Supporting Information

An efficient and facile method towards for producing giant graphene for energy storage application

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Experimental

Synthesisa

A small amount of expandable graphite was sealed in a glass vial. The vial was then heated in a microwave oven for ~15 seconds under ambient atmosphere using a commercial microwave oven to form worm-like graphite (WG).

Giant graphene oxide (GGO) was prepared from WG according to modification of the Hummers. Three grams of WG was added to concentrated sulfuric acid (400 ml) at 0 °C. Then 6 g of $KMnO_4$ was added slowly until dissolved. The reaction was kept at 35 °C for 2 hours. Again, the mixture was added to 400ml de-ionized water and temperature rises to 90 °C for 1h. No H_2O_2 treatment was needed, and the mixture slowly turned to yellow. This solution was very corrosive. It reacts violently with organic material and must be treated with extreme caution. The sediment was decanted and the remaining solution was then centrifugated and washed with a total of 500 ml of 5% HCl solution three times, then washed with DI water 10 times.

The GGO powders made were treated in a microwave oven at ambient conditions for 10 s under high power mode. Upon microwave irradiation, fuming was observed due to a rapid expansion of the GGO powders. A black and fluffy large-sized graphene was yielded and labeled as Microwave-Giant-Graphene (MGG).

Characterisation

Scanning electron microscopy images were obtained by a field-emission scanning electron microscope (FE-SEM JEOL JSM-6700F; JEOL, Tokyo, Japan). The Raman spectra were recorded using a WITEC-CRM200 Raman system (WITEC, Germany). The excitation source was 532-nm laser (2.33 eV). The powder X-ray diffraction (XRD) studies were carried out on a Bruker D8 ADVANCE XRD (Bruker AXS, Germany, k= 1.54056 A °). The AFM measurements were performed with a Digital Instruments Nanoscope. Fourier transform infrared spectroscopy (FTIR)

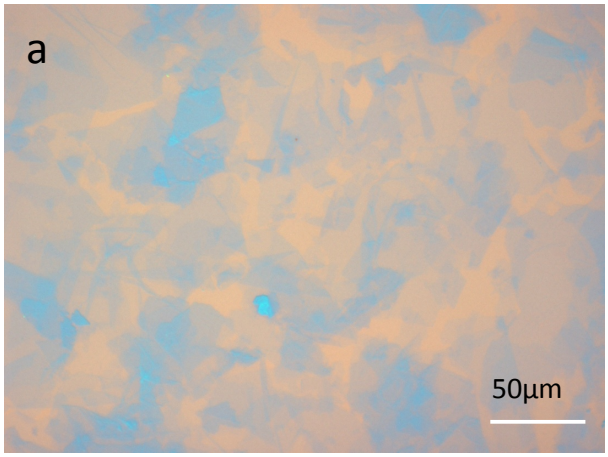
characterization was conducted at ambient temperature with a Perkin Elmer 2000 FTIR spectrometer.

Electrochemical measurements for Li ion battery studies were carried out via CR2032 (3 V) coin-type cell with lithium metal as the counter/reference electrode, Celgard 2400 membrane separator, and 1 M $LiPF_6$ electrolyte solution dissolved in a mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC) (EC/ DMC, 1:1 v/v). The as-prepared MGG was used as the active material for working electrode. The working electrodes were prepared by mixing 80 wt % MGG, 10 wt % acetylene black, and 10 wt % polyvinylidene fluoride binder dissolved in N-methyl-2-pyrrolidinone. After coating the above slurries on Cu foils, the electrodes were dried at 120 °C in vacuum for 2 h to remove the solvent before pressing. Then the electrodes were cut into disks (12 mm in diameter) and dried at 100 °C for 24 h in vacuum. The cells were assembled in an argon-filled glove-box.

For supercapacitor measurements the as-prepared MGG films were used both as the anode and the cathode, with a filter paper separator and a 6M KOH aqueous solution electrolyte. No binder and conducting additives were used in the supercapacitor. Cyclic voltammetry (CV) measurements, gravimetric charge-discharge profiles and electrochemical impedance spectroscopy (EIS, 100 KHz-0.01 Hz) were conducted on a Solartron 1287electrochemical workstation.

The gravimetric capacitance of the MGG was calculated from CV curves according to formula $C = 2 \int IdV/(\gamma mE)$, where I is the current, V the cell voltage, γ the voltage scan rate, E potential window of scanning and m the mass of graphene in the electrode. Capacitance determined by the galvanostatic charge/discharge was measured using $C = I/(dV/dt)$ with dV/dt calculated from the slope of discharge curves.

Supporting Figures



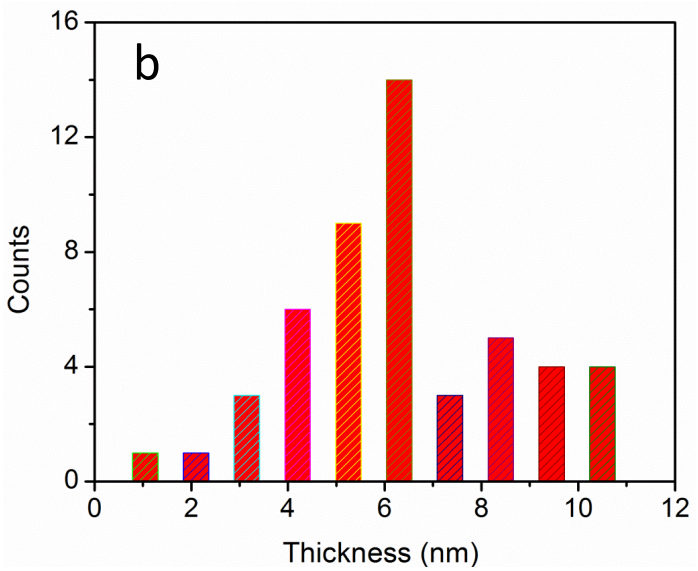


Fig. S1 (a) The optical image of the GGO sheets (b) Statistical AFM measurement results of graphene thickness for randomly selected 40 sheets, where the thickness ranges from 1 to 12 nm

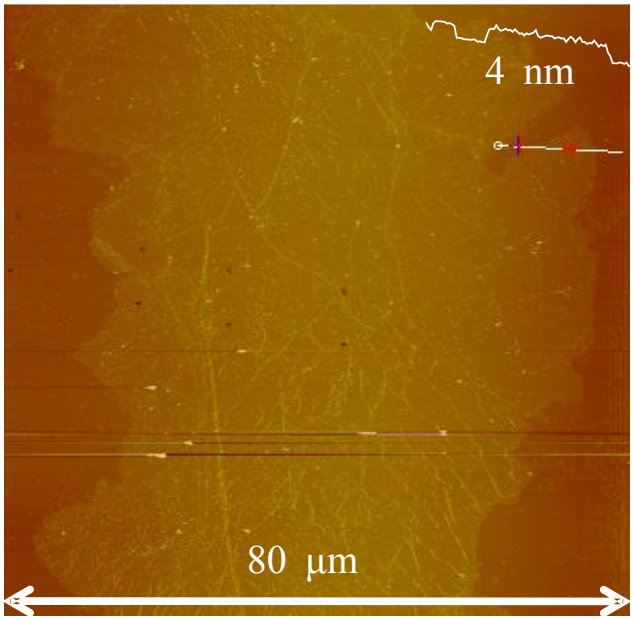


Fig. S2 the AFM image of the MGG sheets

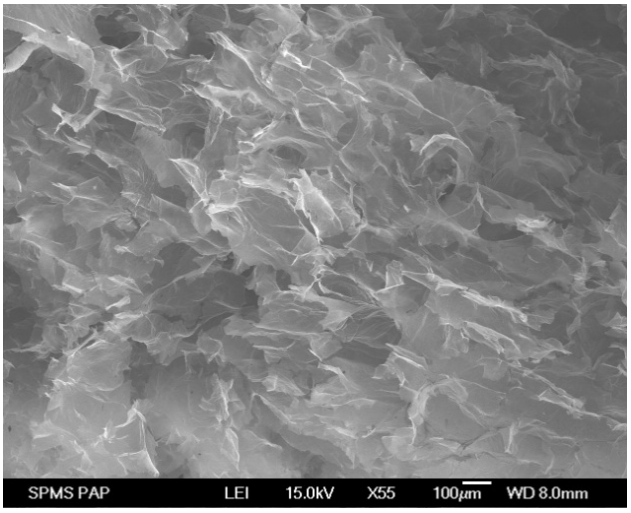


Fig. S3 the SEM image of the MGG sheets

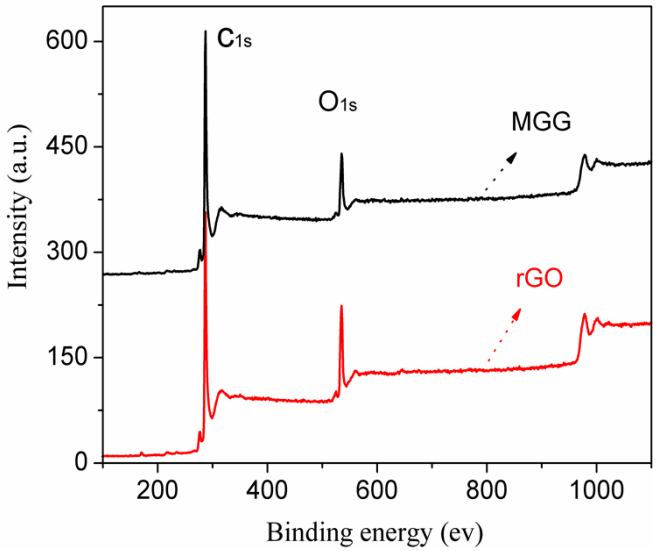


Fig. S4 XPS spectra of the MGG and conventional rGO

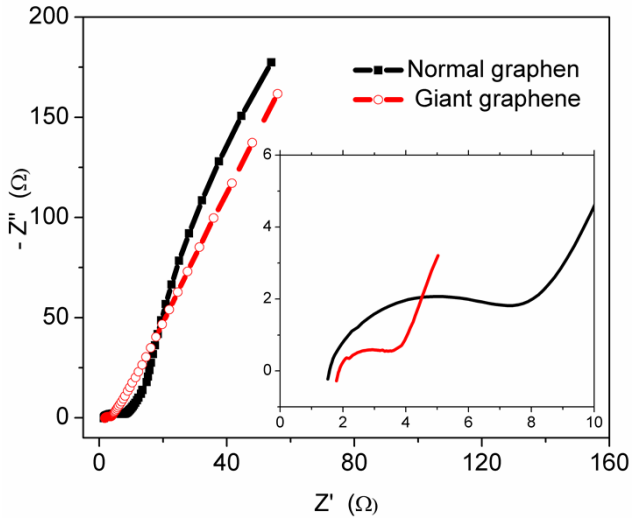


Fig. S5 Nyquist impedance plots of MGG and conventional rGO. The data in the inset show the details at high-frequency ranges.