

## ESI:

# Graphene from electrochemical exfoliation and its direct applications in enhanced energy storage devices

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### Electrochemical exfoliation:

All electrochemical exfoliation experiments were carried out by Autolab potentiostats. The solvent is a 1:1 (v/v) mixture of [BMIM][BF<sub>4</sub>] and deionized (DI) water. It underwent 50 cycles with cyclic voltammetry in the potential range from -2 V to 3V with scan rate of 50mV/s, followed by a 6-hour discharge at a fixed 5 V voltage. The exfoliated solutions were filtrated under vacuum, washed with an ethanol/water mixture several times thoroughly and ultracentrifuged at 13300rpm for 20 mins to separate the carbonaceous products from RTILs.

### Morphology characterization:

Morphology of the resulting graphene flakes was characterized by scanning electron microscopy (SEM) and atomic force microscopy (AFM). The graphene inks made from RTIL-water mixture and from the one containing 0.5 M LiClO<sub>4</sub> were drop casted onto a plasma treated SiO<sub>2</sub> substrate. Electrochemical exfoliation transforms the nearly plain surface of a graphite foil sheet (Fig. 1a) into a wrinkled and curled morphology as shown in Fig. 1b and Fig. 1c. Some small granules can be observed from Fig 3c which may be the crystallized LiClO<sub>4</sub> salts residue on the graphene flakes. The graphene flake exfoliated from 1:1 (v/v) mixture of [BMIM][BF<sub>4</sub>] and DI water containing 0.5 M LiClO<sub>4</sub> was separated and its thickness was measured by AFM as shown in Fig. 1d. AFM imaging of graphene has been carried out in tapping mode over the image area of 2 μm × 2 μm. Various groups reported different results on thicknesses of graphene layer ranging from 0.35nm to 3 nm, relative to the SiO<sub>2</sub> substrate. Novoselov et al. measured platelet thicknesses of 1–1.6 nm<sup>[1]</sup>. Gupta et al. have measured an instrumental offset induced by the AFM, with result of 0.7 nm height for a single layer<sup>[2]</sup>. The variation in the thickness of the single graphene layers may be attributed to change in the tip-sample interaction as the tapping tip scans over the surface. In our case, thickness of the indicated region (Fig. 3d) of the exfoliated graphene is about 2 nm.

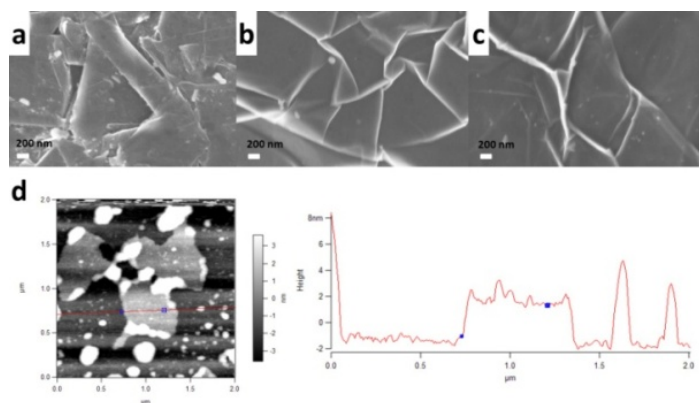


Figure 1 SEM images of a) graphite, b) exfoliated graphene from 1:1 (v/v) mixture of RTIL and water, c) exfoliated graphene from 1:1 (v/v) mixture of RTIL and water containing 0.5 M LiClO<sub>4</sub>, and d) AFM profile measurement image for electrochemically exfoliated graphene.

Ref1: K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov, *Nature*, 2005, **438**,197.

Ref2: A. Gupta, G. Chen, P. Joshi, S. Tadigadapa, and P. C. Eklund, *Nano Lett.*, 2006, **6**, 2667.

#### Assemble of coin cells:

Standard 2032 coin cells were assembled inside an mBraun glovebox (H<sub>2</sub>O < 2ppm, O<sub>2</sub> < 10 ppm). In such primary lithium battery, lithium foil was used as anode with polymer electrolyte composed of high molecular weight poly(ethylene glycol) borate ester. The Lewis acid centers of the borate esters interact with the anion resulting in enhanced lithium ion transport and the resulting performance of such polymer electrolytes is comparable with conventional liquid organic electrolytes at room temperature.