Electronic Supplementary Material (ESI) for Chemical Communications

Supplementary Information

Water can stably disperse liquid-exfoliated graphene

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Experimental and Characterization

100 mg graphite powder (≤300 meshes, Alfa Aesar) was dispersed in 200 mL DMF and then sonicated for 60 min in a low-power ultrasonic bath (45 W, 59 Hz). The dispersions were centrifuged for 45 min at 64g to remove any thick and largish unexfoliated graphitic flakes, eventually resulting in 160 mL homogeneous colloidal dispersions of graphene flakes in DMF. These graphene dispersions were vacuum filtered through a nylon membrane (pore size ~220 nm) and washed several ten times by water to form a graphene cake with negligibly residual DMF. The cake was further dried for 72 h in the vacuum oven at 200 ^{o}C. The dried cake was fragmented into powder. A certain amount of this powder (3 mg) was dispersed in 30 mL deionized water (five samples), followed by sonication for 90 min and subsequent centrifugation for 45 min at 64, 256, 576, 1024, and 1600g. After centrifugation, the top two-thirds of the dispersions were extracted by pipet to obtain graphene dispersions and retained for further characterization.

UV-Vis absorption spectroscopy and optical absorption measurement at a certain wavelength were performed with a Purkinje General TU1901 spectrophotometer using 1 cm quartz with cuvettes. The concentration of dispersions after centrifugation was calculated from Lamber-Beer law, $A/l=\alpha C$, where A is the absorbance measured at 300 nm, l is the path length, C is the concentration, and α is taken as 2460 mL/mg/m.[S1] Thin films were prepared by vacuum filtering the dispersions onto porous mixed cellulose membranes with a nominal pore size of 0.45 μm. Sheet resistance, R_s, was measured by a KDY-1 four-probe resistivity test system (GuangZhou KunDe). Atomic force microscopy (AFM) samples were prepared by dropping several acetone-diluted microliters to the freshly cleaved mica substrates and

drying in the ambient condition. AFM images were captured with a Multimode 8 AFM (Digital Instruments/Bruker Systems) in ScanAsyst Air mode. Transmission electron microscopy (TEM) samples were prepared by drop casting a few drops of dispersions onto holey carbon grids drying in the ambient condition. Bright field TEM imaging and high-resolution TEM (HRTEM) images were taken with a Jeol 2100 operating at 200 KV. The Raman samples were the filtered films. The Raman measurements were made by a Renishaw Rm2000 using a 514 nm laser. X-ray photoelectron spectroscopy (XPS) was obtained by a Thermo Fisher Scientific ESCALAB-250 spectrometer equipped with a monochromatic Al Kα X-rays excitation source (1486.6 eV). Zeta potential measurements were carried out on a Malvern Zetasizer 3000HSa system. All measurements were carried out at 25 ^{o}C. The zeta potential is calculated (in SI units) as $\zeta = \eta\mu/\varepsilon$ by the instrument, where η is the solution viscosity, ε is the solution permittivity, and μ is electrophoretic mobility. Each sample was measured 6 times in a run and the Zeta potential is averaged.

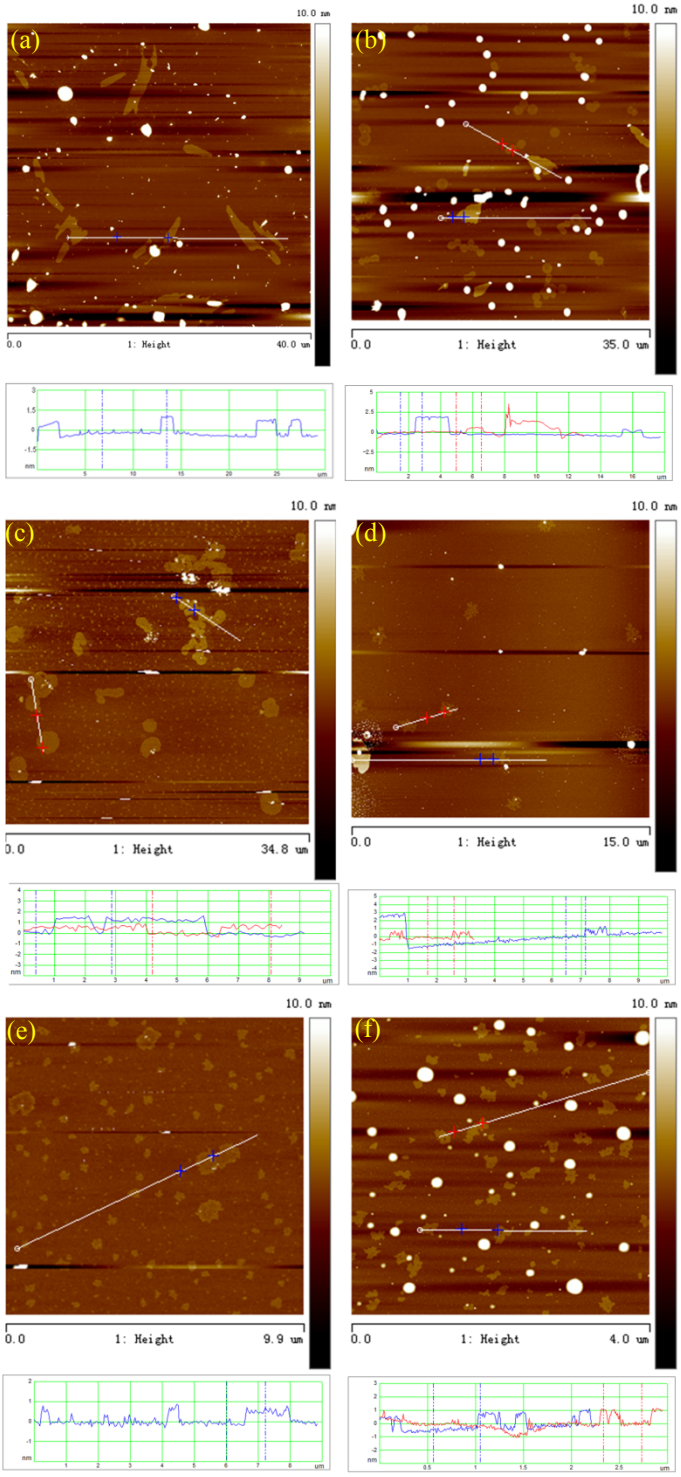

Figure S1. More representative AFM images and the corresponding height profiles of liquid-exfoliated graphene flakes in DMF (a) and graphene flakes dispersed in water after centrifugation acceleration of 64g (b), 256g (c), 576g (d), 1024g (e), and 1600g (f).

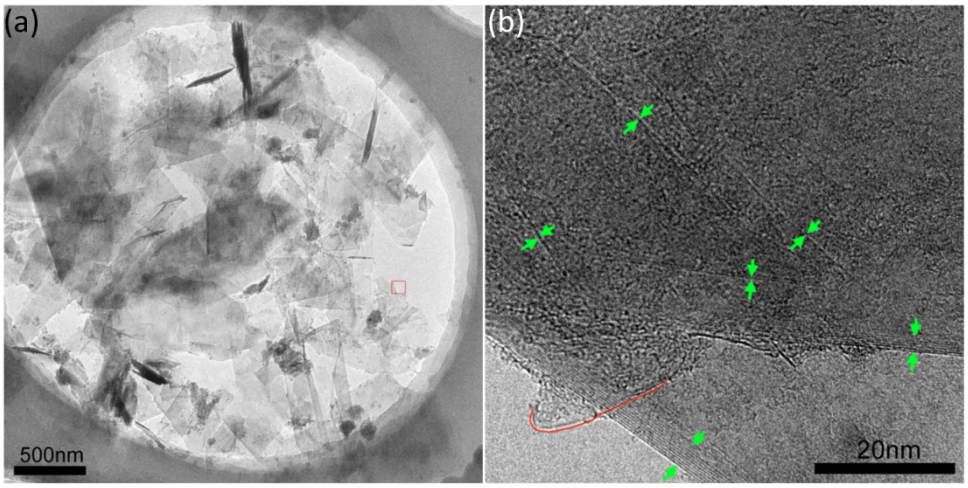

Figure S2. Bright field TEM images of graphene flakes dispersed in (a) water after a centrifugation acceleration of 64g. The centrifugation acceleration is relatively low. Some aggregation happens. The accurate layer number is difficult to determine. (b) Magnified TEM image of the rectangle region in (a), showing lots of folded edges and evidence for monolayers (inset arrows).

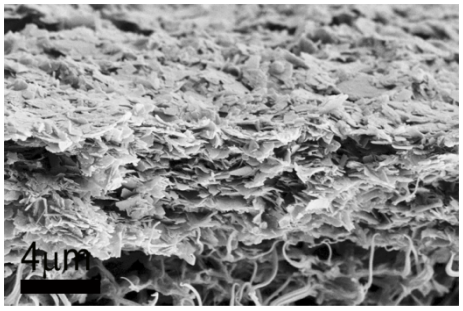

Figure S3. SEM image of the section of a graphene-based film prepared by vacuum filtering water-dispersed graphene. We prepared a film by vacuum filtering these graphene/water dispersions and measured its conductivity by standard four-point probe method. The film (Φ20 mm) is relatively loose and estimated as ~5 μm thick after drying under vacuum at 50 ^{o}C for 24 h (without high-temperature annealing).

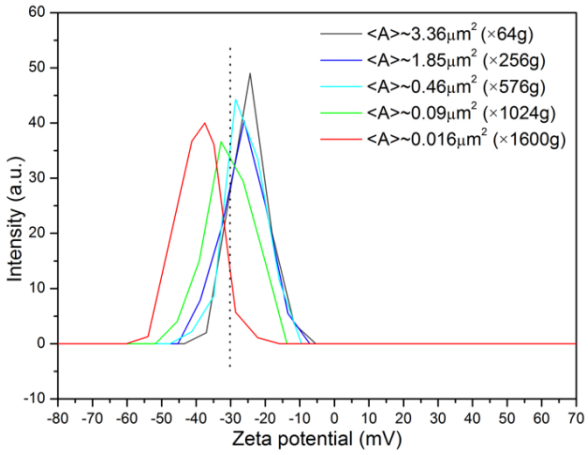

Figure S4. Zeta potential distributions for water-graphene dispersions with graphene flakes of different averaged area.