

SUPPORTING INFORMATION

Pyrrrodiline-Functionalized Fluorine-Containing Graphene Sheets

Jing Dai, Yongjun Li, Zhong Huang, Xiaoyu Huang**

Experimental

Chemicals

All reagents and solvents including colloidal graphite powder (Qingdao Baichuan Graphite Co. Ltd., particle size: 1 μm), *o*-dichlorobenzene (ODCB, Alfa Aesar, 98%), pentafluorobenzaldehyde (Aldrich, 98%), *N*-methyl-2-pyrrolidone (NMP, Alfa Aesar, 99.5+%), sarcosine (Aldrich, 98%), *N,N*-dimethylformamide (DMF, Aldrich, 99.8%), ethanol (Aldrich, anhydrous, 99.5%), dichloromethane (DCM, Aldrich, 99.8%), and toluene (Aldrich, 99.8%) were used as receive.

Preparation of FGS via 1,3-Dipolar Cycloaddition Reaction

Firstly, colloidal graphite powder (100 mg) was dispersed in ODCB (200 mL) by sonication at 53 kHz for 0.5 h to give homogeneous graphite dispersion. This dispersion, pentafluorobenzaldehyde (927 mg), and sarcosine (500 mg) were introduced into a 500 mL sealed flask and the mixture was heated at 150°C with vigorous stirring under N_2 . The initial black mixture turned into brown and a brown

suspension containing *N*-methyl-2-(pentafluorophenyl)pyrrolidine-functionalized graphite flakes was obtained after 4-days 1,3-dipolar cycloaddition reaction. The suspension was then filtered through a polytetrafluoroethylene (PTFE) membrane (0.45 μm) followed by washing repeatedly with ODCB, ethanol, and DCM to remove unreacted organics until the filtrate turned clear. The obtained black solid was dried overnight at 40°C *in vacuo*.

Finally, functionalized graphite was exfoliated in organic solvents such as ODCB, ethanol, and DCM by sonicating at 53 Hz for 0.5 h and then centrifugating in 2000 rpm for 10 min. For XPS measurement, FGS suspension was centrifugated at 8000 rpm for 0.5 h and then dried overnight at 40°C *in vacuo* to afford FGS black solid.

Characterization

Unpolarized Raman scatterings were performed on a Renishaw InVia Raman microscope using a 532 nm argon ion laser and recorded from 500 cm^{-1} to 3500 cm^{-1} . XPS was recorded on a PHI 5000c ESCA photoelectron spectrometer. TGA measurement was run on a TA Q500 system from 50°C to 800°C under N_2 purge with a heating rate of 10°C/min. AFM images were taken by a Veeco Nanoscope IVa MultiMode SPM in the tapping mode. Field emission transmission electron microscope (FE-TEM) images were obtained by a JEOL JEM-2100F instrument operating at an accelerating voltage of 200 kV.

For the sample preparation, FGS was dispersed in different solvents such as ODCB, DMF, NMP, toluene, ethanol, diethyl ether, and DCM. The mixture was then

subjected to low speed centrifugation at 2000 rpm for 10 minutes followed by keeping the black suspension overnight for TEM and AFM measurements. AFM samples were prepared by spin-coating on silicon substrate (washed in a mixture of H_2SO_4 and H_2O_2 prior to use) with a rotation rate of 200 rpm for 6 seconds and 800 rpm for 30 seconds, and TEM samples were prepared by dripping a few drops of the solution onto copper grids with an ultrathin holey carbon film. All samples were dried at ambient condition.

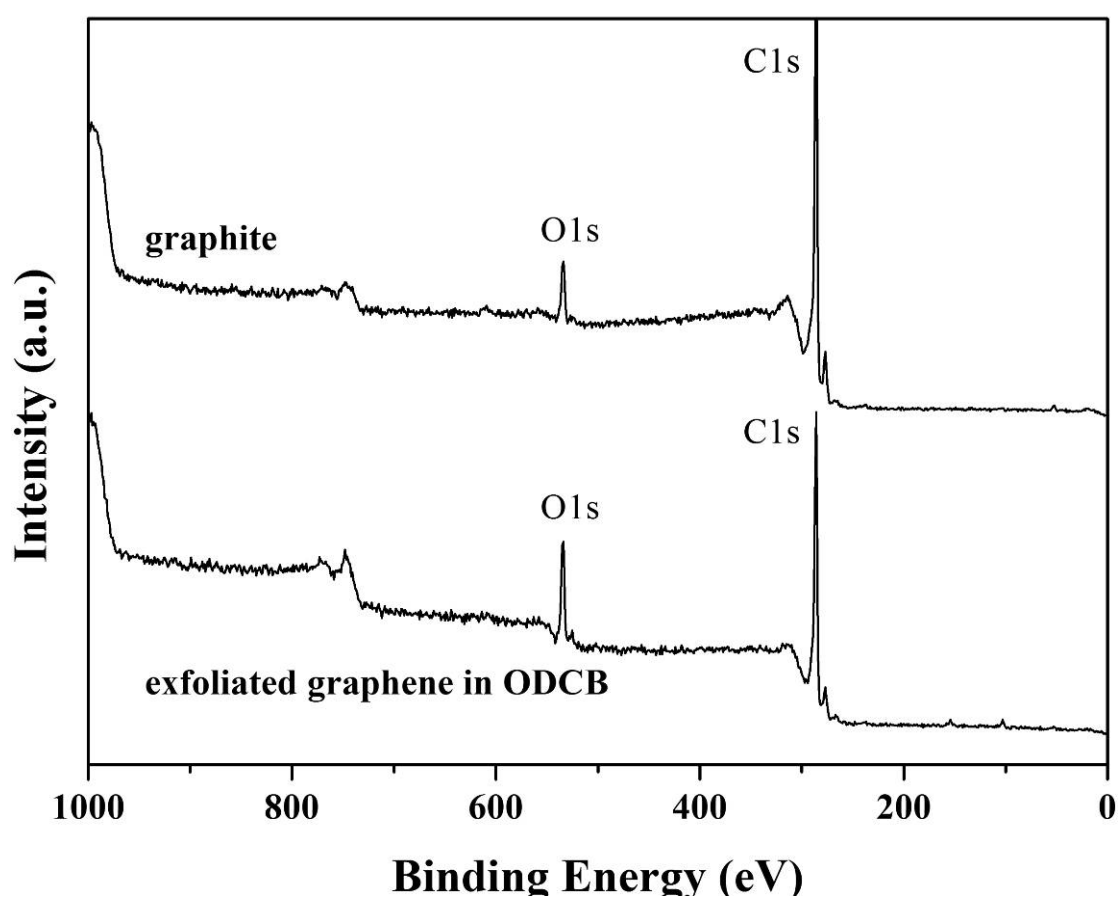


Figure S1. XPS survey scan spectra for graphite and exfoliated graphene in ODCB.

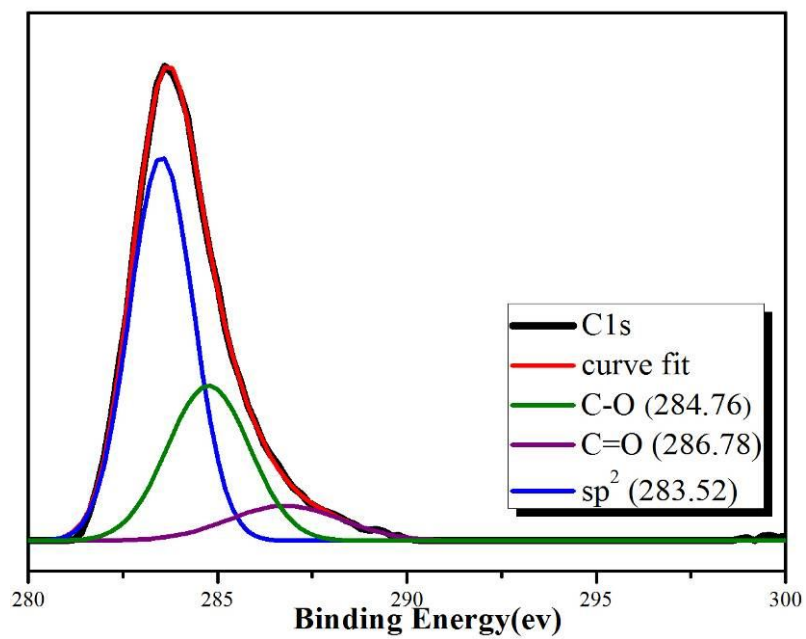
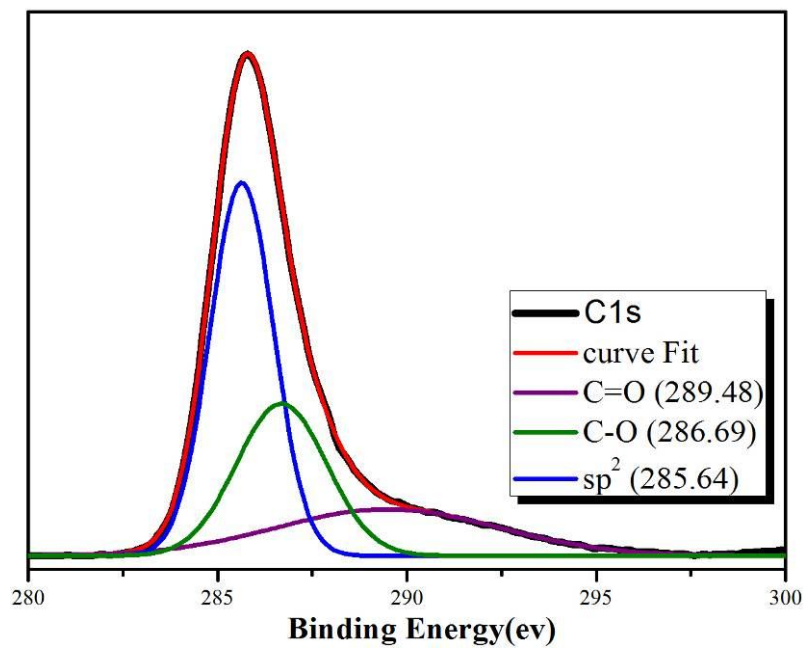


Figure S2. XPS high resolution spectra of C 1s for graphite (up) and functionalized graphite (down).