Supplementary Information

Capacitance enhancement of graphene-based supercapacitors by electrochemically active benzene derivatives

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Fig. S1 N1s spectrum of *p*-rGO. The deconvoluted N1s spectrum contains a single peak centered at 399.5 eV corresponding to the amine group. Neither pyridinic nitrogen (398.2 eV) nor graphitic nitrogen (401.3 eV) is found in the products.



Fig. S2 FTIR spectra of rGO, PPD and *p*-rGO synthesized by hydrothermal method. A distinct new peak at 1567 cm⁻¹ appears in this sample, corresponding to the formation of a new N-H bending vibration that does not exist in *p*-rGO or PPD itself.



Fig. S3 CD curves of p-rGO at charge/discharge current of 1.5, 3.9, 5.9 and 11.9 A/g. The linear and symmetric shape gives a Coulombic efficiency of 97.5% and indicates excellent charge reversibility.

Supplementary Methods Hydrothermal prepared *p*-rGO hydrogel

GO (15 mg) was first dispersed water (15 ml) in a glass vial and ultrasonically exfoliated for 30 minutes. Then PPD (15 mg) was added into the solution and ultrasonicated for another 30 minutes. After that the vial was placed in an oil bath (90 °C, 30 minutes) and a cylinder rGO hydrogel was formed. The as-prepared p-rGO hydrogel 10 liters of deionized water to remove the non-adsorbed PPD molecules.

Calculations

Specific capacitance

For three-electrode configuration, the specific capacitance derived from the galvanostatic discharge curves is calculated with the Equation 1.

$$C = \frac{i\Delta t}{m\Delta V} \qquad (1)$$

where *i* is the discharging current, Δt is the discharging time, ΔV is the voltage range and *m* is the weight of the rGO hydrogel.

Specific capacitance derived from cyclic voltammetry curves based on Equation 2.

$$C_m = \frac{\oint i dV}{2m\Delta V\nu}(2)$$

where *i* is the current, *V* is voltage, *m* is the weight of the rGO electrode, ΔV is the voltage range (0.8V) and ν is the scan rate. Integration is carried out over a whole cycle of CV curve.

For two-electrode configuration, the specific capacitance derived from the galvanostatic discharge curves is calculated with the Equation 3.

$$C = \frac{4i\Delta t}{m\Delta V} \qquad (3)$$

where *i* is the discharging current, Δt is the discharging time, ΔV is the voltage range and *m* is the total weight of both rGO hydrogels.

Specific capacitance derived from cyclic voltammetry curves based on Equation 2.

$$C_m = \frac{2 \oint i dV}{m \Delta V \nu} \, (2)$$

where *i* is the current, *V* is voltage, *m* is the total weight of both rGO electrodes, ΔV is the voltage range (0.8V) and ν is the scan rate. Integration is carried out over a whole cycle of CV curve.

PPD loading estimation

The loading of PPD is estimated according to the XPS survey spectrum of *p*-rGO. The atomic ratio obtained from XPS survey is 82.8:14.3:2.9 for C:O:N. Assuming all the nitrogen element comes from that PPD since the GO sheets does not show any nitrogen peak, the element contribution (C:N:O) from rGO and PPD in *p*-rGO is 74.1:14.3:0, and 8.7:0:2.9, respectively. Based on these data, the weight ratio of PPD to rGO is 145:1118, corresponding to a PPD loading of about 11 wt%.