

Electronic Supporting Information

Electrodeposition of polyaniline on three-dimensional Graphene hydrogel as a binder-free supercapacitor electrode with high power and energy densities†

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1. Preparation of graphene hydrogel (GH)

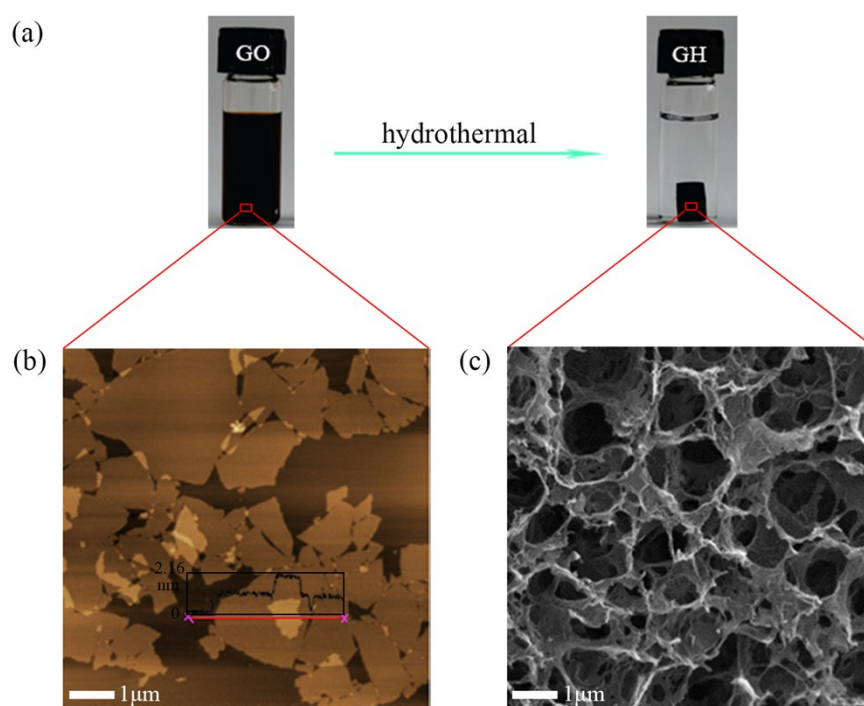


Fig. S1 (a) Preparation of GH via one-step hydrothermal method, (b) AFM image of GO sheets, and (c) SEM image of obtained GH.

2. Raman spectra of GH, PANI, and GH/PANI samples

The Raman spectrum of GH, as expected, displays two prominent peaks at 1600 and 1335 cm^{-1} , corresponding to the G band (in-plane bond-stretching motion of the sp^2 carbon atoms) and D bands (breathing modes of rings or K -point phonons of A_{1g} symmetry) of reduced GO, respectively.^{1,2} The D/G band intensity ratio of GH expresses the atomic ratio of sp^2/sp^3 carbons, is a measure of the extent of disordered graphite.³ The D/G intensity ratio of GH was calculated to be 1.39, indicating an incomplete recovery of graphite structure. In the Raman spectrum of PANI, the bands at 416, 519, 574 and 809 cm^{-1} are attributed to out-of-plane C-H wag, out-of-plane C-N-C torsion, in-plane amine deformation and out-of-plane C-H deformation in the quinonoid ring, respectively. The bands at 1167, 1220, 1594 and 1637 cm^{-1} are associated with C-H bending in the benzenoid ring, C-N stretching mode of the polaronic units, C=C stretching in the quinonoid ring and C-C stretching, respectively. The semiquinone radical structure vibration and the C=N stretching mode of the quinonoid units are situated at 1330-1396 cm^{-1} and 1470-1503 cm^{-1} respectively.⁴⁻⁶ For the GH/PANI composites, the relative intensities of Raman bands associated with PANI, such as 1167 and 1470 cm^{-1} , increased with the increase of deposition time, indicating that more and more PANI was electrodeposited on the GH with the increase in PANI deposition time.

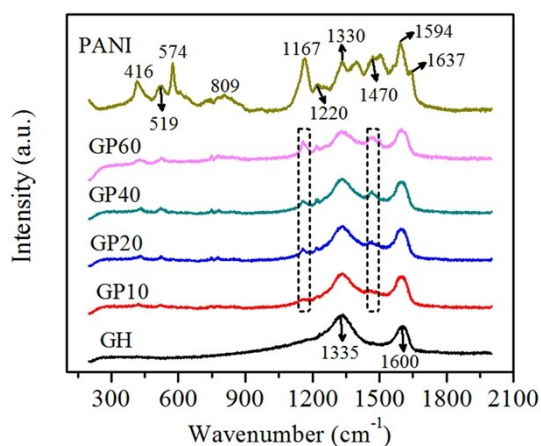


Fig. S2 Raman spectra of GH, PANI, and GH/PANI samples with different electrodeposition time.

3. The morphologies of lyophilized GP10, GP40 and GP60

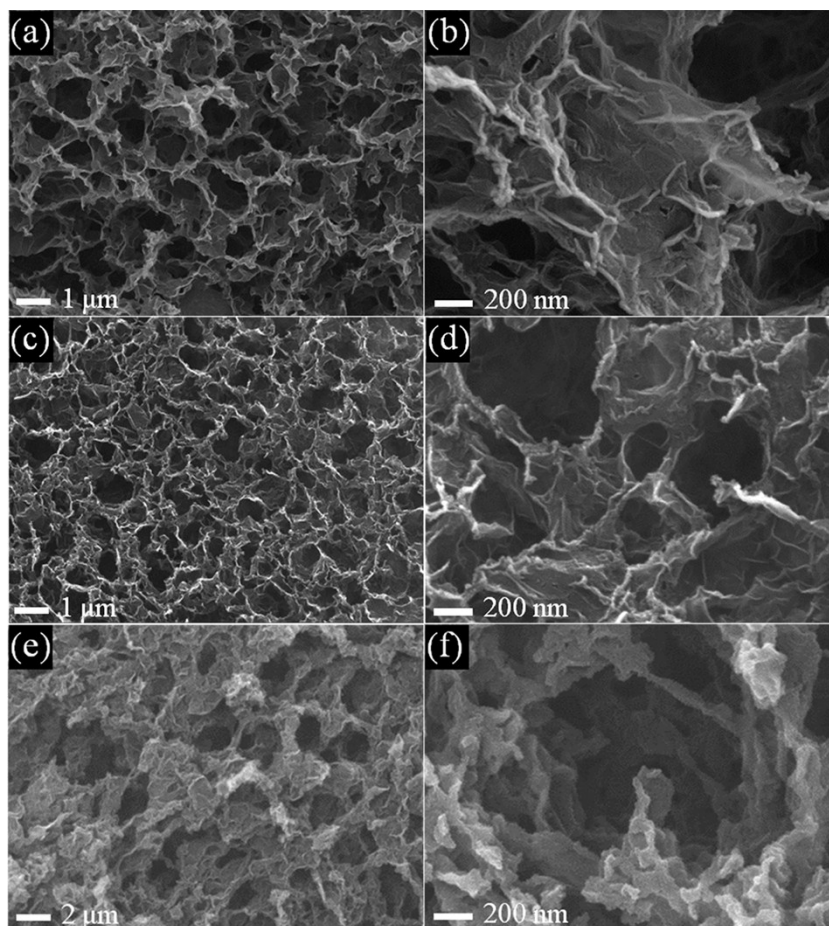


Fig. S3 SEM images of GP10 (a, b), GP40 (c, d) and GP60 (e, f). Low-magnification SEM images (a, c, e) showing the 3D macroporous structure. High-magnification SEM images (b, d, f) showing the morphology of graphene/PANI sheets.

4. Galvanostatic charge/discharge measurement of GH and GP20 based on two-electrode system

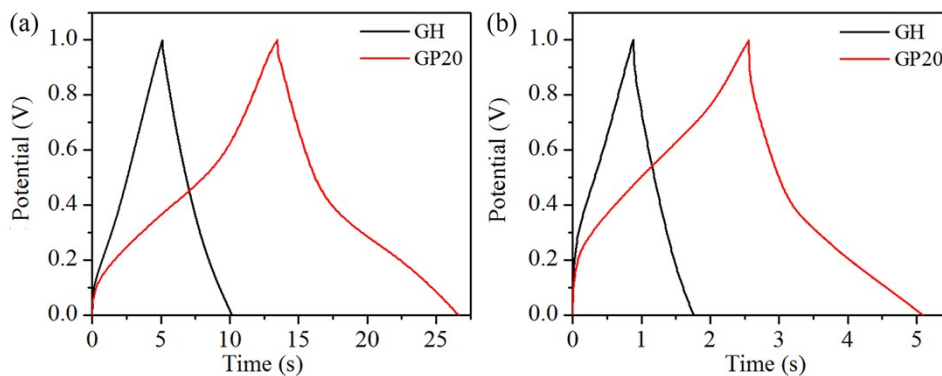


Fig. S4 Galvanostatic charge/discharge curves of GH and GP20 in a symmetrical two-electrode supercapacitor system at a charging/ discharging current density of (a) 20 A g^{-1} and (b) 100 A g^{-1} .

References

- 1 D. D. Xu, Q. Xu, K. X. Wang, J. Chen, Z. M. Chen, *ACS Appl. Mater. Interfaces*, 2014, **6**, 200-209.
- 2 J. Yan, T. Wei, B. Shao, Z. J. Fan, W. Z. Qian, M. L. Zhang, F. Wei, *Carbon*, 2010, **48**, 487-493.
- 3 A. E. Fischer, K. A. Pettigrew, D. R. Rolison, R. M. Stroud, J. W. Long, *Nano Lett.*, 2007, **7**, 281-286.
- 4 H. P. Cong, X. C. Ren, P. Wang, S. H. Yu, *Energy Environ. Sci.*, 2013, **6**, 1185-1191.
- 5 M. Baibarac, I. Baltog, C. Godon, S. Lefrant and O. Chauvet, *Carbon*, 2004, **42**, 3143-3152.
- 6 M. Cochet, G. Louarn, S. Quillard, J. P. Buisson and S. Lefrant, *J. Raman Spectrosc.*, 2000, **31**, 1041-1049.