

Electronic Supplementary Information

One-step synthesis of porous graphene-based hydrogels containing oil droplets for drug delivery

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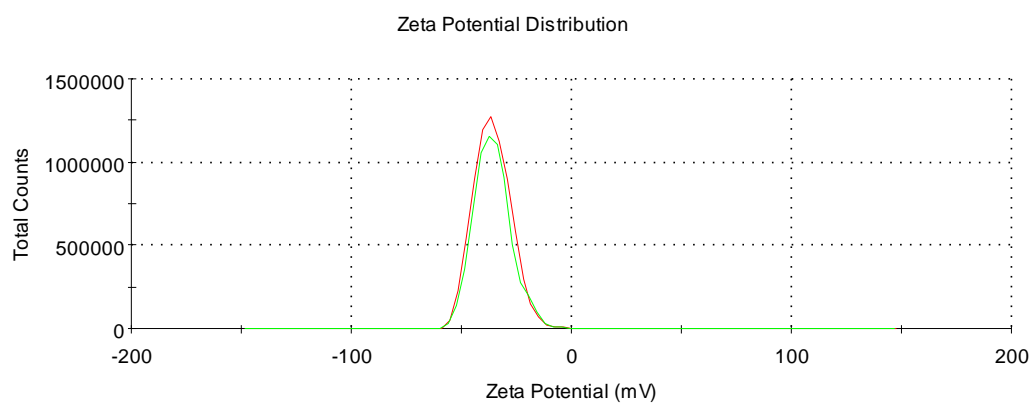


Figure S1. Zeta potential of GO in aqueous solution.

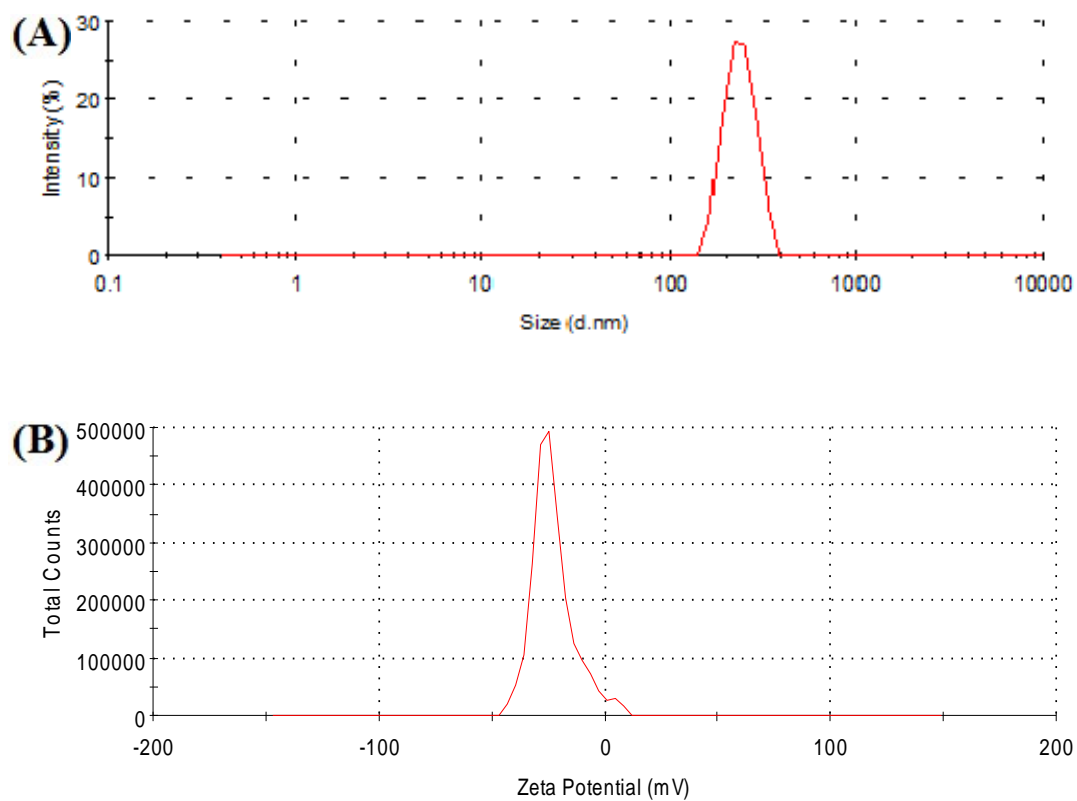


Figure S2. (a) Size and (b) Zeta potential of Fe₃O₄ in water.

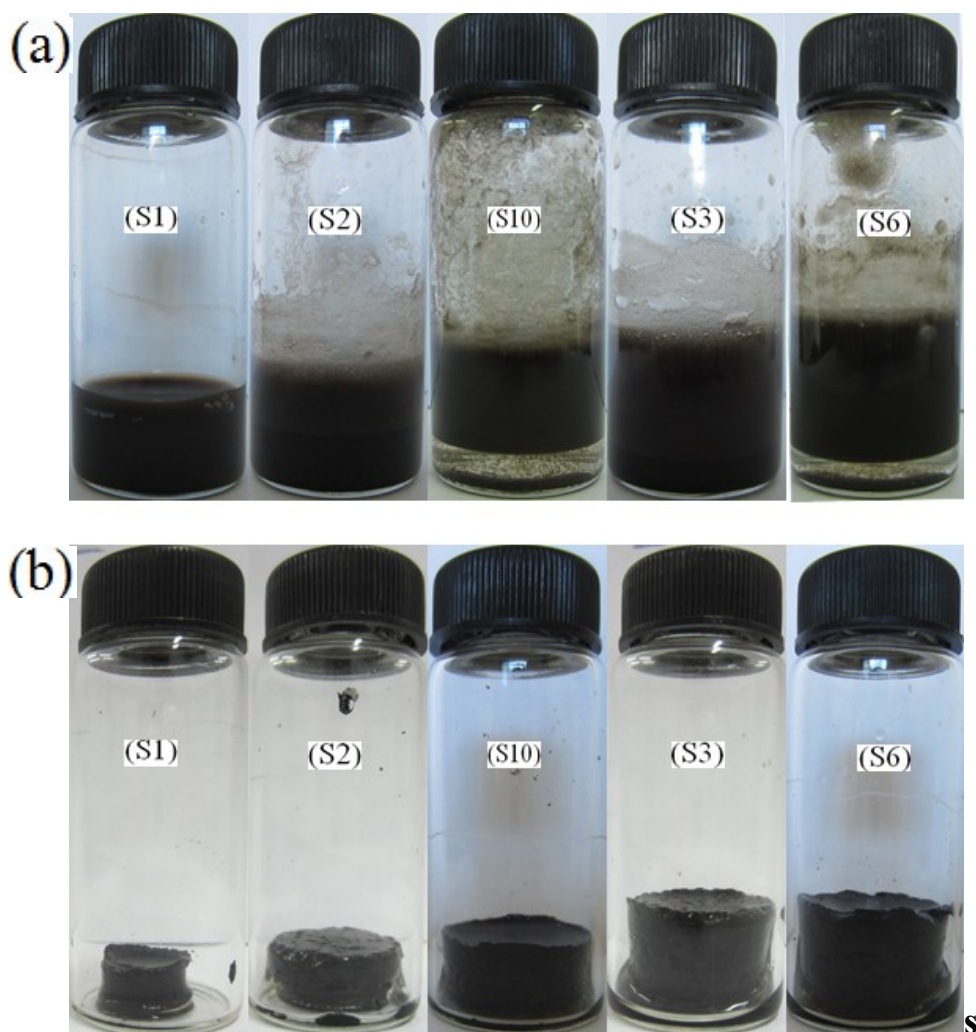


Figure S3. (a) Photos of the GO aqueous dispersion (S1) and typical Pickering emulsions (S2, S3, S6, S10) and (b) the corresponding prepared RGO hydrogels.

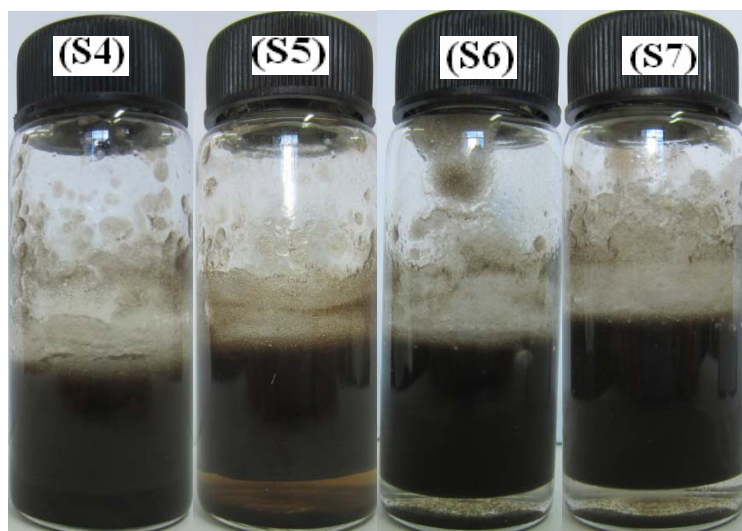


Figure S4. Photographs of emulsions S4, S5, S6 and S7.

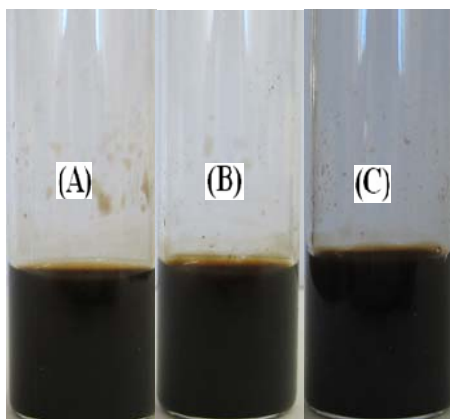


Figure S5. Photos of GO aqueous dispersions with different concentrations of Fe_3O_4 :
(a) 0 mg/mL, (b) 1.5 mg/mL, and (c) 2.5 mg/mL.

(A)

Sample	GO (5 mg/mL)	Fe ₃ O ₄ (20 mg/mL)	b (GO:Fe ₃ O ₄ =10:3)	c (GO:Fe ₃ O ₄ =2:1)
Contact angle (°)	39.2	14.1	35.2	35.1

Note: The ratio of GO and Fe₃O₄ is the weight ratio.

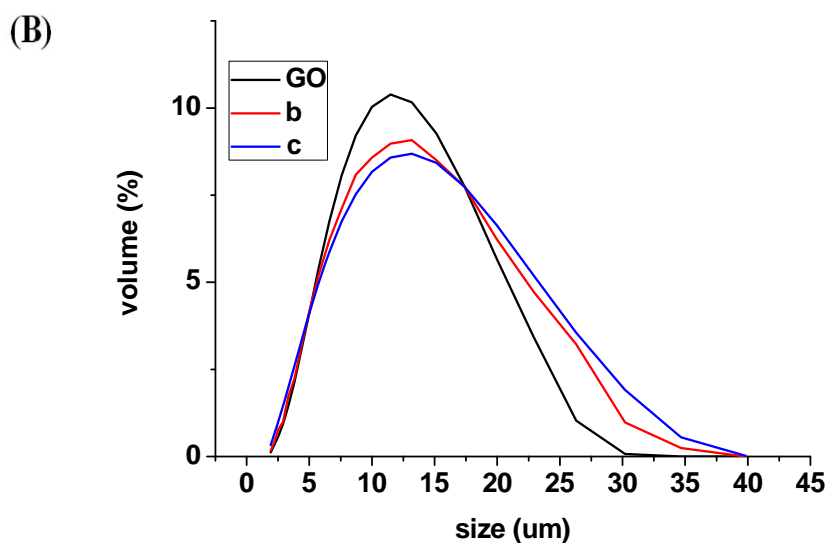


Figure S6. (A) Contact angle of GO, Fe₃O₄, GO and Fe₃O₄ mixture dispersions (b: GO:Fe₃O₄=10:3 w:w; c: GO:Fe₃O₄=2:1, w:w); (B) the size and size distribution of the dispersions.

To study the interaction between GO and Fe₃O₄, we put both in three vials without oil phase (Figure S4). From left to right, the concentrations of Fe₃O₄ were 0, 1.5, 2.5 mg/mL, and the size of mixed solute was 11.1, 12.4, 12.9 μm, respectively (Figure S6B), small increase on size after adding Fe₃O₄ nanoparticles as we can see. It also can attribute to the combination of GO and Fe₃O₄. We observed three vials after one week without stirring, aqueous solution was still homogeneous, no sedimentation occurred even after one week. If GO sheets and Fe₃O₄ nanoparticles formed complexes, which more easily assembled at the interface of water-oil phase, GO

would totally enter into emulsion so that the bottom became clear. Then these three aqueous solutions were used to cast into membranes under room temperature. The contact angles (CA) of them were 39.2, 35.2, 35.1°, respectively (Figure S6A). Bare Fe₃O₄ aqueous membrane had much smaller CA (14.1°), indicating Fe₃O₄ nanoparticles was more hydrophilic than GO sheets. The CA of RGO (49.5°) was much bigger than GO, it assigned to the decrease of oxygen-containing groups when heating. After adding Fe₃O₄, aqueous solution became viscosity and membrane was a little more hydrophilic than bare GO membrane. The mixture of them had a Zeta potential (-38.1 mV) falling in between them, their CA was in accordance with their Zeta potential. Therefore it is reasonable that complexes stably existing in emulsion.

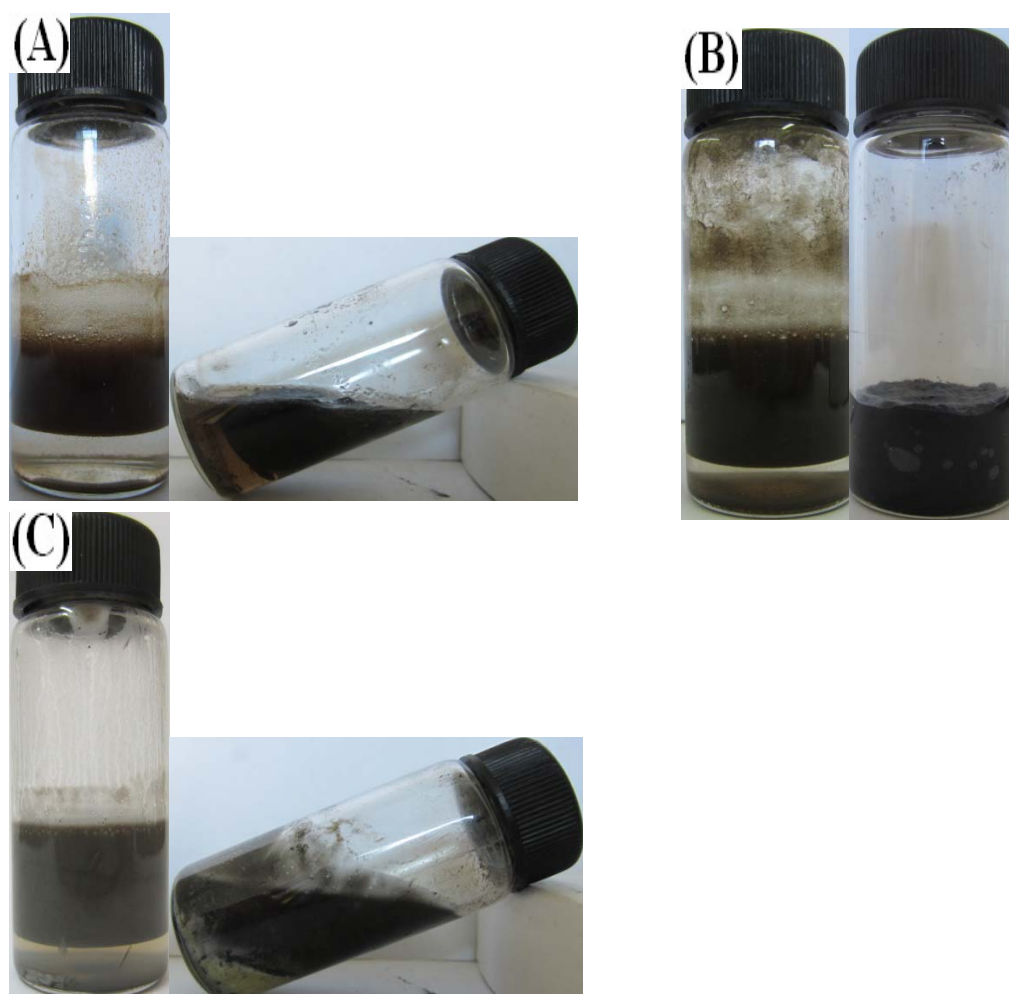


Figure S7. (A) Photos of S8 emulsion and the mixture after heating for 2 h, (B) Photos of S9 emulsion and the prepared hydrogel after heating for 2 h, (C) Photos of the emulsion using Tween 60 as stabilizer and the mixture after heating for 2 h.

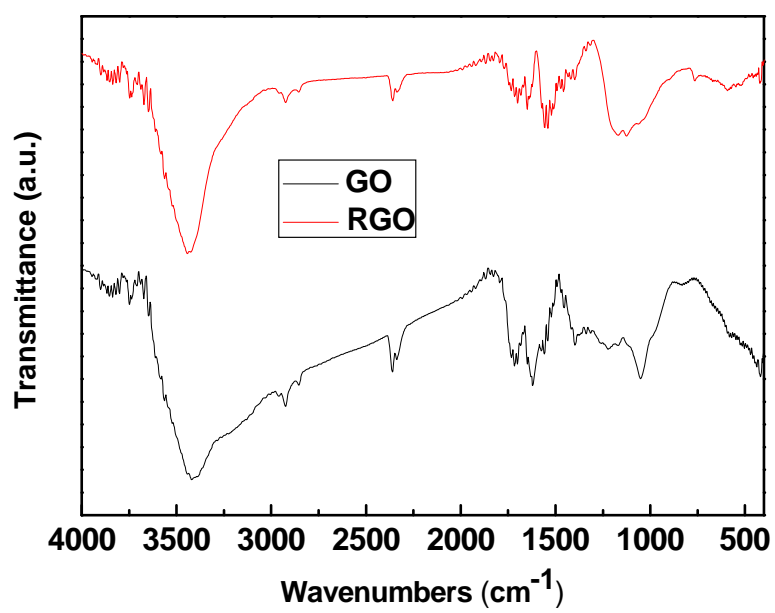


Figure S8. FTIR spectra of GO and RGO

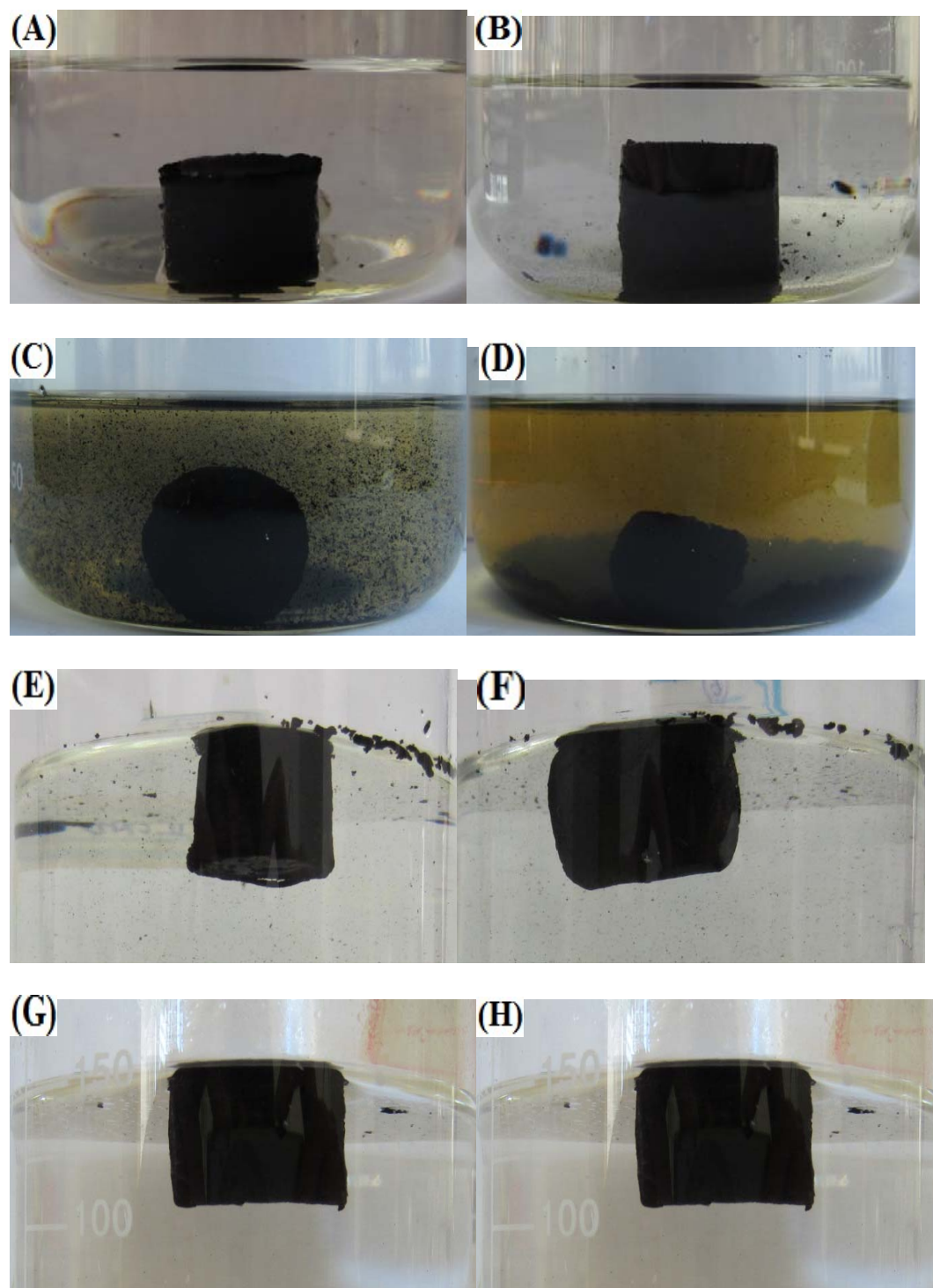


Figure S9. Photos of hydrogels after release in external phases: (A) PN-loaded S10 hydrogel in hexane, (B) PN-loaded S6 hydrogel in hexane, (C) PN-loaded S10 hydrogel in acetone, (D) PN-loaded S6 hydrogel in acetone; (E) IBU-loaded S10 hydrogel in PBS (pH=7.4), (F) IBU-loaded S6 hydrogel in PBS (pH=7.4), (G) IBU-loaded S10 hydrogel in PBS (pH=2.0), (H) IBU-loaded S6 hydrogel in PBS (pH=2.0).