# Electronic Supplementary Information

# High Quality Graphene with Large Flakes Exfoliated by Oleyl Amine

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*Experimental*

A mixture solution of 15 ml 95% sulfuric acid and 5 ml 65% nitric acid was maintained at 20 °C in a water bath. Graphite (1 g) and potassium bichromate (0.01 g) were added to this solution. then, the mixture was stirred for 40 min at 30 °C. During this time, a sulfuric acid/ nitric acid/graphite intercalation compound (SN-GIC) formed. After the reaction was complete, the SN-GIC was washed thoroughly with water, filtered off under vacuum, and dried at 50 °C to give dry intercalation graphite. All the chemicals and solvent are purchased from Aldrich and Alfa Aesar.

The intercalation graphite was dispersed in oleyl amine at a concentration of 3 mg/ml and heated in a sealed reactor vessel at 170 °C for 72 h. The mixture was sonicated in a low power sonic bath for 30 minutes to form a homogeneous suspension. The suspension was centrifuged at 2000 rpm (400 g) for 15 minutes to separate the non-soluble material from the solution. A black suspension with predominantly single layer GS retained in the supernatant was obtained. The concentration was verified by passed the graphite dispersion through a polyvinylidene fluoride (PVDF) membrane (pore size: 400 nm), and carefully measure the filtered

mass. The acid intercalation graphite was sonicated in pure water to remove the inorganic acid. Then the dispersion through a polyvinylidene fluoride (PVDF) membrane to got GS in Fig. S1a. The olyel amine treated GS was filtered from the mentioned “GS suspension”. Both samples was dried in vacuum at 50 °C.

The small flakes graphene solution was got by three times centrifuged at 2500 rmp (650 g) for 10 minutes. The large flakes graphene solution was got after centrifuged at 1000 rmp (100 g) for 10 minutes, the aggregates were then resuspended in fresh oleyl amine by a brief sonication step. Thin films were prepared by vacuum filtration onto polyvinylidene fluoride (PVDF) membranes (pore size: 400 nm), and washed several times by chloroform. This film drying was carried out in a quartz tube at 80 °C a pressure of ~$10^{-3}$ mbar. The centrifugation parameters were decided by our optimized experiments.

The sample was characterized by infrared spectroscopy (IR, Tensor 27), scanning electron microscopy (SEM, Hitachi S-4300, 15 kV), atomic force microscopy (AFM, tapping mode), transmission electron microscopy (TEM, JEM-2010, 200 kV), selected area electron diffraction (SED, equipped on TEM), Raman spectrometer (Lab Ram HR800, with laser excitation at 514 nm), X-ray photoelectron spectroscopy (XPS, SCA Lab220I-XL) measurement.

The interactions between GS and oleyl amine are mainly ascribed to physisorption and ion bond, which could be characterized by IR spectroscopy (Figure S1 ). After treated by oleyl amine, the peak of carboxylic acid vibration (1725 $cm^{-1}$) decreased in intensity, while new peak appeared at 1585 $cm^{-1}$ which corresponds to carbonyl stretch of the $COO^-$ group.[1] This peak confirmed the zwitterions on the surface of GS. It also exhibits bands around 1634 $cm^{-1}$ due to aromatic C=C as well as bands due to carboxy C–O (1412 $cm^{-1}$). The bands located at 1542 $cm^{-1}$ (N–H bending of amide), 1458 $cm^{-1}$ (C–N stretch of amide) appear in Figure S1b.

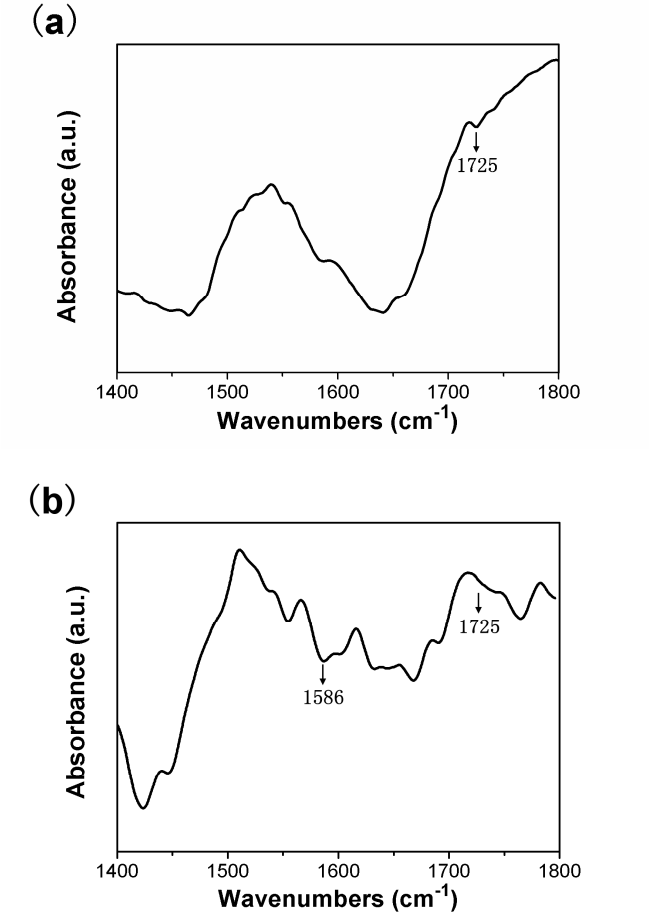

**Figure S1.** (a) IR spectra of acid tread GS (b) IR spectra of oleyl amine tread GS.

The interactions of acid and oelylamine is the key to efficiently exfoliate graphene. After heated in oelylamine, there are 15.8% (by weight) sulfuric acid remained in the intercalated graphite, and the acid could be fully removed after sonication.

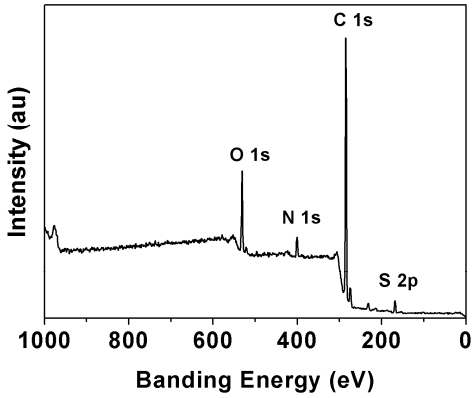


**Figure S2.** Wide survey XPS spectrum of the intercalated graphite after heated in oelyl amine.

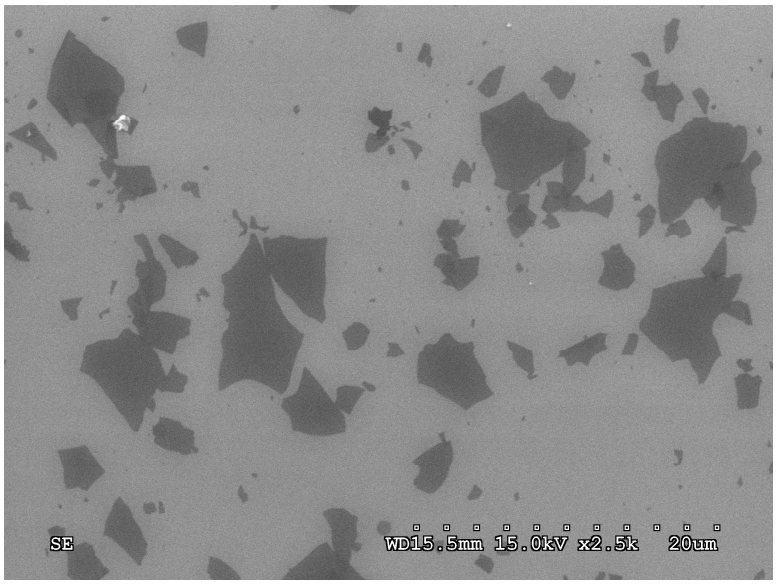


**Figure S3.** SEM images of the graphene sheets on $SiO_2$/Si substrate.

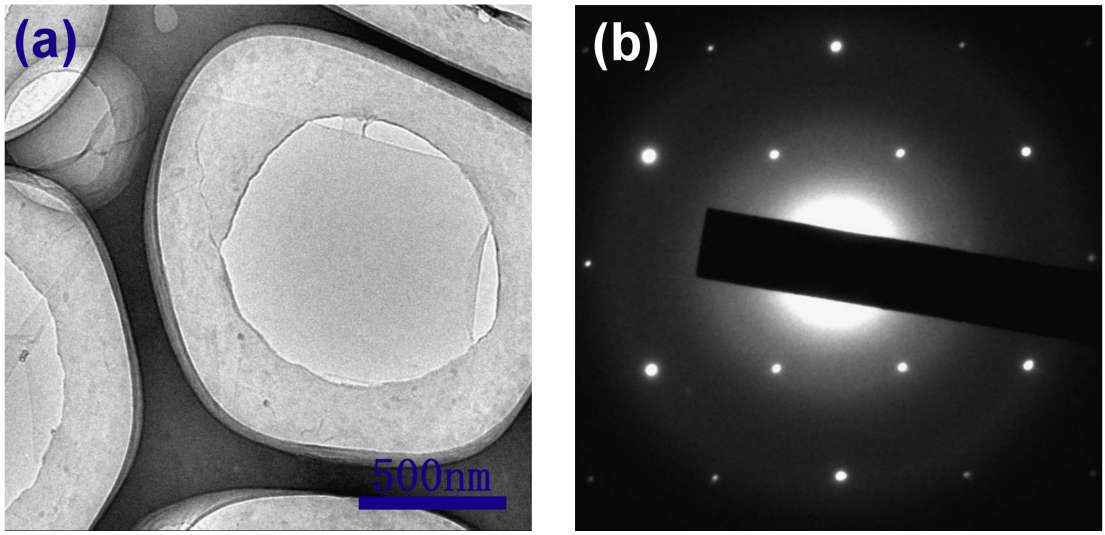


**Figure S4.** (a) Bright-field TEM images of bilayer graphene. (b) Electron diffraction patterns for the several layers graphene shown in (a).

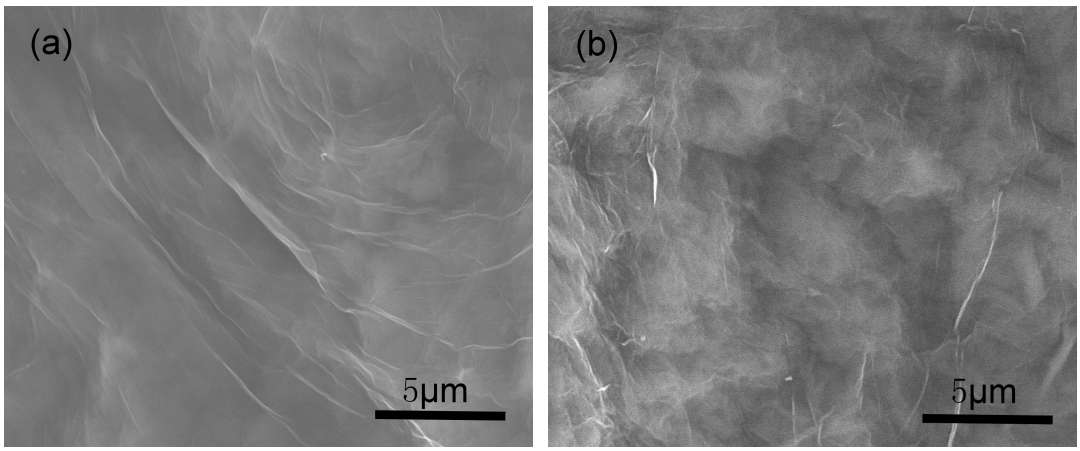


**Figure S5.** SEM images of the surface of a graphene film. (a) Thin film composed of large flakes. (b) Thin film composed of small flakes.

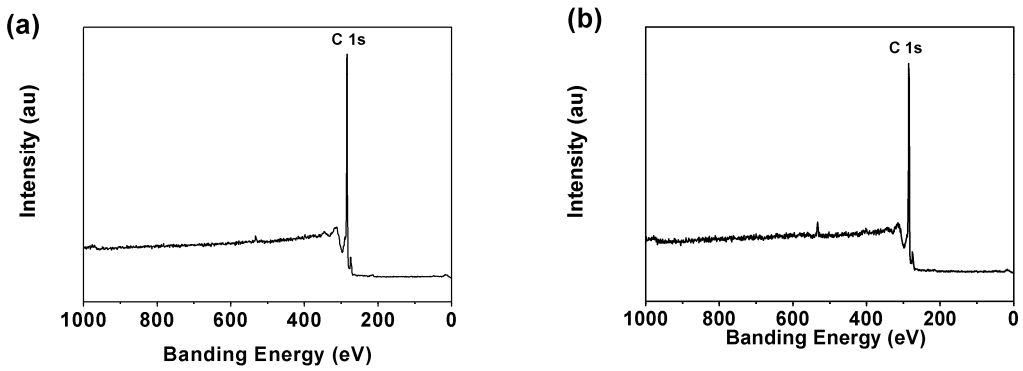


**Figure S6.** Wide survey XPS spectrum of the graphene. (a) Thin film composed of large flakes. (b) Thin film composed of small flakes.

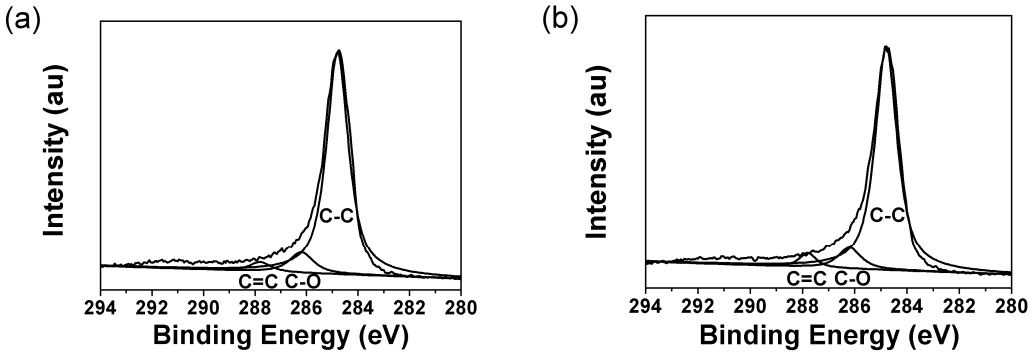


**Figure S7.** High-resolution XPS spectrum of the GS film. a) the large flakes GS film. b) the small flakes GS film.

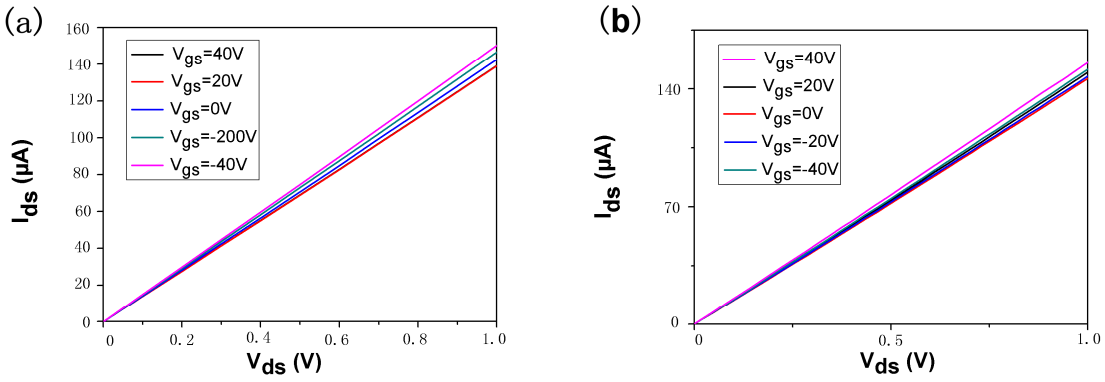


**Figure S8.** (a) $I_{ds}$–$V_{ds}$ characteristics at various $V_g$ for the graphene (Fig. 4) in air. (b) $I_{ds}$–$V_{ds}$ characteristics at various $V_g$ for the graphene (Fig. 4) in nitrogen.

## Notes and references

(1) M. A. Hamon, J. Chen, H. Hu, Y. Chen, M. E. Itkis, A. M. Rao, P. C. Eklund, R. C. Haddon, *Adv. Mater.* **1999**, *11*, 834.