Supporting Information

Hydroxylated-graphene: a promising reinforcing nanofiller

for nano-engineering cement composites

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1. Figures

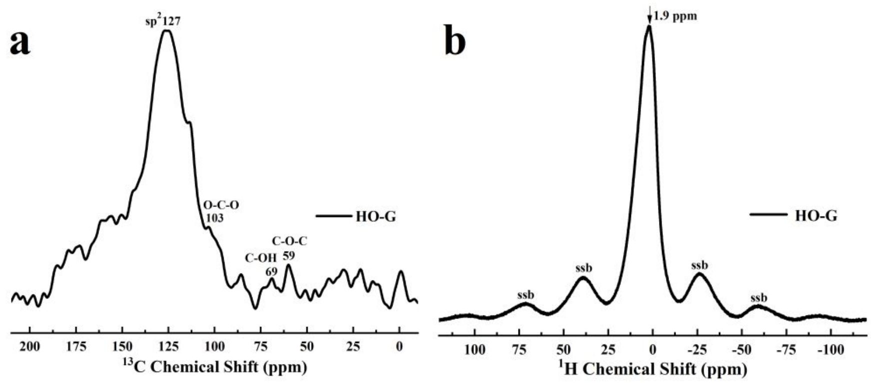

Figure S1. (a) ^{13}C NMR and (b) ^{1}H NMR spectra of HO-G. "ssb" denotes spinning sidebands.

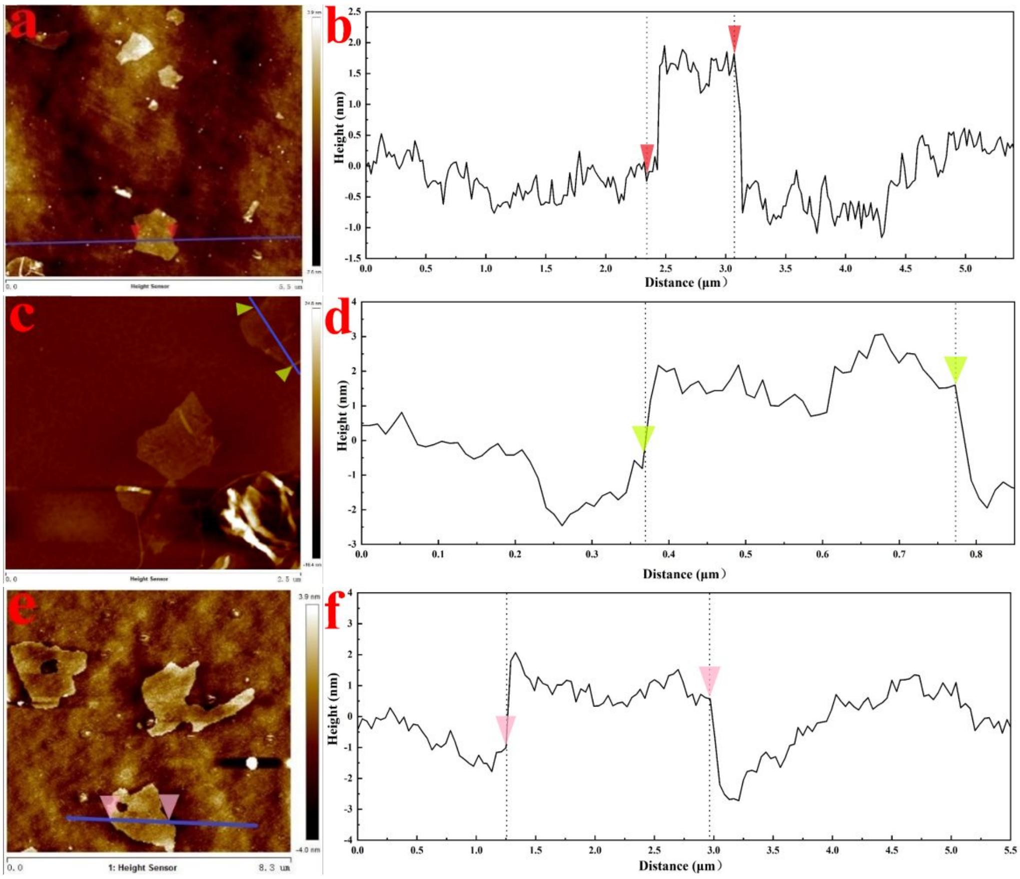

Figure S2. Typical AFM images and the corresponding height profiles of GO(a, b), TRGO(c, d), HO-G(e, f).

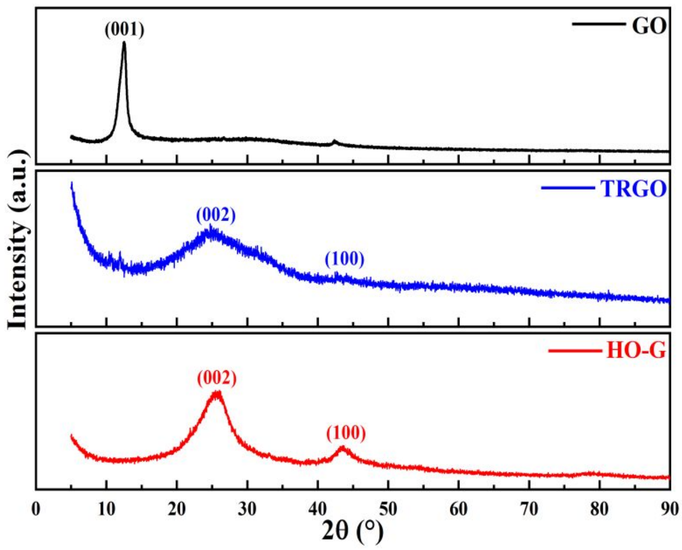

Figure S3. XRD patterns of GO, TTGO and HO-G.

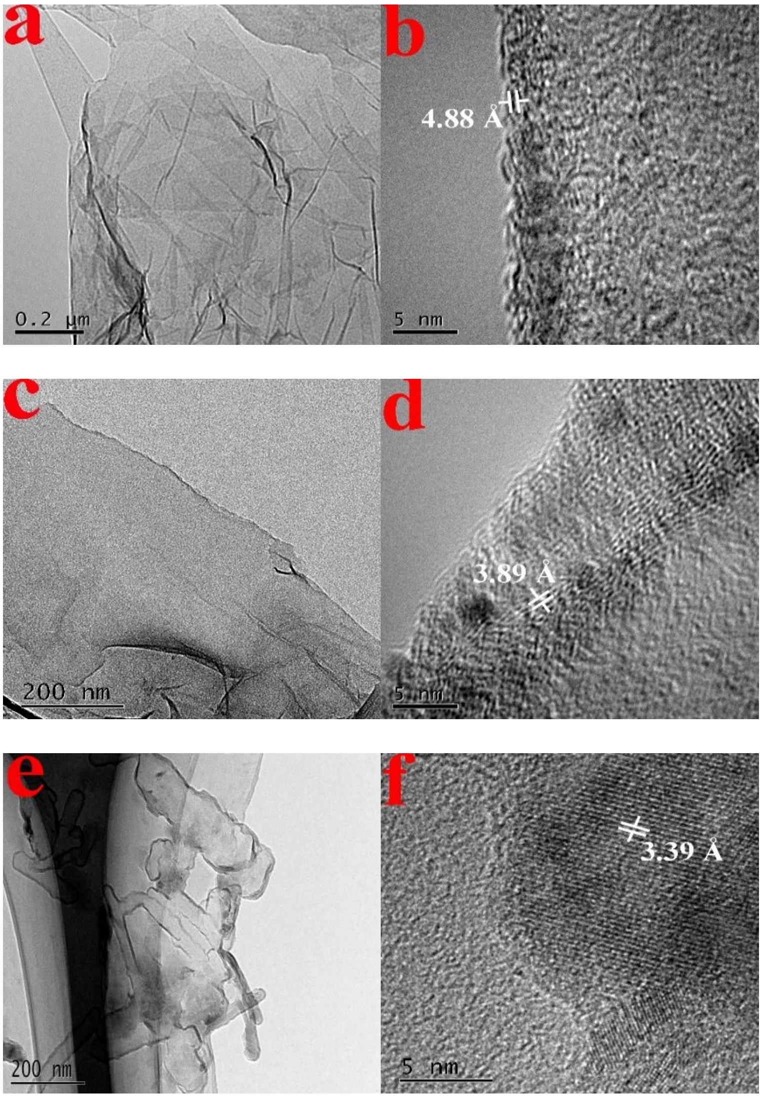

Figure S4. HRTEM images of GO(a, b), TRGO(c, d) and HO-G(e, f).

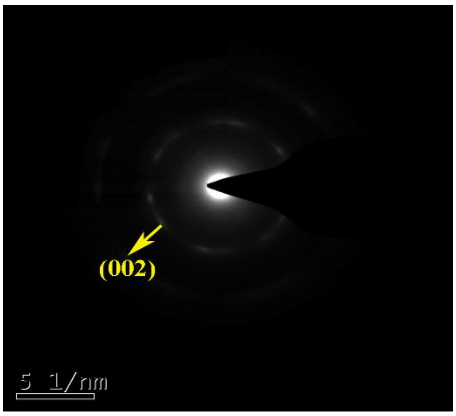

Figure S5. SAED image of HO-G.

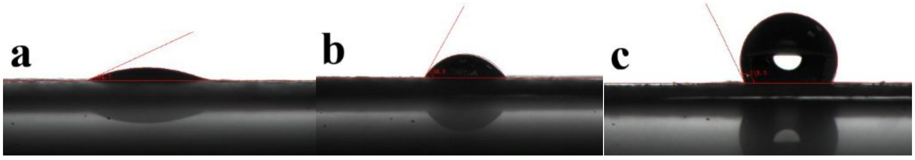

Figure S6. The contact angles of (a) GO, (b) HO-G and (c) TRGO.

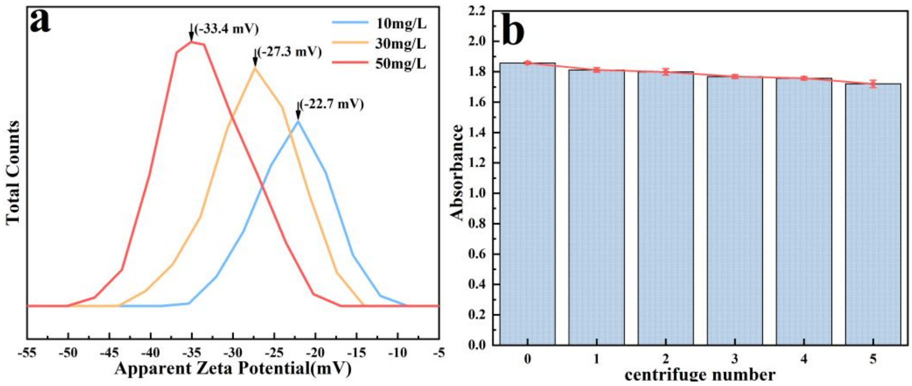

Figure S7. (a) Zeta potentials of aqueous HO-G solutions of different concentrations. (b) The change of absorbance of aqueous HO-G solution (30 mg/L) *vs* centrifuge number.

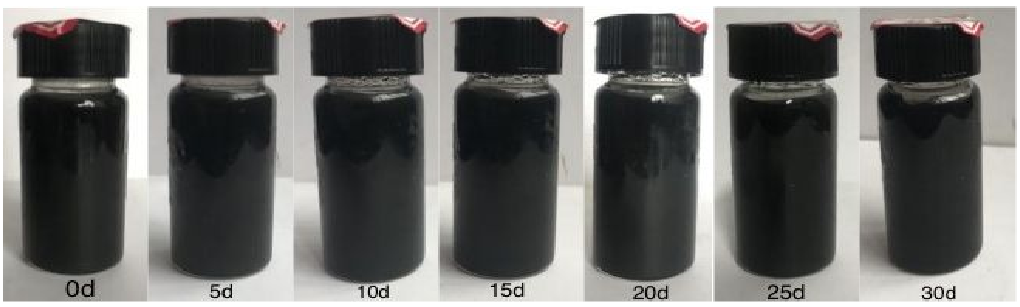

Figure S8. Digital photographs of aqueous HO-G solutions (30 mg/L) with different standing times.

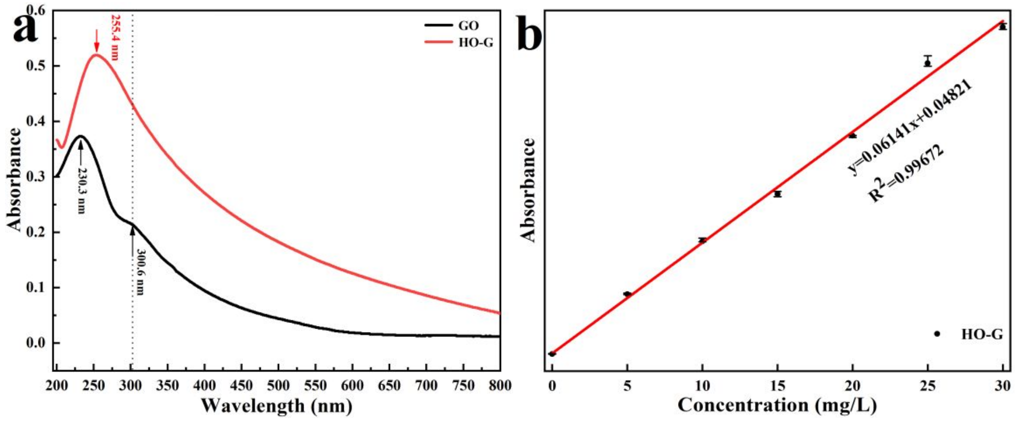

Figure S9. (a) UV-vis spectra of aqueous GO and HO-G solution. (b) The plots of absorbance of aqueous HO-G solutions with different concentrations.

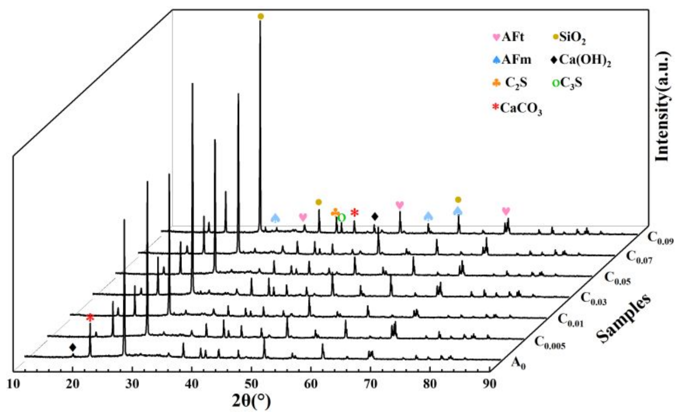

Figure S10. XRD patterns of HO-G blended cement mortars nanocomposites cured at 28 days.

2. Tables

Table S1. Comparisons of HO-G against other nanofillers for the enhancement of cement composites.

Nanofillers	Optimal dosage[a] (%)	Enhancement of the mechanical properties in 28d (%) Compressive strength	Flexural strength	References
Nano silica (nano-SiO_2)	2	10.1	11.0	Shi et al.[1]
Nano-alumina (nano-Al_2O_3)	1	18.46	6.27	Ge et al.[2]
Nano-calcium carbonate (nano-$CaCO_3$)	2	3.23	4.68	Cosentino et al.[3]
Nano-titanium oxide (nano-TiO_2)	0.75	19.33	15.1	Salman et al.[4]
Carbon nanotubes (CNTs)	0.05	6.4	10.1	Du et al.[5]
Graphene oxide (GO)	0.03	19.6	17.54	This study
Hydroxylated-graphene (HO-G)	0.03	21.19	7.89	This study

a. based on the mass ratio of cement weight.

Table S2. Chemical compositions of cement (%).

Chemical composition	Test value (%)
Al_2O_3	4.47
SiO2	21.5
Fe_2O_3	3.37
CaO	65.84
MgO	3.18
SO_3	0.3
NaO	0.49
f-CaO	0.78
C_3S	58.92
C_2S	20.19
C_3A	8.12
C_4AF	8.21

Table S3. Physical properties of cement.

properties	Test value
Fineness (%)	0.6
Density (g/cm^3)	3.15
Specific surface area (m^2/kg)	350
Standard Consistency (%)	25.6
Soundness (mm)	0.5
Initial Setting Time (min)	132
Final Setting Time (min)	198

Table S4. Gradation of standard sand.

Square mesh size (mm)	Remaining on the sieve (%)
2.0	0
1.6	7 ± 4
1.0	33 ± 4
0.5	67 ± 4
0.16	87 ± 4
0.08	99 ± 1

Table S5. Mix proportion of cement mortars.

Sample	Cement (g)	Sand(g)	Water (g)	PCE (%)[a]	GO (%)[a]	HO-G(%)[a]
A_0	450	1350	164.93	0.3	0	0
$B_{0.01}$	450	1350	164.93	0.3	0.01	0
$B_{0.03}$	450	1350	164.93	0.3	0.03	0
$B_{0.05}$	450	1350	164.93	0.3	0.05	0
$B_{0.07}$	450	1350	164.93	0.3	0.07	0
$B_{0.09}$	450	1350	164.93	0.3	0.09	0
$C_{0.005}$	450	1350	164.93	0.3	0	0.005
$C_{0.01}$	450	1350	164.93	0.3	0	0.01
$C_{0.03}$	450	1350	164.93	0.3	0	0.03
$C_{0.05}$	450	1350	164.93	0.3	0	0.05
$C_{0.07}$	450	1350	164.93	0.3	0	0.07
$C_{0.09}$	450	1350	164.93	0.3	0	0.09

a. based on the mass ratio of cement weight.

Reference

(1) Cheng, Y.; Shi, Z. Experimental Study on Nano-SiO2 Improving Concrete Durability of Bridge Deck Pavement in Cold Regions. *Adv. Civ. Eng.* **2019,** *2019*, 1-9, DOI: 10.1155/2019/5284913.

(2) Zhang, A.; Yang, W.; Ge, Y.; Du, Y.; Liu, P. Effects of nano-SiO2 and nano-Al2O3 on mechanical and durability properties of cement-based materials: A comparative study. *J. Build. Eng.* **2021,** *34*, DOI: 10.1016/j.jobe.2020.101936.

(3) Cosentino, I.; Liendo, F.; Arduino, M.; Restuccia, L.; Ferro, G. A. Nano CaCO3 particles in cement mortars towards developing a circular economy in the cement industry. *Procedia Struct Integrity* **2020,** *26*, 155-165, DOI: 10.1016/j.prostr.2020.06.019.

(4) Siang Ng, D.; Paul, S. C.; Anggraini, V.; Kong, S. Y.; Qureshi, T. S.; Rodriguez, C. R.; Liu, Q.-f.; Šavija, B., Influence of SiO2, TiO2 and Fe2O3 nanoparticles on the properties of fly ash blended cement mortars. *Constr. Build. Mater.* **2020**, 258, 119627.1-119627.11, DOI: 10.1016/j.conbuildmat.2020.119627.

(5) Du, M.; Jing, H.; Gao, Y.; Su, H.; Fang, H. Carbon nanomaterials enhanced cement-based composites: advances and challenges. *Nanotechnol. Rev.* **2020,** *9* (1), 115-135, DOI:10.1515/ntrev-2020-0011.