

Supporting Information

A Practical Li-Ion Full Cell with a High-Capacity Cathode and Electrochemically Exfoliated Graphene Anode: Superior Electrochemical and Low-Temperature Performance

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Experimental

Materials synthesis.

LNCM-3. The precursor $[\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}](\text{OH})_2$ was prepared in our previous report¹. $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$, $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$, and $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ were used as raw materials without any purification. A homogeneous aqueous solution of metal sulfates (Ni:Co:Mn=6:2:2, molar ratio) with a concentration of 2 mol L^{-1} was dissolved in distilled water and pumped into the continuously stirred tank reactor (CSTR, 10 L). Meanwhile, a NaOH solution (pH control agent) of 4 mol L^{-1} and the desired amount of NH_4OH solution (chelating agent) were pumped into the CSTR. During the reaction, the temperature, pH, stirring speed were precisely controlled. After the reaction, the hydroxide precursor was obtained after washing with distilled water and finally dried at 110°C for 24 h in a vacuum oven. The obtained precursor was mixed LiOH with molar ratio of 1:1.05, and calcined at 800°C for 15 h in air to obtain LNCM-3.

EG. The EG synthetic process followed the method described in a previous study². Briefly, graphite foil as a working electrode and Pt foil as a counter electrode were immersed in $0.1 \text{ mol L}^{-1} (\text{NH}_4)_2\text{SO}_4$ aqueous solution. A positive voltage (10 V) was applied between the two electrodes to exfoliate the graphite via the two-electrode cell. When the exfoliation was finished, the product was filtered and washed with deionized water and ethanol to remove chemical residues. Finally, the EG powder was obtained through freeze-drying.

Structural characterizations

The chemical compositions of the prepared powders were analyzed by inductively coupled plasma spectrometry (ICP, 6000 Series, Thermo SCIENTIFIC). The synthesized materials of the crystalline phase were characterized by powder X-ray diffraction (XRD, BRUKERD/MAX 2500V/PC) with Cu K α radiation. The morphologies of all samples were observed by SEM (Philips XL30) and TEM (Hitachi 600). Pore size distribution was carried out by a porosity analyzer (Autosorb iQ Station 2) at 77 K. Tap density was determined using a tap-density tester (ZS-201).

Raman spectra were obtained on a confocal Raman microscope system (LABRAM HR 800 UV, Hor-iba Jobin-Yvon) equipped with a Leica microscope, and an excitation wavelength of 633 nm with 2 cm⁻¹ resolution.

Electrochemical measurements

LNCM-3/EG half cell. The cathode electrode was prepared in a LNCM-3, acetylene black, and poly(vinylidene fluoride) with a weight ratio of 8:1:1 in N-methyl 2-pyrrolidone. The obtained slurry was casted on Al foil and dried in a vacuum at 120 °C with area mass loading of about 3.5 mg cm⁻². The anode electrode was prepared in the same way as the cathode electrode with EG as active materials. The average mass loading of anode electrode is about 2.0 mg cm⁻². The cathode and anode electrodes were assembled in a CR2032 coin-type half cells in a glove box (H₂O and O₂ <0.1 ppm). The electrolyte was 1 M LiPF₆ in ethylene carbonate (EC)/diethyl carbonate (DEC) (1:1 in volume). A glass microfiber filter (934-AH, Whatman, UK) was used as the separator. Pure Li foil was used as both counter and reference electrode. The LNCM-3//Li half cells were tested within 3.0-4.4 V. EG//Li half cells were cycled between 0.01-1.2 V. The galvanostatic charge and discharge (GCD) measurements were carried out on a NEWARE battery test system (CT-3008). The cyclic voltammetry (CV) measurements were carried out on a CHI 660E electrochemical workstation at 0.1 mV s⁻¹.

Li-ion full cell. Full cells were fabricated by coupling the LNCM-3 cathode with EG anode in a CR 2032 coin-type cells, and electrochemical properties were assessed on the battery testing system in the voltage window of 3.0-4.4 V. Specific capacity of full cell was calculated based on the mass of cathode material. Energy density is calculated based on the discharge capacity and midpoint potential of cathode material. In order to match the cathode/anode capacity, there is a slight excess capacity of anode compared with cathode, and the mass ratio of EG to LNCM-3 was about 1:1.5. Prior to assembling full cells, pre-lithiation for EG anode was performed. Typically, the anode electrode is in direct contact with the lithium foil, then, it is immersed in the electrolyte for 0.5 h. The pre-lithiation for the EG anode was performed to activate the

material and stabilize the electrode surface³.

Table S1. ICP results of precursor NCM-3 and lithiated LNCM-3

Material	Li	Ni	Co	Mn
NCM-3	0	0.598	0.198	0.204
LNCM-3	1	0.598	0.198	0.204

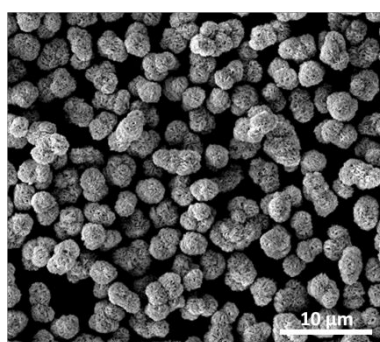


Figure S1. SEM image of the precursor $\text{Ni}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}(\text{OH})_2$ (NCM-3)

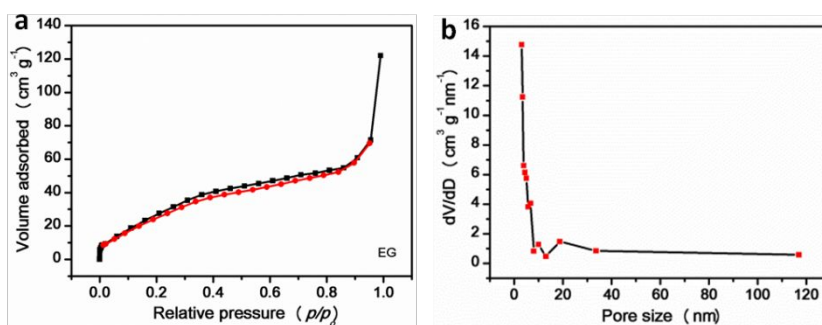


Figure S2. (a) N_2 adsorption-desorption isotherm and (b) the corresponding pore size distribution fitted from the BJH method of the EG.

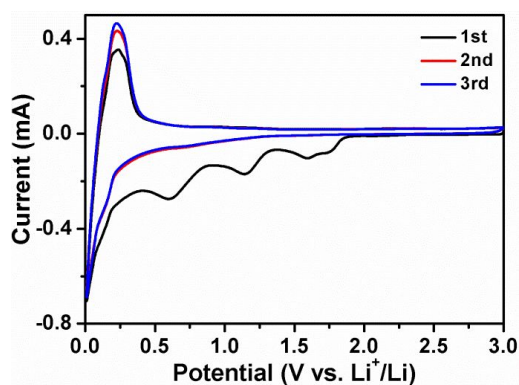


Figure S3. Cyclic voltammetry profiles of the pristine EG at a 0.1 mV s^{-1} scan rate (voltage range: 0.01-3.0 V).

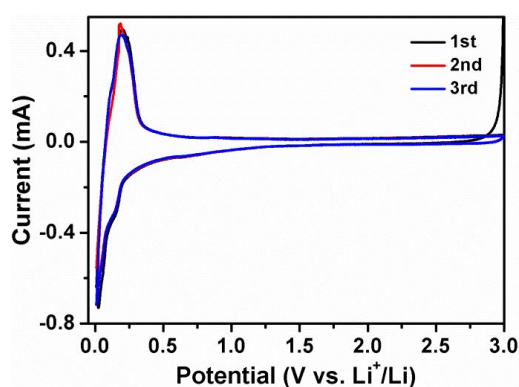


Figure S4. Cyclic voltammetry profiles of the prelithiated EG at a 0.1 mV s^{-1} scan rate (voltage range: 0.01-3.0 V).

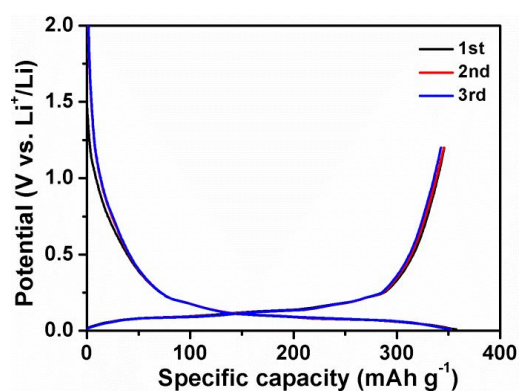


Figure S5. GDC profiles of the prelithiated EG for the first, second, and third cycles at a current density of 0.1 C ($0.1 \text{ C} = 27 \text{ mA g}^{-1}$) between 0.01 and 1.2 V vs. Li^+/Li .

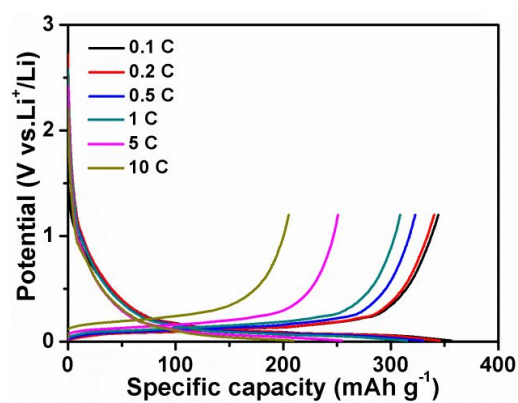


Figure S6. GDC profiles of the prelithiated EG at different current densities between 0.01 and 1.2 V vs. Li^+/Li .

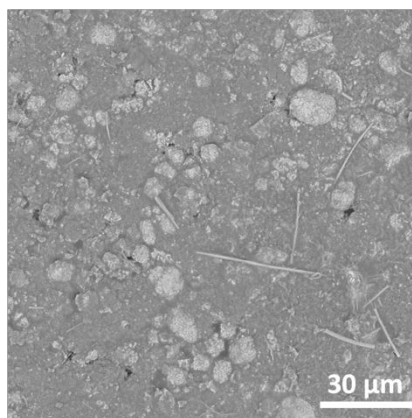


Figure S7. SEM image of the cathode after 300 cycles

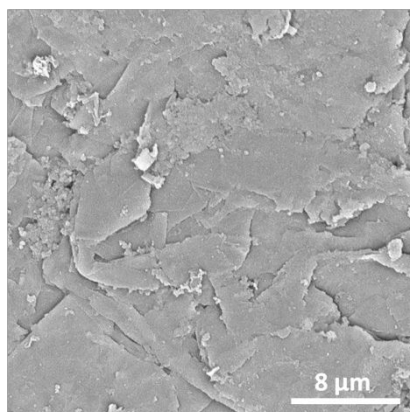


Figure S8. SEM image of the anode after 300 cycles

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