Supporting Information

Synergistic effect of graphene oxide and hydroxylated-graphene on the enhanced

properties of cement composites

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1. Figures



Figure S1. Typical AFM images (a) and the corresponding height profiles (b) of GO.



Figure S2. HRTEM images of GO.

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Figure S3. UV-vis spectra of aqueous solutions of different HO-G to GO ratios.

Solution concentration was kept at 10mg/L.



Figure S4. Digital images of HO-G or GO dispersions (1mg/mL, 5mL) in saturated

 $Ca(OH)_2$ solution (100mL).



Figure S5. The contact angles of (a) GO, (b) HO-G.

2. Tables

Chemical composition	Test value (%)		
Al ₂ O ₃	4.47		
SiO2	21.5		
Fe ₂ O 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 ₃ A	8.12		
C ₄ AF	8.21		

 Table S1. Chemical compositions of cement (%).

Table S2. 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 S3. 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

Samples	Compressive strength ^a (Mpa)/Change ^b (%)			Flexural strength ^a (Mpa) /Change ^b (%)		
	3d	7d	28d	3d	7d	28d
PLAIN	29.6/0	37.5/0	46.8/0	10.6/0	10.9/0	12/0
$P_{\text{HO-G}}$	39.1/+32.1	42.9/+14.4	54.1/+15.6	10.9/+2.8	11.1/+1.8	12.4/+3.3
G1H9	47.5/+60.5	50.1/+33.6	60.1/+28.4	11.1/+4.7	11.4/+4.6	12.8/+6.7
G_2H_8	46.3/+56.4	50.3/+34.1	56.6/+20.9	11.3/+6.6	11.9/+9.2	12.5/+4.2
G_3H_7	50.3/+69.9	55.0/+46.7	62.5/+33.5	11.3/+6.6	12.1/+11.0	12.9/+7.5
G_4H_6	47.4/+60.1	54.0/+44.0	63.1/+34.8	11.3/+6.6	12.1/+11.0	13.2/+10.0
G_5H_5	54.2/+83.1	56.9/+51.7	65.6/+40.2	12.0/+13.2	13.3/+22.0	14.1/+17.5
G_6H_4	52.2/+76.4	57.9/+54.4	63.0/+34.6	11.7/+10.4	13/+19.3	13.4/+11.7
G_7H_3	49.3/+66.6	54.7/+45.9	59.8/+27.8	12.3/+16.0	13.1/+20.2	13.9/+15.8
G_8H_2	51.7/+74.7	57.7/+53.9	58.6/+25.2	12.2/+15.1	13.3/+22.0	14.1/+17.5
G_9H_1	50.5/+70.6	53.6/+42.9	58.2/+24.4	12.1/+14.2	13.2/+21.1	14.0/+16.7
P _{GO}	36.5/+23.3	46.4/+23.7	54.8/+17.1	12.3/+16.0	13.4/+22.9	14.6/+21.7

 Table S4. Compressive and Flexural strength of cement mortars with the hybrids of

various GO: HO-G ratios at 3, 7 and 28d.

a. Average value.

b. Compared to PLAIN.

	Improvement ratio of the compressive strength (%))	
Samples	Compared to P _{HO-G}			Compared to P _{GO}			
	3d	7d	28d	3d	7d	28d	
G ₁ H ₉	+17.1	+16.8	+11.1	+30.1	+8.0	+9.7	
G_2H_8	+15.5	+17.2	+4.6	+26.8	+8.4	+3.3	
G_3H_7	+22.3	+28.2	+15.5	+37.8	+18.5	+14.1	
G_4H_6	+17.5	+25.9	+16.6	+29.9	+16.4	+15.1	
G_5H_5	+27.9	+32.6	+21.3	+48.5	+22.6	+19.7	
G_6H_4	+25.1	+35.0	+16.5	+43.0	+24.8	+15.0	
G_7H_3	+20.7	+27.5	+10.5	+35.1	+17.9	+9.1	
G_8H_2	+24.4	+34.5	+8.3	+41.6	+24.4	+6.9	
G_9H_1	+22.6	+24.9	+7.6	+38.3	+15.5	+6.2	

Table S5. The improvement ratio of the compressive strength of cement mortars withthe hybrids of various GO: HO-G ratios at 3, 7 and 28d.

	Improvement ratio of the flexural strength (%)						
Samples	Compared to P _{HO-G}			Compared to P _{GO}			
	3d	7d	28d	3d	7d	28d	
G ₁ H ₉	+1.8	+2.7	+3.2	-9.8	-14.9	-12.3	
G_2H_8	+3.7	+7.2	+0.8	-8.1	-11.2	-14.4	
G_3H_7	+3.7	+9.0	+4.0	-8.1	-9.7	-11.6	
G_4H_6	+3.7	+9.0	+6.5	-8.1	-9.7	-9.6	
G_5H_5	+10.1	+19.8	+13.7	-2.4	-0.7	-3.4	
G_6H_4	+7.3	+17.1	+8.1	-4.9	-3.0	-8.2	
G_7H_3	+12.8	+18.0	+12.1	0	-2.2	-4.8	
G_8H_2	+11.9	+19.8	+13.7	-0.8	-0.7	-3.4	
G_9H_1	+11.0	+18.9	+12.9	-1.6	-1.4	-4.1	

Table S6. The improvement ratio of the flexural strength of cement mortars with thehybrids of various GO: HO-G ratios at 3, 7 and 28d.