Electronic Supplementary Information

A Green Approach to the Synthesis of High-Quality Graphene Oxide Flakes via Electrochemical Exfoliation of Pencil

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Experiment Section

Materials characterizations. Transmission electron microscopy (TEM, JEM-2010, 200 kV), Atomic Force Microscopy (AFM, Nanoscope V, diDimension) and Scanning electron microscopy were used to investigate the morphology of exfoliate GO flakes. Fourier transform infrared spectroscopy (FTIR) characterization was conducted at ambient temperature with a Perkin Elmer 2000 FTIR spectrometer. X-ray diffraction (XRD, Bruker D-8 Avance) and Raman system (Renishaw, 532 nm excitation laser) were used to character samples.

Electrochemical Measurements. Cyclic voltammerty (CV) measurements were performed in a standard three-electrode cell using an Autolab PGSTAT302 electrochemical test system (Eco Chemie, Netherland). A platinum foil and Ag/AgCl electrode were used as the counter and the reference electrode, respectively. The working electrode was prepared with a similar method to that reported previously[S2]. Generally,10 mg of exfoliated GO was ultrasonically dispersed into1 ml of 2-propanl containing Nafion solution (5 wt%, dupont) to form a catalyst ink.10 μl of the catalyst ink was coated on the glassy carbon disk (5 mm in diameter) and dried at 80 for 15 min. Pt-loaded carbon catalyst (Pt/C 20% on Vulcan XC-72R, E-TEK division, PeMEAS Fuel Cell Technology) with the same amount was also studied for comparison by using the same electrode configuration. Oxygen reduction reaction (ORR) measurement was conducted in a 0.1 M, 0.5 M and 1 M KOH aqueous electrolyte saturated with oxygen at a scan rate of 10 mV/s on the rotating-disk electrode system (Eco Chemie, Netherland). The bare glassy carbon electrode was tested without catalyst as comparison. Methanol toxicity test was conducted at a constant potential -0.5 V (vs Ag/AgCl). All measurements were carried out at room temperature.

In our experiment, we used saturated Ag/AgCl electrode as reference electrode. Koutecky-Levich plots (J^{-1} vs $\omega^{-1/2}$) were analyzed at various electrode potentials. The slops of their linear fit regression were used to calculate the number of electron transferred (*n*) according to the Koutecky-Levich equation:

$$\frac{1}{J} = \frac{1}{J_L} + \frac{1}{J_K} = \frac{1}{B\omega^{1/2}}$$

$$B = 0.62nFC_0(D_0)^{2/3}v^{-1/6}$$

$$J_L = B\omega^{1/2}$$

$$J_K = nFkC_0$$

where J is the measured current density, J_K and J_L are the kinetic and diffusion limiting current densities, □ is the angular velocity, *n* is transferred electron number, *F* is the Faraday constant, C_0 is the bulk concentration of O_2, v is the kinematic viscosity of the electrolyte, and k is the electron-transfer rate constant. For the Tefol plots, the kinetic current was calculated from the mass-transport correction of RDE by:

$$J_K = \frac{J \times J_L}{(J_L - J)}$$

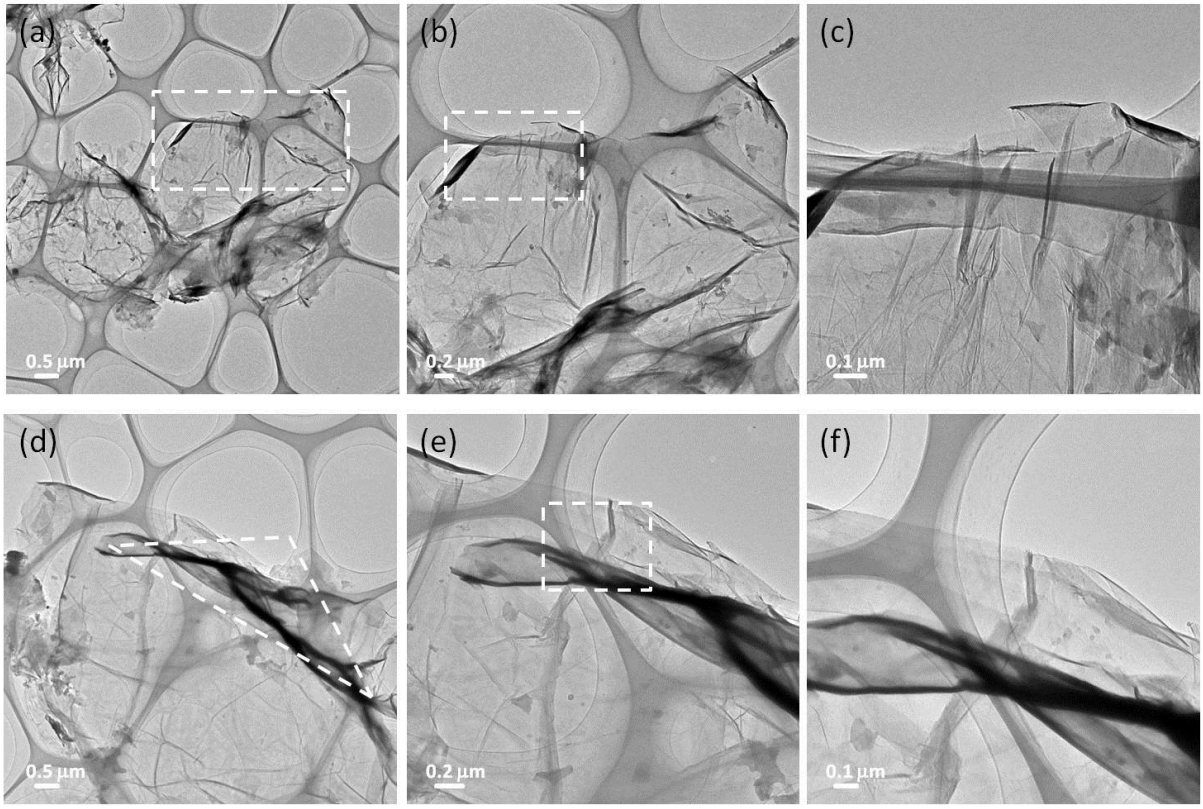

Figure S1. Representative TEM images of exfoliated GO flakes.

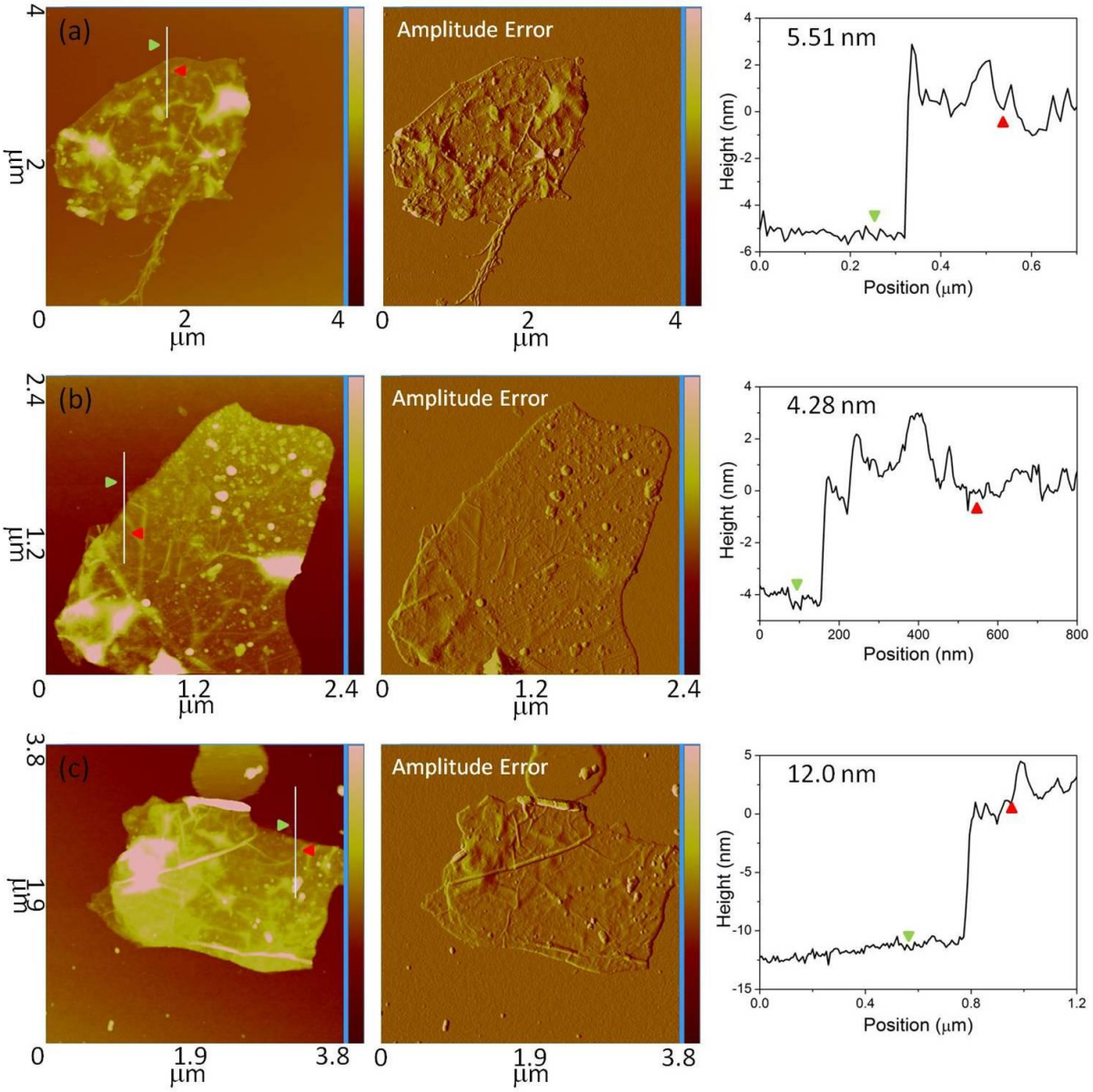

Figure S2. Representative AFM images of exfoliated GO flakes.

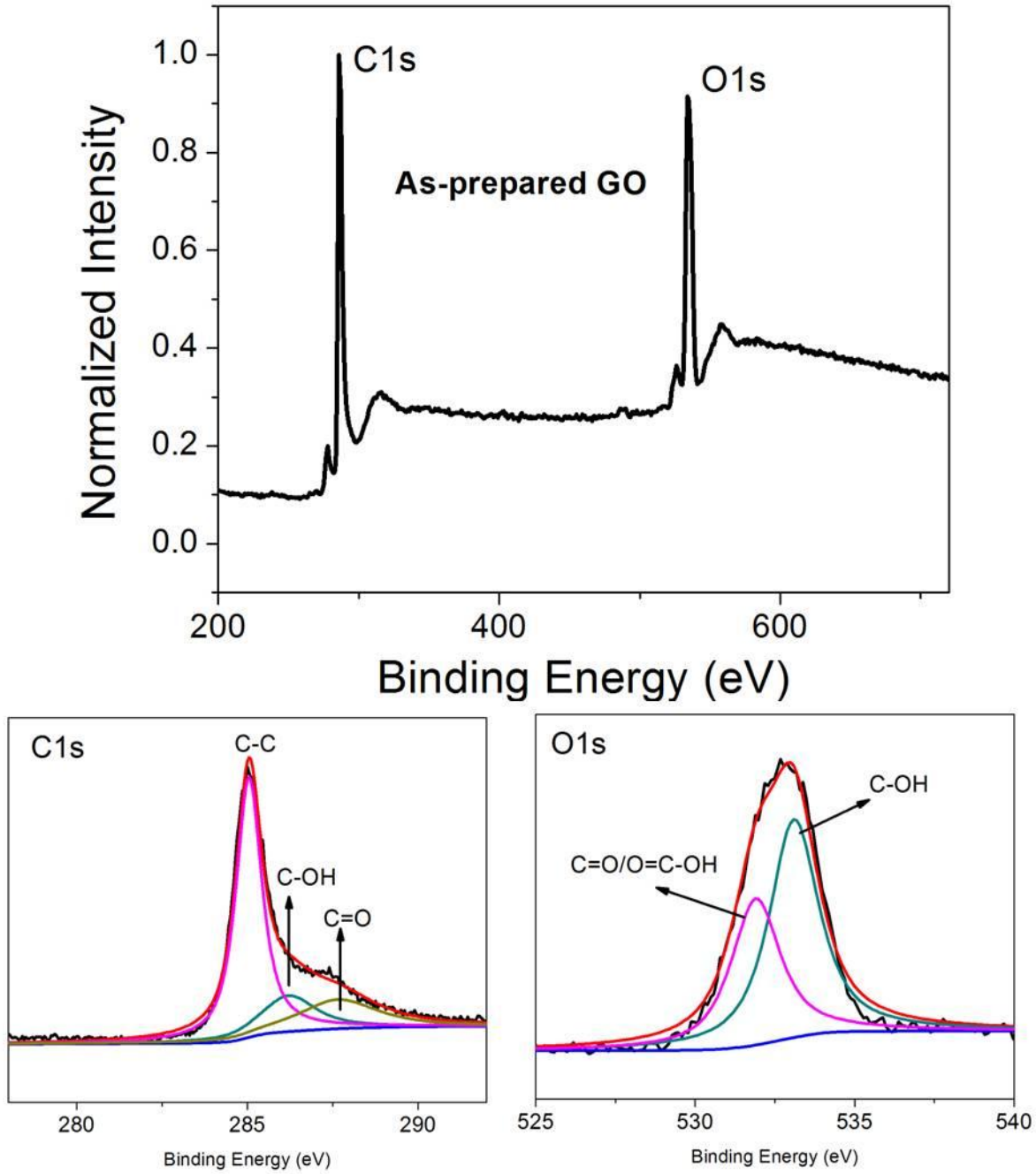

Figure S3. XPS wide scan, C1s and O1s spectra of as-prepared Graphite Oxide flakes.

References

[S1] F. B. Su, Z. Q. Tian, C. K. Poh, Z. G. Wang, S. H. Lim, Z. L. Liu, J. Y. Lin, *Chemistry of Materials* **2010**, *22*, 832-839.