Supplementary Information

High-energy-density lithium-ion battery using carbon-nanotube-Si composite anode and compositionally graded Li[Ni$_{0.85}$Co$_{0.05}$Mn$_{0.10}$]O$_2$ cathode

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Fig. S1 (a) SEM, (b) TEM, and (c) HR-TEM images of nanoporous Si.
Fig. S2 (a), (b) SEM, and (c), (d) HR-TEM images of the CNT-Si anode after ball milling.
Fig. S3 Impedance spectra measured of porous Si, milled Si, and milled Si with CNT.
<table>
<thead>
<tr>
<th>Material</th>
<th>Li/M</th>
<th>Ni/M</th>
<th>Co/M</th>
<th>Mn/M</th>
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<tbody>
<tr>
<td>Precursor TSFCG</td>
<td>0.000</td>
<td>0.844</td>
<td>0.058</td>
<td>0.098</td>
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<tr>
<td>Lithiated TSFCG</td>
<td>1.012</td>
<td>0.849</td>
<td>0.054</td>
<td>0.097</td>
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<tr>
<td>Lithiated NCA</td>
<td>1.051</td>
<td>0.842</td>
<td>0.110</td>
<td>0.048</td>
</tr>
</tbody>
</table>

**Table S1** ICP results of precursor TSFCG precursor $\text{[Ni}_{0.85}\text{Co}_{0.05}\text{Mn}_{0.15}]\text{(OH)}_2$, lithiated TSFCG $\text{Li[Ni}_{0.85}\text{Co}_{0.05}\text{Mn}_{0.15}]\text{O}_2$ and Li[$\text{Ni}_{0.85}\text{Co}_{0.11}\text{Al}_{0.04}]\text{O}_2$. 
Fig. S4 EPMA line scan of the integrated atomic ratio of transition metals as a function of the distance from the particle center to the surface of TSFCG Li[Ni_{0.85}Co_{0.05}Mn_{0.10}]O_2.
**Fig. S5** Before and after 100 cycled electrodes XRD patterns of (a) TSFCG Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2 and (b) Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2. (c) Variation of the lattice parameters (a and c) and volume of TSFCG and NCA electrodes about before and after cycled.
**Fig. S6** (a) 100 zone HR-TEM image of the TSFCG Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2 after 100 cycles in a half cell with Li counter electrode including a Fourier filtered image of the marked region, (b) HR-TEM images of Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2 after 100 cycles (Insets in (b) show local Fourier transformed images of the marked regions to illustrate the presence of the NiO-like cubic phase on the surface of the cycled Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2 cathode).
Fig. S7 Amount of dissolved Ni, Co, and Mn present after charging the TSFG Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2 and Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2 to 4.3V versus Li^+. (a) before and (b) after 100 cycled.
**Fig. S8** (a),(b) Voltage profiles and cycling performances of CNT-Si anode in EC:DEC = 1:1 with FEC 10 wt.% electrolyte. (c),(d) Pre-lithiation voltage profiles and cycling performances of CNT-Si anode in EC:EMC = 3:7 with VC 2 vol.% electrolyte. CNT-Si half cells were operated within voltage range of 0.01 – 1.5 V at current density of 400 mA g\(^{-1}\). (e),(f) Electrochemical performance of TSFCG Li\([\text{Ni}_{0.85}\text{Co}_{0.05}\text{Mn}_{0.15}]\)O\(_2\) cathode in different electrolyte within voltage range of 2.7 – 4.3 V at 0.1 C-rate.
Schematic diagram of the pouch cell used for the weight fraction calculation.

**Fig. S9** Mass fraction of individual components used in the pouch full-cell test.
Fig. S10 EPMA line scan of the integrated atomic ratio of transition metals as a function of the distance from the particle center to the surface for the TSFCG Li[Ni$_{0.85}$Co$_{0.05}$Mn$_{0.15}$]O$_2$ cathode: pristine and after 500 cycles.