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Synergistic Impact of Sustainable Graphene Derivative and Dune Sand on Cement Mortars

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Abstract **-** This study investigates the impact of a sustainable graphene derivative, designated as D-GSH, on the compressive strength development of cement mortars. The research evaluates the effects of D-GSH addition by 0.25%, by weight of cement, and dune sand-to-cement ratios on compressive strength at 1, 7, and 28 days. Compressive strength tests reveal that the addition of 0.25% D-GSH significantly enhances strength across all age intervals, with increases of 13%, 55%, and nearly 50% at days 1, 7, and 28, respectively. Conversely, increasing the dune sand-to-cement ratio from 1:1 to 1:3 adversely affects early strength development. While the higher sand content initially boosts strength, its long-term performance is hindered, highlighting the necessity for optimal dune sand content in cement mortars. The findings demonstrate that D-GSH effectively accelerates early strength gain, making it a viable alternative for improving the performance of cement-based composites.

Keywords**:** Sustainable Graphene Derivative, D-GSH, Cementitious Materials, Nanomaterials, Compressive Strength

1. Introduction

Concrete's quasi-brittle nature presents significant challenges in structural applications, including low resistance to crack formation, poor tensile and flexural strength, and limited strain capacity [1]. Traditional methods, such as reinforcing concrete with steel rebars, as well as the addition of mineral admixtures and fibers, have provided partial solutions to these issues. However, brittleness remains a critical limitation in cement-based composites, and advancements like geopolymer technology have not fully addressed this challenge [2-4]. Recent research has shifted towards using nanotechnology to enhance the mechanical properties and durability of cementitious composites, with promising results [5, 6].

Among the various nanomaterials, graphene derivatives—specifically graphene oxide (GO) and graphene nanoplatelets (GNPs), have attracted significant attention due to their exceptional mechanical properties [7]. Graphene's high tensile strength, large surface area, and excellent dispersibility make it a suitable candidate for reinforcing concrete. When added to cementitious matrices, graphene derivatives refine the microstructure, reduce porosity, and inhibit crack formation, thereby improving both the strength and durability of the material [8]. Additionally, graphene's ability to accelerate the growth of cementitious crystals through its cation-attracting properties creates a stronger and more interconnected network within the composite [9]. However, the global availability and high cost of producing high-quality graphene derivatives limit their widespread use [10].

To address these challenges, sustainable approaches to graphene production have been explored [11]. A recent study [12] demonstrated the successful synthesis of a graphene derivative, the waste date syrup-based graphene sand hybrid (D-GSH), using abundant natural resources from the Middle East, specifically date syrup and dune sand. This process offers a cost-effective and eco-friendly alternative to conventional graphene production methods, which typically rely on graphite reserves and energy-intensive processes. The development of D-GSH presents a promising opportunity for incorporating advanced nanomaterials into construction, particularly in regions where sustainable resources are available [13, 14].

As such, this study is aimed to focus is on investigating the impact of a sustainable graphene derivative, referred to as D-GSH, on the compressive strength development of cement mortars. Additionally, this research explores how increasing the amount of dune sand in cement mortar mixes containing the D-GSH additive affects compressive strength development, providing insights into optimizing mix design for enhanced performance.

2. Materials

Type I Ordinary Portland cement (CEM-1 42.5N) was used to form the cement matrix of the composites, combined with tap water and dune sand. The chemical composition of the materials revealed that Type I OPC was primarily composed of calcium oxide (CaO) and silica (SiO_2), while dune sand contained a higher proportion of SiO_2 compared to CaO. The particle sizes of Type I OPC ranged 1-50 µm, while dune sand particles measured in the range of 200-300 µm.

The preparation process of D-GSH additive involves using a waste date syrup and a thermal treatment procedure. This method was adapted from Khan et al. [12]. As graphically described in Figure 1, dune sand and waste date syrup were mixed at a ratio of 2:5 followed by consistent stirring for 1 hour. Subsequently, the mix was left for approximately 12 hours at a constant temperature of 80 °C to dry and eliminate excess moisture. The dry mixture was then placed in a tube furnace for carbonation under a nitrogen (N_2) infused environment. The gas inside the tube furnace was replaced with N_2 before starting the heating protocol described in Figure 1. The heating protocol aimed to transform the dry mix of wasted date syrup covering dune sand particles into graphene-based carbon through carbonization. The temperature within the tube furnace was increased gradually, starting with a heating rate of 2.5 °C/min until reaching 100 °C, then the dry mix was kept at 100 °C for 30 minutes. Then, the heating rate was increased to 3.3 °C /min until reaching 200 °C, where the dry mix was kept at 200 °C for 1 hour. Finally, the heating rate was increased to 9.1 °C/min to achieve a heating temperature of 750 °C, where the dry mix remained under 750 °C for almost 3 hours, to ensure a complete graphitization of date syrup. The final product, i.e. D-GSH, was then collected after cooling of the furnace tube.

Although D-GSH has a much lower surface area (22.4 m²/g) compared to typical graphene sheets, it retains active surface characteristics due to its rough, micro-porous texture, while also displaying hydrophobic properties due to the complete coating of hydrophilic sand particles. Elemental analysis of D-GSH additive confirmed the existence of carbon, oxygen, silica, and calcium.

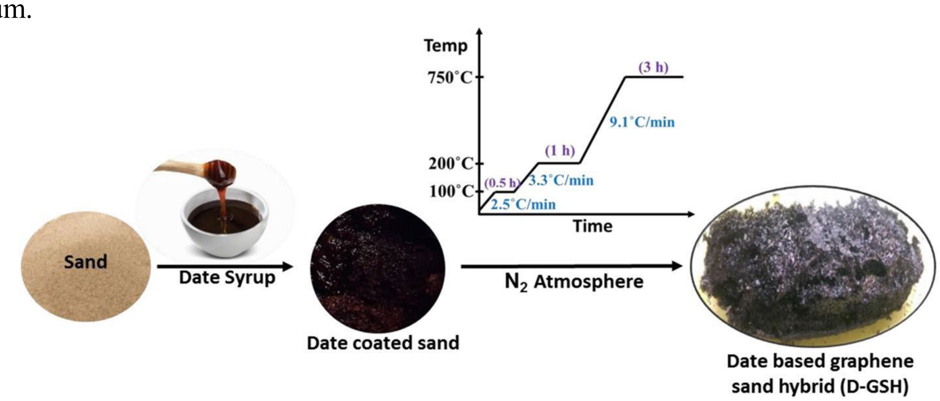

Figure 1: Synthesis process of D-GSH involves mixing sand with date syrup, then subjecting the mixture to a controlled N_2 atmosphere at accelerated temperatures until reaching 750°C, adopted from Khan et al. [12].

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3. Sample Preparation

To evaluate the effects of D-GSH addition and increased dune sand content in cement mortars, three mixes were prepared with a constant water-to-cement ratio of 0.5 and dune sand-to-cement ratios of 1:1 and 1:3 as shown in Table 1. The reference mix (M-D-G0-S1), made without D-GSH additive and a dune sand to-cement ratio of 1:1, was used as a benchmark for the remaining mixes. The second mix was made with 0.25% D-GSH additive, by weight of cement, and a dune sand-to-cement ratio of 1:1. The third mix consisted of 0.25% D-GSH and a dune sand-to-cement ratio of 1:3. The specific mixes were labelled as M-D-Gx-Sy, where 'x' indicates the D-GSH concentration and 'y' represents the dune sand ratio. For instance, the mix containing 0.25% D-GSH at a 1:1 dune sand ratio was designated as M-D-G0.25-S1.

Table 1: Mixture proportions of mortar mixes in kg/m^3.

Mix ID	D-GSH (%)	Water (kg/m^3)	Cement (kg/m^3)	Dune Sand (kg/m^3)
M-D-G0.00-S1	0	432	864	864
M-D-G0.25-S1	0.25	432	864	864
M-D-G0.25-S3	0.25	432	864	2592

The sample preparation involved dispersing D-GSH in water through a 30-minute sonication process, incorporating intermittent pauses for 2 minutes after each 10 minutes of sonication to prevent deterioration of the additive. A magnetic stirrer was employed to enhance circulation during sonication. The dry ingredients, including cement and dune sand, were mixed for 2 minutes. Water or the D-GSH solution was then gradually added, followed by an additional 2-3 minutes of mixing. The fresh mortar was cast into 50 mm cube molds, demolded after 24 hours, and cured in water until testing, all under ambient laboratory conditions of 23 ± 2 °C and 50 ± 5% relative humidity.

4. Results and Discussion

The compressive strength of both plain and D-GSH-reinforced cement mortars was evaluated using a static Material Testing System (MTS) machine with a capacity of 300 kN, following ASTM C109/C109M-20 [15]. The compressive strength was determined at 1, 7, and 28 days, with axial loads applied at a rate of 7 kN/sec. A total of three replicate specimens were used to calculate an average compressive strength at each age.

Figure 2 presents the compressive strength of cement mortars at 1, 7, and 28 days. The strength development trends further illustrate these findings. The reference mix (M-D-G0-S1) showed a modest early strength gain of 19% over the first 7 days, followed by a substantial 93% increase between days 7 and 28. In contrast, M-D-G0.25-S1 exhibited a more rapid early gain of 64 in the first 7 days and an 86% increase between days 7 and 28, indicating that the D-GSH additive accelerates early hydration. The mix with a higher dune sand content (M-D-G0.25-S3) experienced an exceptional early strength development of 544% between 1 and 7 days but only a 2.9% increase between days 7 and 28, suggesting that rapid early strength may exhaust hydration potential at later ages. These results indicate that while D-GSH effectively enhances early strength, excessive dune sand may hinder long-term performance. Therefore, optimizing dune sand content is essential to balance early and long-term strength development.

The addition of 0.25% D-GSH significantly enhances the compressive strength across all time intervals. At day 1, the control mix (M-D-G0-S1) exhibited a compressive strength of 12.97 MPa, while the D-GSH mix (M-D-G0.25-S1) improved this to 14.65 MPa, reflecting a 13% enhancement. By day 7, M-D-G0-S1 increased to 15.43 MPa, whereas M-D-G0.25-S1 showed a substantial rise to 23.96 MPa, marking a 55% increase due to the D-GSH addition. At day 28, the control mix achieved 29.80 MPa, while M-D-G0.25-S1 reached 44.67 MPa, indicating a nearly 50% increase attributable to D-GSH, which likely facilitated better hydration and bonding within the cement matrix. Similar enhancement percentages across all ages were found when incorporating conventional graphene derivatives in cement mortars [16, 17].

In contrast, increasing the dune sand to cement ratio from 1 to 3 in mix M-D-G0.25-S3 adversely affected compressive strength, particularly in the early age. At day 1, M-D-G0.25-S3 demonstrated a significantly lower compressive strength of 5.37 MPa, indicating that the higher dune sand content negatively impacted initial strength development due to disrupted particle packing and hydration efficiency. By day 7, M-D-G0.25-S3 exhibited a notable increase to 34.63 MPa, surpassing the strength of M-D-G0.25-S1, yet this rapid gain does not compensate for its much lower initial value. By day 28, the compressive strength of M-D-G0.25-S3 was not affected and remained the same, highlighting the adverse effects of excessive dune sand on long-term strength development. Comparable to conventional graphene derivatives, the addition of 0.25% D-GSH is thought to have significantly contributed to the early development of compressive strength of M-D-G0.25-S3, attributed to its role as a nucleation site [18].

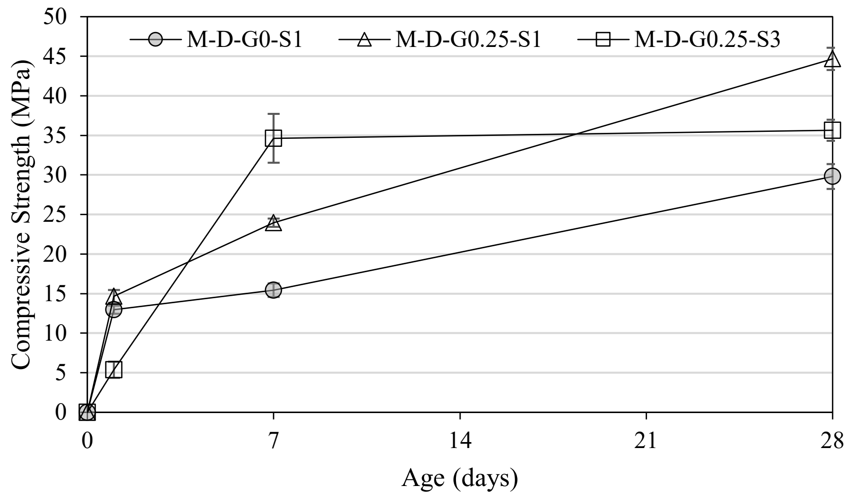

Figure 2: Compressive strength of mortar mixtures across time.

5. Conclusion

This study successfully demonstrates the potential of D-GSH in enhancing the compressive strength of cement mortars. The results indicate that incorporating D-GSH not only accelerates early strength development but also improves long-term performance when balanced with the appropriate dune sand content. However, excessive dune sand in cement mixes poses challenges that can compromise strength development over time. The addition of 0.25% D-GSH significantly improves the compressive strength of cement mortars across all curing ages, with increases of 13%, 55%, and nearly 50% at days 1, 7, and 28, respectively. Conversely, increasing the dune sand-to-cement ratio from 1:1 to 1:3 adversely affects early strength development, resulting in a notable initial strength gain that ultimately hinders long-term compressive strength performance. Future studies should focus on further optimizing mix designs and exploring additional applications of D-GSH in various construction contexts to maximize its benefits for enhanced structural performance and sustainability.

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