Supporting Information

Influence of the coffee-ring effect and size of flakes of graphene oxide films on its electrochemical reduction

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Materials and Methods

Graphite (3061) was obtained from Asbury Carbon. A Nikon Eclipse LV150-NL with differential interference contrast was used to visualize materials. Statistical Raman spectroscopy (SRS) was recorded using a Horiba Explora spectrometer with a 532 nm laser for excitation combined with a 100x magnification objective. For the preparation of Langmuir-Blodgett films, we used a Kibron µThroug system with water as subphase. The surface tension of water = 72.8 mN m^{-1} was set to zero. The films were formed at a surface tension value of 3 mN m^{-1}. AFM images were recorded on a JPK Nanowizard 4 equipped with NSG10/Au probes, and intermittent contact mode was used with a Tap300-G AFM Probe. We used a USC300T (Supply: 230 V 50Hz 150 VA. Output: HF 45kHz 80W) for sonication. Electrochemical experiments were performed with a Keithley 2460 SourceMeter in phosphate buffered (PB) solution as a supporting electrolyte. The pH is 7.4. The material modified glassy carbon electrode with a diameter of 5mm is used as the working electrode, Ag/AgCl is used as the reference electrode, and platinum wire is used as the counter electrode. The images in this test are taken from video screenshots, using a camera to capture the electrochemical reduction process and control the capture's synchronization with the electrochemical reaction..

Synthesis of Oxo-Functionalized Graphene (oxoG)

OxoG was prepared by low-temperature oxidation of graphite according to our previously developed method. [1-3] Accordingly, 2 g of graphite (type 3061, Asbury Carbon Mills) were mixed with 50 mL of sulfuric acid (97.5%) in a Teflon reactor under mechanical stirring at a temperature below 10 °C. After that, 4 g of $KMnO_4$ were slowly added within 4 h and further stirred for 16 h. Then, 40 mL of cold dilute sulfuric acid (20 wt%) and followed by 100 mL of cold double distilled water were slowly continuously added through a programmed pump within 4 h and 16 h, respectively. Then, 40 mL of H_2O_2 (5 wt%) were added into the reaction mixture to solubilize manganese species. Then, the dispersion was washed with cold double distilled water by repeated centrifugation and redispersion in double distilled water six times. The suspension is adjusted to 1 mg/mL.

Formation of films of flakes of oxoG on Si/300 nm SiO_2 wafer.

Flakes of oxoG were deposited on a Si/300 nm SiO_2 wafer by Langmuir Blodgett technique. First, a light yellowish oxoG dispersion was prepared by dilution of water with methanol in equal volume. The dispersion was dropped on the water surface of the Langmuir-Blodgett trough and the barriers were compressed until a surface tension of 3 mN m^{-1} was reached.

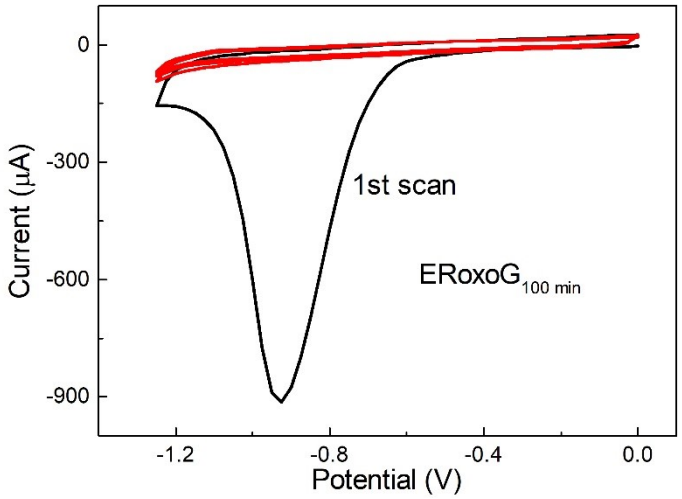

Figure S1. Profiles of cyclic voltammetry were obtained by electrochemical reduction of $ERoxoG_{100\ min}$. The black line is the first scan of electrochemical reduction. The other 4 scans are represented by the red line.

Referents

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3. P. Feicht, J. Biskupek, T. E. Gorelik, J. Renner, C. E. Halbig, M. Maranska, F. Puchtler, U. Kaiser and S. Eigler, *Chem. Eur. J.*, 2019, **25**, 8955-8959.