

## Supplementary Information

### Soft-template synthesis of 3D porous graphene foams with tunable architectures for lithium-O<sub>2</sub> battery and oil adsorption applications

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#### Experimental

**Preparation of graphene oxide aqueous suspension:** Graphite oxide was synthesized from graphite, using a modified Hummers method. In a typical reaction, graphite (0.5g), NaNO<sub>3</sub> (0.5 g), and H<sub>2</sub>SO<sub>4</sub> (23 ml) were stirred together in an ice bath. KMnO<sub>4</sub> (3g) was added slowly. After mixing for 30 min, the suspension was transferred to a water bath (35 °C) and stirred for about 1 h. Then, H<sub>2</sub>O (40 mL) was added, and the mixture was stirred for 30 min while the temperature was raised to 90 °C. Finally, H<sub>2</sub>O (100 mL) was added, followed by the slow addition of 3 mL of H<sub>2</sub>O<sub>2</sub> (30%). The final suspension was then filtered and washed with water to obtain graphite oxide. The as-synthesized graphite oxide was dispersed into distilled water, and sonicated for 2 h to form homogeneous graphene oxide/H<sub>2</sub>O suspension (5.0 mg ml<sup>-1</sup>).

**Synthesis of PGF-3:** DMDMS (6.0 ml) was added into HCl solution (2.0 M, 100 ml) and kept stirring for 30 minutes at 25 °C. Then, the mixture was sonicated for 30 minutes to form a cloudy suspension. Graphene oxide solution (60 ml, 5.0 mg ml<sup>-1</sup>) and distilled H<sub>2</sub>O (40 mL) were mixed into the DMDMS emulsion suspension and the whole system was left to stir for 5 minutes at room temperature. After then, the precipitate was collected by vacuum filtration. The obtained precipitate was firstly sintered at 350 °C in argon for 5 h and further heated at 900 °C for another 5 h under argon atmosphere to obtain the final product PGF-3.

**Synthesis of PGF-4:** N-hexadecane (4.88 ml) and CTAB (27 mg) was mixed in distilled water (300 ml) and kept stirring for 2h minutes at 25 °C. Then, the mixture was sonicated for 30 minutes to form a cloudy suspension. Graphene oxide solution (90 ml, 5.0 mg ml<sup>-1</sup>) and distilled H<sub>2</sub>O (10 mL) were mixed with the emulsion suspension at room temperature. After then, the precipitate was collected by vacuum filtration. The obtained precipitate was firstly sintered at 350 °C in argon for 5 h and further heated at 900 °C for another 5 h under argon atmosphere to obtain the final product PGF-4.

**Synthesis of PGF-5:** F108 (1 g) and TMB (1 g) was mixed in HCl solution (2.0 M, 30 ml) and kept stirring overnight at 25 °C. Graphene oxide solution (180 ml, 5.0 mg ml<sup>-1</sup>) was added into the emulsion suspension and mixed at room temperature. Then, the precipitate was collected by vacuum filtration. The obtained precipitate was firstly sintered at 350 °C in argon

for 5 h and further heated at 900 °C for another 5 h under argon atmosphere to obtain the final product PGF-5.

***Oil adsorptions:*** Hexadecane, toluene and olive oil were chosen as the model oil molecules. Firstly, hexadecane, toluene and olive oil were labeled by Sudan Black B (10 mg ml<sup>-1</sup>). Then, 250 μl dyed hexadecane, toluene and olive oil were injected into water, respectively. PGF-2 monoliths (6 mg) were immersed to start adsorption. PGF-2 recycling was conducted by directly burning the monolith using a Bunsen burner.

The mechanism of oil adsorption relies on the hydrophobic interaction and  $\pi$ - $\pi$  affinity between oil molecules and hierarchical graphene foams. In practical diesel spill and crude oil leaking, light and heavy hydrocarbon components usually exhibit hydrophobic natures and/or contain large  $\pi$ -conjugations. The hierarchical porous graphene foam has several special features that make them ideal for oil cleanup. The graphene skeletons are compatible with hydrocarbons, because graphene are hydrophobic and consisted by  $\pi$ -conjugated sp<sup>2</sup> carbon atoms. Furthermore, the massive pores with hierarchical structure can provide capillary force and large free spaces to collect oils.

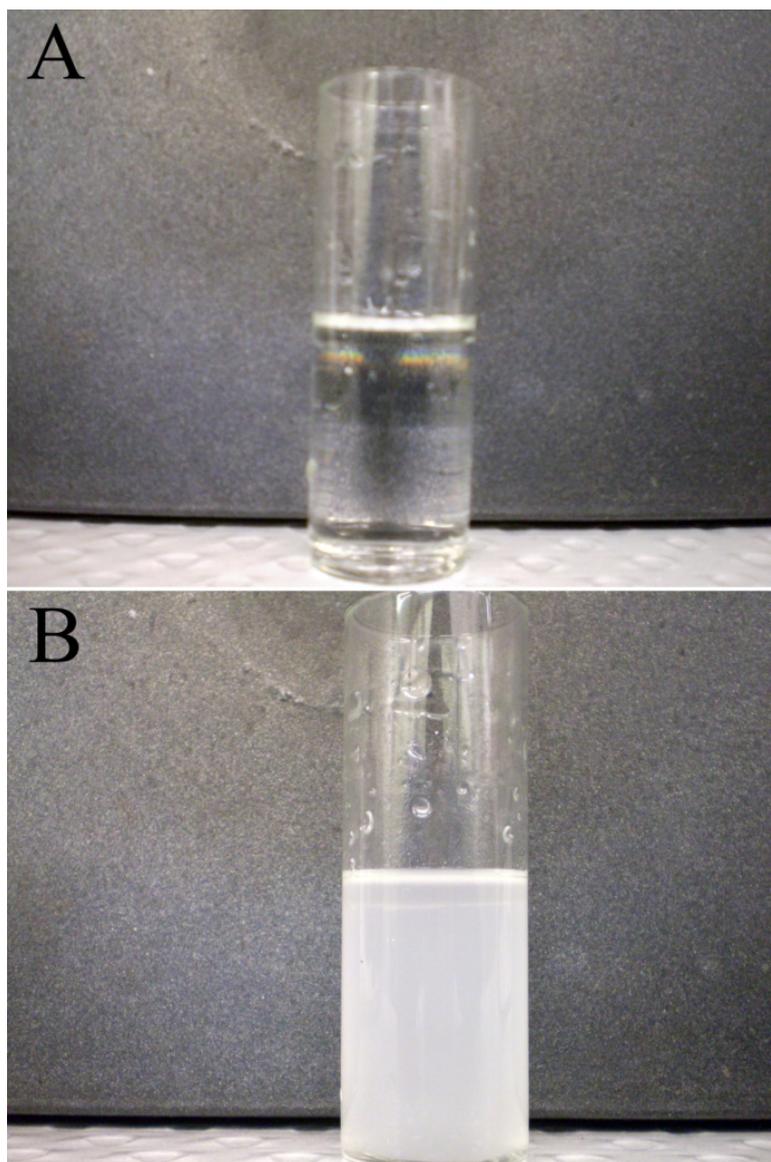


Figure S1. Digital photos of TMB emulsion suspension. (A) Before stirring and sonication, TMB was separated and floated on surface of the aqueous solution. (B) After agitation by stirring and sonicating, a cloudy suspension was formed.

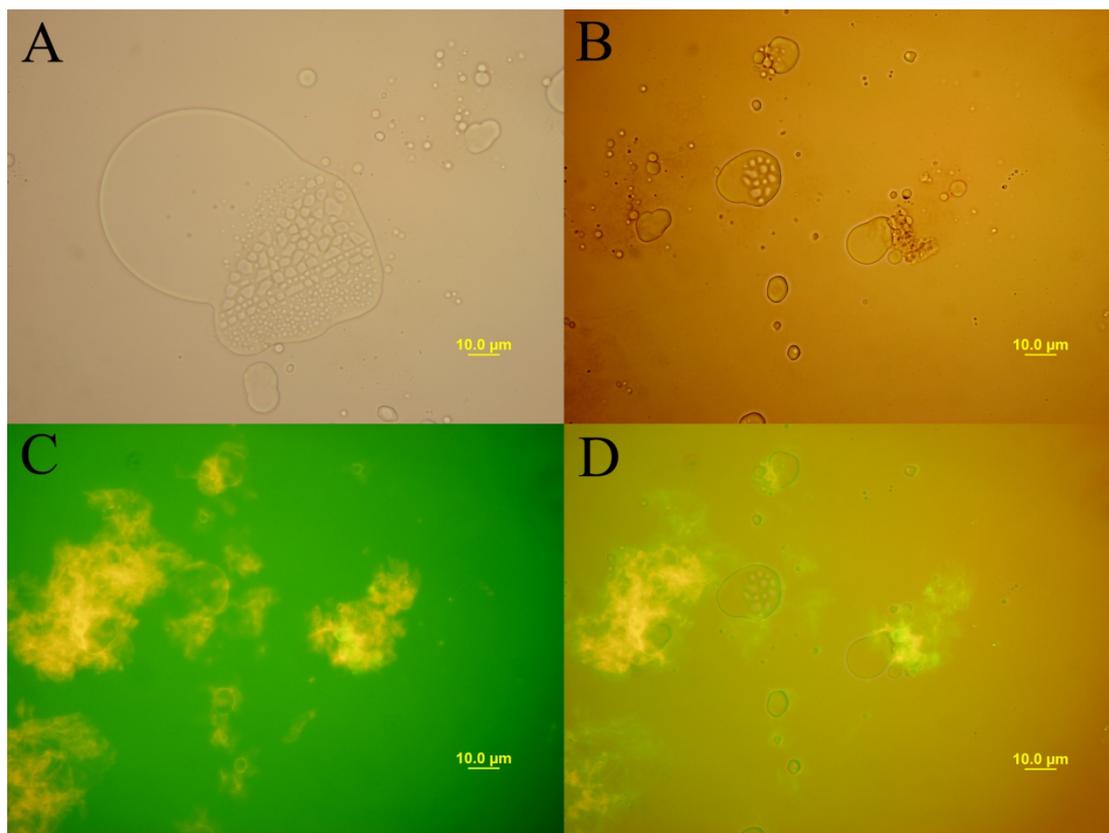


Figure S2. Self-assembly of TMB emulsions and GO nanosheets under the optical microscopy observation. (A) TMB emulsion suspension under bright field observation. (B) TMB emulsion/GO composite suspension under bright field observation. (C) Fluorescent view of TMB emulsion/GO composite suspension, with an excitation light of 450-480 nm. (D) The combined view of (B) and (C).

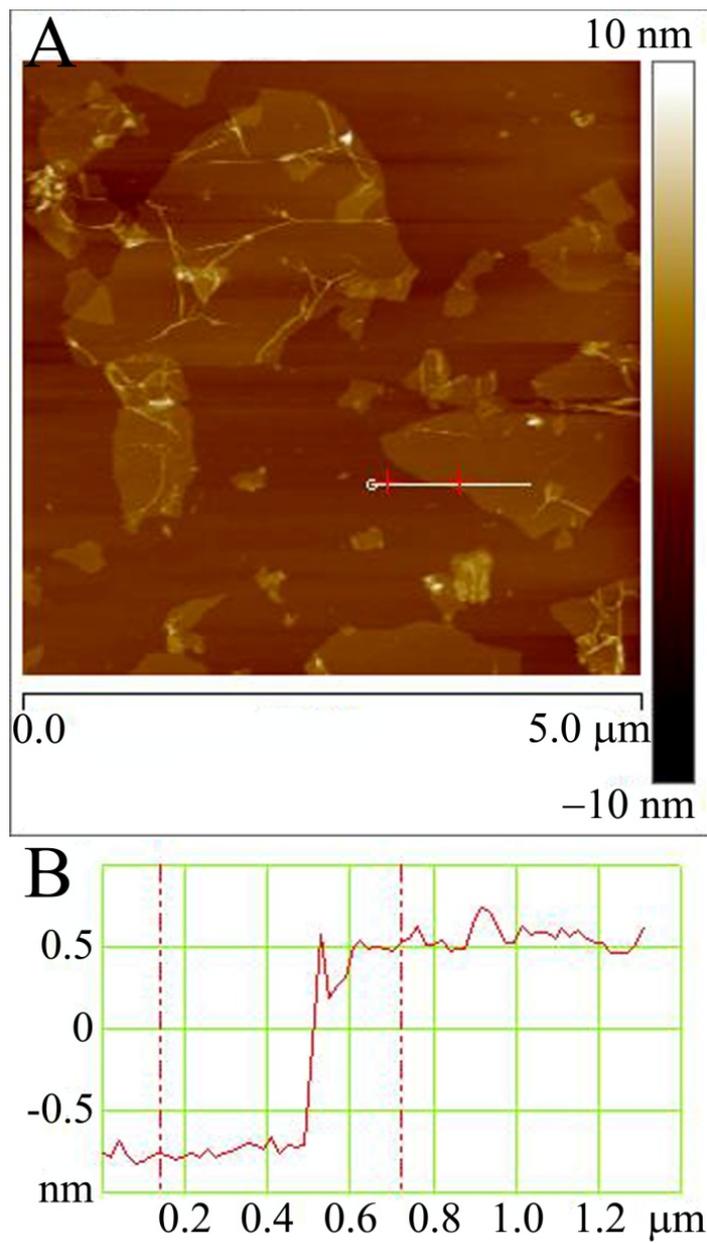


Figure S3. AFM analysis results of GO.

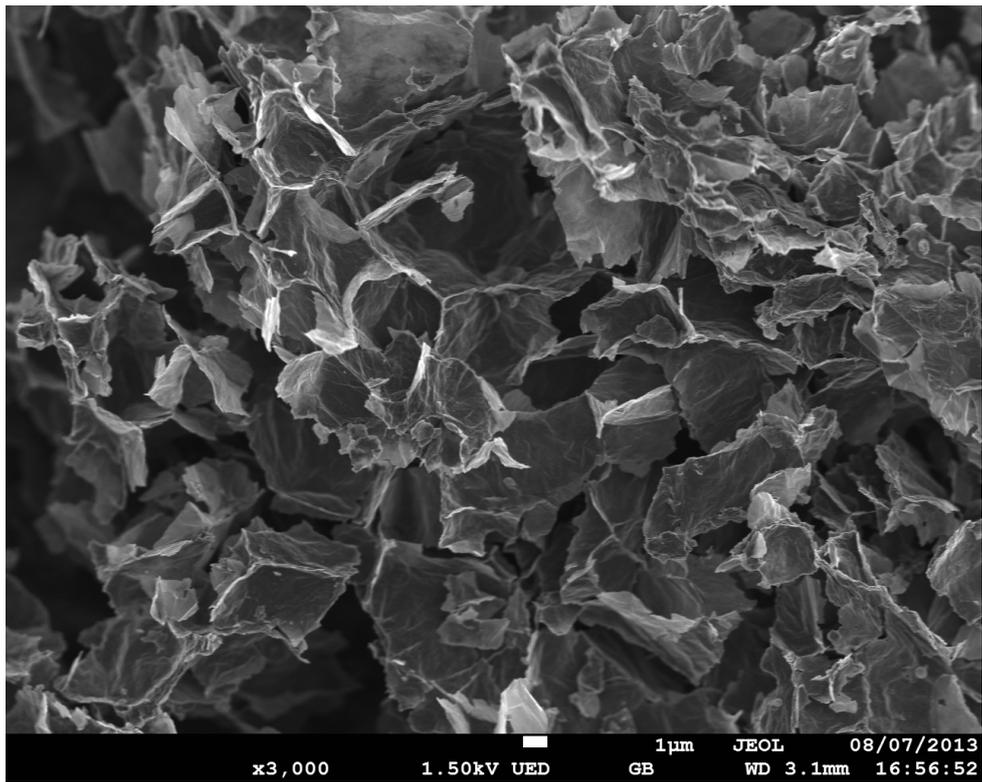


Figure S4. Low magnification SEM view of the PGF-1 product.

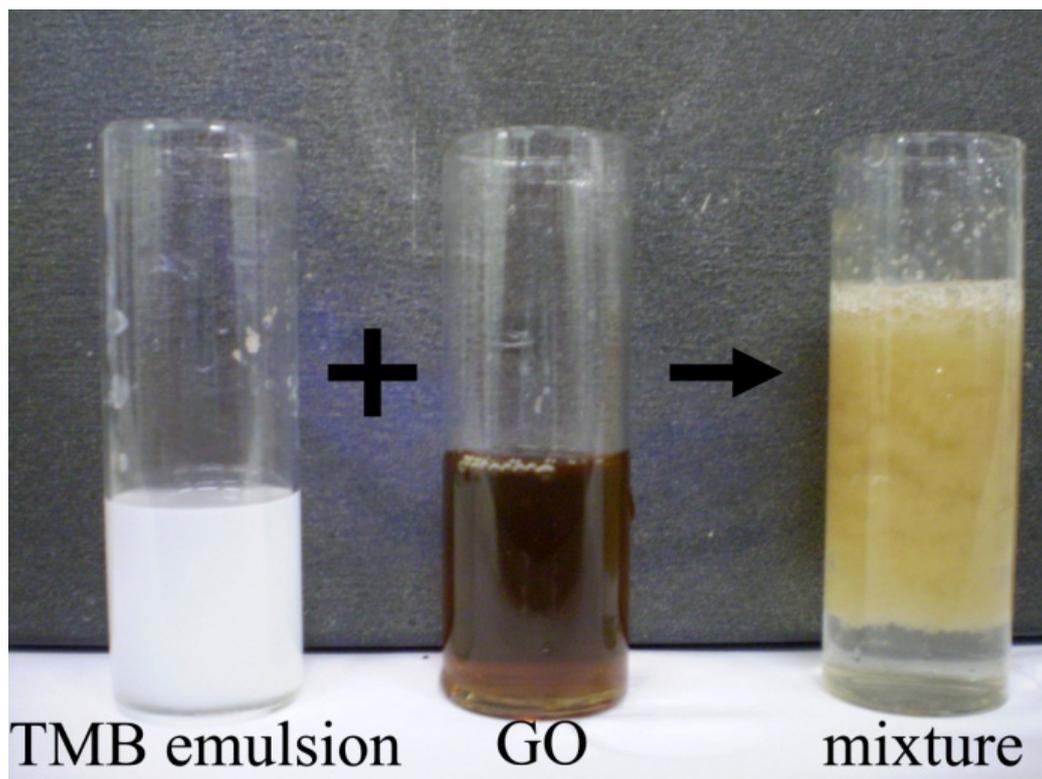


Figure S5. Digital photos of TMB emulsion suspension, GO solution and their mixture.

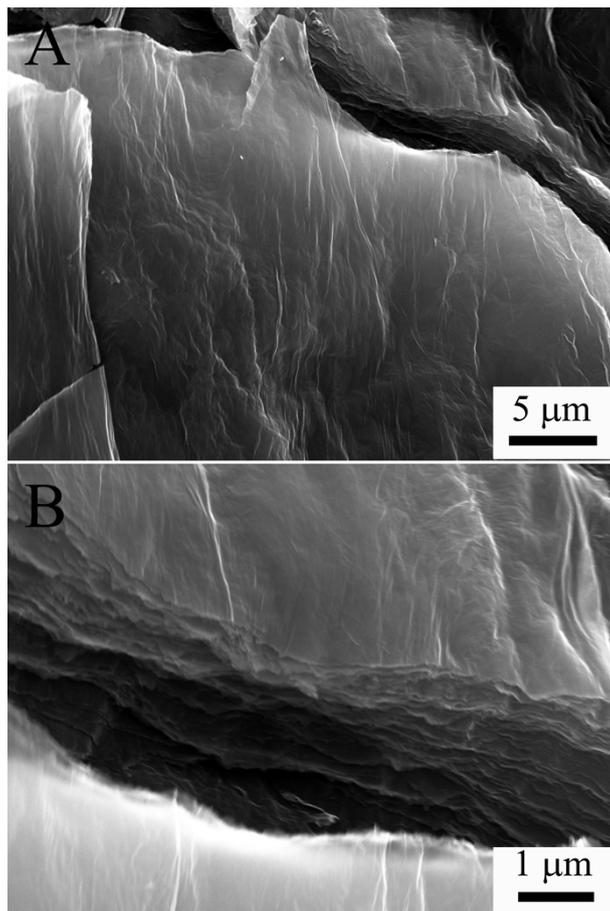


Figure S6. SEM images of graphene sample, which was synthesised without adding TMB emulsion during the preparation process, showing the compact (non-porous) feature.

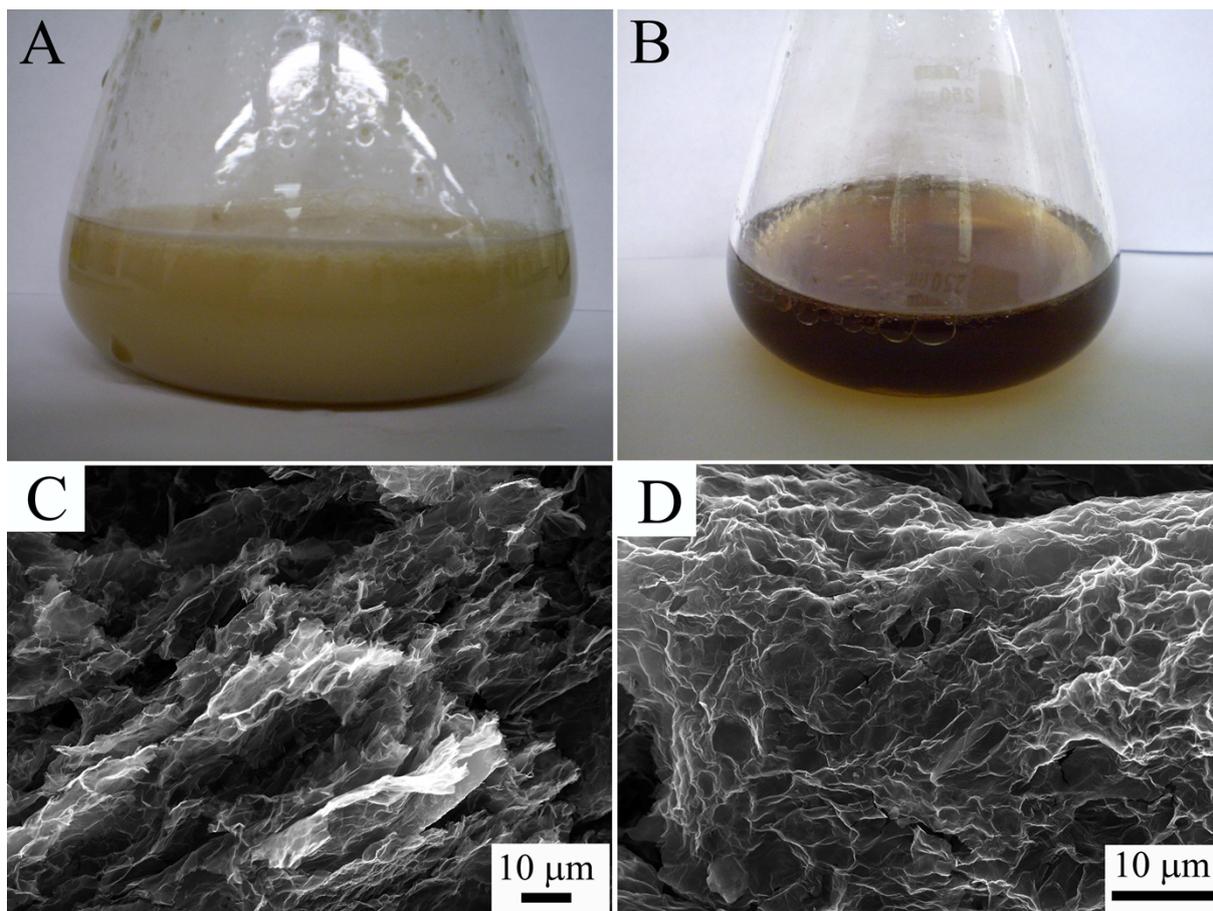


Figure S7. TMB and GO in natural solution. (A) A homogeneous suspension was formed in the beginning. (B) After 2 hours, severe phase separation occurred. TMB emulsions aggregated and form large droplets. GO solution became transparent. (C) and (D) SEM images of the product collected in the initial filtration (after calcination). A highly porous morphology can be observed.

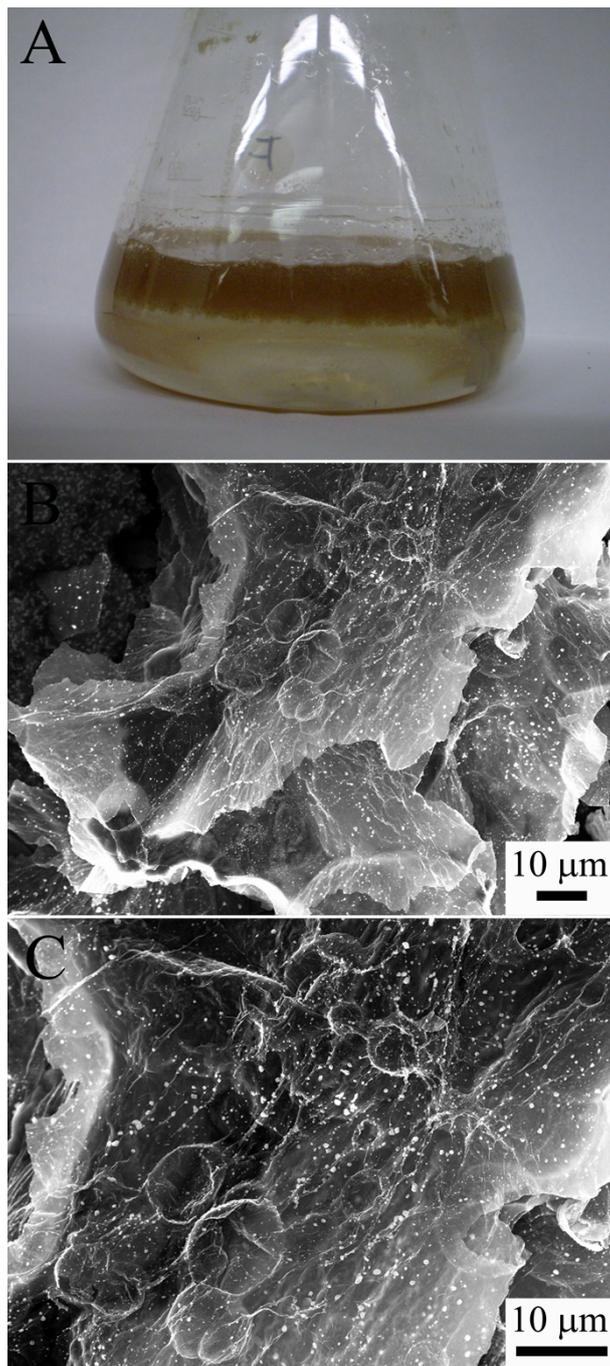


Figure S8. TMB and GO in KCl solution. (A) Digital photo of the composite, floating on the suspension. (B) and (C) SEM images of the product after calcination. Both porous graphene foam and salt residues were observed.

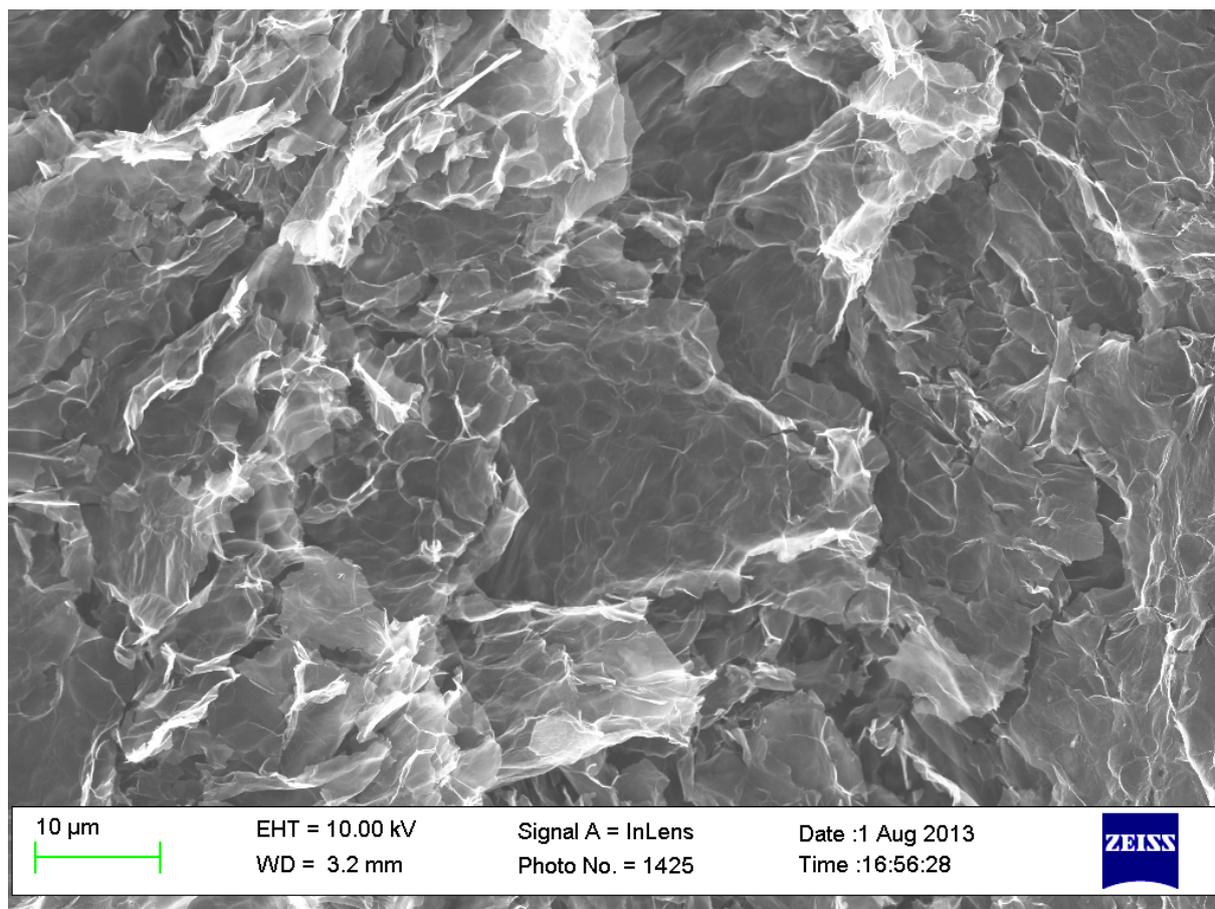


Figure S9. SEM images of H<sub>2</sub>SO<sub>4</sub> acid product after calcination. Highly porous graphene foam was presented.

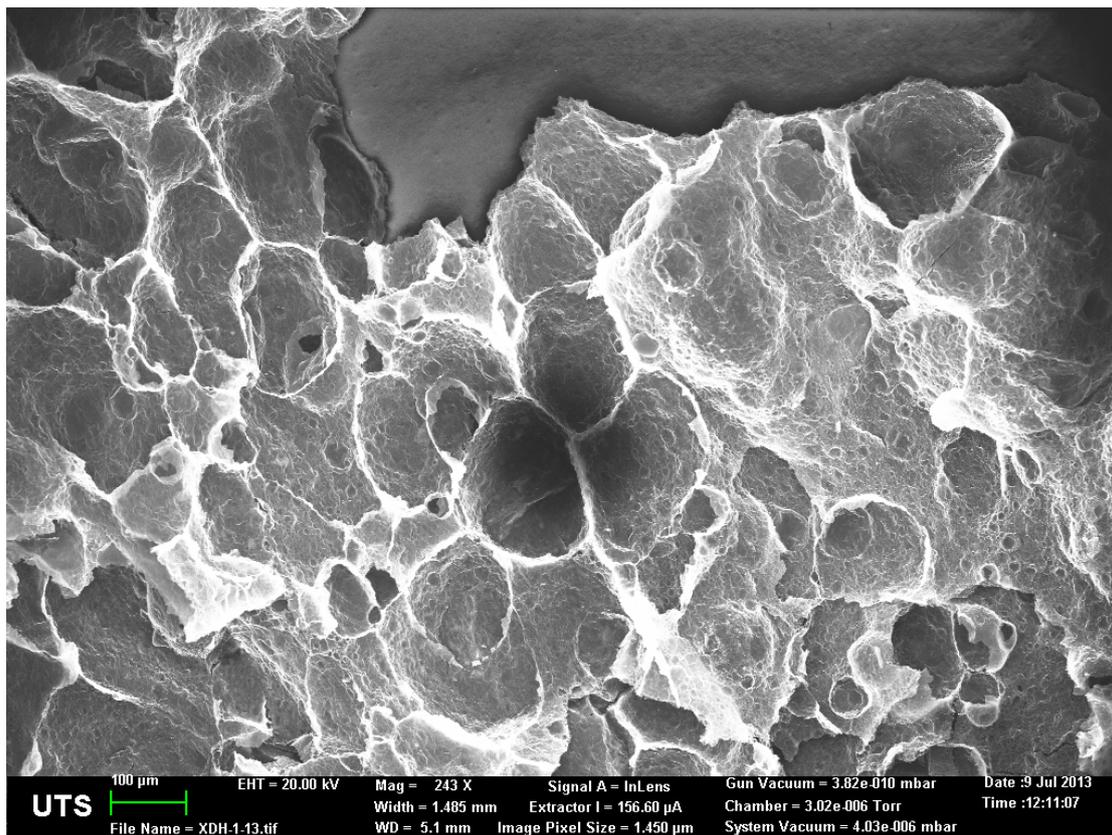


Figure S10. Low magnification SEM view of PGF-2 product.

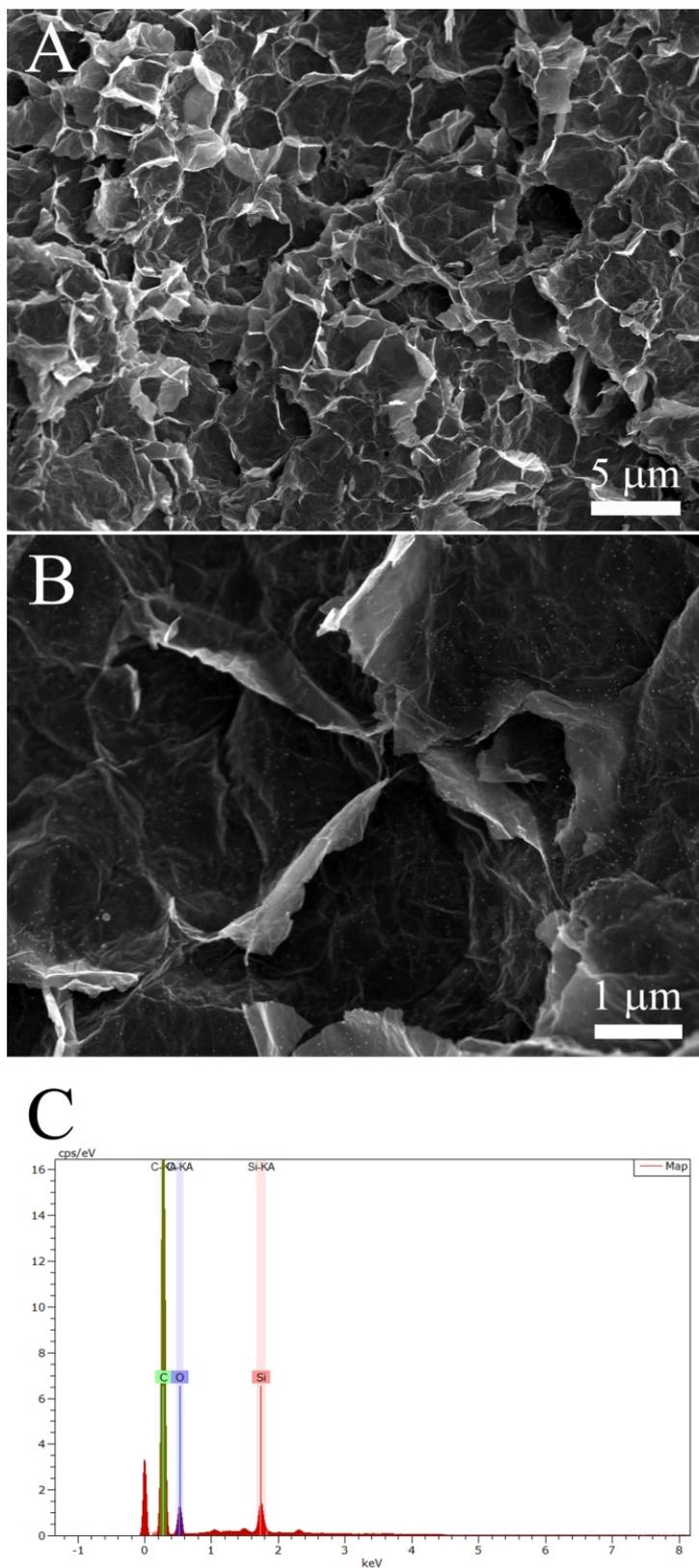


Figure S11. SEM characterization of PGF-3. (A) and (B) SEM images of PGF-3. (C) EDS analysis result of PGF-3. Porous graphene foams with an uniform distribution of silica nanoparticles on the surface of graphene.

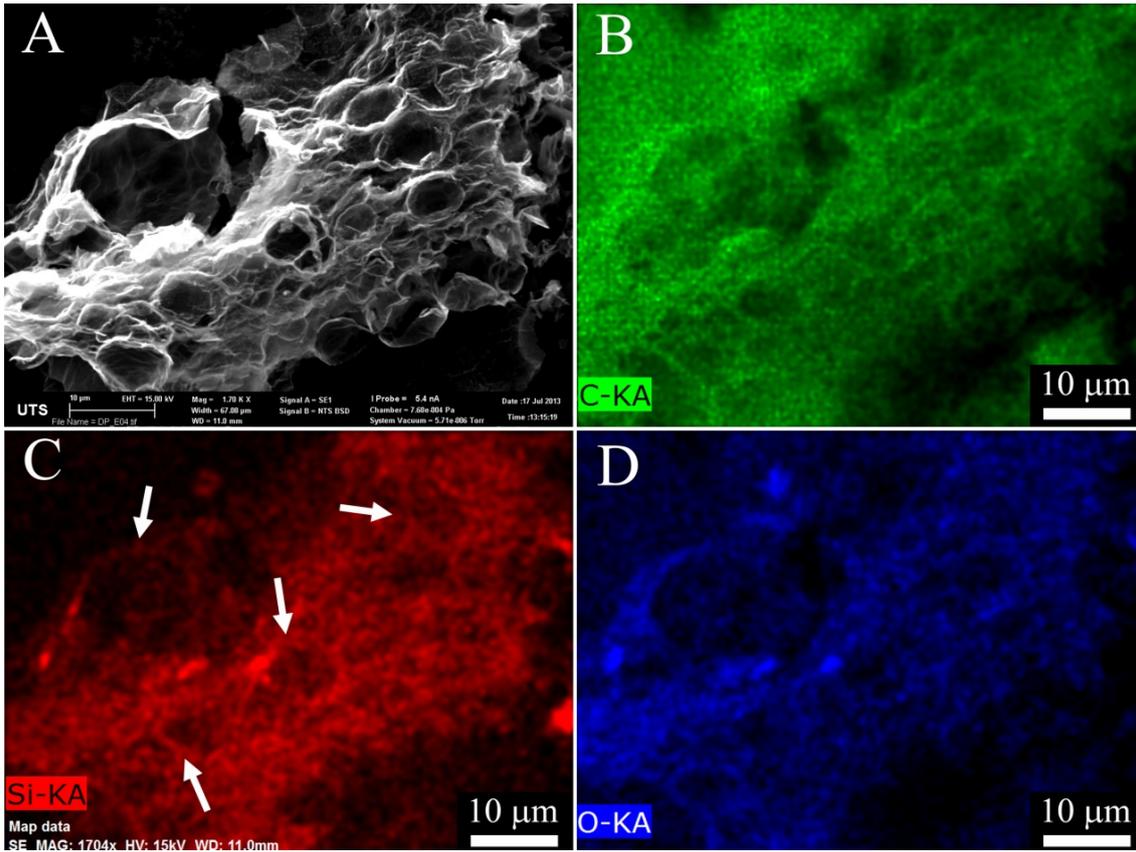


Figure S12. Element mapping results of PGF-3. (A) SEM image of PGF-3. (B) carbon, (C) silicon and (D) oxygen element mapping images, showing a homogeneous distribution of silica nanoparticles on graphene foam. Marked by arrows, pore fringes can be clearly identified.

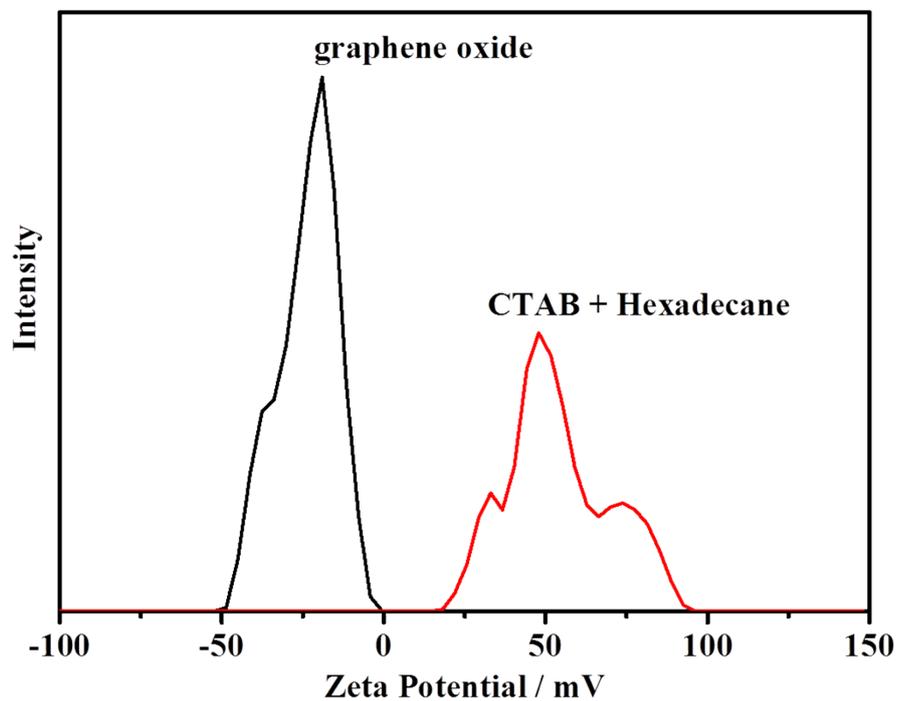


Figure S13. Zeta-potential measurement result of GO (centered at -19.0 mV) and CTAB/Hexadecane emulsion (main peak at 47.9 mV).

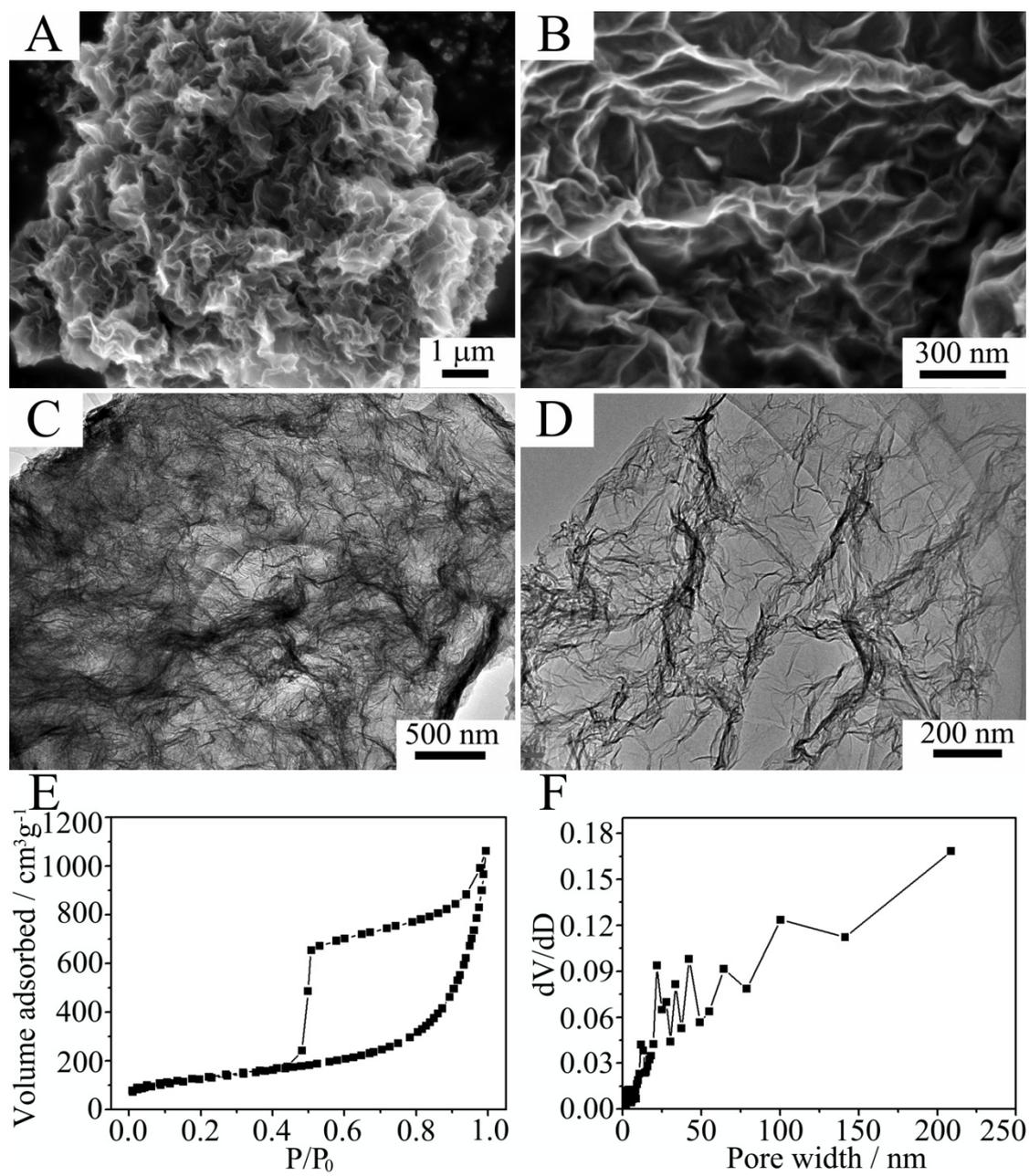


Figure S14. Structural characterization of PGF-4. (A) and (B) SEM images with different magnifications. (C) and (D) TEM images with different resolutions. (E) Nitrogen-sorption isotherms and (F) the corresponding pore diameter distribution.

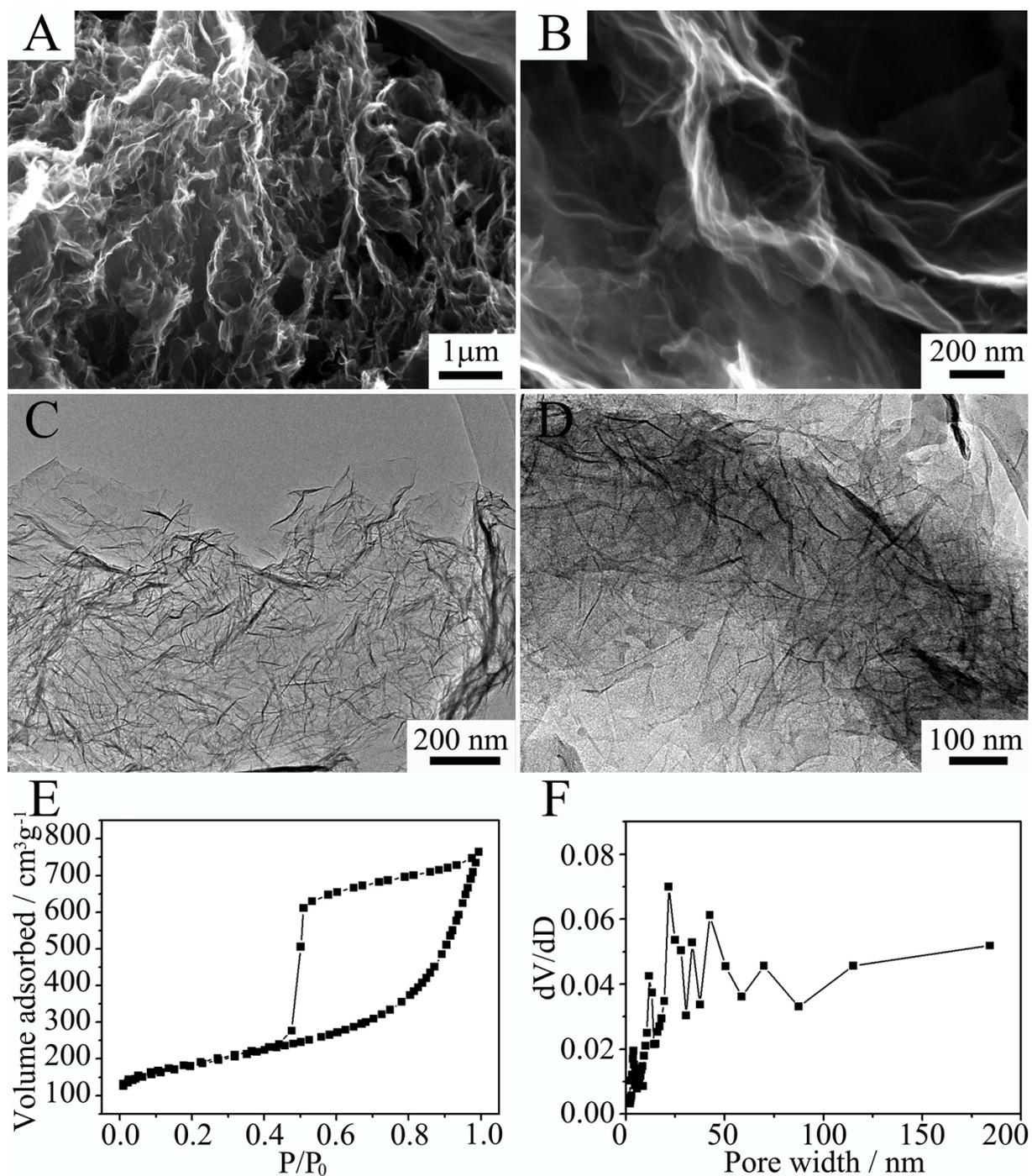


Figure S15. Structural characterizations of PGF-5. (A) and (B) SEM images with different magnifications. (C) and (D) TEM images with different resolutions. (E) Nitrogen-sorption isotherms and (F) the corresponding pore diameter distribution.

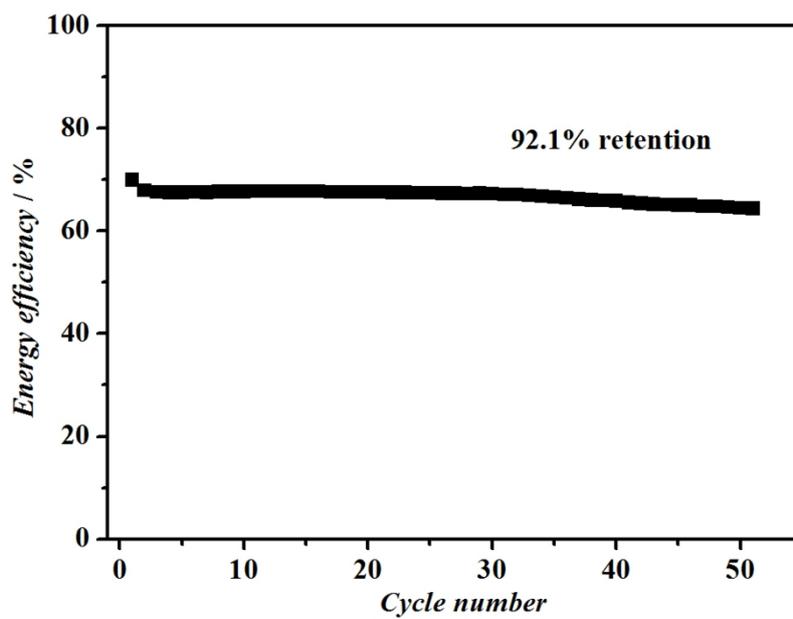


Figure S16. Roundtrip electrical efficiency of of PFG-1 at 200 mA g<sup>-1</sup> with a curtailing capacity of 1000 mAh g<sup>-1</sup>.

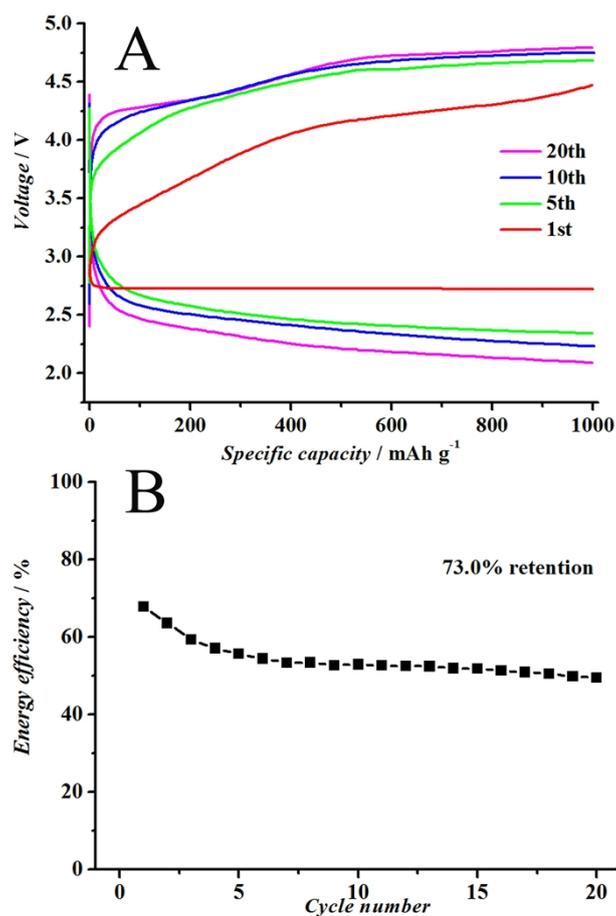


Figure S17. (A) Charge-discharge profiles and (B) roundtrip electrical efficiency of the non-porous graphene cathode cell at various cycle numbers at 200 mA g<sup>-1</sup> with a curtailing capacity of 1000 mAh g<sup>-1</sup>.

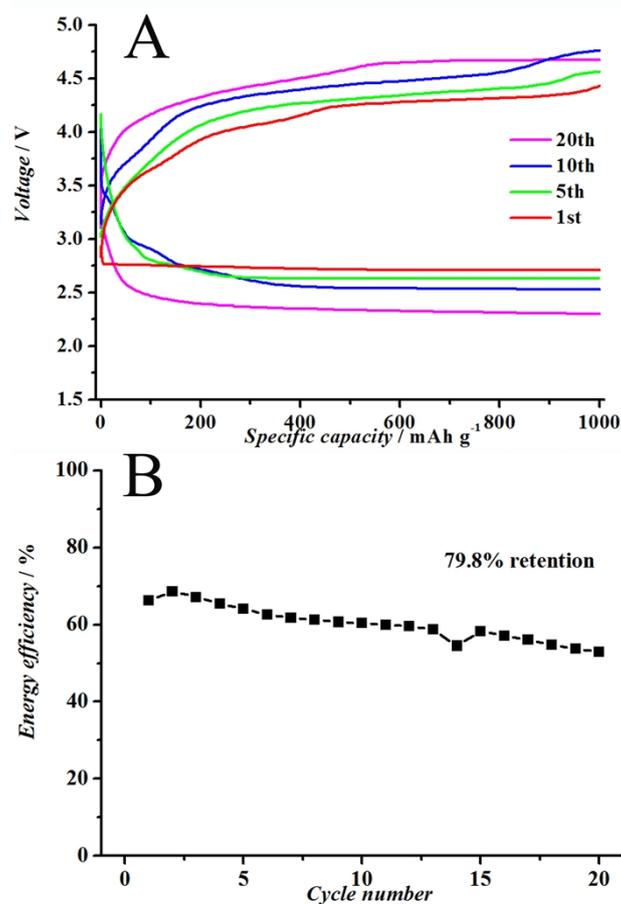


Figure S18. (A) Charge-discharge profiles and (B) roundtrip electrical efficiency of the non-porous graphene cathode cell at various cycle numbers at 200 mA g<sup>-1</sup> with a curtailing capacity of 1000 mAh g<sup>-1</sup>.

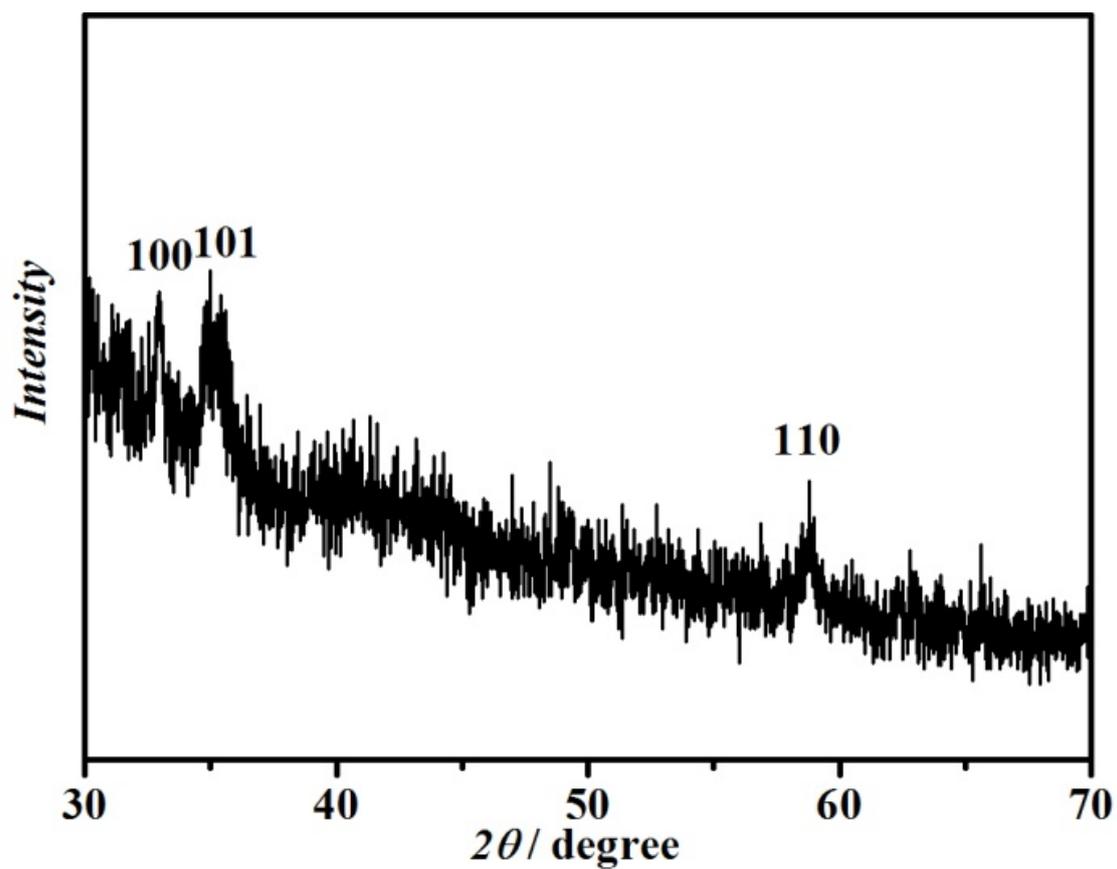


Figure S19. XRD pattern of PGF-1 electrode after discharge in the first cycle. Diffraction peaks can be identified to Hexagonal phase of lithium peroxide (JCPDS 09-0355).