

Supporting Information

Few-Layer Graphene Island Seeding for Dendrite-Free Li Metal Electrodes

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Experimental: Hydrothermal exfoliation and spin coating

Water, ethanol, and ammonia molecules were flowed and filled within the interspace of the expanded graphite basal planes during heating while using a magnetic stirring process. In the high temperature and pressure assisted hydrothermal reduction reaction, H^+ and NH_4^+ ions provided a dehydration effect for the hydrogen and carboxyl group-terminated ExG. The ions can increase the surface zeta potential and electrostatic repulsion force and thus exfoliated graphene sheets underwent plane separation and dispersion with only weak van der Waals bonds. During the process, a few nitrogen atoms from NH_4^+ ions can be doped or absorbed onto the exfoliated graphene sheets. Finally, the exfoliated few-layer graphene sheets had sizes that varied from hundreds of nanometers to micrometers.

The graphene sheets were dispersed with PVDF in an N-methyl-2-pyrrolidone solution by sonication. In the solution, the concentration of PVDF was 0.02 wt% and the ratio of N-FLG to PVDF was 9:1. Then, the solution was coated onto a Cu foil by spin-coating. During the process, PVDF is subjected to centripetal forces that promote the deposition of the graphene sheets as well as the formation of lateral interconnections between the artificial graphene islands on the Cu substrate. Conversely, highly massive particles that were not exfoliated from ExG and highly stacked multilayer graphene were subjected to high centrifugal forces and thus tended to fly off the substrate. As a result, few-layer graphene islands with dimensions of only a few hundred micrometers covered the surface of the Cu foil. Meanwhile, an extremely thin PVDF layer was also coated onto the remaining area of the Cu foil.

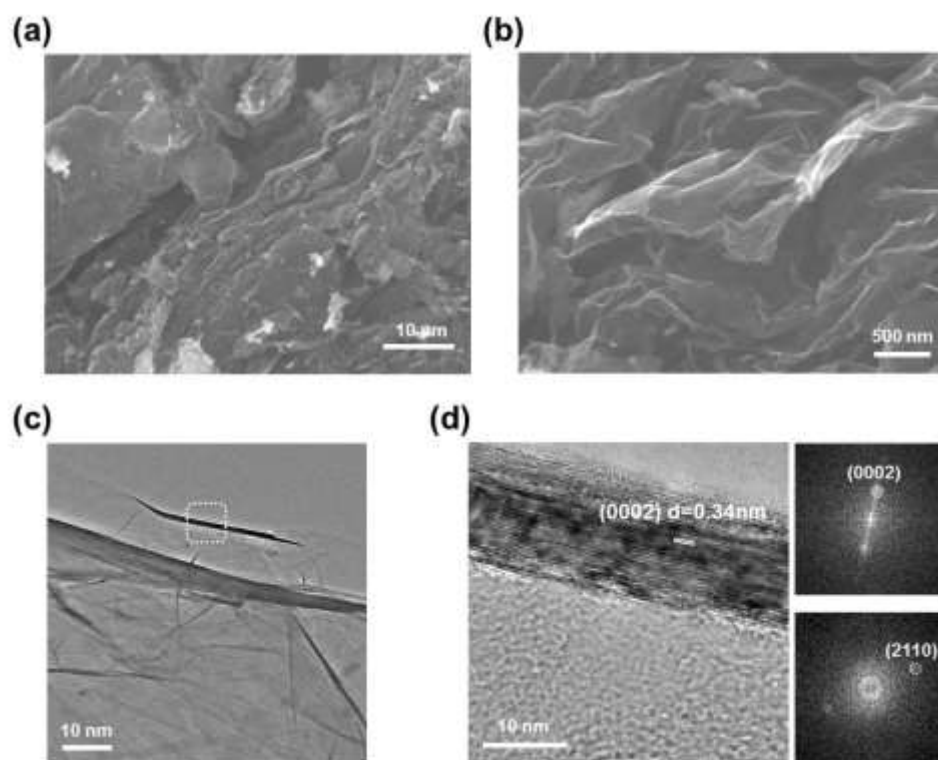


Figure S1. (a) low-magnification and (b) high-magnificaiton FE-SEM images, (c) TEM image, and (d) HR-TEM image and corresponding FFT patterns for pristine EXG.

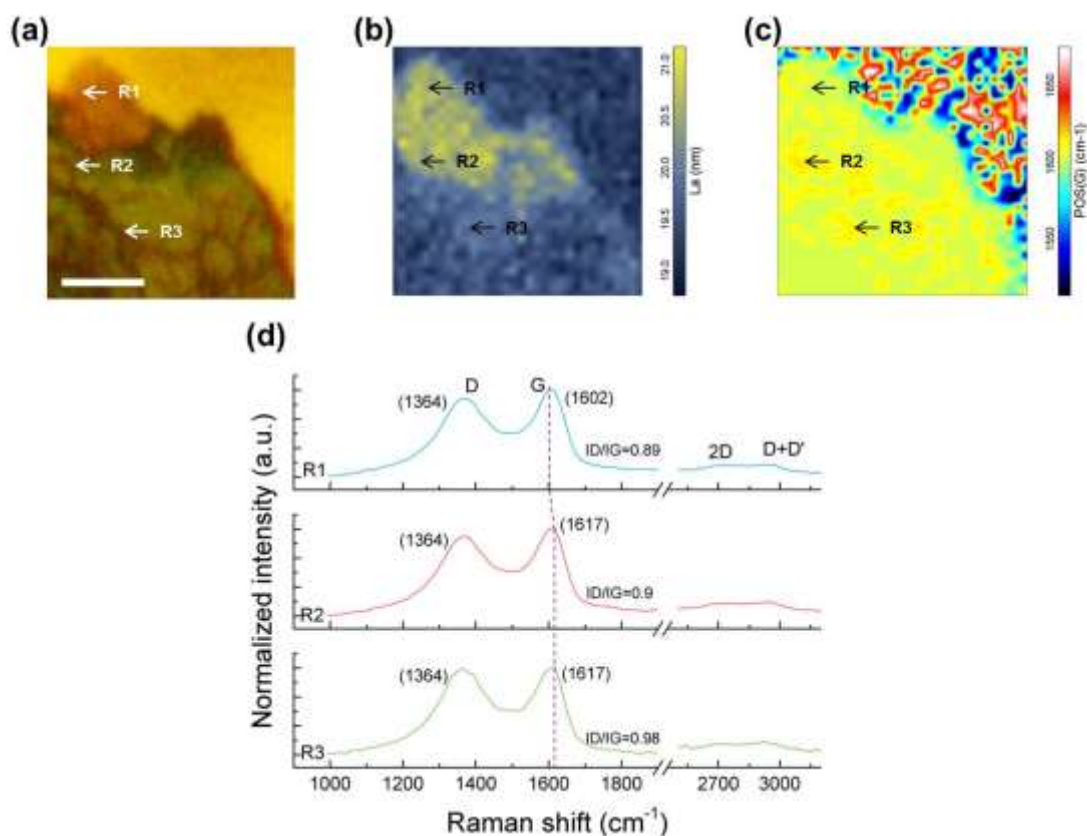


Figure S2. (a) CCD camera image, (b) L_a : calculated crystal size, (c) POS(G): position of G band and (d) Raman spectra at selected points in the mapping image of N-FLG/Cu (scale bar: 5 μm).

Figure S2 shows the Raman spectroscopy analysis results. The pronounced D and G bands at around 1364 cm^{-1} and $1602/1617\text{ cm}^{-1}$ are the characteristic peaks corresponding to crystal imperfections and C-C bond stretching, respectively.^{1,2} The ratio of $I(\text{D})/I(\text{G})$ represents the degree of disorder, which is interpreted as the ordered crystal size (L_a) based on a calculation according to the following equation.³

$$L_a(\text{nm}) = (2.4 \times 10^{-10}) \lambda_1^4 \left(\frac{I_D}{I_G} \right)^{-1}$$

As shown in the calculated L_a mapping (Figure S2b), relatively small crystal sizes and local variations of the N-FLG on Cu were found. The up-shifted G band was commonly observed at the top surface (Figure S2c and Figure S2d); this band can be induced by both electron and hole doping.⁴ Therefore, the N-FLG should have some defects caused by N-doping, which can contribute to the initial Li nucleation.

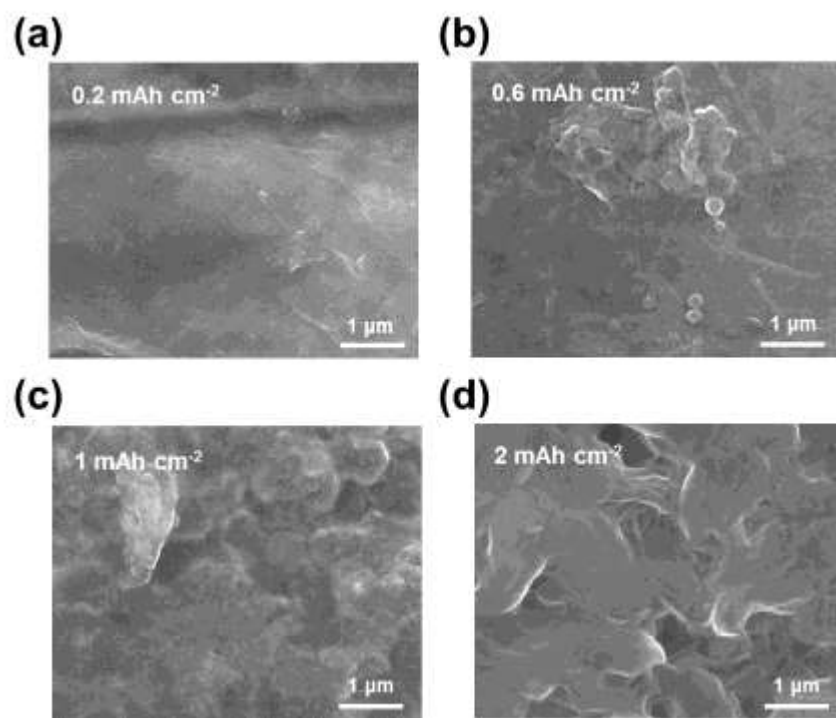


Figure S3. FE-SEM images at various charge stages for (a) 0.2 mAh cm^{-1} , (b) 0.6 mAh cm^{-1} , (c) 1 mAh cm^{-1} , and (d) 2 mAh cm^{-1} .

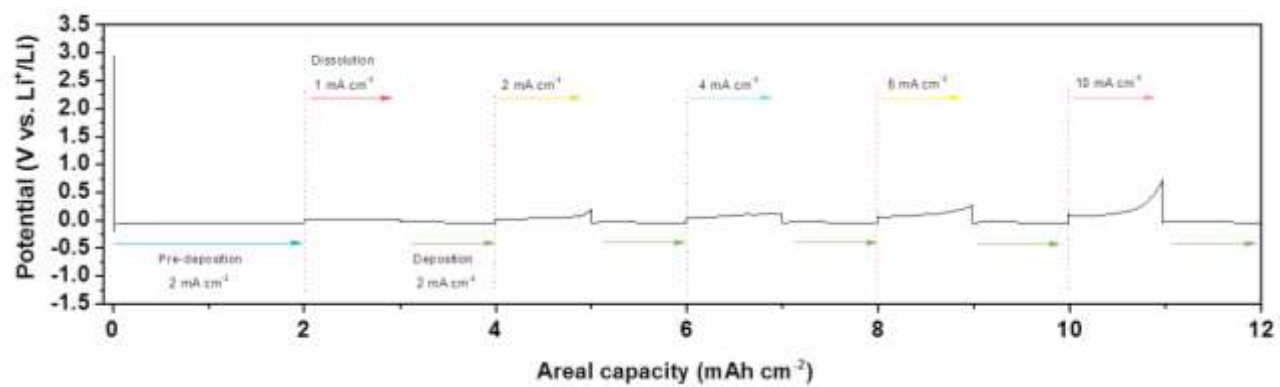


Figure S4. Voltage profiles at various rates (constant currents).

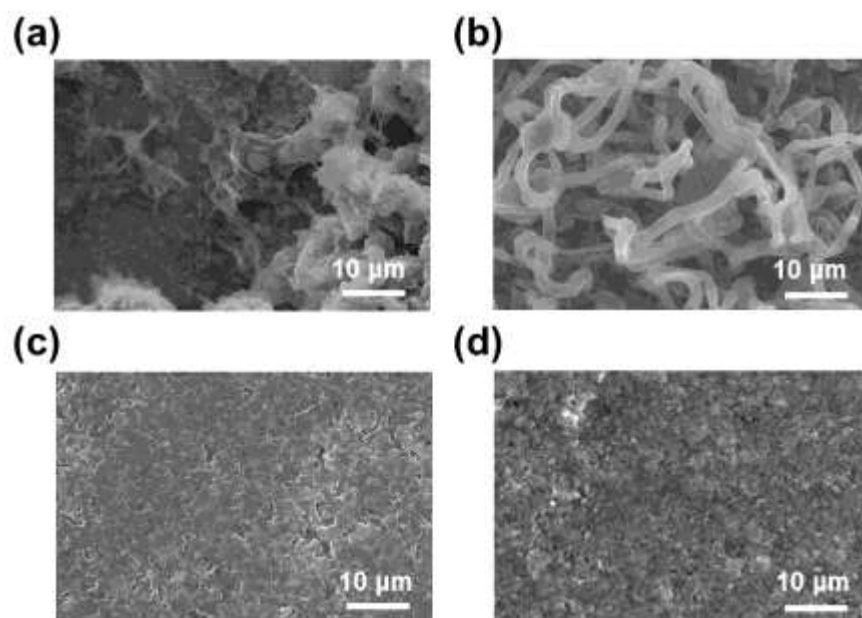


Figure S5. FE-SEM images for pure Cu electrodes (a) after the first deposition and (b) after the 40th deposition and for N-FLG electrodes (c) after the 40th deposition and (d) after the 120th deposition.

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