

Supporting Information

Simultaneously Electrochemical Synthesis of Few-layer Graphene Flakes on both Electrodes in Protic Ionic Liquids

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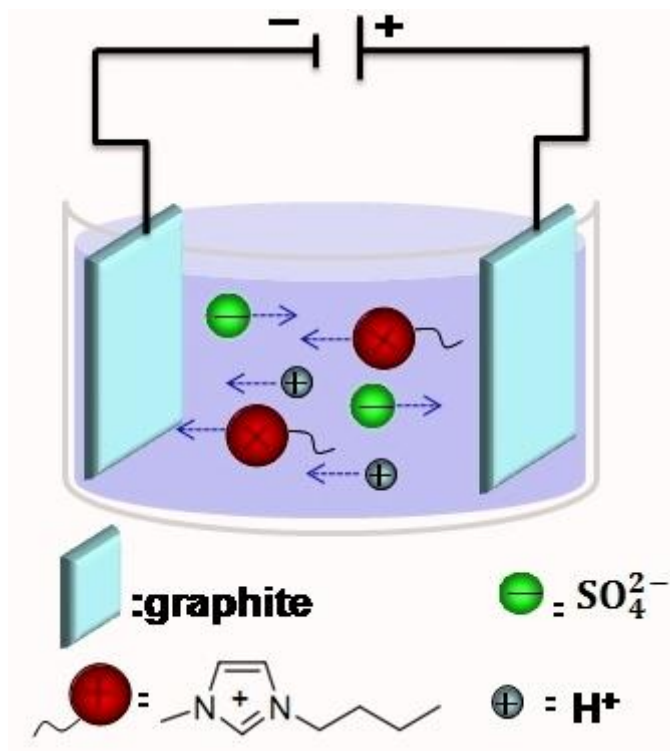
Experimental processes

Graphite powders of 15 mg were packed into graphite plates (7 mm × 5 mm) in stainless steel meshwork through direct compression process. The self-made graphite plates were used as both negative and positive electrodes, and a protic ionic liquid 1-butyl-3-methyl imidazolium hydrosulfate (BMIMHSO₄) was used as electrolyte. A voltage of 3.0 V is carried out between the graphite/graphite cell for 7 hrs (Scheme S1). The distance between the cathode and anode was 20 mm. The anodic and cathodic expanded graphite powders were separately transferred into two agate mortars and ground with additional 0.1 mL protic ionic liquid (BMIMHSO₄) for 2.5 hrs. The mixtures were centrifugated at 13000 rpm for 10 min in a 1:1 (vol/vol) solution of DMF and acetone to remove the ionic liquid for 3 times. The suspensions dispersed in 50 mL DMF and centrifugated at 2000 rpm for 5 min, the supernatants were separated and stored for use. All the experiments were operated under 20-22 °C in ambient atmosphere with 50% relative humidity.

Characterizations

Atomic Force Microscopy (AFM, NanoMan VS) was used to investigate the thickness of the products. The sample suspension was dropped on clean mica for observation. High-resolution Transmission Electron Microscope (TEM, JEOL, JEM-2100F) was used to investigate the layers of the samples. The sample suspension was dropped on lacy carbon-coated Cu grid for TEM observation. Fourier-transformed Infrared (FT-IR) spectra were taken on a Nicolet 6700 FT-IR Spectrometer by using pressed KBr pellets. Raman spectra were recorded on a LabRAM HR800 Raman spectrometer using a 532 nm excitation laser. The sample suspension was dropped on clean mica for observation. RST-9 4-Point Probes Resistivity Measurement System (Guangzhou four-probe Technology Co., Ltd., China) was used to measure the sheet resistivity of samples. The sample films with the thickness of $\sim 20 \mu\text{m}$ were coated on an insulated glass pane ($15 \times 15 \text{ mm}^2$) by dropping and drying. The electrochemical measurements were performed on a CHI model 660D electrochemical workstation (Shanghai Chenhua Instruments, China). The graphene modified glass carbon electrode as the working electrode, a platinum wire electrode as counter electrode and an Hg/HgO electrode as reference electrode, and $6 \text{ mol}\cdot\text{L}^{-1}$ KOH solution as the electrolyte. The potential range for cyclic voltammetry was from 0.3 to -0.7 V .

Additional Data



Scheme S1. Diagrammatic drawing of “graphite/graphite cell” using BMIMHSO₄

PIL electrolyte

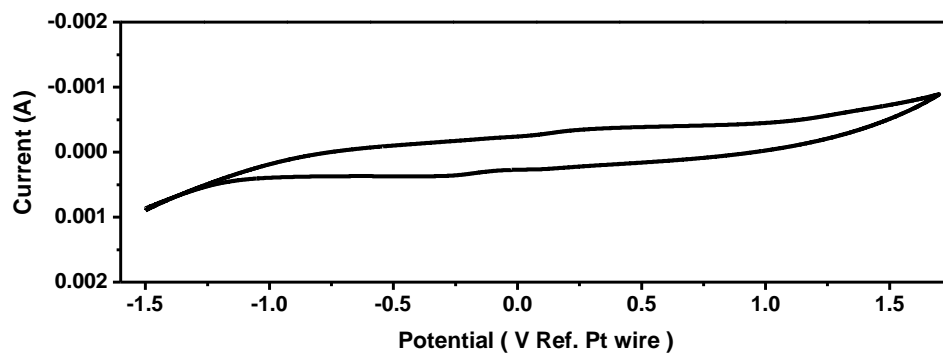


Figure S1. Electrochemical windows of BMIMHSO₄ electrolyte
(Working electrode: graphite, Counter electrode: platinum foil, Reference electrode:
platinum wire)



Figure S2. Both CFLGs and AFLGs dispersed in DMF (~ 0.5 mg/mL) present dark color. The dispersions have been stored more than 5 months without any sediment.

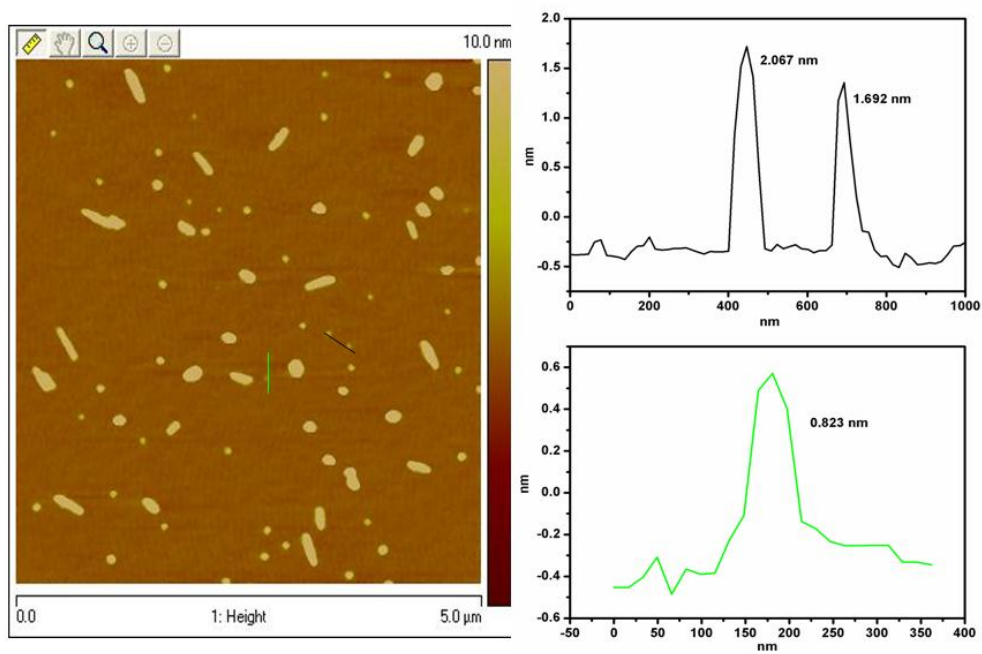


Figure S3. AFM shows different thickness of AFLG flakes.

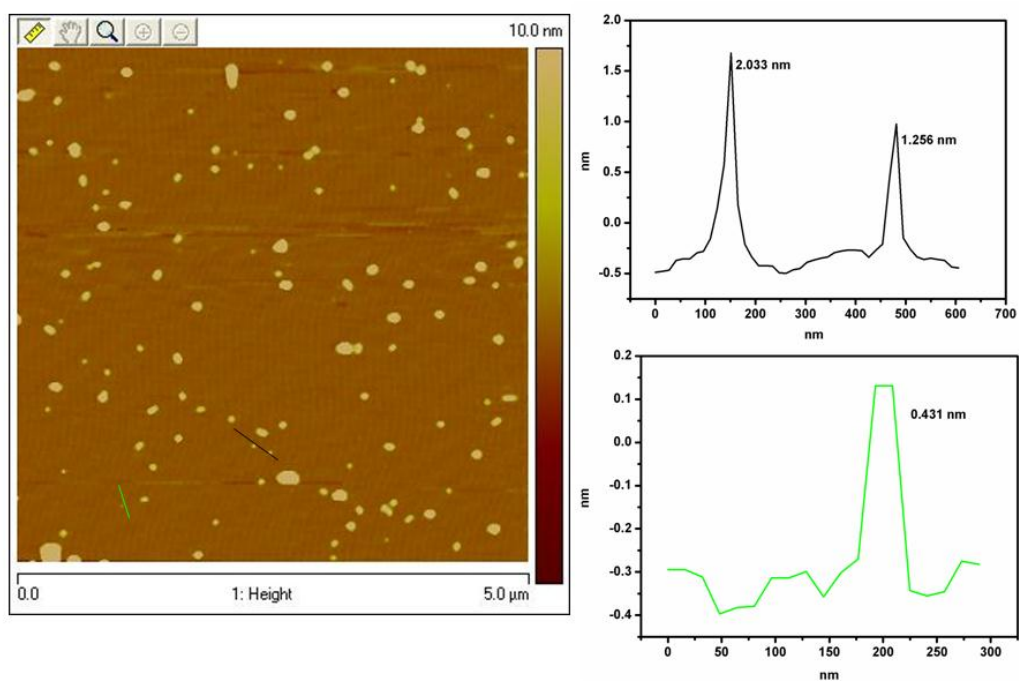


Figure S4. AFM shows different thickness of CFLG flakes.

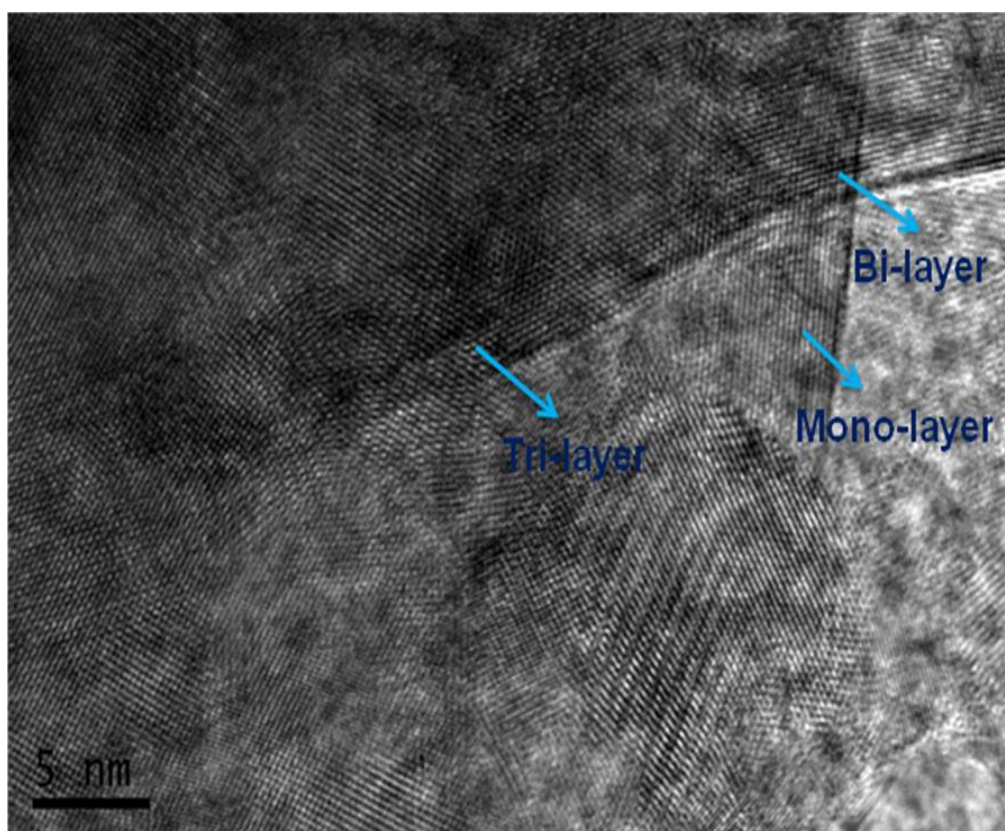


Figure S5. HRTEM of CFLG flakes shows different layer numbers, and crystal lattices of graphene are clearly resolvable.

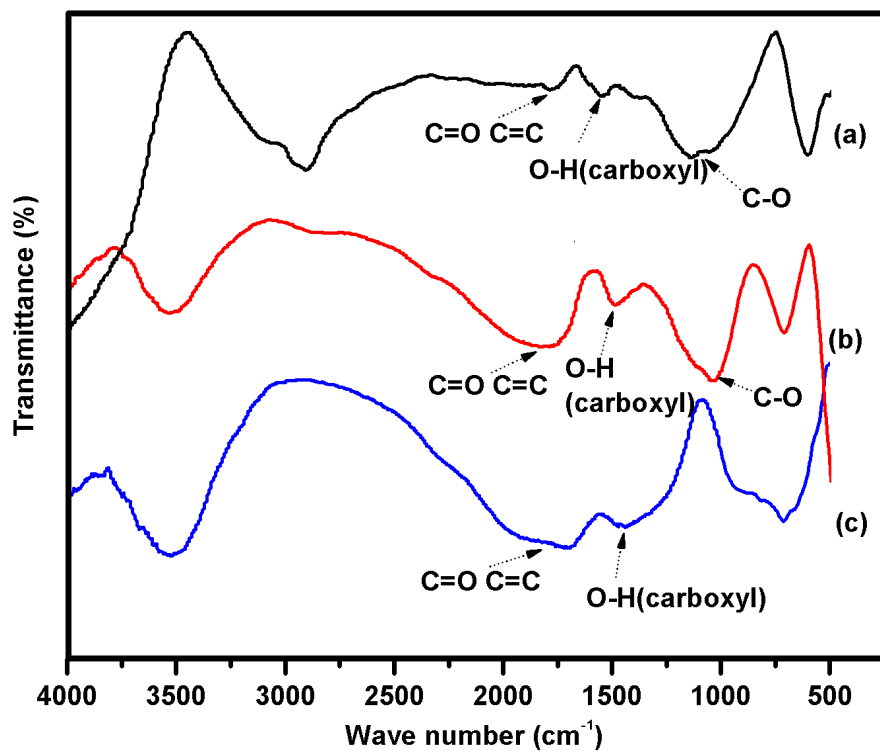


Figure S6. FT-IR spectra of pristine graphite (a), AFLGs (b), and CFLGs (c). The functional groups were identified as follows: C=O stretching vibrations from carbonyl and carboxylic groups and C=C skeletal vibrations from unoxidized graphitic domains partly overlapped at 2100-1700 cm^{-1} , O-H originated from carboxylic acid at $\sim 1400 \text{ cm}^{-1}$ and C-O from epoxy and alkoxy at $\sim 1100 \text{ cm}^{-1}$.

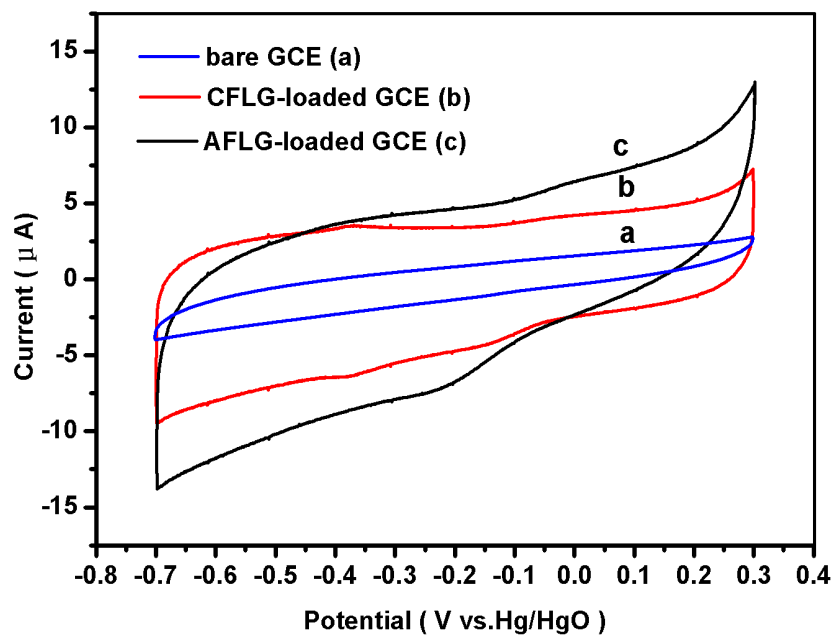


Fig S7. Cyclic voltammograms of bare GCE, CFLG-loaded GCE, and AFLG-loaded GCE in 6 mol·L⁻¹ KOH aqueous solutions at a scan rate of 100 mV·s⁻¹.