

## SUPPLEMENTAL INFORMATION

### High Yield preparation of macroscopic graphene oxide membranes

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#### EXPERIMENTAL

**Chemicals.** Acid intercalated expandable graphite (Grafguard 160-50N, mean particle size: 350 $\mu$ m, the graphite was intercalated with organic acid and sulfuric acid and nitric acid.<sup>1)</sup>) was kindly provided by Graftech Inc. Concentrated sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), Potassium peroxodisulfate (K<sub>2</sub>S<sub>2</sub>O<sub>8</sub>), Phosphorus(V) oxide (P<sub>2</sub>O<sub>5</sub>), Potassium permanganate (KMnO<sub>4</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), anhydrous hydrazine (N<sub>2</sub>H<sub>4</sub>) was purchased from Aldrich. All chemical are used as received unless otherwise stated.

**Microwave assisted expansion of expandable graphite.** A small amount of expandable graphite was sealed in a glass vial and purged with high purity nitrogen for 2 hours. The vial was then heated in a microwave oven (Panasonic, model NN-S547WA, 60Hz) for less than 2 s. Graphite that had not expanded fully was carefully picked out.

**Oxidation and exfoliation of expanded graphite.** Graphene oxide (GO) was prepared according to Hummers method.<sup>2,3</sup> K<sub>2</sub>S<sub>2</sub>O<sub>8</sub> (5 g) and P<sub>2</sub>O<sub>5</sub> (5 g) were mixed with concentrated sulfuric acid (30 ml) at 90 °C. Then 150 mg of microwave-expanded graphite was carefully added to the solution while stirring. The reaction was keep at 80 °C for 4.5 hours. On completion of the reaction, the mixture was added to excess

de-ionized water, followed by filtration through a 0.2 micron Nylon Millipore filter and washing with excess water until the pH of the filtrate was close to neutral. The fluffy graphite oxide was dried in air at 60°C for 3 hours. The dried sample was then added to concentrated sulfuric acid (100 ml) at 0 °C.  $\text{KMnO}_4$  (60 g) was then added slowly until dissolved. The reaction was kept at 35 °C for 2 hours. Again, the mixture was added to 1L de-ionized water and stirred for 2h. Then 50 ml of 30%  $\text{H}_2\text{O}_2$  was added very slowly, and the mixture allowed to settle for 24 hours. *Warning: this solution is very corrosive and must be treated with extreme caution; it reacts violently with organic material.* The sediment was decanted and the remaining solution was then centrifuged and washed with a total of 1 L of 10% HCl solution three times, then centrifuged again and washed with 1 L of DI water 10 times. The final graphene oxide solution is then concentrated to ~230 ml, yielding a very viscous, brownish transparent solution with concentration 0.30% (w/w). Trace amounts of ions are then removed by dialysis against deionized water for 60 days, using tubing with a 12,000MW cutoff (Fisher Scientific).

**X-ray diffraction.** A small amount of graphene oxide membrane solution was dried at room temperature under vacuum for 12 hours until no further weight loss was observed. The x-ray diffraction (XRD) of this material was performed on a Rigaku GiegerFlex D/Max-B X-ray diffractometer at 40 kV and 20 mA for monochromatized  $\text{Cu K}\alpha$  ( $\lambda = 0.15416$  nm) radiation. For graphene oxide film, an INEL CPS 120 powder diffractometer at 40kV, 20mA with Cu target (1.5416 angstroms) was used. This is equipped with a position-sensitive detector covering 120 degree range of  $2\theta$ . The

graphene oxide sample was rotated continuously during data accumulation to average out the effects of preferred orientation in the diffracting plane. Basal distances of (002) planes were calculated according to the Bragg equation.

**Deposition of graphene membrane on SiO<sub>2</sub>/Si substrate.** Alignment markers were fabricated on the oxidized silicon substrate (SiO<sub>2</sub> thickness of 300 nm) using photolithography and thin film evaporation of 3 nm Chromium and Gold. 20 µl of sample solution was diluted with 2 ml of DI water. 100 µl of the diluted solution was spin-coated onto the substrate at 3000 r.p.m. for 1 minute. The GO/substrate and an open vial of 200 µl hydrazine was then sealed in large glass container and kept at 50°C for 24 hours to reduce the graphene oxide.

**Atomic Force Microscope (AFM).** Tapping mode AFM was used to acquire the images under ambient conditions (Digital Instruments Multimode AFM).

**High resolution Scanning Electron Microscope (HRSEM).** SEM images were acquired with a JSM-7500F (JOEL), using a 2 kV accelerating voltage in gentle beam mode (bias voltage applied to the specimen stage).

**Fabrication of GO transistor using electron beam lithography.** Source and drain electrodes were patterned by electron beam lithography using polymethylmethacrylate PMMA as the e-beam resist. Single layer graphene oxide membranes on an oxidized silicon substrate (300 nm SiO<sub>2</sub> thickness with pre-fabricated alignment markers; see above) were first identified and located by optical microscopy. Then, 100 µl of 5% PMMA (relative molecular mass, 495K) chlorobenzene solution was applied to the

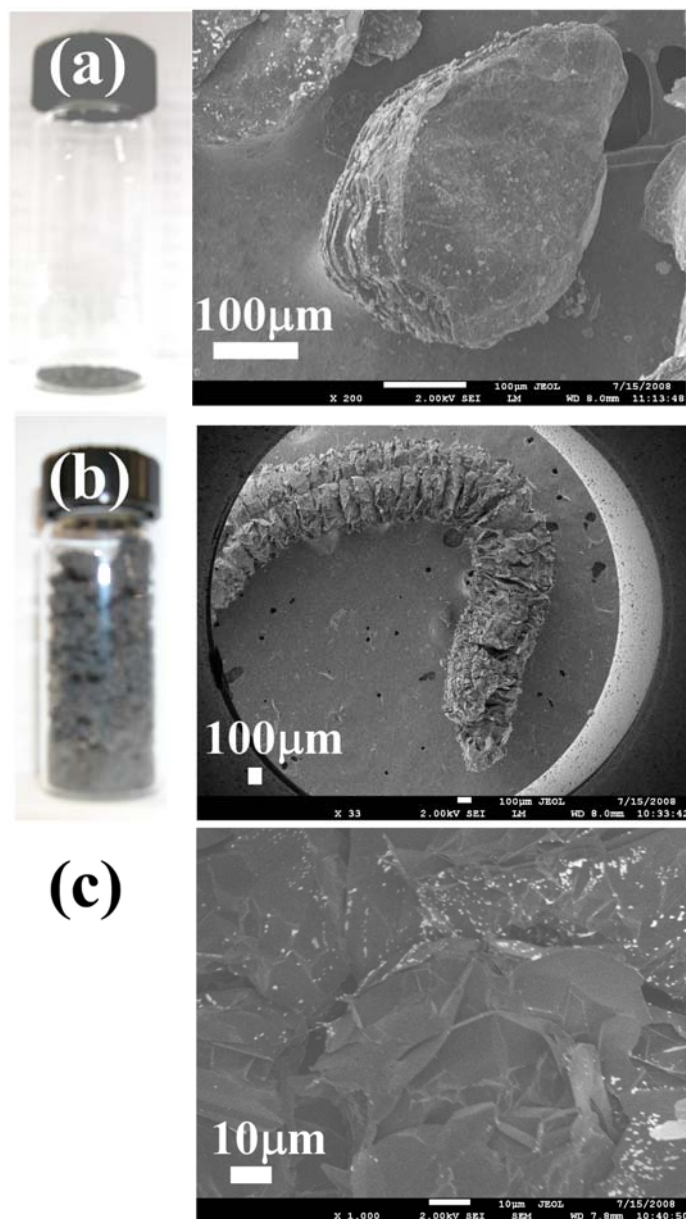
substrate at 3000 rpm, followed by baking at 180°C for 2 minutes. Based on spin curves provided by the manufacturer (Microchem Corp., [www.microchem.com](http://www.microchem.com)), the PMMA thickness is about 300 nm. Electron-beam patterning was done using a JEOL SEM 6400 operated at 30 kV with a Raith Elphy Plus controller, with an exposure dosage 500 $\mu$ A/cm<sup>2</sup>. The exposed PMMA was developed with a methyl isobutyl ketone (MIBK) and isopropyl alcohol (1:3) solution. Chromium (3nm from R.D. Mathis company) and Gold (50 nm) were then deposited onto the substrate in a thermal evaporator with a cryopump producing a base pressure of 10<sup>-7</sup> Torr. The deposited films were then lifted off in acetone bath for 12 h at 70°C and rinsed extensively with isopropyl alcohol.

**Electronic transport measurement.** The bulk of the heavily doped p++ (conducting) silicon substrate was used as the gate electrode, with contact made to the chip with silver paint. Source, drain and gate electrodes were contacted using three individual probes in a homemade, small-signal probe station, which is controlled using Labview. A DAC card (National Instruments) installed in a PC (Dell) was used to output source-drain voltage, and a Keithley 6517A current meter was used to read the source-drain current and to output the gate voltage.

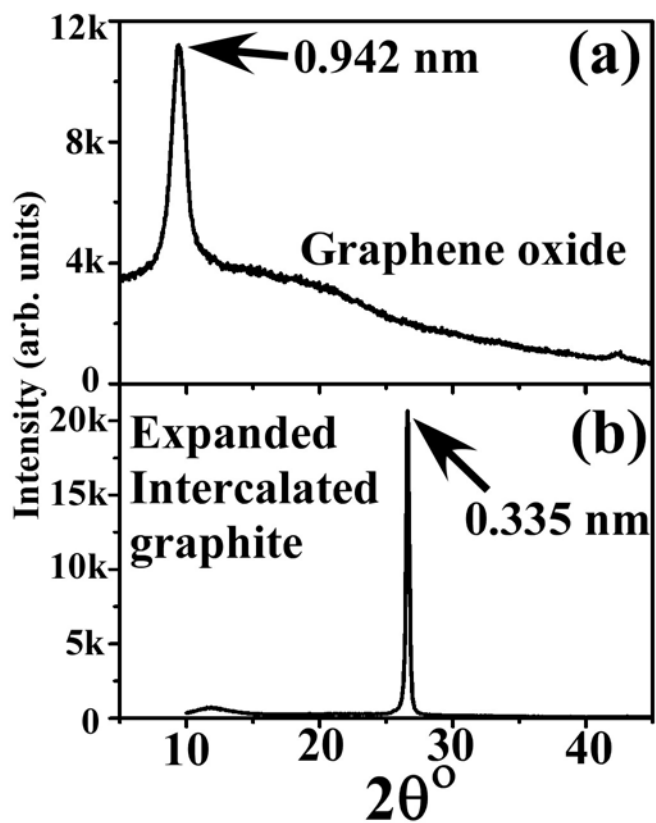
## **CITED REFERENCES**

- <sup>1</sup> R. A. Greinke, ((Graftech Inc., USA). Application: US, 2002), p. 8 pp.
- <sup>2</sup> W. S. Hummers, Jr. and R. E. Offeman, J. Am. Chem. Soc. **80**, 1339 (1958).
- <sup>3</sup> S. Gilje, S. Han, M. Wang, et al., Nano Lett. **7**, 3394 (2007).

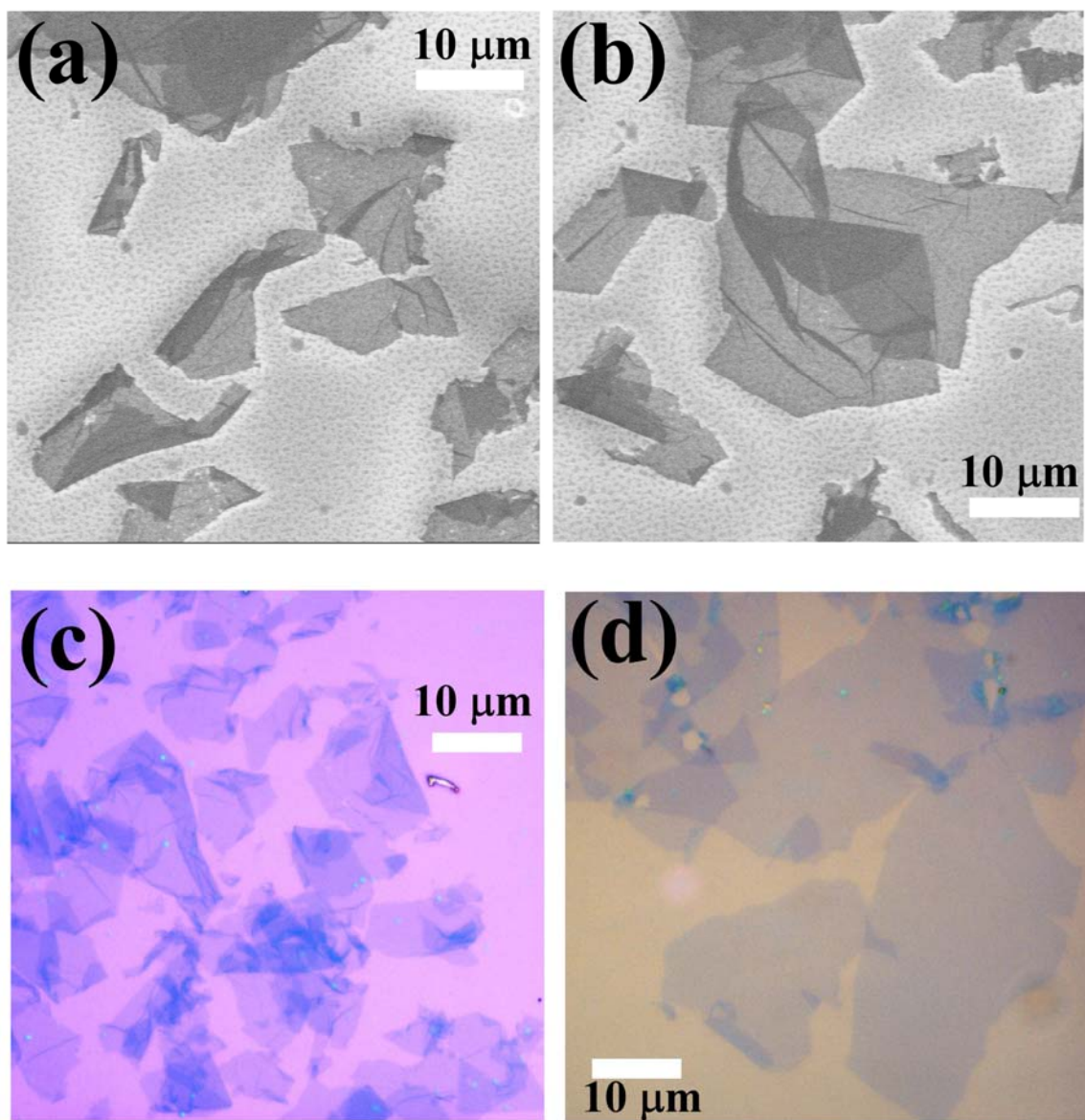
**Figure S1.** (a & b) Pictures and SEM image of graphite samples before and after microwave expansion. (c) Zoomed region in (b).



**Figure S2.** (a & b) X-ray diffraction pattern of graphene oxide membranes and their microwave-assisted expanded graphite.



**Figure S3.** SEM(a&b) and optical (c&d) image of graphene oxide membranes on  $\text{SiO}_2/\text{Si}$  substrate.



**Figure S4.** AFM image of typical graphene oxide Field effect transistor.

