Electronic Supplementary Material (ESI) for Energy & Environmental Science.

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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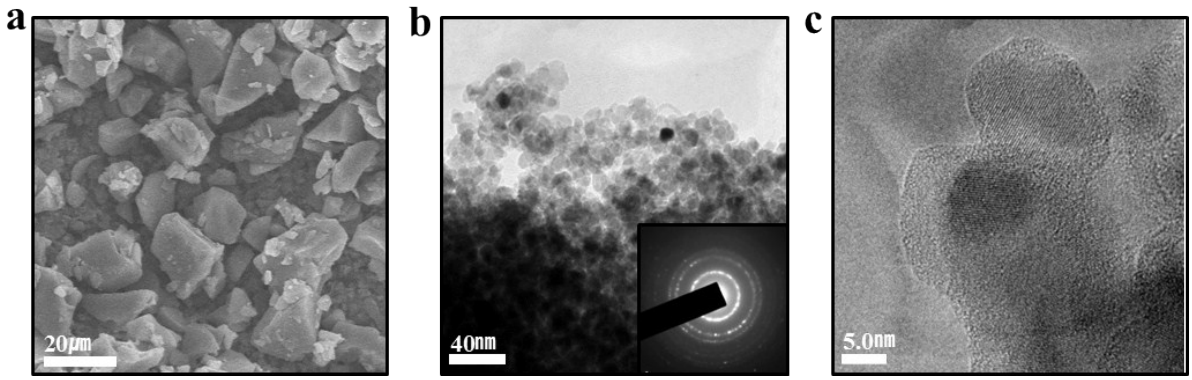


Fig. S1 (a) SEM, (b) TEM, and (c) HR-TEM images of nanoporous Si.

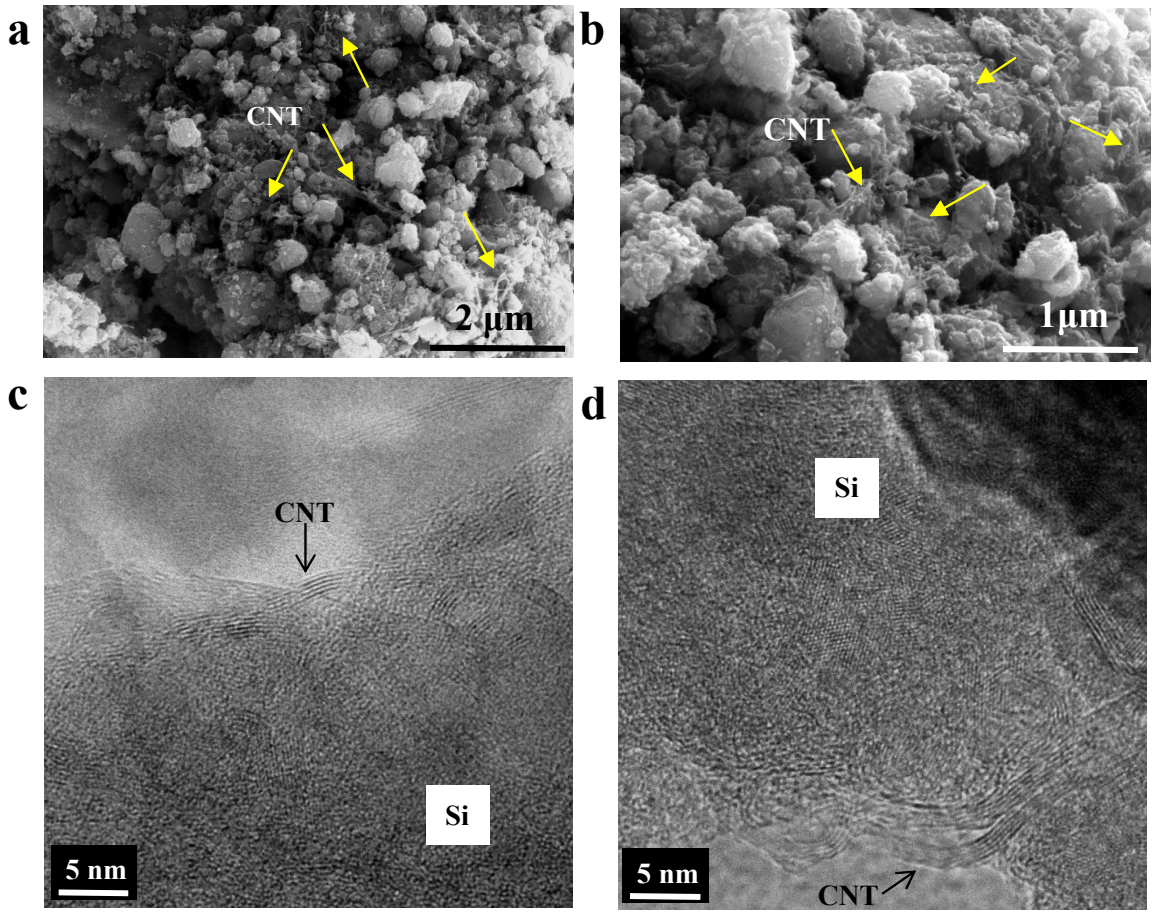


Fig. S2 (a), (b) SEM, and (c), (d) HR-TEM images of the CNT-Si anode after ball milling.

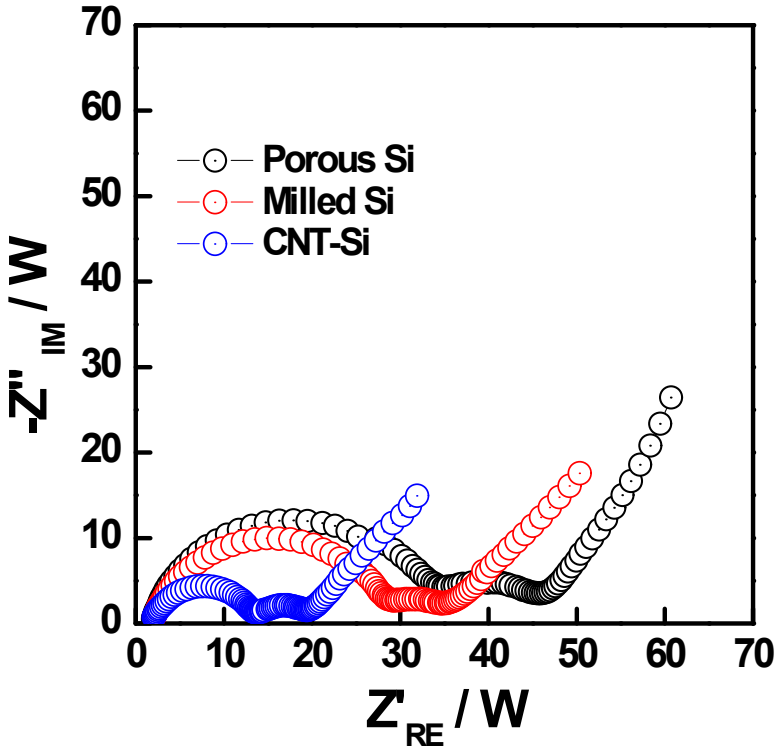


Fig. S3 Impedance spectra measured of porous Si, milled Si, and milled Si with CNT.

Material	Li/M	Ni/M	Co/M	Mn/M
Precursor TSFCG	**0.000**	**0.844**	**0.058**	**0.098**
Lithiated TSFCG	**1.012**	**0.849**	**0.054**	**0.097**
Lithiated NCA	**1.051**	**0.842**	**0.110**	**0.048**

Table S1 ICP results of precursor TSFCG precursor $[Ni_{0.85}Co_{0.05}Mn_{0.15}](OH)_2$, lithiated TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2$ and $Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2$.

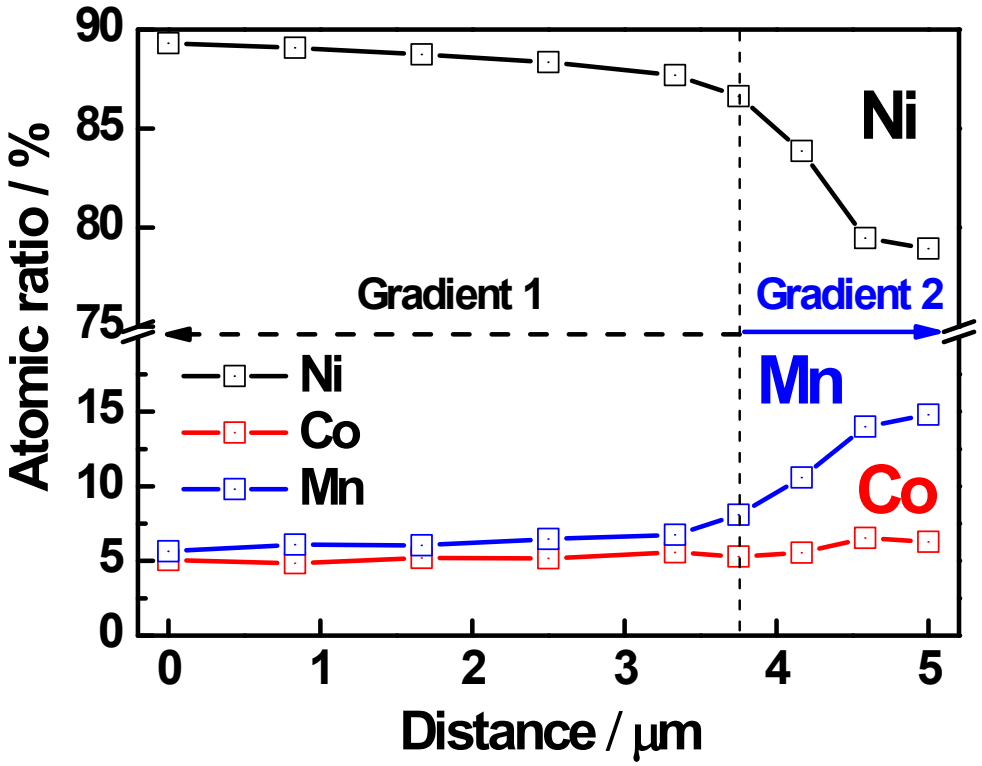


Fig. S4 EPMA line scan of the integrated atomic ratio of transition metals as a function of the distance from the particle center to the surface of TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.10}]O_2$.

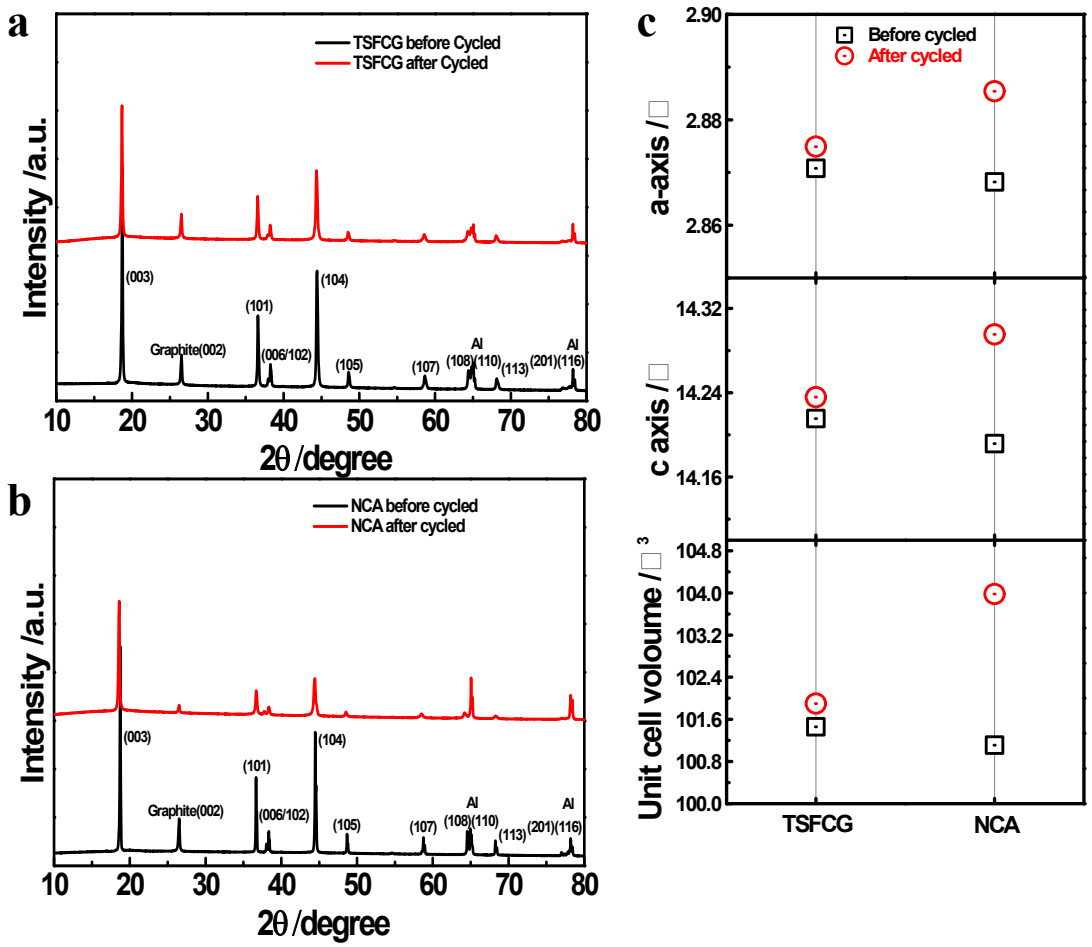


Fig. S5 Before and after 100 cycled electrodes XRD patterns of (a) TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2$ and (b) $Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2$. (c) Variation of the lattice parameters (a and c) and volume of TSFCG and NCA electrodes about before and after cycled.

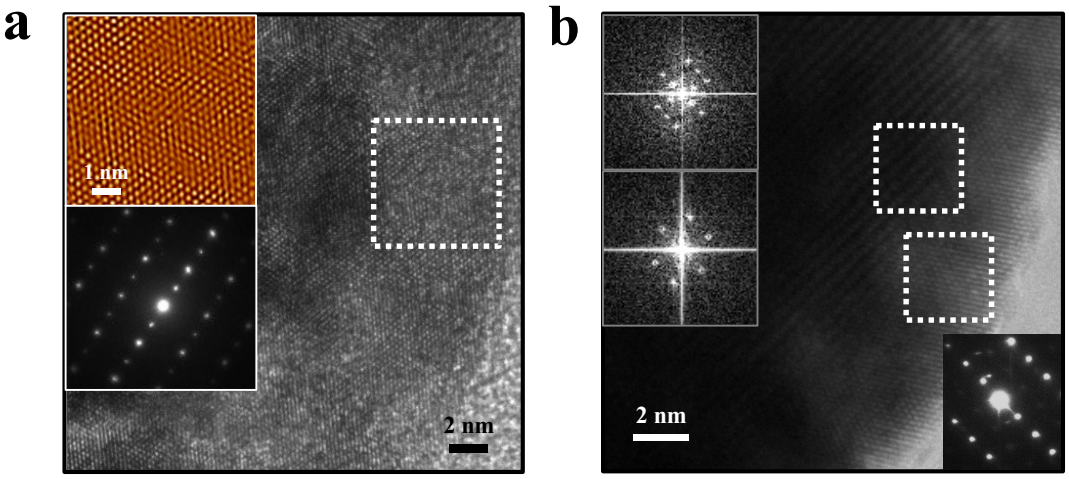


Fig. S6 (a) 100 zone HR-TEM image of the TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2$ after 100 cycles in a half cell with Li counter electrode including a Fourier filtered image of the marked region, (b) HR-TEM images of $Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2$ after 100 cycles (Insets in (b) show local Fourier 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) .

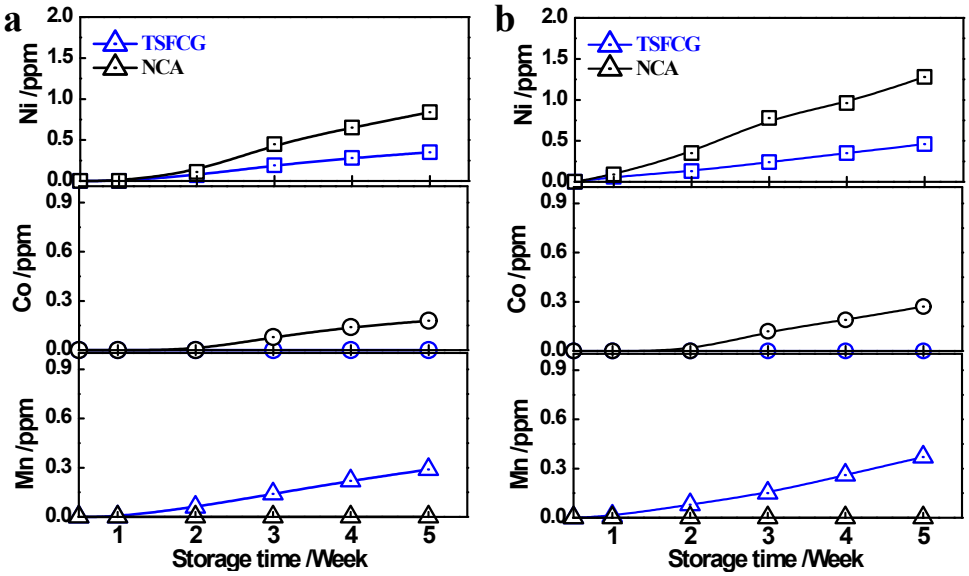


Fig. S7 Amount of dissolved Ni, Co, and Mn present after charging the TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2$ and $Li[Ni_{0.85}Co_{0.11}Al_{0.04}]O_2$ to 4.3V versus Li^+. (**a)** before and (**b)** after 100 cycled.

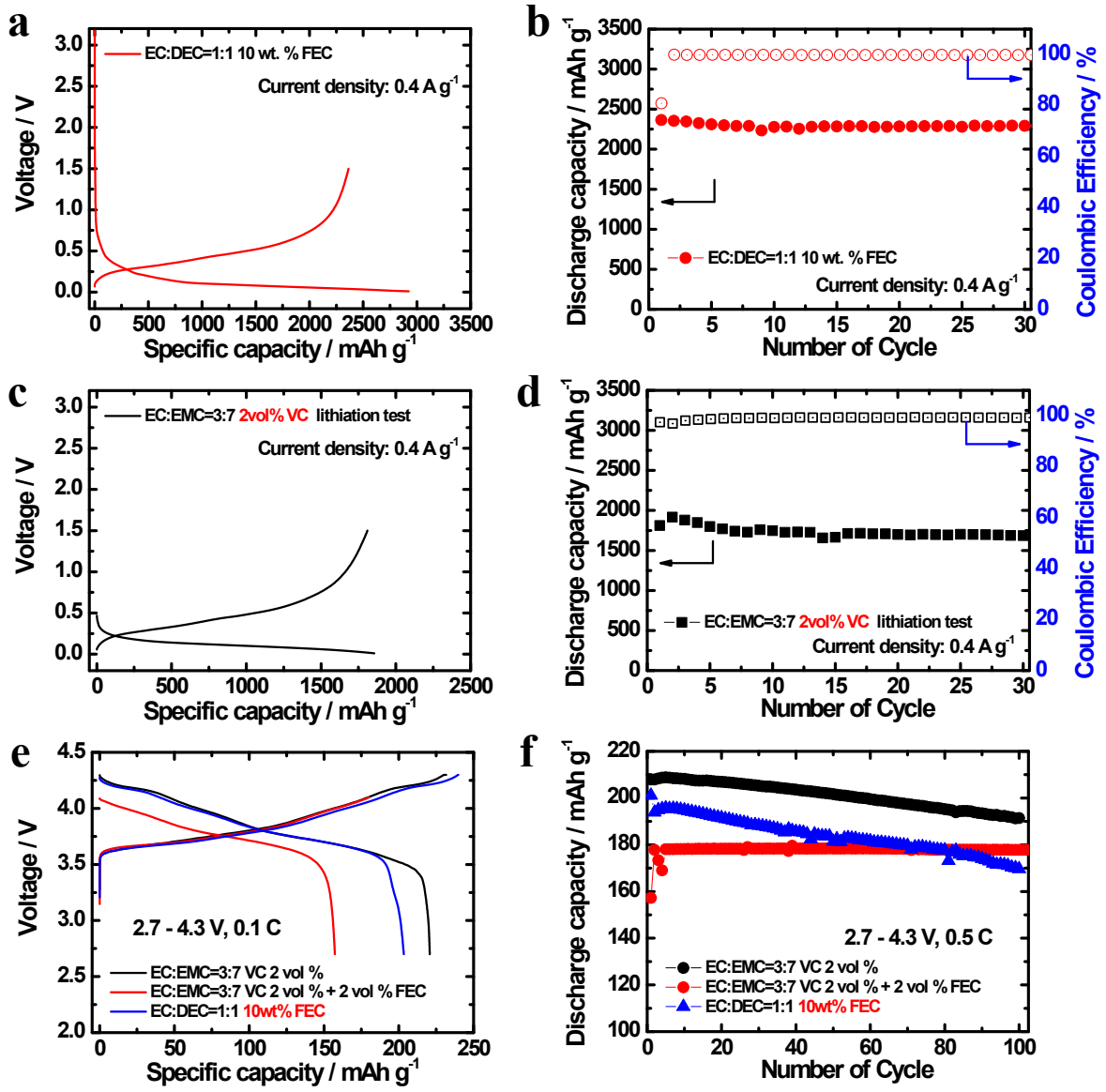
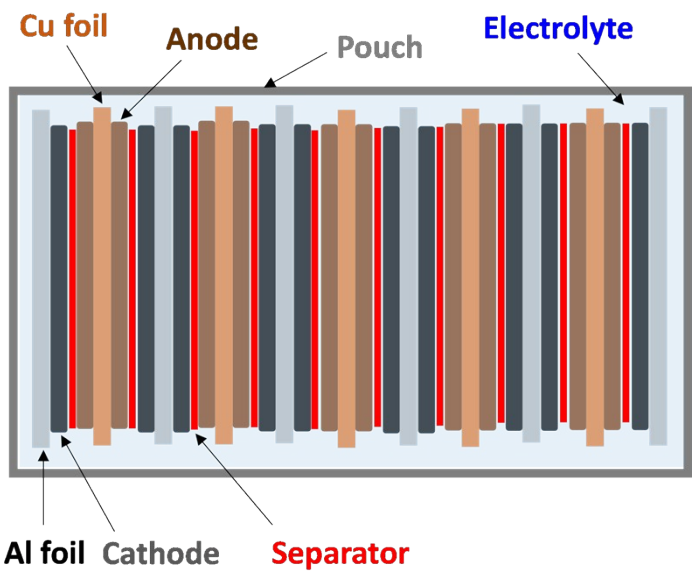
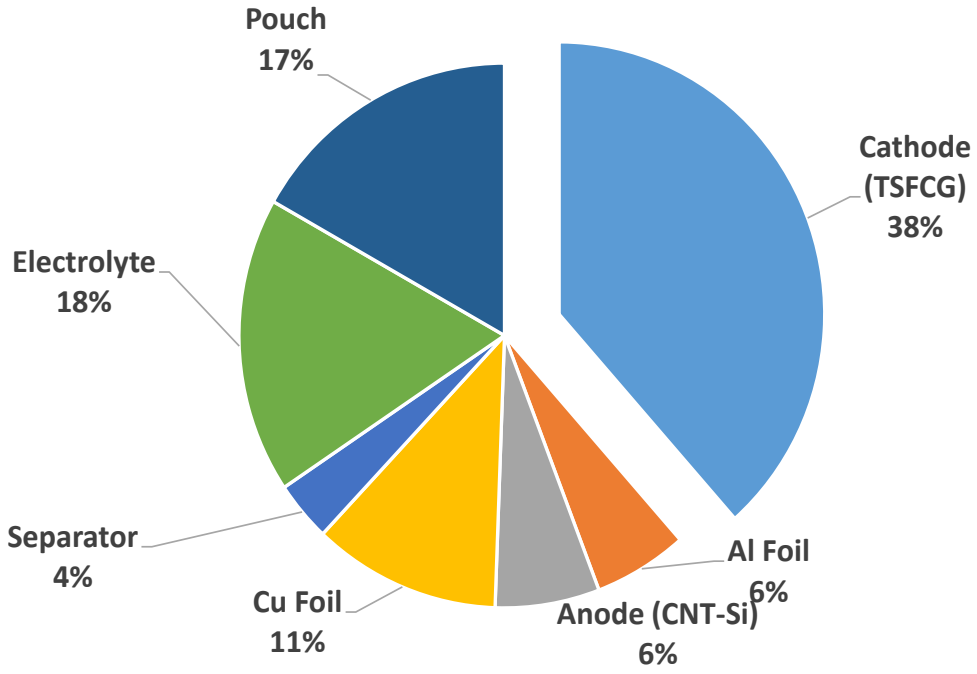


Fig. S8 (a),(b) Voltage profiles and cycling performances of CNT-Si anode in EC:DEC = 1 : 1 with FEC 10 wt.% electrolyte. (c),(d) Pre-lithiation voltage profiles and cycling performances of CNT-Si anode in EC:EMC = 3 : 7 with VC 2 vol.% electrolyte. CNT-Si half cells were operated within voltage range of 0.01 – 1.5 V at current density of 400 mA g^{-1}. (e),(f) Electrochemical performance of TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2$ cathode in different electrolyte within voltage range of 2.7 – 4.3 V at 0.1 C-rate.



Calculation component	
Stack	5
Electrode Area (3cm x 5cm)	15 cm^2
Cathode	11.68 mg cm^{-2}
Anode	1.3 mg cm^{-2}
Al Foil (12 μm)	6 ea
Cu Foil (8 μm)	5 ea
Electrolyte	0.9 g
Separator	0.183 g
Pouch	0.86 g

Schematic diagram of the pouch cell used for the weight fraction calculation.



Component	Cathode	Al Foil	Anode	Cu Foil	Separator	Electrolyte	Pouch	Total
Weight (g)	**1.968**	**0.292**	**0.325**	**0.585**	**0.183**	**0.9**	**0.86**	**5.114**
Mass fraction (%)	**38.5**	**5.7**	**6.4**	**11.4**	**3.6**	**17.6**	**16.8**	**100**

Fig. S9 Mass fraction of individual components used in the pouch full-ell test.

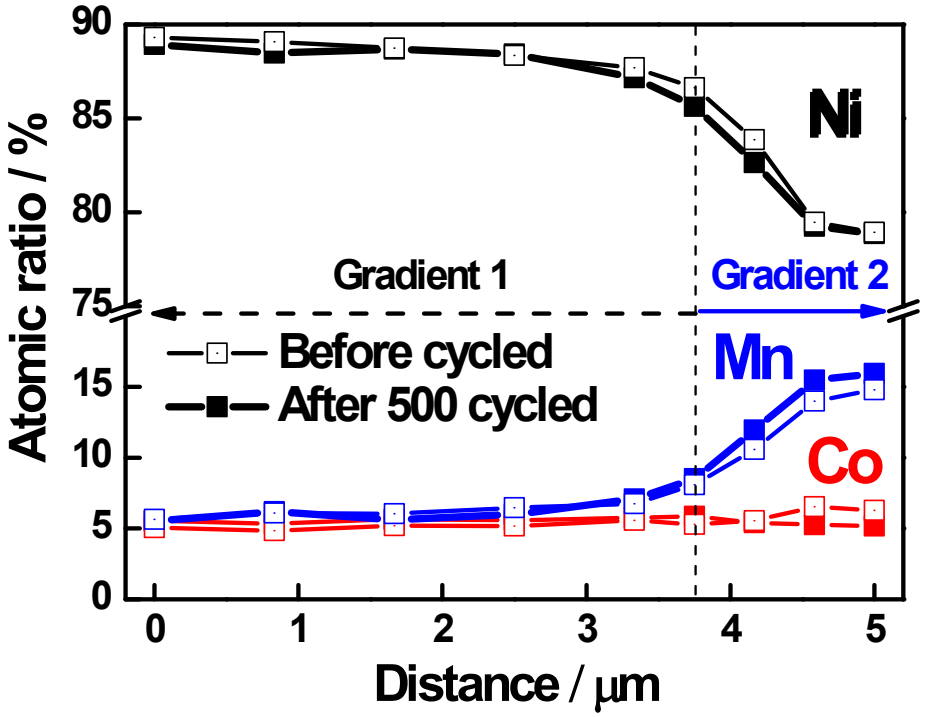


Fig. S10 EPMA line scan of the integrated atomic ratio of transition metals as a function of the distance from the particle center to the surface for the TSFCG $Li[Ni_{0.85}Co_{0.05}Mn_{0.15}]O_2$ cathode: pristine and after 500 cycles.