [19]. Apart from that, durability can be characterized by acid attack test, sulphate attack tests and the resistance to freeze and thaw. However, it was identified that resistance to sulphate attack has increased with the addition of graphene oxide but conclusions on acid attack were not clear enough. In the same study, Chen et al. (2019) investigated about the freeze and thaw resistance of graphene-based concrete and concluded that it performs better compared than plain concrete under freeze and thaw [23]. Considering this, above all, graphene can be anticipated as a suitable material to use in CRCP for its performance improvements and cost cutting, such as smaller slab thicknesses and low amount of steel rebars.

It is important to mention about a few future potential applications of graphene-based nanofillers to the transport sector, including CRCP. Being a carbon allotrope, graphene and graphene-based compounds, such as graphene oxide, offer various multifunctional advantages to the concrete industry in terms of its electrical and thermal conductivity, which may be very helpful in future smart infrastructures, such as smart pavements. Three main multifunctional characteristics of graphene-based nanofillers are electrical conductivity, thermal conductivity and electromagnetic interference.

Uniform dispersion of graphene in concrete or other cementitious composites offers a continuous path for electrical conductivity. This gives more insights into pseudo-resistivity that can be used in various damage-sensing applications in health monitoring of infrastructures [43,101,102], including concrete pavements. Li et al. [103] introduced an equation

as a function of thickness and diameter of graphene to find the minimum nanoparticle volume fraction needed to establish a continuous path, which is the backbone of electrical conductivity. It is usually defined as the percolation threshold in the percolation theory that comes under mathematics. It can be believed that the benefits of electrical conductivity can be used in future smart pavements for wireless charging, vehicle counting, health monitoring, etc.

Like the electrical conductivity benefits, complying to the same reason that graphene can form a continuous path due its uniform distribution, this path or network can be used as a heat passage which offers greater benefits. When these two properties, namely electrical and thermal conductivity, amalgamate together, features such as de-icing and temperature monitoring can be introduced to pavements. Moreover, the thermal diffusivity can be greatly improved due to the extraordinary heat sink capacity of graphene. In other words, graphene can act as a heat reservoir while absorbing heat without showing significant changes in the temperature. This has been effectively investigated in terms of increased resistance to thermal cracking in mass concrete structures where internal heat can be catastrophic [99,100].

Electromagnetic interference is one such multifunctional benefit graphene holds and, when the graphene is mixed in the cement composites, excellent benefits in terms of security and health can be achieved. For instance, this property can reduce the electromagnetic emissions harmful to human health and be can used for military purpose to secure information from other threats. Graphene microstructures reduce the energy of external electromagnetic sources to destroy them or convert to other frequencies with less energy [16].

Even though these smart technologies discussed above are not fully practiced in the industry nowadays, a large amount of research is being carried out in different fields of science. It can be anticipated that these properties might be effective for the transport sector to enhance current practices.

5. Challenges in Application

Although graphene and graphene oxide seemed to be a promising addition to cementitious composites, including CRCP, they simultaneously bring a few challenges as well. Lack of design standards, particularly for graphene-based cementitious composites, is one of the dominant challenges. The improved performance characteristics, such as mechanical strengths are hard to employ into the pavement design of CRCP due to lack of specific guidelines [104–108]. For an example, the increased post-crack strength of concrete or, in other words, toughness resulting from graphene-based additions does not usually count when the slab thickness is determined according to the traditional design guidelines. Furthermore, most of the performance enhancements rely on the uniform dispersion of graphene and graphene oxide in the cement matrix, which should be carefully performed. Overdose or selection of superlattices in the wrong manner can cause serious problems. Subsequently, flowability and the constructability should be carefully addressed, which might be a great concern when it comes to pavements. Moreover, graphene can indulge various health risks, especially due to extremely smidgen particle sizes [16]. Research regarding toxicology of graphene-based nanofillers should be carried out and insights should be provided. Eventually, the cost of construction is one of the greatest concerns in construction projects. Adding graphene and processing them chemically before fabrication should be carefully analyzed using a cost benefit ration, which is very limited in research. A life cycle analysis should be holistically conducted to check the compliance of graphene-based nanofillers in the pavement construction.

6. Summary and Conclusions

Most of the past research conclude that graphene and graphene oxide can be used in cementitious composites effectively due to their performance enhancements in terms of compressive, flexural, tensile strengths and fatigue resistance. Following that, it can be

anticipated that this might be a suitable addition to CRCP to overcome its inherent flaws attributed to concrete.

Relationships between compressive strength, flexural strength and tensile strength of graphene-based and graphene-oxide-based cement composites were derived using statistical analysis of past experimental data. The derived equations were then compared to the particular equations in AS 3600 and ACI 318 guidelines given for plain concrete.

Dispersion methods and addition of admixtures should be carefully selected depending on the application. Moreover, the suitable dosages should also be cautiously selected because overdose can cause serious issues in the range of small to catastrophic.

Comprehensive design guidelines should be developed incorporating the increased toughness and strengths to receive the full benefit of adding graphene-based nanofillers to CRCP.

According to the past research, addition of graphene into the concrete does not improve the fatigue performance considerably. Fatigue, being one of the dominant durability performance indicators for concrete pavements, should be thoroughly evaluated by means of more research.

Field monitoring of crack widths and crack spacings in CRCP constructed with graphene-based nanofillers should be conducted to observe the real picture. A long-term performance monitoring should be conducted to draw conclusions on these performance indices as well, otherwise it would not be possible to understand the applicability for CRCP.

Apart from the strength improvements, addition of graphene can bring in various multifunctional benefits due to its electrical conductivity, thermal conductivity and electromagnetic wave reflection properties, which might be absolutely suited for future smart pavements.

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

Appendix A

Table A1. Key details and findings of past studies on graphene/graphene-oxide-based cement composites.

Ref	Graphene/GO/Additions (Dosage Range)	Strength Properties			Overall Highlights
		Compressive Strength	Flexural Strength	Tensile Strength	
[19]	GO (0–0.2%) GO (0–0.2%) & silica fume	(14% increase) (14% increase)	(1.7% increase) (8.2% increase)	(13% increase) (31% increase)	GO-based concrete containing silica fume was tested for concrete pavements. Mix with steel fibers gave denser hydration products indicating strong bonding.
[14]	GO (0–0.2%)	(12.7% increase)	(14.5% increase)	(13.68% increase)	Workability gradually decreased, while compressive, flexural, and tensile strengths increased. SEM results depicts dense microstructure and less pores
[53]	GO nano sheets (0.05%)	(7.82% increase)	(12.60%) increase		Better hydration characteristics were observed. Compressive strength and flexural strength were increased. SEM showed good dispersion. Fluidity reduced
[23]	GNP (0–0.4%)	(22.4% increase)			Freeze and thaw resistance was observed to be increased with GNP dosage. Compressive strength was increased, while the workability was reduced
[24]	GNP (0–2.5%)				Pore refinement was observed. Average void diameter and porosity were reduced. Improved resistance to water penetration and chloride attack. Tortuosity was improved.
[25]	GNP (in terms of graphite)	(146% increase)	(79.5% increase)		Reducing the use of cement content can reduce the CO ₂ emissions. Heat capacity increased. Resistance to water penetration increased.
[67,81]	GNP (0–0.3%)	(Slight increase)	(59% increase)	(45% increase)	Energy absorption capacity and hardness were improved with GNP content. Compressive, tensile and flexural strengths were improved
[58]	GNP (0–0.3%)				Observed accelerated early hydration products. Sorptivity was reduced. Improved abrasion resistance was reported.
[13]	GO (0–2%)			(48% increase)	Tensile strength was increased. Improved growth of C-S-H hydrates.
[60]	0.5 SWCNT + 1.5% GO GO (1.5%)		(73% increase) (51.2% increase)		Effect of multi-carbon nanomaterials was observed. Flexural strength increased. When the CNT was added, the compressive strength increase was larger.
[64]	GO (1%)	(77% increase for 1%)		(37.5% increase for 0.25%)	Combination of GO and FCNT improved the strength properties compared to their individual performance.
[62]	1% GO as synthesized and 1% GO ball milled	(63% increase and 86% increase)			Commercial GO and Ball-milled GO were tested in cement. Ball-milled GO gave better improvements in terms of compressive and flexural strengths.

Table A1. Cont.

Ref	Graphene/GO/Additions (Dosage Range)	Strength Properties			Overall Highlights
		Compressive Strength	Flexural Strength	Tensile Strength	
[65]	Pure GO & 0.1% GO functioned with NH ₂	(38.4% increase)	(84% increase)		NH ₂ functionalized GO and pure GO were tested on cement mortars and functionalized GO observed to perform better. In both, compressive and flexural strengths increased. Porosity was reduced. Further increase in GO led to a decrease in strength.
[66]	GO (0.5%) with Microwave curing	(26.6% increase)			Increased hydration due to GO was observed due dense structure. Increase in compressive strength. Further increase was observed when cured in microwave.
[93]	GO (0–0.06%) + SiO ₂ (0–2%) GO (0–0.06%)	(39.89% increase at 0.045% GO) (32% increase at 0.03%)			Use of hybrid nanofibers were tested with GO and SiO ₂ . Hybrid composites gave better strength improvements. Also, hybrid seemed to accelerate the hydration and pozzolanic reaction. Flexural toughness observed to be increased more with hybrid approach which assures more durability.
[41]	GO (0–0.066%)	(22.59% increase at 0.022% GO + 0.22% PC)	(24.56% at 0.022% GO + 0.22% PC)		Compressive and flexural strengths increased. Flexural toughness was increased. PC concentration was found to be sensitive. Fluidity was increased with the use of increasing PC concentrations. SEM showed thin cracks and high tortuosity.
[74]	GO (0–0.066%) + PC modified	(17.68% increase)	(22.55% increase)		Proper dispersion characteristics were observed with the mixing of PC. Load transfer efficiency, compressive and flexural strengths were improved. Better crack resistance was observed.
[77]	GO (0–1%)	(46.5% increase)	(15% increase)	(35% increase)	Measured the optimum dosage as 0.05–0.1% and strength improvement was told to be between 20 and 32% generally. Reason for strength improvement in terms of the bridging effect of hydrates with GO making covalent bonds stronger.
[68]	GO (0–0.5%)	(78% increase)		(37.5% increase)	Optimum dosage was 0.1% and found that further increase in dosage reduces the strength characteristics. Improvement in early age hydration characteristics.
[78]	GO (0–2%)			(48% increase)	Growth of CSH hydrates was evident. Nucleation of CSH along with GO flakes considered to be the reason for the improved bond strength.
[85]	GO (avg size = 100 μm) Ball milled GO (avg size = 780 nm)	(63.3% increase) (77.8% increase)			Commercial GO and Ball-milled GO were tested in cement. Ball-milled GO gave better improvements in terms of compressive and flexural strengths. The reason was ball-milled GO is smaller in size which allows more dispersion and surface area.
[69]	GO (0–0.12%) + PVA (2% vol)	(24.8% increase)	(37.7% increase)	(80.6% increase)	Too much GO causes PVA fiber to rupture before the pull-out failure. Proposed mechanism diagram for improved mechanical behavior. Increased the bonding between PVA and cement matrix.

Table A1. Cont.

Ref	Graphene/GO/Additions (Dosage Range)	Strength Properties			Overall Highlights
		Compressive Strength	Flexural Strength	Tensile Strength	
[40]	(FGN)[Functionalized Gr. (0–0.05%) Graphene with Nitric acid ammonium hydroxide)	(20% increase)	(32% increase)		Fluidity reduced with increased FGN dosages. The optimum dosage was found to be 0.02%. Strength improvement was considered to be a result from fine pore structure and compacted hydrated crystals.
[57]	GNP (0–0.12%)	20% increase	17% increase		Ultrasonication time was recommended to be less than 2 h. Concluded that enhancement in strength is not significant enough for industry purposes. Observed to be highly conductive when graphene is added.
[79]	Graphen (Multi-layer graphene sheets)	(54% increase at 1% vol)	(21% increase at 1% vol)		reasons for the improved reinforcing mechanism were stated; better distribution of graphene, decreased w/c ratio, self-curing by adsorption, lowering crystal orientation index and crack bridging.
[80]	Graphene (0–0.4%)	(42% decrease)	(13% increase)		Acoustic emission monitoring was performed to characterize graphene cement attributed to fracture energy.
[83]	Graphene (0–1%)	(8% increase)	(16% increase)		observed a decrease in total porosity. Piezoresistive behaviors considered to be governed by the filling and interfacial effects.
[84]	Graphene	(19.9% increase)			Size of graphene/GO particles found to be important in strength enhancement. Chemical attack was found to be decelerated. GO showed improved freeze and thaw performance. AFM and SEM revealed the better bonding characteristics and dense structure.
	GO	(13.2% increase)			
[90]	Graphene + (0–1% rice husk)	(75% increase)			Risk husk was synthesized with graphene. Conductivity and strength were improved.
[15]	GO (only 0.03%)	(46% increase)		(53% increase)	Results revealed enhanced degree of hydration when GO is added. Reduced the total porosity.
[40]	GO (0 & 0.02%) with silica fume (0–10%)				Shear mixing can be used to reduce the size of GO. Desirable amount of silica fume made good strength improvements while the over usage of silica fume decrease the performance.
[61]	GO (0.05%)	(15–33% increase)	(41–59% increase)		Air voids were characterized by ultrasonic measurements and recorded a lesser time in GO based samples. Workability was reduced. Elastic modulus increased. Surface area and pore structure was greatly refined.
[55]	0.02 Reduced GO (rGO)	(22% increase)	(23% increase)		Porosity was decreased. Compressive and flexural strengths were increased.

Table A1. Cont.

Ref	Graphene/GO/Additions (Dosage Range)	Strength Properties			Overall Highlights
		Compressive Strength	Flexural Strength	Tensile Strength	
[63]	GO 0.02% + CNT 0.04% (Hybrid)	(23.9% increase)	(16.7% increase)		Total porosity was reduced with addition of GO and CNT. Hybrid form of CNT and GO increased strength characteristics than their individual performance.
[87]	GO (0.05%)	(6.8% increase)	(8.3% increase)		Low setting time and low fluidity. 0.05% GO refined the pore structure better and further increase in dosage increased the porosity and decreased the mechanical performance.
[70]	GO (0.2%)	(10% increase)			SEM demonstrated that GO adhere to hydration products to make the structure stronger. However, the research concluded that GO does not influence the CSH structure and stated that mechanical strength improvement is due to the accelerated hydration.
[71]	GO (0.04%)	(14% increase)			Workability was reduced. Hydration was accelerated. Pore structure was strengthened.
[72]	GONP (0–0.05%)	(13% increase)		(41% increase)	0.02–0.03% of dosage was recommended. Refined the pore structure by making it more compact. Increased the thermal stability of the hydration products.
[88]	GO (0.01–0.05%)	(47.9% increase)	(60.7% increase)	(78.6% increase)	Proposed hydration regulation mechanism that forms flower like hydration crystals. Research concluded that GO reduces the brittleness while improving the toughness.
[76]	GO (0.05%)	(40.4% increase)	(90.5% increase)		Viscosity was increased while fluidity was decreased. Setting time also got shortened. Concluded that GO effectively interacts with hydration reaction of cement while aligning crystals in a compact and desirable pore structure. Early strength gain was evident.
[75]	GO (0.02%)	(197.2% increase at 25.45% oxygen)	(184.5% increase at 25.45% oxygen)	(160.1% increase at 25.45% oxygen)	Strength improvement was considered to be due to the flower-like crystals in the hydration mechanism. The results were believed to be beneficial for cracking resistance and improved toughness.
[89]	GO (0.05%)	(11% increase)	(16.2% increase)		GO and CNT hybrid form of reinforcement was tested. Hybrid type showed better strength improvements compared to the individual effect.
	GO (0.025%) + CNT (0.025%)	(20% increase)	(24% increase)		
[28]	GO (0 and 0.05%)	(15–33% increase)	(41–59% increase)		Surface area increase of the samples was observed (27.3–64.9 m ² /g), compressive and flexural strengths increased.

Table A1. Cont.

Ref	Graphene/GO/Additions (Dosage Range)	Strength Properties			Overall Highlights
		Compressive Strength	Flexural Strength	Tensile Strength	
[46]	GNP (0 & 0.05%)	(3–8% increase)	(15–24% increase)		Optimum GNP/MC was found to be 1:7. Cement hydration has promoted at early age.
[59]	GNP (0–0.1%)	(28% increase)	(13% increase)		Optimum GO dosage was found to be 0.05%. Positive influence on early hydration was stated. Porosity was reduced.
[47]	Graphene (0–0.05%)	(10% increase)	(16% increase)	(Slight increase)	Optimum ratio for graphene to SDBS was found to be 1:6. Optimum dosage of graphene was found to be 0.025% and the strength characteristics decreased with increasing dosages thereafter.
[82]	Graphene (0–0.09%)	(11% increase)	(27.8% increase)		0.06% of dosage was found to be optimum. Pore structure refinement was observed.
[54]	GNP (0–0.05%)	(18.8% increase)	(56.6% increase)		Observed accelerated hydration process and more hydration products. Different dispersants were checked. C-S-H gel seemed to be increased. Total porosity was decreased.
[94]	GNP (0–0.05%)	(25.28% increase)	(56.6% increase)		3D network structure for hydration products was proposed and stated to be the reason for strength enhancements.

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