Supplementary information

Thermal processing to modulate surface chemistry and bulk charge distribution in nickel-rich layered lithium positive electrodes

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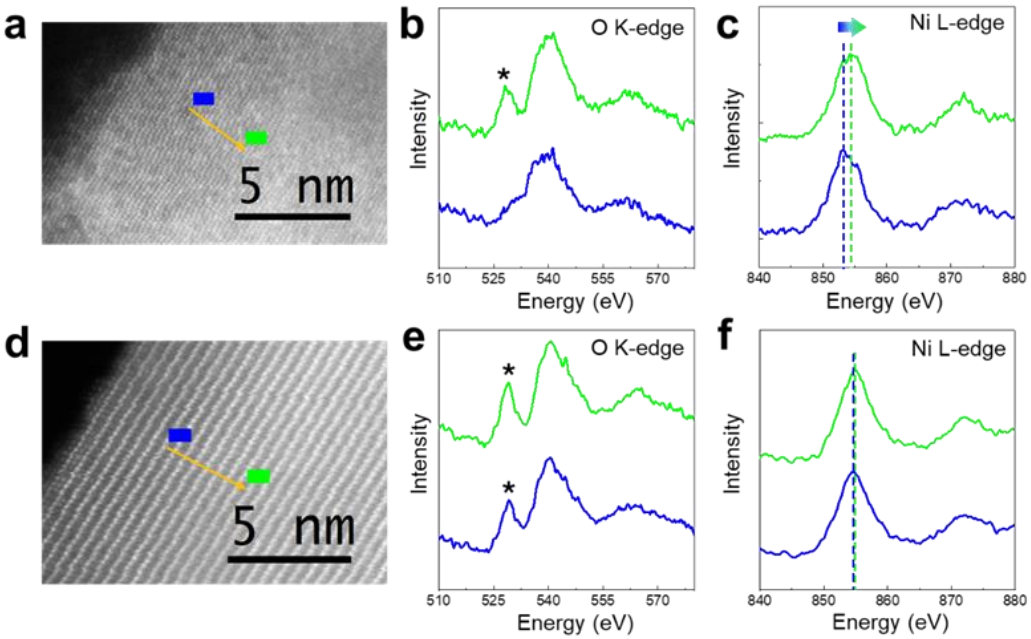
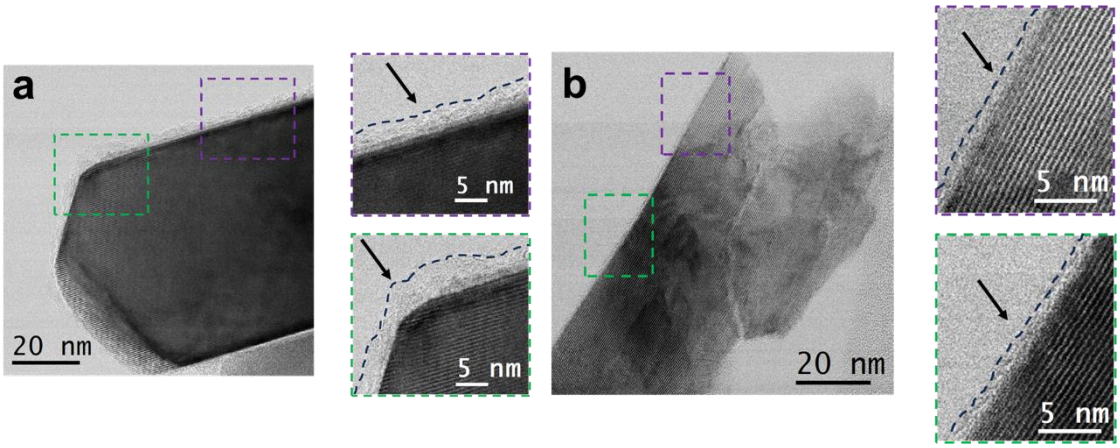
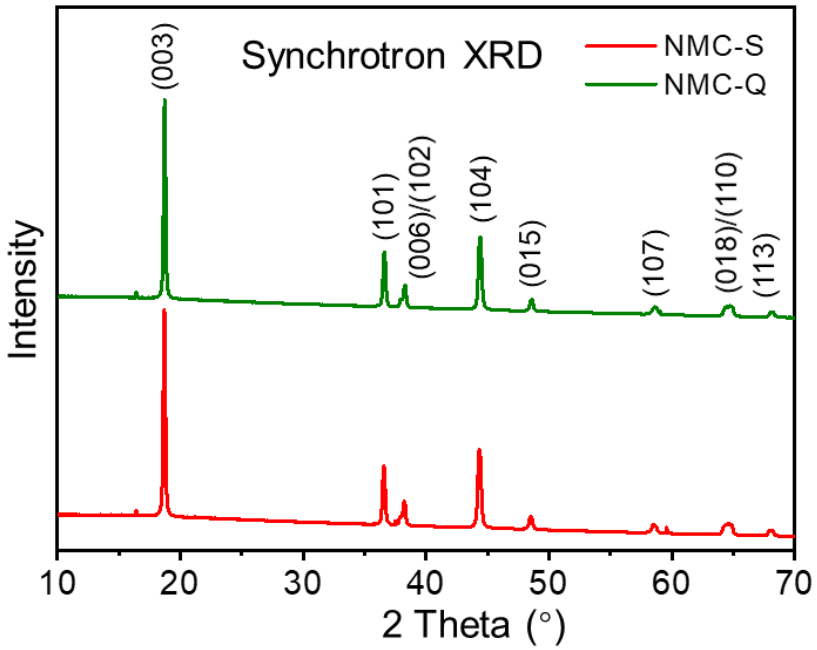


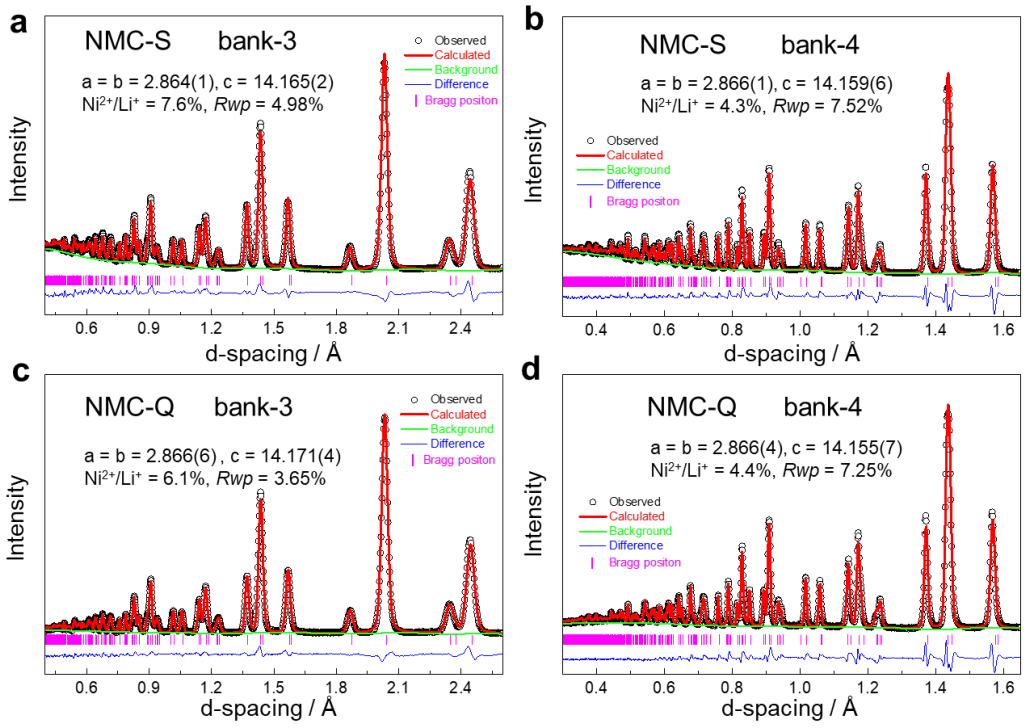
Supplementary Figure 1. STEM images to show the selected regions for EELS acquisition for (a) NMC-Q and (d) NMC-S. EELS O K-edge spectra (b,e) and Ni L-edge spectra (c,f) for NMC-Q and NMC-S corresponding to the regions in a and d.



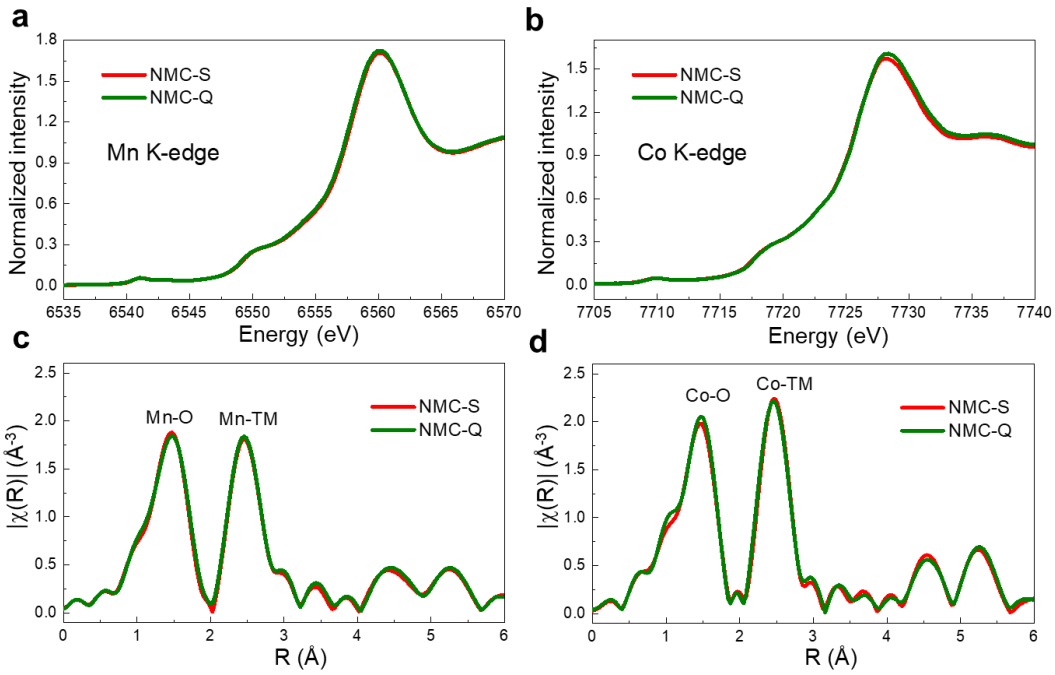
Supplementary Figure 2. BF-STEM images of (a) NMC-Q and (b) NMC-S with magnified regions.



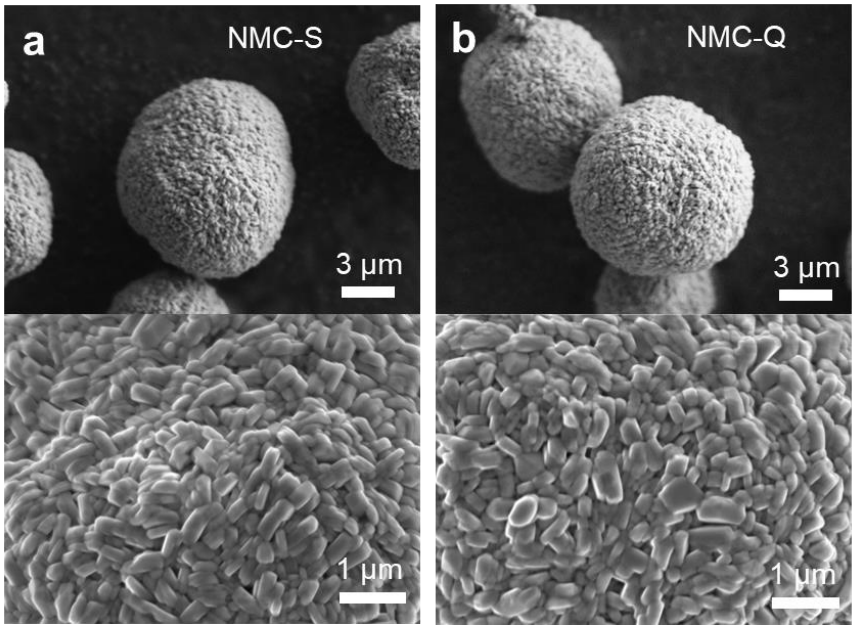
Supplementary Figure 3. Synchrotron XRD for NMC-S and NMC-Q powder samples (calibrated to X-ray wavelength of 1.5406 Å).



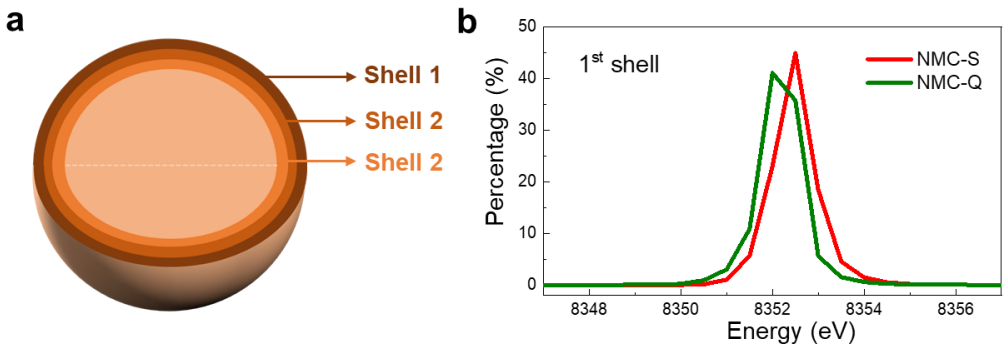
Supplementary Figure 4. Neutron patterns and refinement for NMC-S and NMC-Q powder in different banks. Bank 3 for (a) NMC-S and (c) NMC-Q; bank 4 for (b) NMC-S and (d) NMC-Q.



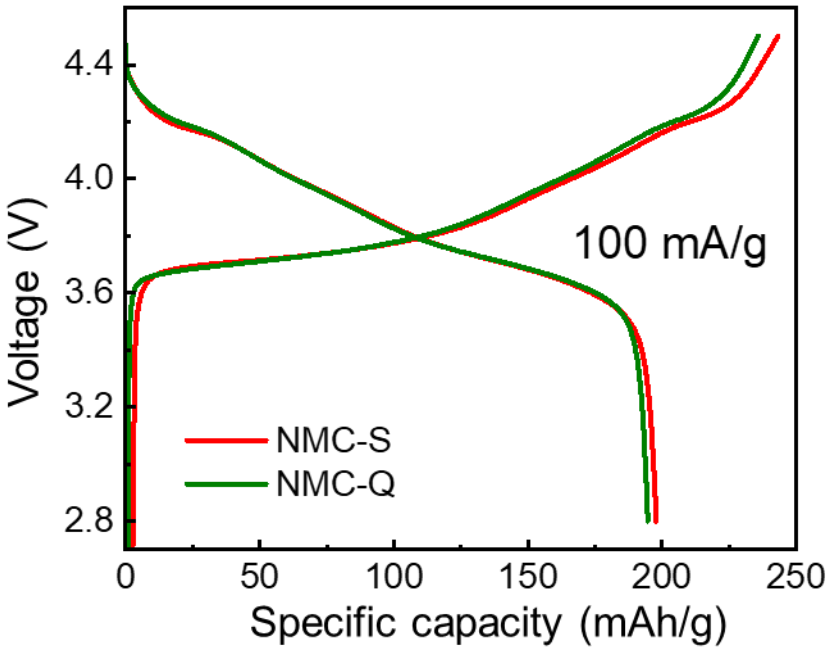
Supplementary Figure 5. Hard XAS XANES spectra for (a) Mn K-edge and (b) Co K-edge. EXAFS spectra of (c) Mn K-edge and (d) Co K-edge for NMC-S and NMC-Q powders.



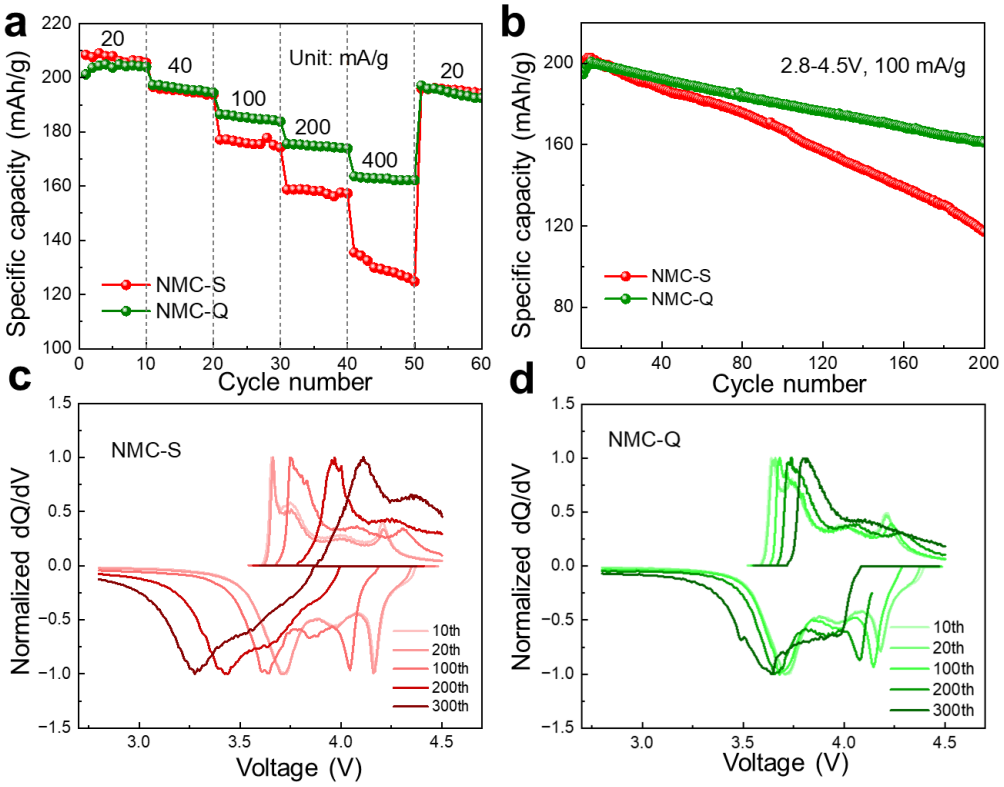
Supplementary Figure 6. SEM images for (a) NMC-S and (b) NMC-Q powders in various magnification.



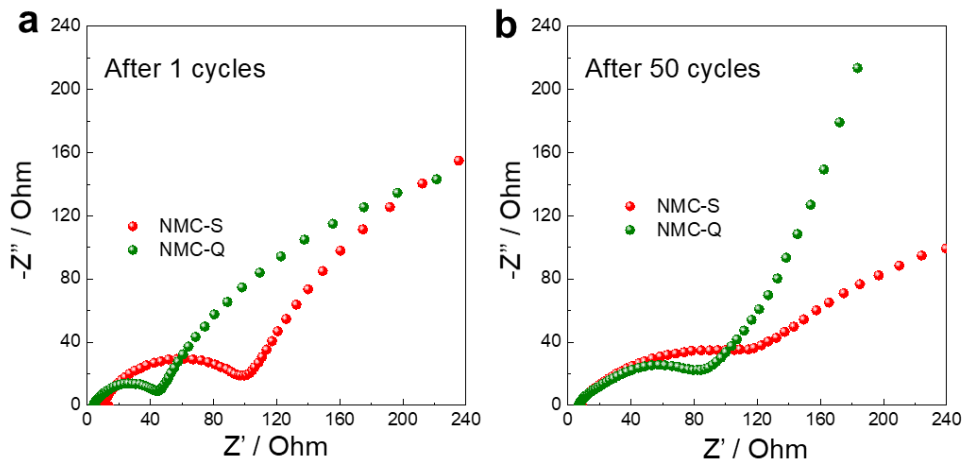
Supplementary Figure 7. (a) Schematic illustration of TXM depth profiling analysis. Each shell is 20 nm away from its outer shell. (b) The first shell (20 nm away from particle surface) Ni K-edge energy distribution of NMC-S and NMC-Q at the pristine state.



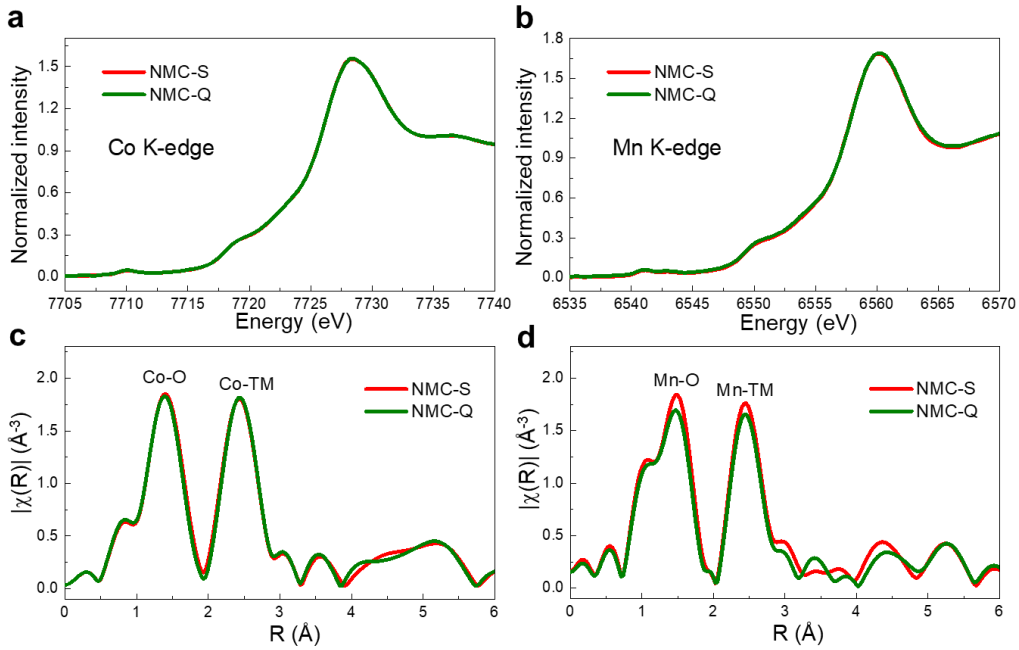
Supplementary Figure 8. The charge/discharge profiles of NMC-Q and NMC-S for the first cycle at a specific current of 100 mA/g with a voltage range of 2.8-4.5 V at 22 °C.



Supplementary Figure 9. (a) Rate capability at different specific current, and (b) cycling performance at a specific current of 100 mA/g of NMC-S and NMC-Q positive electrodes in Li metal cells with a voltage range 2.8-4.5 V at 22 °C. dQ/dV curves of 10th, 20th, 100th, 200th and 300th cycle at a specific current of 100 mA/g for (c) NMC-S and (d) NMC-Q.



Supplementary Figure 10. Nyquist plot of EIS performed with fully discharged cells at a specific current of 100 mA/g with a voltage range of 2.8-4.5 V at 22 °C after (a) 1 cycle and (b) 50 cycles of NMC-S and NMC-Q positive electrodes.



Supplementary Figure 11. XANES spectra of (a) Co K-edge and (b) Mn K-edge for NMC-S and NMC-Q after 100 cycles in the discharge state at a specific current of 200 mA/g with a voltage range of 2.8-4.5 V at 22 °C. EXAFS spectra of (c) Co K-edge and (d) Mn K-edge for NMC-S and NMC-Q after 100 cycles.

Supplementary Table 1. Neutron diffraction Rietveld refinement results for NCM-S and NCM-Q.

Samples		a / Å	b / Å	c / Å	Ni^{2+}/Li^{+} / %	R_{wp} / %
	bank-2	2.868(9)	2.868(9)	14.152(4)	4.6	5.54
NCM-S	bank-3	2.864(1)	2.864(1)	14.165(2)	7.6	4.98
	bank-4	2.866(1)	2.866(1)	14.159(6)	4.3	7.52
	bank 2	2.870(0)	2.870(0)	14.150(3)	3.8	4.64
NCM-Q	bank-3	2.866(6)	2.866(6)	14.171(4)	6.1	3.65
	bank-4	2.866(4)	2.866(4)	14.155(7)	4.4	7.25