

Reducing Graphene Oxide with Modified Birch Reaction (Supporting Information)

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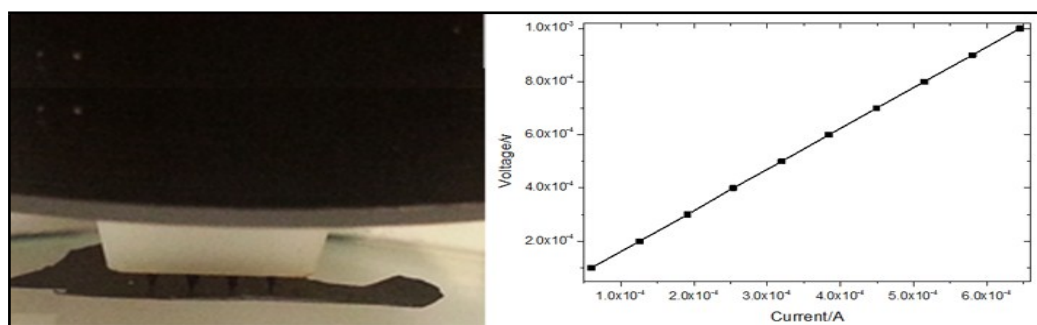


Fig. S1 Visual appearance of RGO film and electrical conductivity of RGO film measured with four-probe equipment

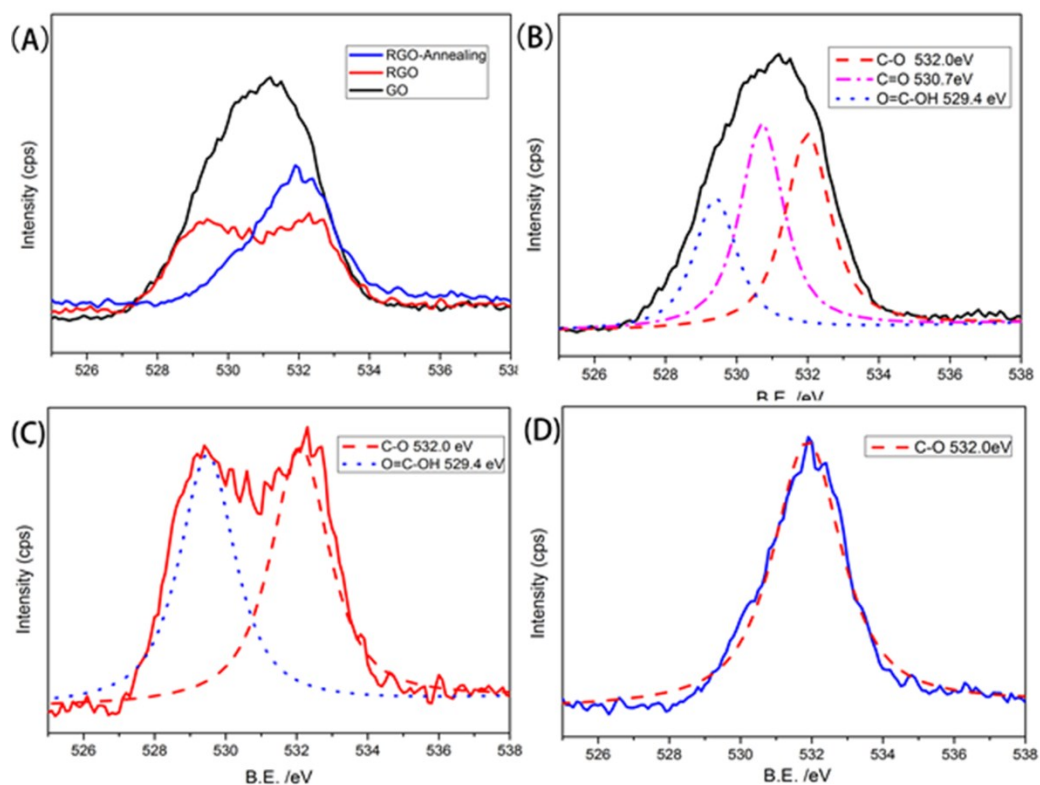


Fig. S2 XPS survey scans of O_{1s} peak of GO, RGO and (A)RGO with annealing. decompositions of O_{1s} peaks of GO (B), RGO (C) and annealed RGO (D)

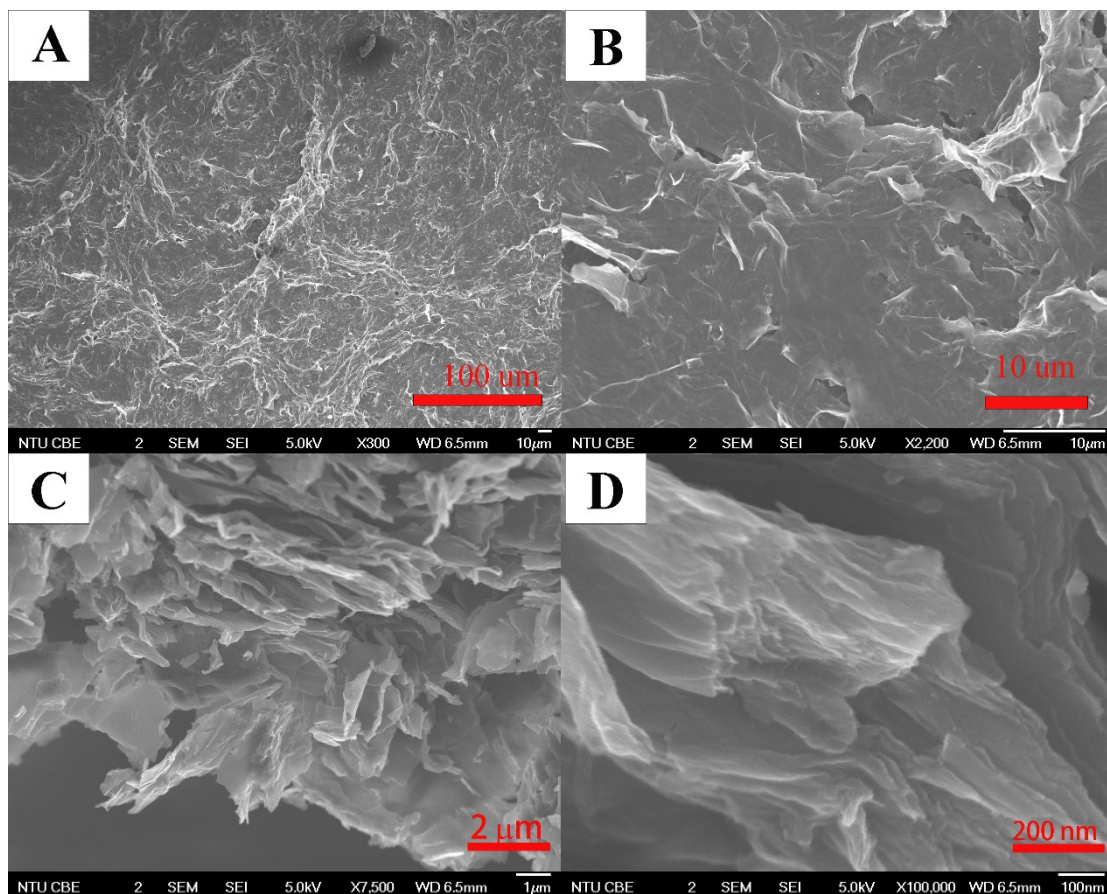


Fig.S3 SEM images of RGO film

Experiment section

Materials and methods

Natural graphite flakes (300-mesh in size) were obtained from Qingdao Haida Graphite Co. Ltd (Shandong, China). Potassium permanganate, Hydrogen peroxide (30 wt%) and Sulfuric acid (98%), Tris(2-aminoethyl) amine and sodium were brought from Sigma-Aldrich (Milwaukee, WI) and used without further treatment. The water in the experiment was ultrapure deionized water (18.2 M Ω ·cm) (D.I. water) from a Milli-Q System (Millipore, Billerica, MA). The Teflon reactor vessel employed in the experiment was a 50 mL stainless steel autoclave. All other solvents were used as received unless otherwise stated.

Preparation of graphene oxide (GO)

Graphene oxide (GO) was prepared by a pressurized oxide process according to a previous report¹⁹. Briefly, all the starting materials and Teflon reactor were cooled to 0-4°C and kept totally dry. Then graphite (0.6 g) and potassium permanganate (3.0 g) were poured into the Teflon reactor. Cooled sulfuric acid (30 mL) was added to the mixture and the reactor was sealed. The Teflon reactor was kept at 0°C for 1.5h and then the temperature was increased to 100°C for 2h. The resulting materials was put in to 1L deionized H₂O. Under vigorous stirring, H₂O₂ was added into the mixture. When the colour changed to golden yellow, the addition was stopped. Then hot HCl

and DI water were employed to wash the residual until there was no more dissociation H^+ using repeated centrifugation method. The GO suspension was sonicated in a sonication bath for 24h to exfoliate the graphite oxide to graphene oxide, and then dried in a freeze drier for 48h, resulting in a sponge-like dry GO powder.

Reduction of graphene oxide (RGO) using Sodium in amine

40 mg of GO was suspended in 30 mL of tris(2-amineethyl) amine under argon atmosphere with magnetic stirring. This suspension was stirred at room temperature until the GO powder uniformly dispersed in the amine, and then 80 mg of sodium was added to the mixture. Within 1h, the colour of the suspension turned completely black from light yellow indicating reduction of the GO. To ensure complete reduction, the mixture was reacted for 24h. After the 24h, the temperature of the reaction bottle was increased to 90°C and the mixture was kept at this temperature for a further 24h. The temperature of the obtained suspension was decreased to 0°C in an ice-water bath. The remaining sodium was eliminated with slowly dripped ethanol. The RGO product was isolated by membrane filtration, washed with deionized water and DMF and dried.

Characterization of graphene

MFP 3D microscope (Asylum Research, Santa Barbara, CA) in ac mode was used to record AFM. Scanning electron microscopy (SEM) was conducted on JEOL JSM-6700F and LEO 1550 Gemini field-emission scanning electron microanalyzer at an accelerating voltage of 5 kV. X-ray diffraction (XRD) patterns were measured with a Bruker AXS D8 Advance, equipped with a Cu K α photon source (40 kV, 20 mA, $\lambda=0.15406$ nm) scanned at the rate of $1.0^\circ \text{ min}^{-1}$ over the range of 5° - 60° (2θ). Raman spectra were recorded with a WITec CRM200 confocal Raman microscopy system with 633 nm excitation and an air cooled charge coupled device (CCD) as the detector (WITec Instruments Corp, Germany). X-ray photoemission spectroscopy (XPS) measurements were carried out using a Kratos AXIS Ultra spectrometer with a monochromatized Al K α X-ray source (1486.71 eV) operated at 15 kV and 10 mA. Samples for XPS were prepared by loading the films on silicon wafer. The binding energy (B.E.) scale of the XPS spectrum was calibrated with the C1s peak (peak at 284.5 eV). Thermogravimetric analyses (TGA) were recorded on a Mettler Toledo TGA/DSC at a heating rate of $1^\circ \text{ C min}^{-1}$ in nitrogen atmosphere for GO and $10^\circ \text{ C min}^{-1}$ for RGO. The resistance of the RGO film was measured with a four-probe KEITHLEY 2636A system. To detect the functional groups on the materials during the process of reduction, Fourier transform infrared spectroscopy (FTIR, Perkin-Elmer Perkin-Spectrum one FT-IR spectrometer) in the frequency range 450 - 4000cm^{-1} was employed.