Supplementary Information

Boosting Stability in Ni-Rich Cathodes: A Synergistic Approach to Surface and Bulk Modifications for Advanced Lithium-Ion Batteries

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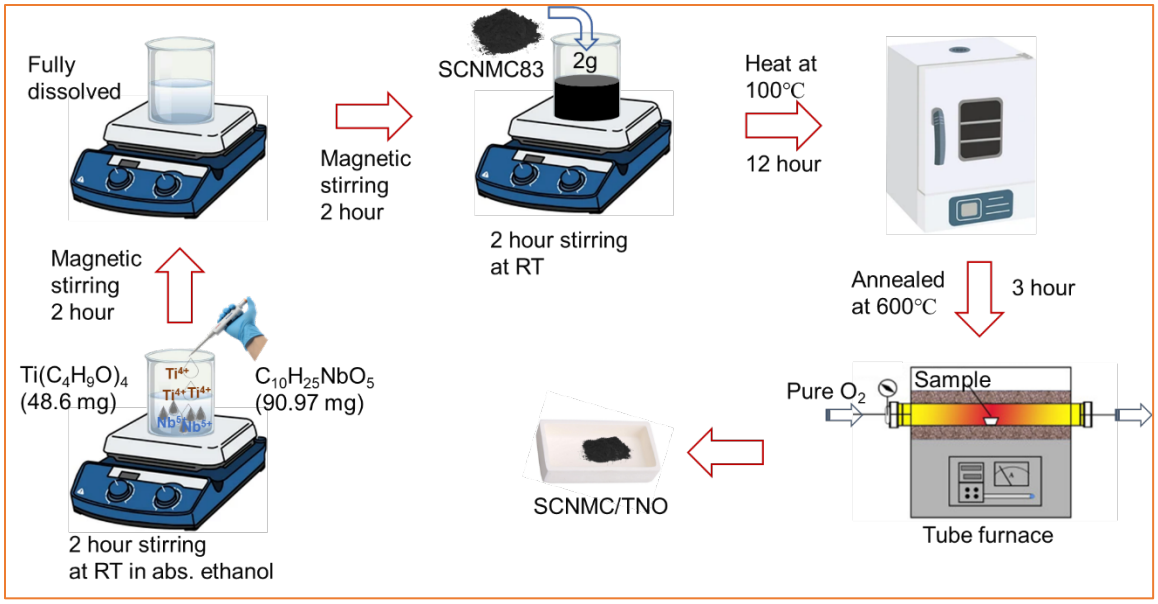

Figure S1. Experimental procedure to prepare SCNMC/TNO powder.

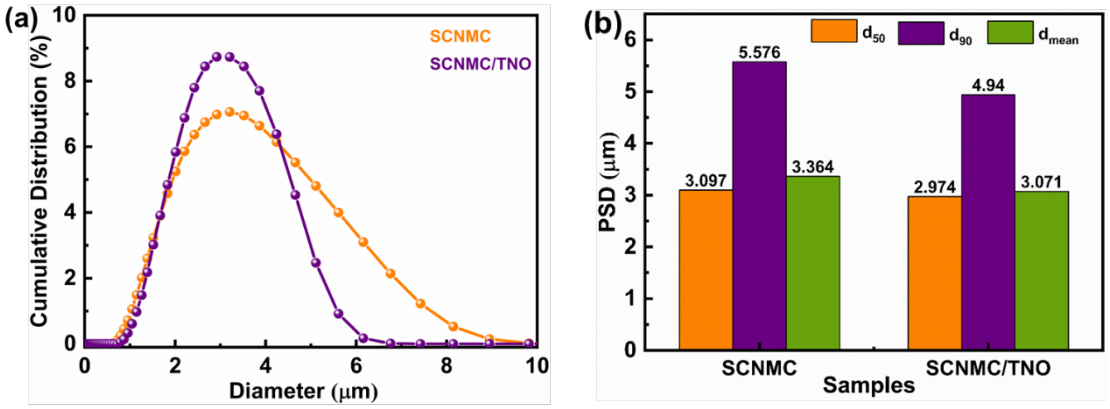

Figure S2. (a) Particle size distribution for SCNMC and SCNMC/TNO by Dynamic Light Scattering (DLS), (b) d_{50}, d_{90}, and d_{mean} values (μm).

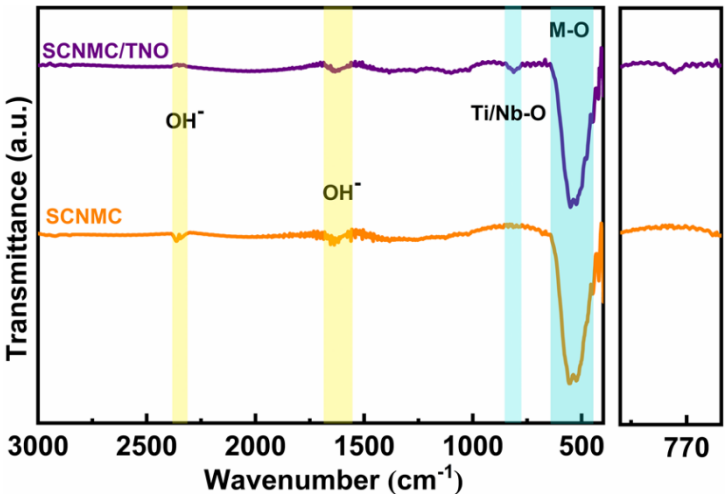

Figure S3. FTIR spectra for SCNMC and SCNMC/TNO.

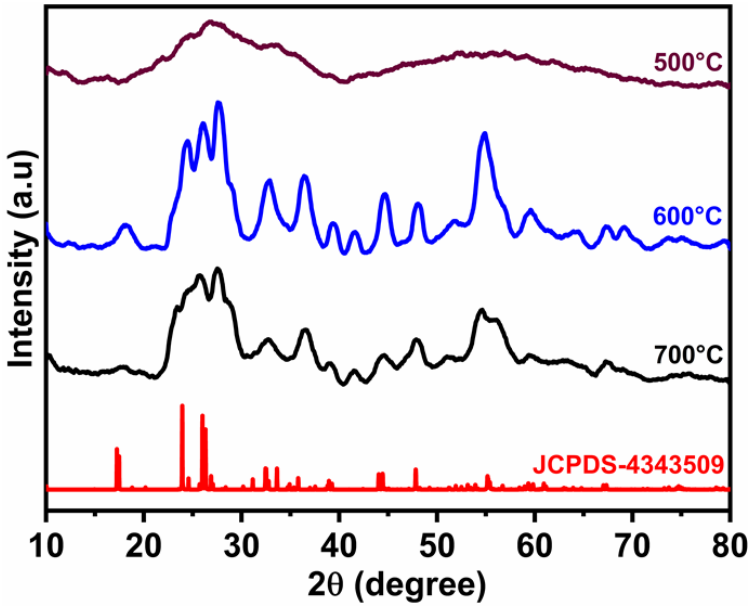

Figure S4. XRD patterns of synthesized pure $TiNb_2O_7$ at 500 ℃, 600 ℃, and 700 ℃.

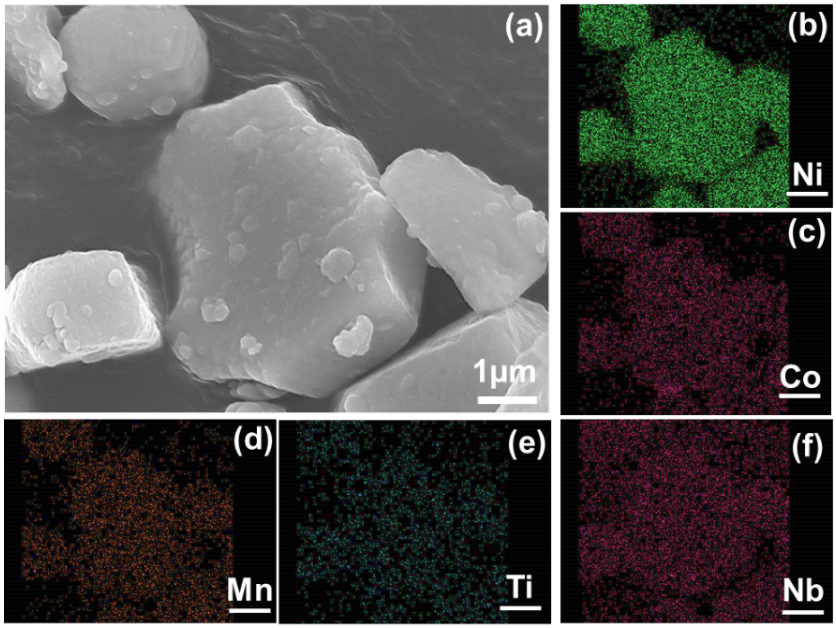

Figure S5. (a) Single particle of SCNMC/TNO, SEM-EDS mapping of (a) Ni, (c) Co, (d) Mn, (e) Ti, and (f) Nb.

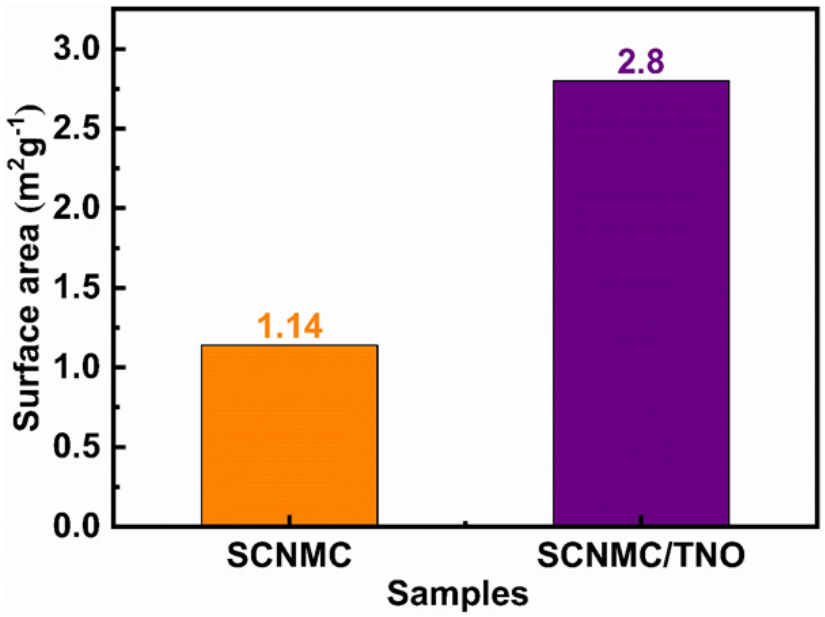

Figure S6. Specific surface area value for SCNMC and SCNMC/TNO.

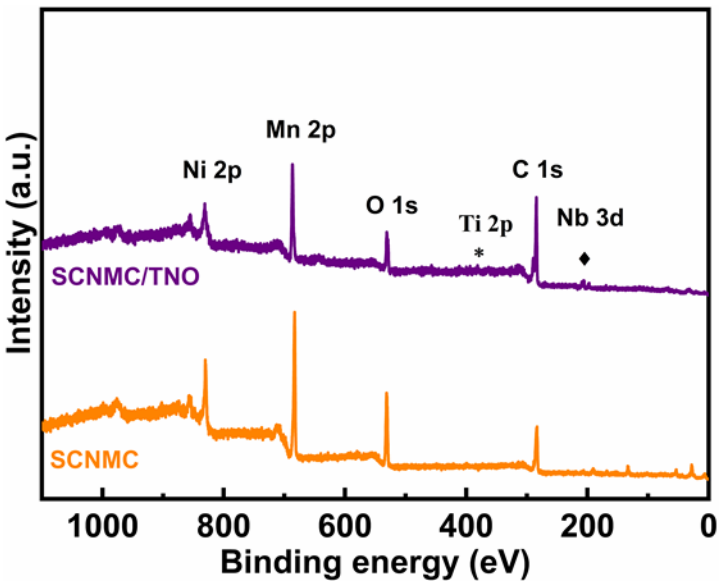

Figure S7. XPS survey spectra of SCNMC and SCNMC/TNO

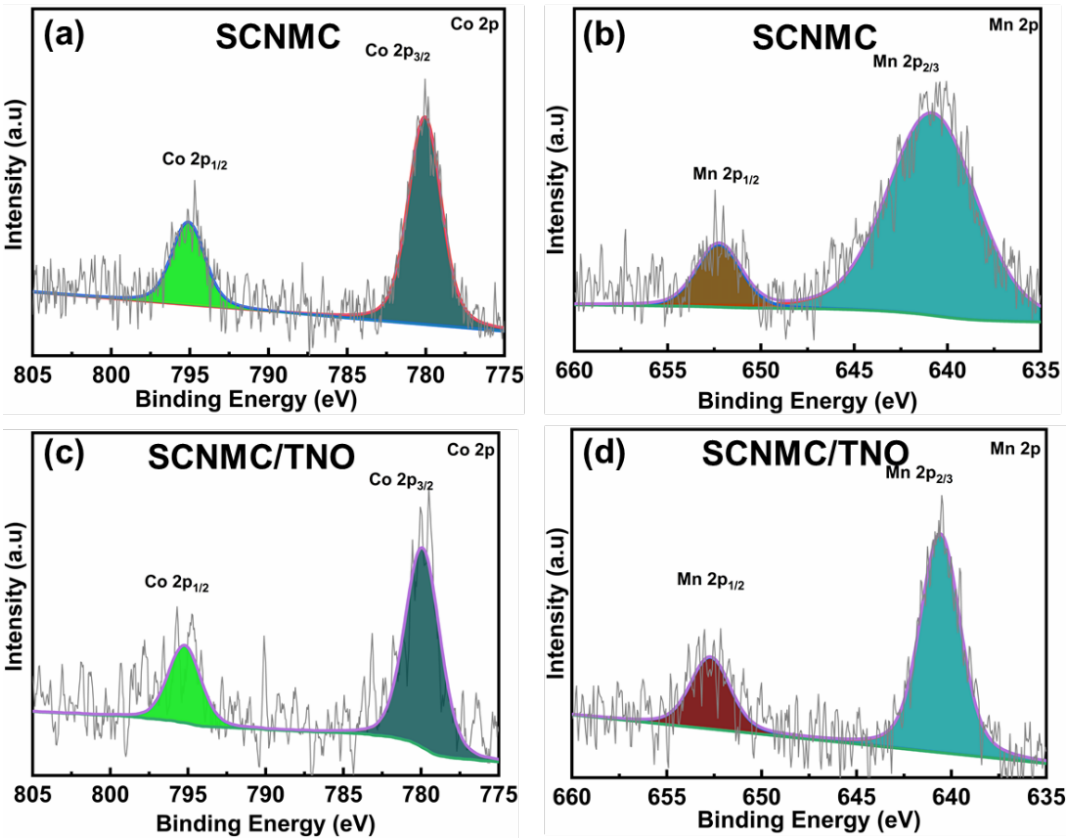

Figure S8. (a) and (c) shows Co 2p spectra with fitted components for SCNMC and SCNMC/TNO, (b) and (d) shows Mn 2p spectra with fitted components for SCNMC and SCNMC/TNO samples.

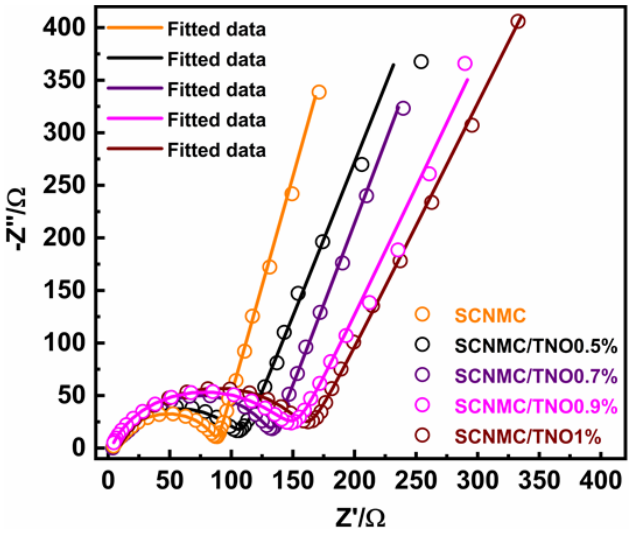

Figure S9. EIS profile of various mole % of TNO coated SCNMC

Table S1 Corresponding EIS fitting data from various mole % of coated SCNMC

Samples	**R_b**	**R_{ct}**
SCNMC	3.92	83.98
SCNMC/TNO0.5%	4.015	103.73
SCNMC/TNO0.7%	3.84	123.47
SCNMC/TNO0.9%	3.9	146.79
SCNMC/TNO1%	3.51	160.09

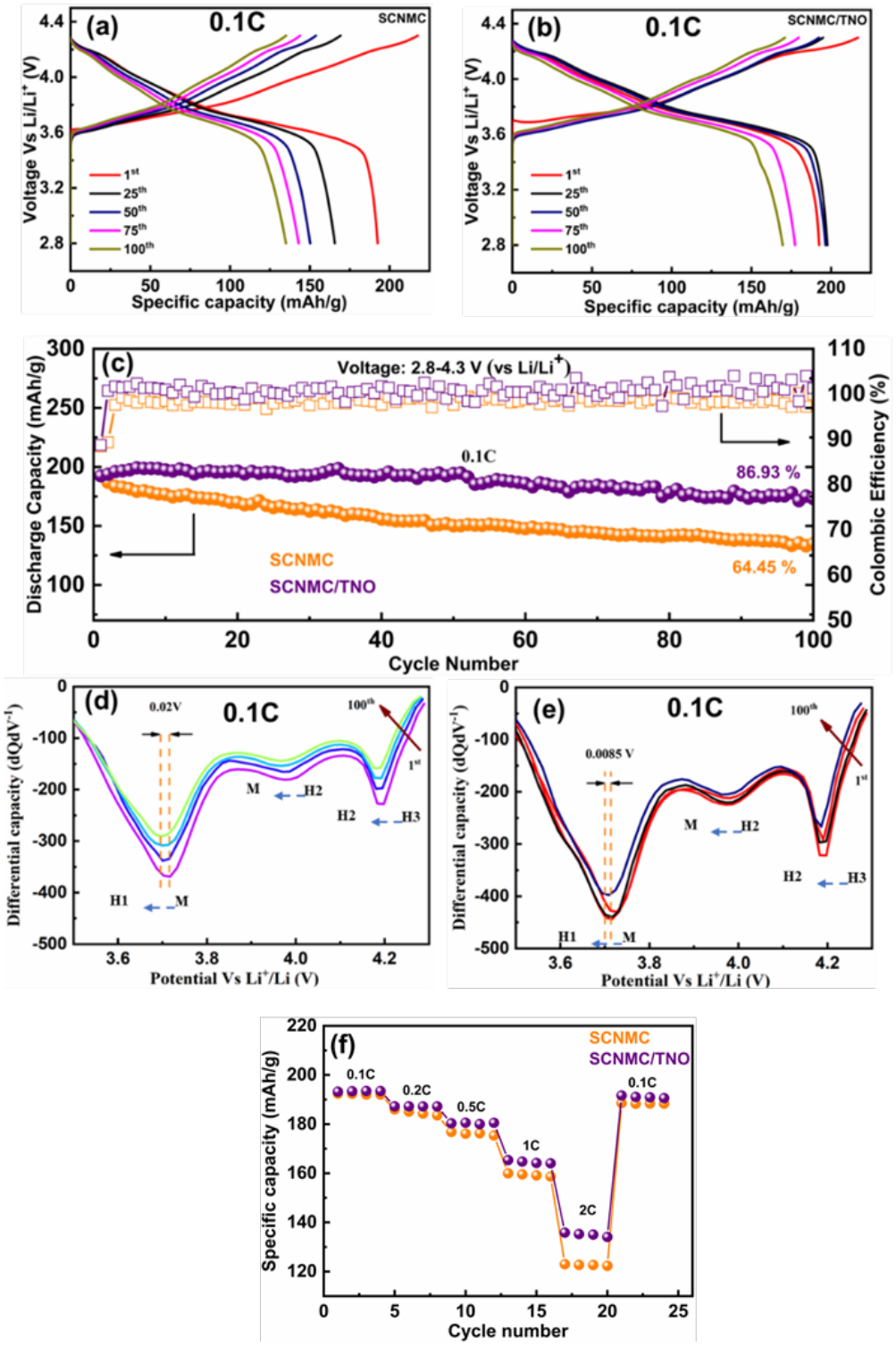

Figure S10. Li//NMC charge-discharge profiles with 1M $LiPF_6$ in EC: EMC (3:7, v:v) from 2.8 - 4.3 V at 0.1 C for (a) SCNMC, (b) SCNMC/TNO, c) corresponding discharge capacity vs coulombic efficiency plots; dQ/dV curves derived from the 1st to 100th cycle curve at 0.1 C for (d) SCNMC and (e) SCNMC/TNO, respectively, (f) rate capability.

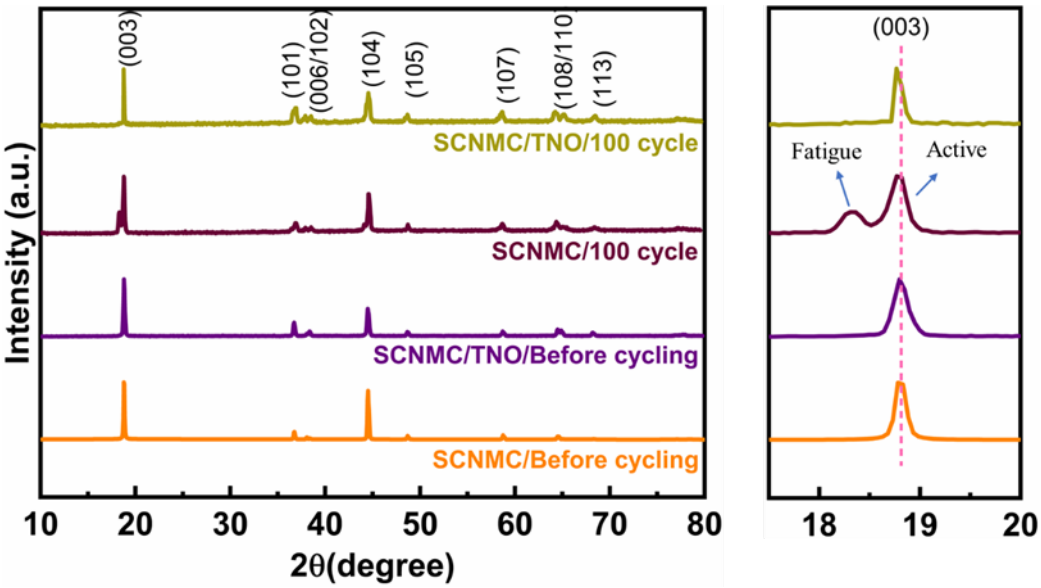

Figure S11. XRD of SCNMC and SCNMC/TNO after 100 cycles and zoom view of (003) indexed peak.

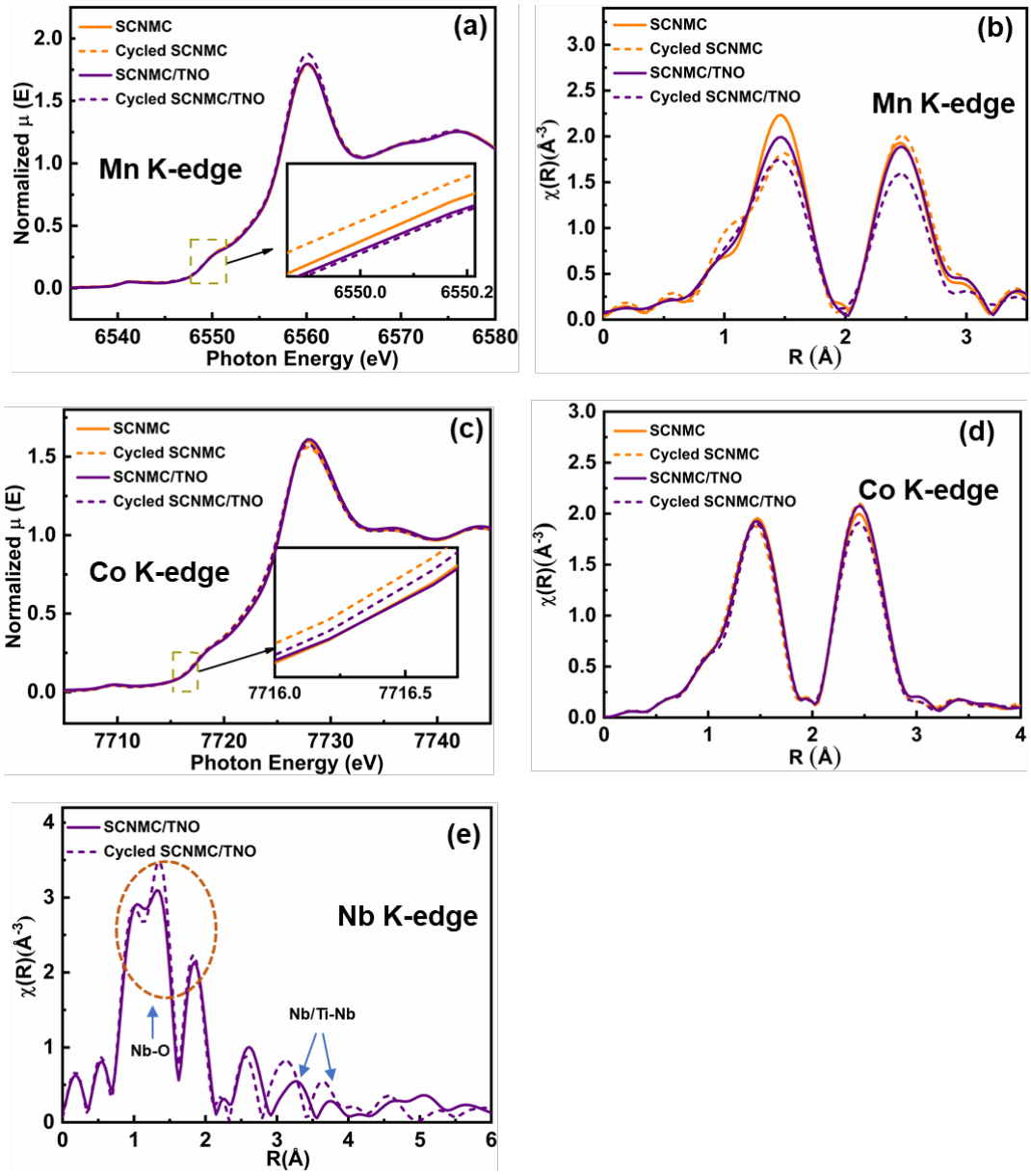

Figure S12. XANES spectra of SCNMC and SCNMC/TNO before and after 100 cycles for (a) Mn, and (c) Co; FT-EXAFS spectra for (b) Mn, (d) Co, and (e) Nb- K edge.

Table S2. Overview of performance metrics for Nickel-rich cathodes reported in literatures.

Coating/doping compound	Electrolyte	Cycles	C-rate	Capacity retention (%)	Improvement due to modification (%)	Ref.
Pristine/ 4-CPBA/SCNMC	EC:EMC:DMC (1:1:1)	200	0.5C	61/ 82.5	21.5	[1]
Pristine/ SiO_2/NMC811	EC:EMC:DMC (1:1:1)	100	0.5C	73.4/ 87.3	13.9	[2]
Pristine/ MgO/NMC811	EC:EMC:DMC (1:1:1)	100	1C	74.5/ 90.1	15.6	[3]
Pristine/ ZrO2/NMC82	EC:DEC (1:1)	100	0.3C	64.2/ 89.4	25.2	[4]
Pristine/ LTZO/NMC88	EC:EMC:DMC (1:1:1)	150	1C	87.2/ 92.64	5.44	[5]
Pristine/PVP-PANI/NMC8	EC:EMC:DMC (1:1:1)	100	1C	66.3/ 88.2	21.9	[6]
TNO/SCNMC (single-crystalline NMC83)	EC:EMC(3:7)	100	1C	58.55/ 81.92	23.37	This work

Table S3. Rietveld refinement data of Powder X-ray diffraction for (a) SCNMC and (b) SCNMC/TNO.

(a) SCNMC						**(b) SCNMC/TNO**					
a = 2.8614 Å, c = 14.2431 Å, V = 100.997 Å^3, c/a = 4.977, Rwp = 4.27%						a = 2.8621 Å, c = 14.2489 Å, V = 101.086 Å^3, c/a = 4.978, Rwp = 4%					
Elements	**Site**	**x**	**y**	**z**	**Occupancy**	**Elements**	**Site**	**x**	**y**	**z**	**Occupancy**
Li	3a	0	0	0	0.9792	Li	3a	0	0	0	0.9819
Ni	3a	0	0	0	0.0208	Ni	3a	0	0	0	0.0181
Li	3b	0	0	0.5	0.0208	Li	3b	0	0	0.5	0.0181
Ni	3b	0	0	0.5	0.8092	Ni	3b	0	0	0.5	0.8119
Co	3b	0	0	0.5	0.11	Co	3b	0	0	0.5	0.11
Mn	3b	0	0	0.5	0.06	Mn	3b	0	0	0.5	0.06
O	6c	0	0	0.259	1	O	6c	0	0	0.259	1