Supplementary Information

Hybrid Lithium-Ion Capacitors with Asymmetric Graphene Electrodes

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Figure S1 Electrochemical performance of AC in AC//AC device: a) Galvanostatic chargedischarge curves at various current densities, b) Rate performance. Electrochemical performance of SG in SG//SG device under various voltage: 2.7 V, 2.8 V and 3 V: c) Cyclic voltammetry at the scan rate of 200 mV s⁻¹, d) Rate performance.

Figure S1 compared the electrochemical performance of AC and SG in an EDLC device (AC//AC and SG//SG respectively). The specific capacitance of SG was up to 145 F g⁻¹ under a current density of 50 mA g⁻¹. It was slowly reduced to 101 F g⁻¹ when the current density was increased to 1 A g⁻¹. This value is much larger than that of the symmetric capacitor device AC//AC, as shown in Figure S1a and 1b. The specific capacitance of AC was only 60 F g⁻¹, and quickly dropped to 31 F g⁻¹, keeping only 53% of its initial capacitance compared to 70% for SG. When the voltage window increased slightly to 2.8 V and 3 V, the SG//SG kept its EDLC performance as shown in Figure S1c and 1d.



Figure S2 a) Cyclic voltammograms in different scan rates, b) Galvanostatic charge-discharge curves at various current densities, c) cycling performances and d) rate performance measured in a SG//Li device.

As shown in Figure S2a, the voltammetry characteristics curves of the SG//Li device exhibit near rectangular shapes under increasing scan rates. This result, indicative of the excellent non-faradaic behavior of SG, can be confirmed by the nearly straight galvanostatic charge-discharge curves under increasing current densities in Figure S2b. The anions were adsorbed and desorbed quickly within the potential range from 2 V to 4V vs. Li/Li⁺. The cycling performance of the SG//Li device is shown in Figure S2c. The capacitance retention and coulombic efficiency were 91% and 96% after 250 cycles, respectively. As shown in Figure S2d, the SG exhibited excellent rate performance in the SG//Li device.



Figure S3 Electrochemical performance of Graphite anode: a)Cyclic voltammograms at a potential scan rate of 0.1 mV s⁻¹, b) Galvanostatic charge-discharge curves at 50 mA g⁻¹, inset are the photos of graphite electrode before and after lithium- intercalation.

After pre-lithiation, the color of reference graphite anode (Figure S3) change from black to golden, indicating a high pre-lithiation degree through this method. Compared with graphite, which has been commonly used as the anode material in LIB and LIC, the cyclic voltammogram (CV) curve of Li-SG (Figure 3a) exhibits a typical feature of nano-size carbonaceous materials.