

The Extended Growth of Graphene Oxide Flakes Using Ethanol CVD

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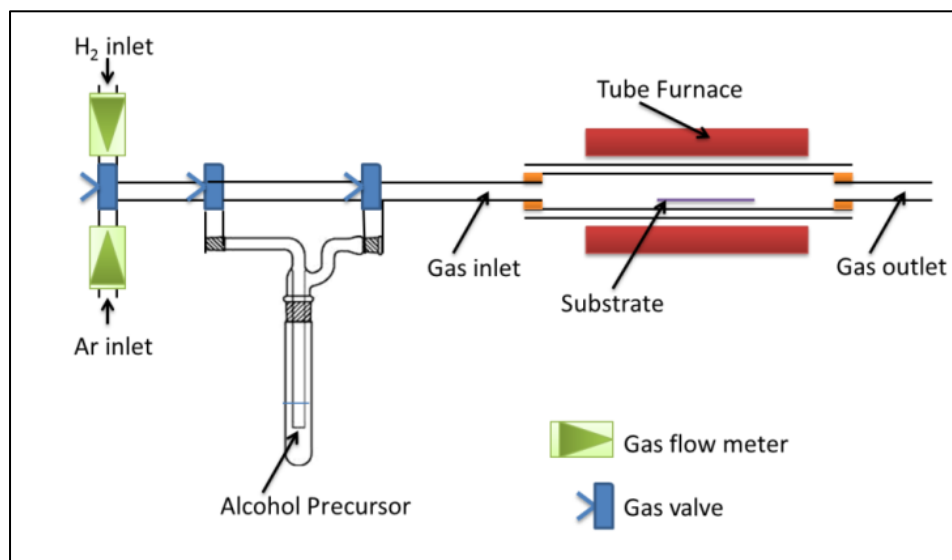


Figure S1. An atmospheric pressure CVD setup with alcohol vapors for growth of Reduced Graphene Oxide (RGO).

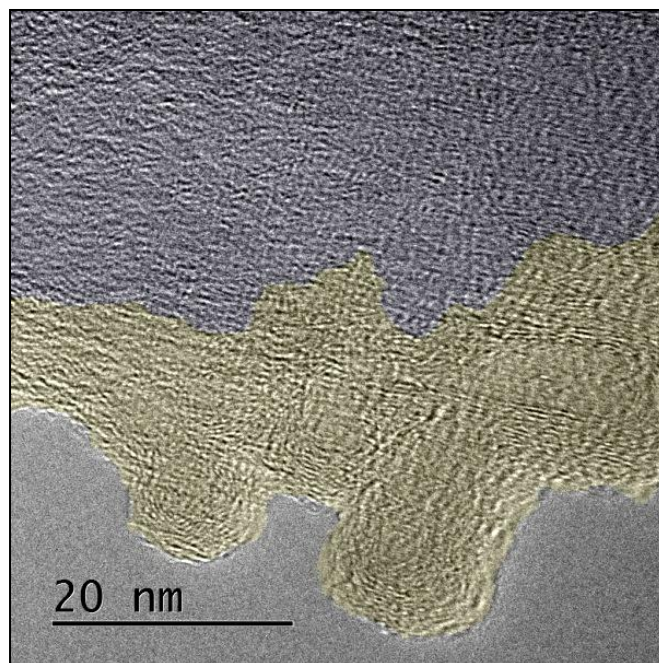


Figure S2. TEM image of pre-existing RGO (darker) and new grown RGO (highlighted in yellow).

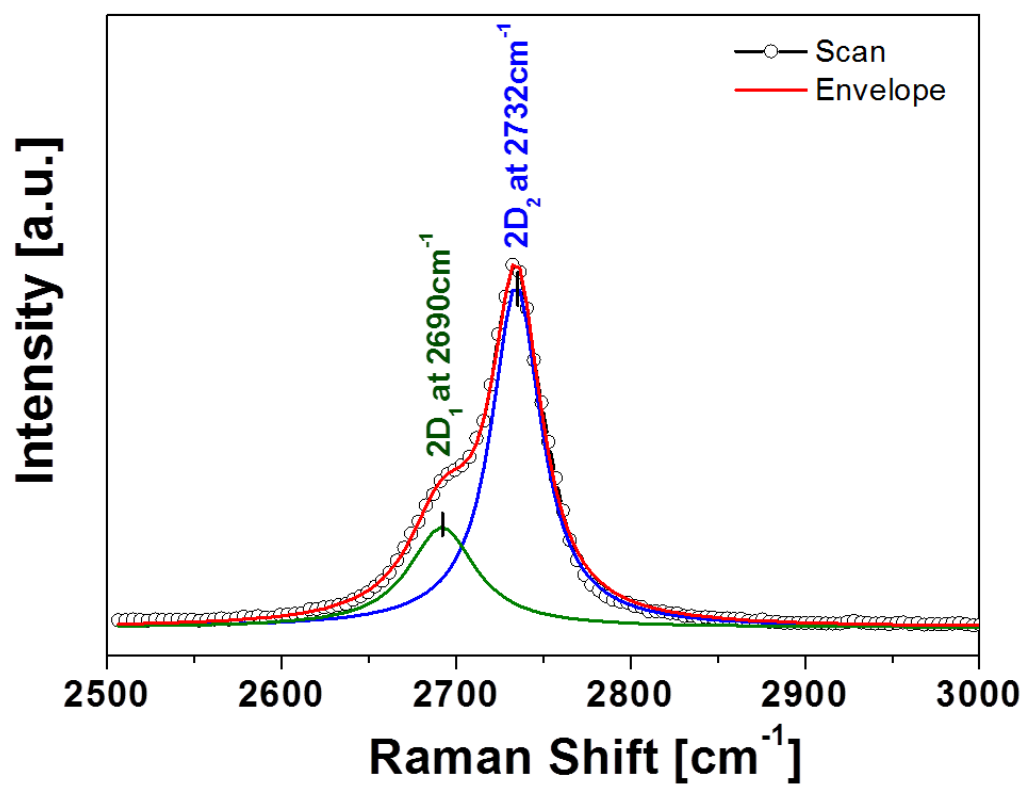


Figure S3. Raman spectra of 2D band of HOPG with its 2D₁ and 2D₂ components deconvoluted.

1. GO Synthesis

To extend the size of RGO, a modified Hummers method was used to obtain Graphene Oxide (GO) sheets. Briefly, 2 grams of graphite flakes (3-5mm, NGS) were mixed with concentrated H_2SO_4 (12ml) and stirred for 4 hours at 80°C . After cooling to room temperature, the flakes were ultra-sonicated for 1 hour and diluted with DI water (500ml) and left overnight. The mixture was then filtered over a $0.2\mu\text{m}$ filter to obtain pre-oxidized graphite powders. The product was dried overnight. Dried graphite powder was then added into concentrated H_2SO_4 (120ml) and KMnO_4 (15g) and stirred for 2 hours. The solution was then put into an ice bath and diluted slowly with DI water (250ml). After 2 hours, a DI water (700ml) and H_2O_2 (20ml) was added into the mixture, resulting in a color change to light yellow. The obtained mixture was dialyzed and centrifuged to obtain GO.

2. Transfer of GO to substrates

SiO_2 substrates are RCA cleaned two times, dried and then rinsed in ethanol before being placed into 1% v/v 3-APTES ethanol solution for 1 hour. The 3-APTES SiO_2 substrates were then placed in oven for 1 hour to complete the functionalization process. After the substrates cooled, prepared GO solution was drop-casted and allowed to stand for 1 hour. Thereafter, the substrates were rinsed, blow-dried and kept ready for further treatment.

3. Reduction of GO by Hydrazine

Prepared GO substrates are placed in a glass petri-dish with $400\mu\text{l}$ of hydrazine monohydrate dropped in the center of the dish and then sealed. The petri dish was heated to 70°C and left overnight for complete reduction. Thereafter, the substrates are rinsed and blow-dried.

4. Characterizations

SEM: Field Effect SEM images were taken using a JEOL JSM-7600F instrument operating in gentle beam mode at 1KV.

TEM: Transmission Electron Microscope images were taken using a JEM-2100F instrument operating at 200KV.

AFM: AFM images were obtained by using Asylum AFP-3D (Asylum Research, US) in tapping mode using AC240TS Silicon tip (Olympus, Japan) under ambient conditions with a scanning rate of 0.2Hz, scanning line of 256 and analyzed using WSxM Develop 3.3 software. As it is not possible to transfer the GO layers to another substrate, the bare substrate is taken as the lowest point in all the images.

Raman: Raman spectra were recorded on a WITec CRM200 confocal Raman microscopy system using excitation source from a 488 nm laser with an air-cooled charge-coupled device as detector (WITec Instrument Corp, Germany). Before measurements, the instrument was calibrated by silicon wafer. Sample areas of 10 μ m x 10 μ m were mapped using a precision xyz stage with 100 points per line and 100 lines per image.

XPS: XPS spectra was carried out using a VG ESCA 220i-XL Imaging XPS (Thermo Scientific, US). Mg Ka X-ray source from twin-anode X-ray gun was employed for analysis with photoelectron take-off angle of 90°. “Large Area” lens mode was used without turning on the flooding gun. The analysis area is about 4mm x 4mm. C1s and C KLL spectra were captured with pass energies of 20eV and 100eV, respectively.

Electrical measurements: Electrical output characteristics were measured with a 4200-SCS semiconductor parameter analyzer (Keithley, US). Gold (100nm) electrodes with titanium (20nm) adhesion layer were fabricated on the samples via thermal evaporation with hard-masks of 100 μ m channel length.