Electronic Supplementary Information

3D Flower-structured Graphene from CO$_2$ for Supercapacitors with Ultrahigh Areal Capacitance at Large Current Density

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Experimental Methods:

3D hierarchical porous graphene was synthesized as follows: Lithium (Li) particle (Aldrich) was loaded into a ceramic tube batch reactor and then CO$_2$ was introduced into the reactor with initial pressure of 50 psi at room temperature, followed by heating the reactor to 550°C and then remaining at the temperature for 48 hours. The obtained graphene was separated from other solid products by the treatment of 36.5 wt% hydrochloric acid (HCl), de-ionized (DI) water washing (more than 10 times), and centrifugation separation. The obtained graphene was dried overnight at 80°C to obtain a graphene sample. The structure of the graphene sample was characterized by Hitachi-4700 field emission scanning electron microscope (FESEM) with energy dispersive spectroscopy (EDS), and JEOL JEM2010F transmission electron microscope (TEM) with electron energy loss spectroscopy (EELS). Element analysis was carried out via the combustion approach with an instrument (Model 240XA, Control Equipment Corporation). The pore size distribution and surface area of the sample were measured by N$_2$ adsorption at liquid nitrogen temperature (77 K) with ASAP 2000 instrument.

The symmetric supercapacitor cell was fabricated with the following approach: The 3D CFG powder was mixed with carbon black and poly(tetrafluoroethylene) (weight ratio of 80:10:10) in isopropyl alcohol to form a homogeneous slurry. The slurry was rolled to a strip and pressed onto a nickel foam current collector (about 1 cm × 1 cm), followed by drying at 100°C for 24 h. The two-electrode configuration was used for the measurement of electrochemical properties of CFG. The obtained electrode was used as both anode and cathode to assemble a symmetric cell with a separator (glassy microfiber filter GF/F) and an electrolyte (2M KOH aqueous solution) between them. Cyclic voltammetry (CV) and galvanostatic charge/discharge cycling were
carried out at room temperature (or 55 °C) with the electrochemical workstation (Princeton Potentiostat/Galvvanostat Model 273A).

**Supporting Figure**

Fig. S1. Energy density vs. powder density of 3D graphene-based supercapacitor at room temperature.