Electronic Supplementary Information

Facile preparation of n-type reduced graphene oxide field effect transistor at room temperature

Luyang Wang, Younghun Park, Peng Cui, Saemi Lee and Hyoyoung Lee*

National Creative Research Initiative, Center for Smart Molecular Memory, Department of Chemistry and Energy Science and SAINT, Sungkyunkwan University, 2066 Seoburo, Jangan-Gu, Suwon, Gyeonggi-Do 440-746, South Korea. Fax: (+) 82-31-299-5934; Tel: (+) 82-31-299-4566;

*Corresponding author: hyoyoung@skku.edu

1. Experimental methods

1.1 Materials

Natural graphite (Bay Carbon, SP-1 graphite), sulfuric acid (95-97%), hydrogen peroxide (30 wt. %), potassium permanganate, sodium nitrate, hydroiodic acid (57 wt. %), hydrazine monohydrate, sodium bicarbonate, lithium, ethylenediamine (EDA) and acetic acid were obtained from commercial sources and used as received.

1.2 Characterization

Raman spectroscopy measurements were taken using a micro-Raman system (Renishaw, RM1000-In Via) with an excitation energy of 2.33 eV (532 nm). All X-ray photoemission spectroscopy (XPS) measurements were made by a Sigma probe (ThermoVG, U.K.) with a monochromatic Al-K X-ray source at 100 W. All I-V curves were measured by a probe-station (Keithley, U.S) at room temperature after the devices were dried in a vacuum oven at 60 °C for more than 12 h.

1.3 Preparation of graphene oxide (GO)

GO was prepared from natural graphite powder by the modified Hummer's and Offenman's method using sulfuric acid, potassium permanganate, and sodium nitrate.[1]

1.4 Fabrication of GO- field effect transistor (FET) device and substrate

A GO solution of a concentration of 0.5 mg/ml in DI water was sonicated in a Branson 1510 ultrasonic bath cleaner for 1 h and then shaken at 200 rpm for 1 h. After standing for 1h, the resulting solution was dropped onto the device (substrate) surface with a spin-coating

method.[2]

1.5 Reduction of GO FET to rGO FET

In a typical procedure, GO FET device was placed on a device-stander in a closed jar that contains 2.0 ml of HI and 5.0 ml of acetic acid. The cover glass was sealed with vacuum grease and placed in an oil bath at 40 °C for 24 h (In hydrazine condition, it was 400 ul hydrazine and 90°C for 4 h.). This product was rinsed with a saturated sodium bicarbonate solution, water and acetone, and then dried in vacuum oven prior to the next steps in the synthesis.[3]
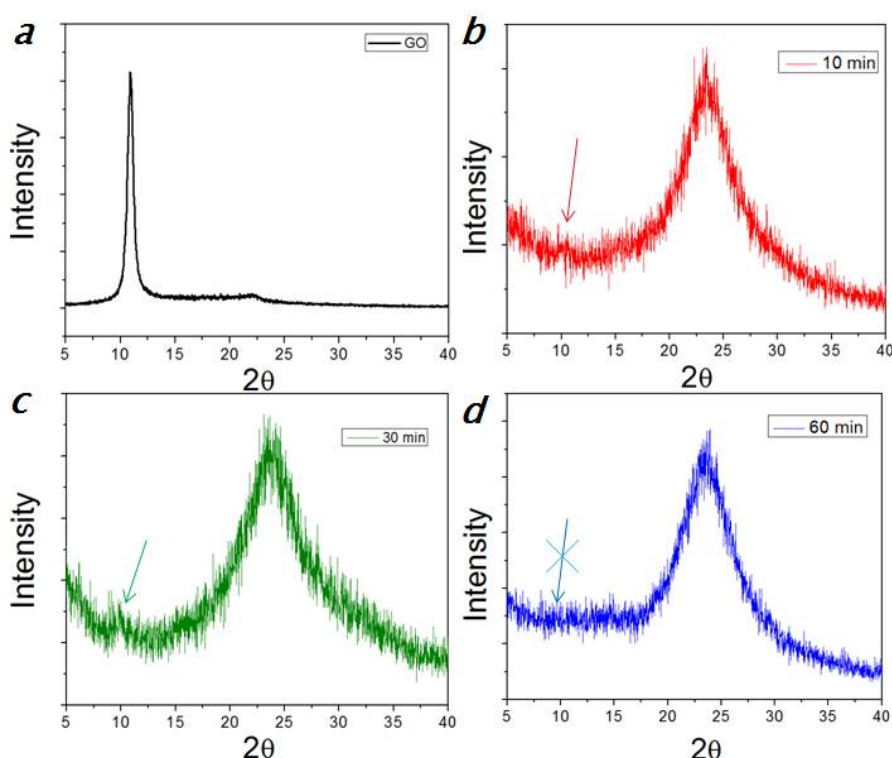
2. Characterization data

Figure S1. Time dependence of the reaction in X-ray diffraction (XRD). a) GO; b) reaction time: 10 min; c) reaction time: 30 min; d) reaction time: 60 min.

From Fig. S1a, we could clearly infer that the GO was well oxidized during the oxidation. The peak of GO shifted to 10.89° indicating that the spacing between the grap

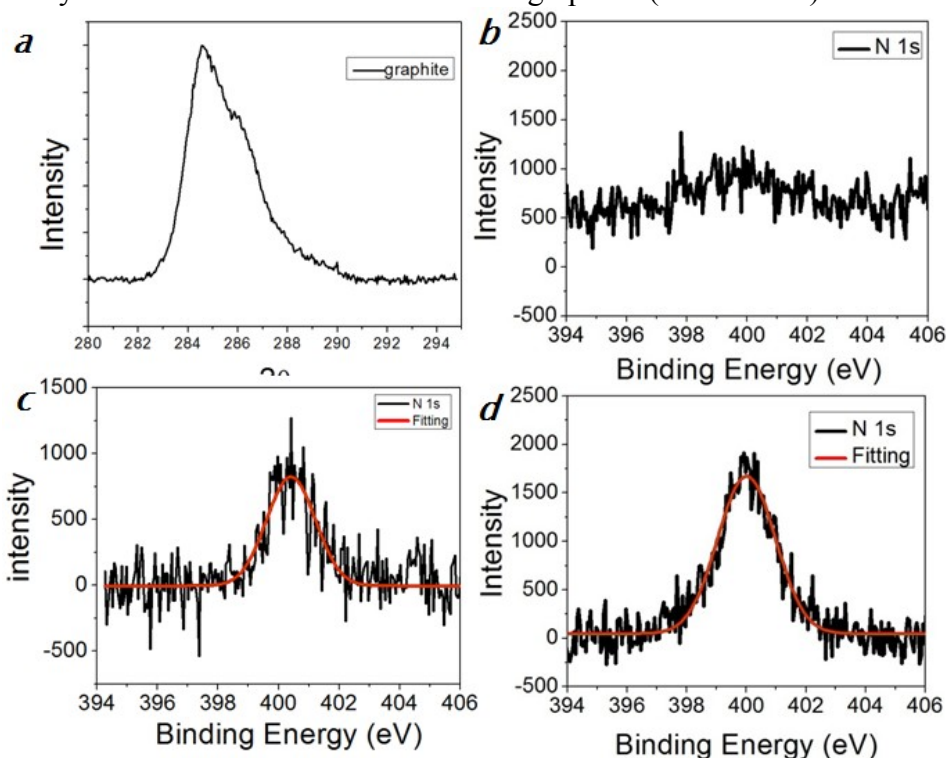hene oxide layers was increased from the raw graphite (2θ= 26.34°).

Figure S2. a) C 1s X-ray photoemission spectroscopy (XPS) of graphite. b) N 1s XPS of Li-rGO (Li concentration: 1.67 mg/ml) c) N 1s XPS of Li-rGO (Li concentration: 3.33 mg/ml) d) N 1s XPS of Li-rGO (Li concentration: 6.67mg/ml)

Fig. S2a was taken from the raw graphite which was bought from the Bay carbon company. Compare the GO, Li-rGO and raw graphite XPS data, we conclude that the Li-rGO was reduced from the GO by remove the C-O bond which was located at around 286 eV in XPS. Fig. S2a was taken from the Li-rGO which was reduce from the GO by a concentration of Li at 1.67 mg/ml. Fig. S2c was taken from the Li-rGO which was reduced at the Li concentration of 3.33 mg/ml. Fig. S2d was taken from the Li concentration of 6.67 mg/ml.

Table S1. Li amount and the N atoms content from XPS

Different concentration of Li (mg/ml)	1.67	3.33	6.67
N atom (percentage)	trace	1.1	2.1

We could observe that when the concentration of the Li solution was quite lower than 1.67 mg/ml, we could not obtain the N doping from the reduction. Surprisingly after increase the concentration of Li, we could dope the N atoms into the graphene plane during the reaction which is a very strong evidence to support our explanation of the reaction mechanism. The doping was related to the amount (concentration) of the alkali metal, Li.
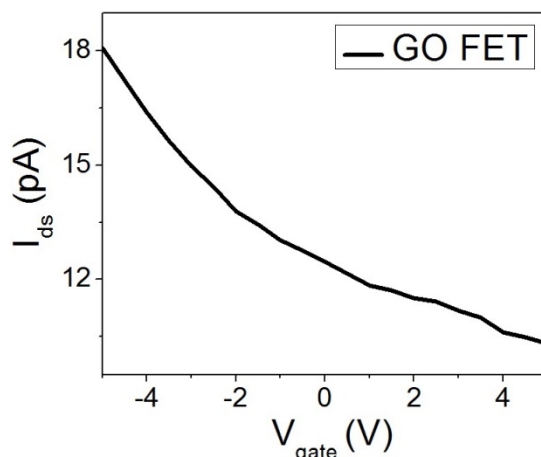
Figure S3. I-V characterization of the EDA treated GO FET device.
We could observe that the GO FET shows almost insulating which indicates the reduction could effectively improve the mobility and conductivity of graphene channels.
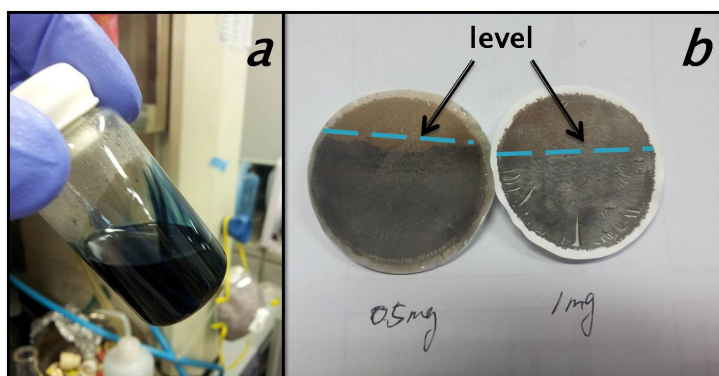

Figure S4. a) Blue color Li-EDA solution showed the stability at room temperature. (before the reduction) b) The color of GO paper was changed by the reduced of the Li-EDA solution. (The GO paper was 0.5 mg and 1 mg individually.) The significant color change shows the level of the Li-EDA solution.
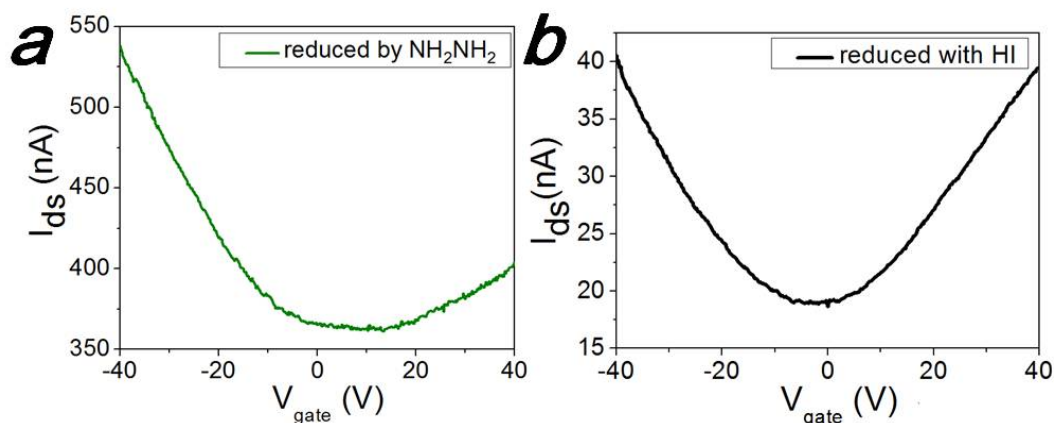

Figure S5. a) rGO FET reduced by hydrazine b) rGO FET reduced by HI method
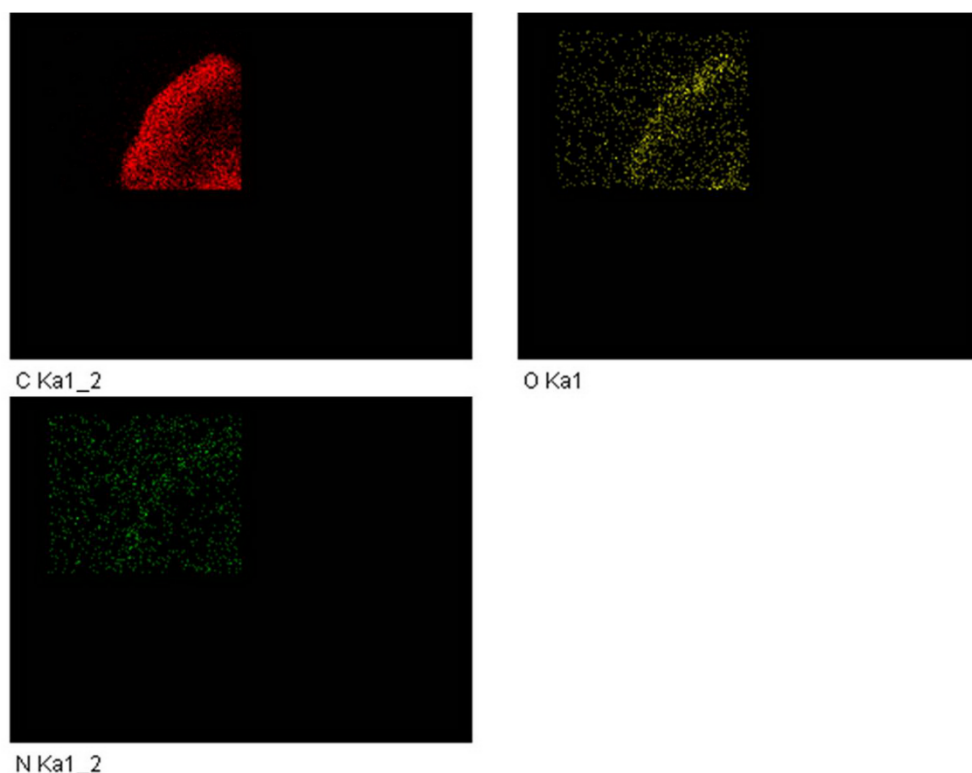**References**

C Ka1_2

O Ka1

N Ka1_2

Figure S6. Element mapping images of the Li-rGO. a) C atoms b) O atoms c) N atoms. From the element mapping we could obviously catch the distribution of the carbon, oxygen and nitrogen atoms. There is a significant boundary line between the Li-rGO and the gold substrate.
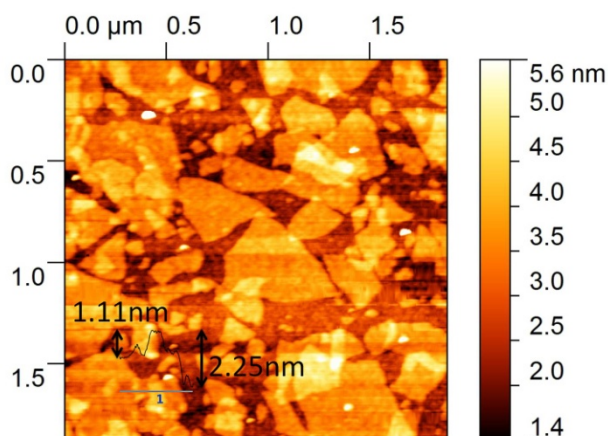

Figure S7. AFM image of GO. We could observe the GO pieces on the SiO_2 surface. From the image, we got a height of single layer piece GO was around 1.1 nm.

1.   W. S. Hummers and R. E. Offeman, *J Am Chem Soc*, 1958, **80**, 1339-1339.
2.   P. Cui, S. Seo, J. Lee, L. Wang, E. Lee, M. Min and H. Lee, *Acs Nano*, 2011, **5**, 6826-6833.
3.   I. K. Moon, J. Lee, R. S. Ruoff and H. Lee, *Nat Commun*, 2010, **1**, 73.