Electronic Supplementary Information

Facile and economical exfoliation of graphite for high-quality graphene sheets with mass production

Tianquan Lin,^{*a,b*} Jian Chen,^{*a*} Hui Bi,^{*a*} Dongyun Wan,*^{*a*} Fuqiang Huang,*^{*a,b*} Xiaoming Xie,^{*c*} and Mianheng Jiang^{*c*}

 ^a CAS Key Laboratory of Materials for Energy Conversion, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050, China;
^b State Key Laboratory of Rare Earth Materials Chemistry and Applications, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, China;
^c State Key Laboratory of Functional Materials for Informatics, Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Shanghai 200050, China.

*Correspondence should be addressed to <u>huangfq@mail.sic.ac.cn</u>

Characterizations. The sheet resistances and electron mobility of wafer-sized graphene films were measured by the four-probe Van Der Pauw method with an Accent HL5500. Raman spectra were collected on a Thermal Dispersive Spectrometer using a laser with an excitation wavelength of 532 nm at laser power of 10 mW. TEM were conducted using a JEOL 2100F microscope, operating at 200 kV. XPS experiments were carried out on a RBD upgraded PHI-5000C ESCA system (Perkin Elmer) with Mg K α radiation (hv = 1253.6 eV). Fourier transform infrared (FTIR) spectra were recorded on Perkin-Elmer Spectrum 100 using KBr disks. Zeta-potential values were determined using a Malvern Zetasizer (Nano ZS, Malvern Instruments).



Figure S1. Additional TEM images of three different graphenes for comparion. (a–b) HRTEM image of chemically modified graphene sheets (CMG) and its corresponding to Fast Fourier Transform (FFT). (b–c) TEM images of graphene sheets obtained by planetary milling with NaCl (NaCl-G) and MgO (MgO-G), respectively.



Figure S2. The thickness statistics of graphene sheets *via* ball mill with oxalic acid by analyzing a large number of HRTEM images for the flake edges and AFM height profiles.



Figure S3. Micro-Raman spectra recorded at the edge and within the basal plane of graphene before (a) and after annealing (b) at 600 °C for 5 min. Insets are optical microscopy images.



Figure S4. (a) Photograph of freestanding flexible graphene paper and its cross-sectional SEM image (b).



Figure S5. Raman spectra from within the basal plane of exfoliated graphene with assistance of NaCl (NaCl-G) and MgO (MgO-G).



Figure S6. XPS survey spectra of chemically modified graphene (CMG), as-made graphene sheets (GS).