Supplementary information

Synthesis of superior conductive chemically converted graphene xerogel †

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Descriptions of chemicals, GO synthesis, electrode fabrication, electrochemical measurements.

1. Chemicals

Expandable graphite (Grade 1721) was supplied by Asbury Carbon. Concentrated sulfuric acid (H2SO4), potassium permanganate (KMnO4), hydrochloric acid (HCl), hydrogen peroxide (H2O2), iodine, hypophosphorous acid (H3PO2, 50%) were purchased from Aldrich. All chemicals were used as received without further purification.

2. Experimental

2.1. Synthesis GO:

A small amount of expandable graphite was charged into a 1-L beaker and heated for 10 s in a microwave oven (Panasonic, NN-5653A). The graphite expanded to about 150 times its original volume. Graphene oxide (GO) was synthesized from expanded graphite according to a
modified Hummer method [1]. Typically, 500 mL of concentrated H₂SO₄ was charged into a 3-L, three-necked flask equipped with a mechanical stirrer (Teflon impeller). The flask was put into an ice bath to chill to 0°C. Five grams of expanded graphite were gradually added under stirring to make a suspension. Then, 30 g of KMnO₄ was slowly added so that the temperature did not exceed 20°C. The temperature was then elevated to 35°C, and the suspension was stirred for 2 h. The flask was then chilled again in the ice bath, and 1 L of deionized water was slowly added to maintain a temperature below 70°C. The mixture was stirred for 1 h and subsequently diluted with 5 L of deionized water. Fifty milliliters of H₂O₂ (30 wt%) was slowly added, and vigorous bubbles appeared as the color of the suspension changed from dark brown to yellow. The suspension was centrifuged and washed with 10% HCl solution four times, followed by centrifuging at 10,000 rpm and washings with deionized water to completely remove the acid until the pH of the GO dispersion reached 6. The as-synthesized GO dispersion was a paste. The concentration of GO was 1.0 wt%, which was determined after drying the GO dispersion at 80°C under vacuum for 24 h.

2.2. Synthesis of CCGX:

While keeping the weight ratio of GO:HPA:I₂ with 1:100:10, GO (4 mg/mL) was dispersed into an aqueous solution of HPA-I, whose final volume was 40 mL. The mixture was sonicated for 5 min and then placed into a water-bath maintained at 90°C. After 12 h, the black gel-like cylinder was formed and it was washed by alcohol and water in a soxhlet extractor for 12 h to get graphene hydrogel. The wet gels were freeze-dried in two days to archive xerogel.

3. Characterization
The CCGX structures were characterized by scanning electron microscopy (SEM, JOEL JSM-6500FE) and high resolution transmission electron microscope (HRTEM, JOEL JEM-2100F). X-ray photoelectron spectra were recorded using a K-alpha (Thermo Fisher). Raman spectra were measured using a WITEC confocal Raman microscopy (WITec, Alpha300S) at 532 nm wavelength incident laser light. Thermal gravimetric analysis was performed under nitrogen atmosphere at heating rate of 10°C per minute (TA Instruments, Q50). X-ray diffraction (XRD) pattern were characterized using a high power X-Ray diffractometer (Rigaku, D/MAZX 2500V/PC) with Cu-Kα radiation (35 kV, 20 mA, λ = 1.5418 Å) at a scan rate of 2° (2θ) per minute. A cylindrical CCGX was sandwiched between two silver paste electrodes and the electrical conductivity of CCGX was calculated via the sheet resistance which was determined by fitting current-voltage (I-V) curves obtained from two probes station using Keithley 4200 semiconductor character system (MS Tech). The mechanical properties of CCGX were measured by texture analyzer (TAplus – LLOYD instruments, AMETEK Company) controlling the strain ramp rate with 2 mm/min.
Figure S1. SEM images of CCGX synthesized at different GO concentrations.

Figure S2. Nitrogen adsorption-desorption isotherms of CCGXs (top) and the variations of pore size and specific surface area (SSA) of CCGXs with GO concentration (bottom).
Figure S3. Stress-strain curves of GGCXs prepared at different GO concentrations.

Figure S4. XPS survey plots of CCGXs.

Reference