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Microstructural Dynamics in Cement Mortars Fortified with a Sustainable Graphene Derivative

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Abstract **-** This study explores the influence of a sustainable form of graphene derivative, Date Syrup Graphene Sand Hybrid (D-GSH), on the microstructural evolution of cement mortars. Two mortar mixtures were prepared: plain and D-GSH-modified cement mortar. D-GSH-modified cement mortar mix had a concentration of 0.5% by weight of cement, while maintaining a water-to-cement ratio of 0.5. The synthesized D-GSH was ultrasonicated in water to ensure dispersion before incorporation. Microstructural changes were assessed using Scanning Electron Microscopy (SEM) and X-ray Diffraction (XRD) at 1 and 7 days. The results revealed that D-GSH significantly accelerated the hydration and enhanced the microstructure. The mixture made with 0.5% D-GSH exhibited a denser matrix, refined crystalline morphology, and reduced porosity, attributed to the nucleation effect of D-GSH. Unique flower-like structures on the surfaces of calcium silicate hydrate (C-S-H) crystals were observed in the D-GSH-modified mixes, which were absent in the plain mix, resulting in reduced pores in the microstructure of the D-GSH-modified mixtures. XRD analysis confirmed the enhanced hydration kinetics with accelerated C_3S consumption and higher portlandite ($Ca(OH)_2$) formation. The findings suggest that D-GSH could improve the performance and longevity of cement mortar, offering a sustainable solution for developing advanced building materials.

***Keywords*:** Scanning Electron Microscopy, X-ray diffraction, sustainable graphene derivative, D-GSH, microstructure, hydration

1. Introduction

The incorporation of graphene and its derivatives as two-dimensional (2D) nano-reinforcements in cementitious composites has sparked enhanced reactions, leveraging their nucleation effect and extensive surface area [1], [2]. Various studies exploring diverse graphene derivatives in cementitious composites have consistently identified them as pivotal for macro-level performance enhancement and have displayed positive impacts on microstructural aspects, unlike macro-reinforcements [3], [4], [5], [6]. In an early-age cement hydration investigation by Li et al. [7], varying graphene oxide (GO) dosages resulted in significantly higher hydration rates in GO-reinforced cement paste than in plain cement, accelerating the hydration process and indicating enhanced nucleation sites. However, the formation of a water barrier by the functional groups of GO poses challenges [8]. Zhu et al. [9] reported increased hydration products with graphene addition, implying potential flexural strength improvements due to 2D structure of graphene derivatives. Gladwin and Alex [10] observed microstructure changes, transitioning to denser flower-like and polyhedron crystals with increased GO content, correlating with enhanced compressive strength. The persistent occurrence of hydroxide crystals in the GO-reinforced composites, with a proportional increase in size, signifies improved compressive strength.

Conventional graphene derivatives are effective but have hurdles like high costs and complex production methods [11]. To mitigate these concerns, the utilization of sustainable graphene derivatives presents sustainable and economically feasible substitutes [11]. Among these, the date syrup graphene-based sand hybrid (D-GSH) developed by Khan et al. [8] represents a sustainable bio-derived graphene form synthesized from date syrup and dune-sand particles. This study aims to investigate the microstructural dynamics of plain and D-GSH-modified cementitious mortar over curing ages of 1 and 7 days. The modified cement mortars had a D-GSH concentration of 0.5% by cement weight. The microstructure was analyzed using Scanning Electron Microscopy (SEM) and X-Ray Diffraction (XRD) to elucidate the role of D-GSH in modifying the microstructure of cement mortar mixes. By focusing on the early- and mid-term microstructural behavior, this study aims to provide insights into the interplay between D-GSH addition and the evolution of the microstructure of modified cement mortars.

2. Materials & Methods

Type I Ordinary Portland Cement (OPC) was used as the main binder to form the cement matrix with tap water mixed with abundant dune sand. Their chemical composition consists mainly of calcium oxide (CaO) and silica (SiO_2) for OPC, while dune sand has higher SiO_2 than CaO. Their particle sizes range from to 1-50 µm for OPC and to 200-300 µm for dune sand. D-GSH is a bio-derived sustainable graphene derivative that graphitizes waste date syrup via pyrolysis, as described by Khan et al. [12]. Figure 2 illustrates the synthesis of a bio-graphene derivative (D-GSH) from waste dates and dune sand. Waste dates were washed, de-pitted, soaked, blended into a paste, boiled at 105 °C to extract sugars, and concentrated into a syrup [13]. This syrup was mixed with dune sand in a 2:5 ratio, stirred for 1 h, and dried at 80 °C for 12 h to remove moisture. The dried mixture was carbonized in a tube furnace under a nitrogen atmosphere. The heating protocol involved gradually increasing the temperature to 100 °C (held for 30 min), 200 °C (held for 1 h), and 750 °C (held for 3 h) to achieve graphitization. The final D-GSH product was collected after 24 h of cooling. The synthesized D-GSH yielded a singular crystalline peak at 26 degrees in XRD spectrum.

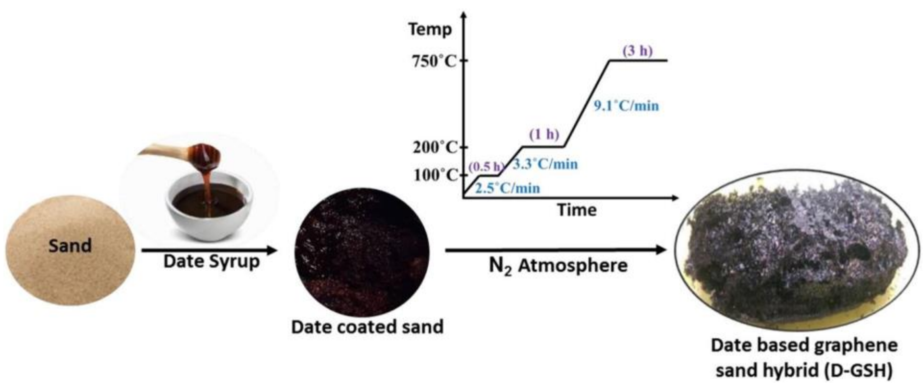

Fig. 1. Synthesis process of D-GSH involves mixing sand with date syrup, then subjecting the mixture to a controlled N2 atmosphere at accelerated temperatures until reaching 750°C, adopted from Khan et al. [12].

Plain and D-GSH-modified mortar mixes were prepared with a water-to-cement ratio of 0.5 and a dune sand-to-cement ratio of 1:1, to ensure maximum interaction with the cement [14]. The modified mix is denoted as 'M-G-*x*,' where *x* indicates the d-GSH concentration percentage by weight of cement. The plain mix did not contain D-GSH and was used as the benchmark. The second mix contained 0.5% D-GSH by cement weight, denoted as M-G-0.5. D-GSH was ultrasonicated in water for 30 min to create a dispersed solution, as recommended in previous studies, to avoid agglomeration [1]. The dry ingredients were mixed, followed by tap water for the plain mix and a D-GSH dispersed solution for the modified mixes. Cube samples (i.e., size of 50-mm) were cast and tested for compressive strengths at 1 and 7 days. Samples were taken from the fractured surfaces of the tested cube specimens and analyzed using SEM and XRD. XRD analysis was conducted on powdered samples over a 2θ range of 5°–80°, with a Cu tube (λ = 1.54056 Å) at 30 kV and 10 mA, using a step size of 0.5°, providing qualitative insights into hydration products.

3. Results & Discussion

3.1 Scanning Electron Microscopy

The microstructural evolution of cement mortars incorporating D-GSH was examined using SEM at 1 and 7 days, revealing notable impacts on the hydration product morphology and development. The plain mix exhibited typical hydration progression, transitioning from loose and flaky crystalline formations at day 1 to a compact structure with needle-like crystals by day 7, accompanied by visible micro-cracks and voids. In contrast, the mortar modified with D-GSH demonstrated accelerated and refined microstructural evolution. The inclusion of 0.5% D-GSH significantly reduced micro-pores and voids, indicating advanced early-age crystal organization, enhanced hydration kinetics, and minimized defects by day 7. These enhancements are attributed to the nucleation effect of D-GSH, which promotes a denser, inter-connected matrix. Additionally, unique flower-like structures were observed on the surface of the C-S-H crystals in the D-GSH-modified mortars, as shown in the magnified image of Fig. 2(d), but were absent in the plain mix, underscoring the influence of D-GSH on microstructural refinement.

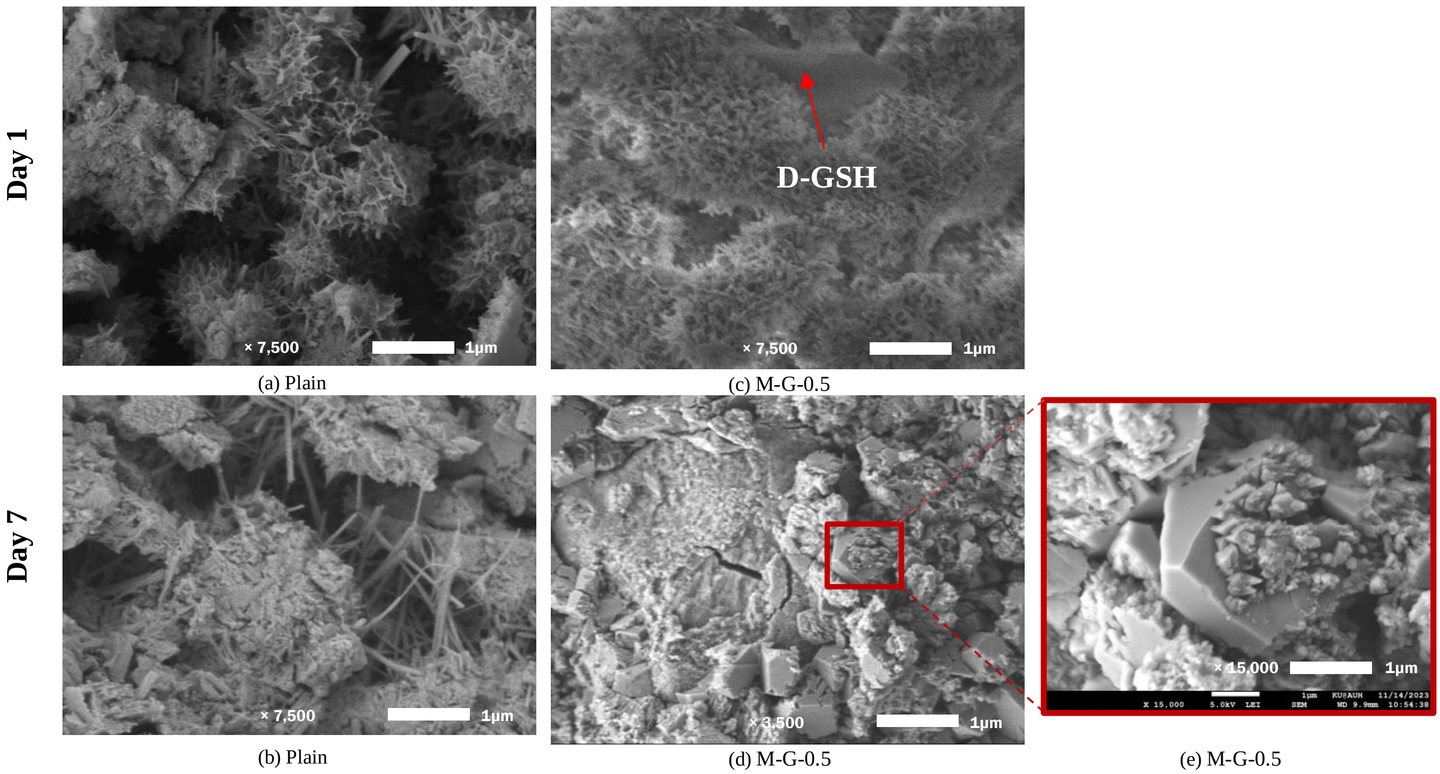

Fig. 2. SEM images of Plain (a-b) and D-GSH reinforced Mortars (c-d) at various curing ages of 1-, and 7-days, and (e) magnified SEM image of C-S-H crystal with a flower-like structure due to the interaction of D-GSH

3.2 X-ray diffraction (XRD)

The XRD analysis in Figure 3 reveals time-dependent changes in the hydration products of the plain cement mortars and those modified with the addition of 0.5% D-GSH. In the plain mix, tricalcium silicate (C_3S) at 29.8° showed high peak intensities at day 7, reflecting rapid early hydration and the formation of calcium silicate hydrate (C-S-H) and portlandite ($Ca(OH)_2$) [15]. Ettringite (at 9.5°) initially exhibited moderate formation but declined by day 7, likely due to partial transformation into monosulfate phases. The intensity of the $Ca(OH)_2$ peak at 18.2° increased steadily, indicating continuous hydration and $Ca(OH)_2$ release. Quartz at 27.1°, an inert phase, remained consistent and served as a reference. These findings align with the typical hydration process of plain mortars, which is characterized by rapid early reactions, peak activity at intermediate stages, and reduced rates at later stages [16].

The addition of D-GSH significantly altered hydration. The addition of 0.5% D-GSH accelerated C_3S hydration, with higher and sharper peak intensities on day 1 compared to plain mortars, attributed to the nucleation and bonding effect of D-GSH. Ettringite peaks remained stable by day 7 but doubled that of the plain mix, indicating sustained stabilization within the matrix. Furthermore, the formation of $Ca(OH)_2$ increased, with greater crystallinity, suggesting enhanced calcium hydroxide precipitation, resulting in the formation of C-S-H crystals. The additional formation of C-S-H crystals yielded a denser microstructure with enhanced mechanical and durability properties of the cementitious composite, as proven by the SEM images. Similar findings were obtained with the addition of GO to cement mortar [17], [18].

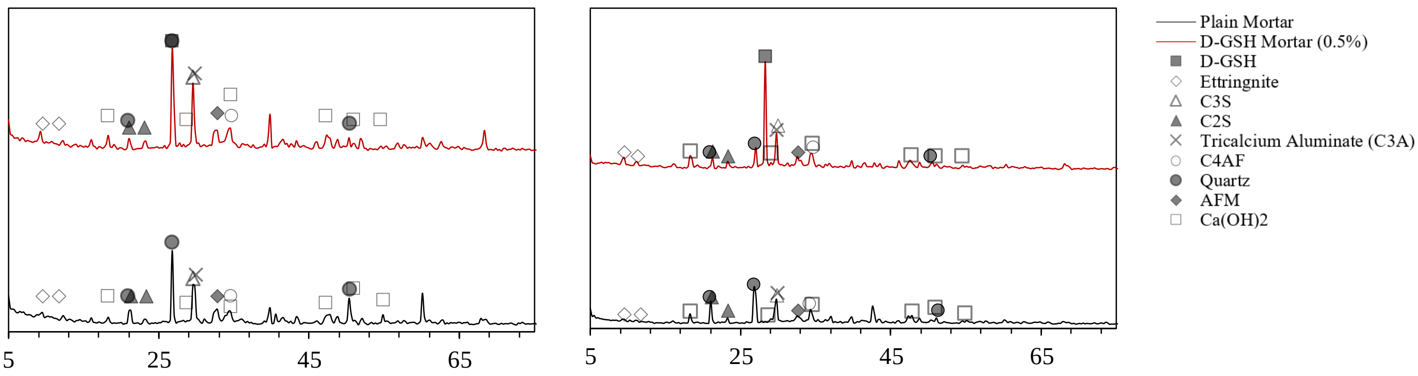

Fig. 3. XRD patterns for all mixes at (a) day 1, and (b) day 7

4. Conclusion

This study investigated the impact of a Date Syrup Graphene Sand Hybrid (D-GSH) on the microstructural evolution of cement mortars. The findings are summarized as follows:

- SEM analysis revealed that D-GSH-modified mixes exhibited accelerated hydration. Compared to the plain mix, the 0.5% D-GSH mix showed faster early-age hydration, well-organized crystal structures, and minimal micro-cracks and pores by day 7. Unique flower-like structures on the C-S-H surfaces were present in the D-GSH mixes but absent in the plain mix.
- Needle-like ettringite structures were barely noticeable in the SEM images of the D-GSH-modified mixes by day 7, indicating accelerated C-S-H formation.
- XRD analysis confirmed that D-GSH accelerated C_3S consumption, increased portlandite formation, and promoted the transformation of ettringite into monosulfate phases, attributed to the nucleation and bonding effects of D-GSH within the cement matrix.

Further research is necessary to explore the long-term durability and performance of D-GSH-modified mortars under different environmental conditions. This will help optimize their practical applications in cementitious materials.

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