Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information

Recycling with Precision: Engineering Ni-Rich NMC Cathodes through Impurity Management

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Table S1. ICP-MS result of NMC811 samples.

Element	Li (wt.%)	Co (wt.%)	Ni (wt.%)	Mn (wt.%)
NMC-R1	11.73	9.42	69.67	9.18
NMC-R2	12.02	8.87	69.97	9.14
NMC-CS	11.97	10.17	73.33	4.53
NMC-BASF	11.55	10.43	73.66	4.36

Table S2. Composition of aqua regia digested black mass (provided by Altilium).

Element	Al	Co	Li	Mn	Ni	Ca	Cu	Fe	Mg	Na	Pb	Zn
Composition (wt.%)	3.5	7.6	3.9	9.5	16.5	0.2	1.2	0.2	0.03	0.03	0.04	0.1

Table S3. Material properties of NMC811 samples.

Sample	Unit	NMC-R1	NMC-R2	NMC-CS	NMC-BASF	Standard
Skeletal density	g cm^{-3}	3.43±0.01	3.72±0.01	4.10±0.01	4.26±0.01	≥3
Tapped density	g cm^{-3}	2.53±0.03	2.59±0.02	2.67±0.01	2.89±0.03	≥2.2
D10	µm	3.32±0.06	6.03±0.20	5.28±0.08	7.01±0.10	≥5
D50	µm	9.27±0.26	12.70±0.35	11.27±0.16	11.32±0.21	11±2
D90	µm	20.74±1.68	22.89±1.19	19.33±0.23	18.36±0.51	≤25
Specific surface area	m^2 g^{-1}	1.29	0.81	0.37	0.46	0.2-0.7
pH of water extract	-	11.76	11.73	11.59	11.70	11.4±0.3

Table S4. Statistical variation in charge/discharge capacities and initial Coulombic efficiency for NMC811||Li half-cell

Sample	Standard deviation of charge capacity (mAh g^{-1})	Relative standard deviation of charge capacity	Standard deviation of discharge capacity (mAh g^{-1})	Relative standard deviation of discharge capacity	Standard deviation of initial Coulombic efficiency (%)	Relative standard deviation of initial Coulombic efficiency
NMC-R1	1.03	0.57%	0.43	0.27%	0.74	0.84%
NMC-R2	2.93	1.55%	1.03	0.59%	0.71	0.76%
NMC-CS	0.59	0.30%	2.16	1.24%	0.83	0.95%
NMC-BASF	3.69	1.73%	2.92	1.56%	0.88	1.00%

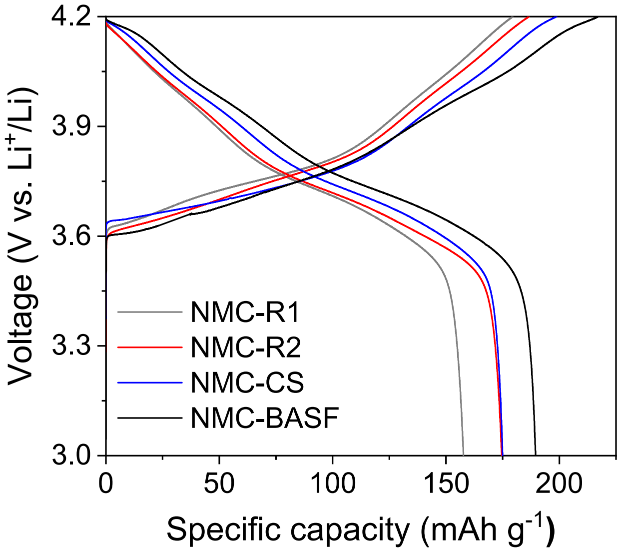

Figure S1. Charge-discharge profiles of the initial cycle at 0.05C of NMC811||Li half-cell measurements.

Table S5. Comparison of Regenerated NMC Cathodes: Methods, Feedstocks, and Performance

No	NMC types	Method	Feedstock source	Performance	Ref
1	NMC 811	Hydrometallurgy to precursor to cathode	Industrial black mass. Pilot scale.	175 mAh g^{-1} at 0.05C. (initial CE 93%); 165 mAh g^{-1} at 0.2 C. 95.1% after 50 cycles at 0.2C.	This work
2	NMC 111	Hydrometallurgy to cathode	Spent EV batteries (manually disassembled): Metal ratio limited by variability in commercial feed. Requires monitoring and adjustment	158 mAh g^{-1} at 0.1C.	[1]
3	NMC 811	Simulated hydrometallurgy: Co-precipitation	Pristine precursors with introduced additional Na impurities. Lab-scale.	172, 178, and 196 mAh g^{-1} at 0.1C. (initial CE ~89–91%)	[2]
4	NMC 622	Simulated hydrometallurgy: Co-precipitation	Pristine precursors with introduced additional Al impurities. Lab-scale.	180.2, 175.7, and 141.6 mAh g–1 at 0.05C.	[3]
5	NMC 811	Simulated hydrometallurgy: Co-precipitation	Pristine precursors with introduced additional Cu impurities. Lab-scale.	189.7, 226.4, 226.7, and 213.9 mAh g–1 at 0.1C. (initial CE 77.25%, 82.47%, 81.81% and 76.3%)	[4]
6	NMC 811	Direct recycling-solid-state sintering	Spent EV batteries (manually disassembled). Lab-scale.	162 mAh g^{-1} at 0.25C, 78% after 100 cycles at 0.25C	[5]
7	NMC 811	Direct recycling-molten-salt fluxing	Spent EV batteries (manually disassembled). Lab-scale.	180.1 mAh g^{-1} and retained 86.5% of the initial capacity after 200 cycles at 1C.	[6]
8	NMC 811	Defect-healing	Pristine NMC811 material, assembled into laboratory pouch cells	Capacity increases by 10% from 6.49 to 7.14 Ah at 1C.	[7]

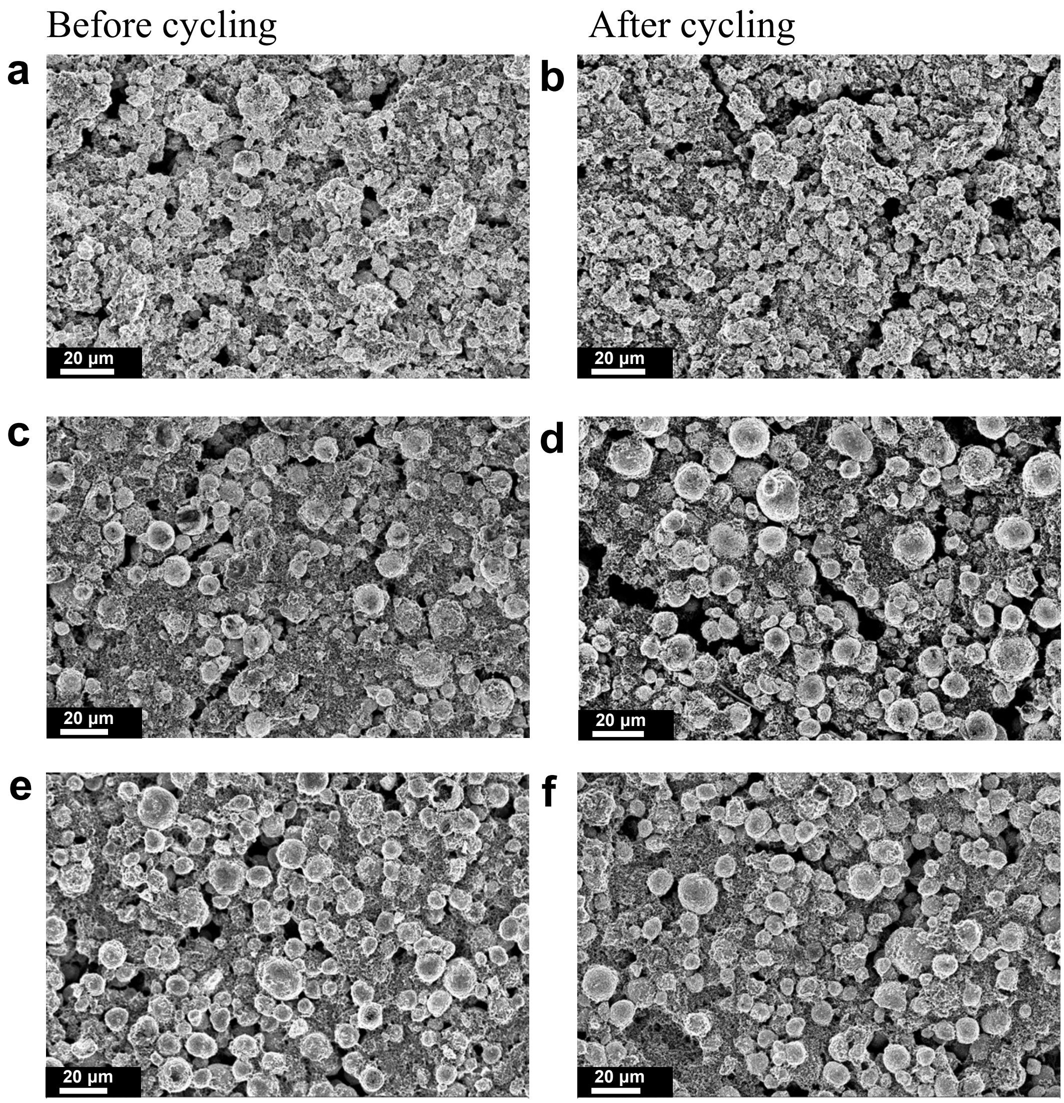

Figure S2. Bulk SEM images of NMC811 electrodes before and after 50 cycles. (a-b) NMC-R2. (c-d) NMC-CS. (e-f) NMC-BASF.

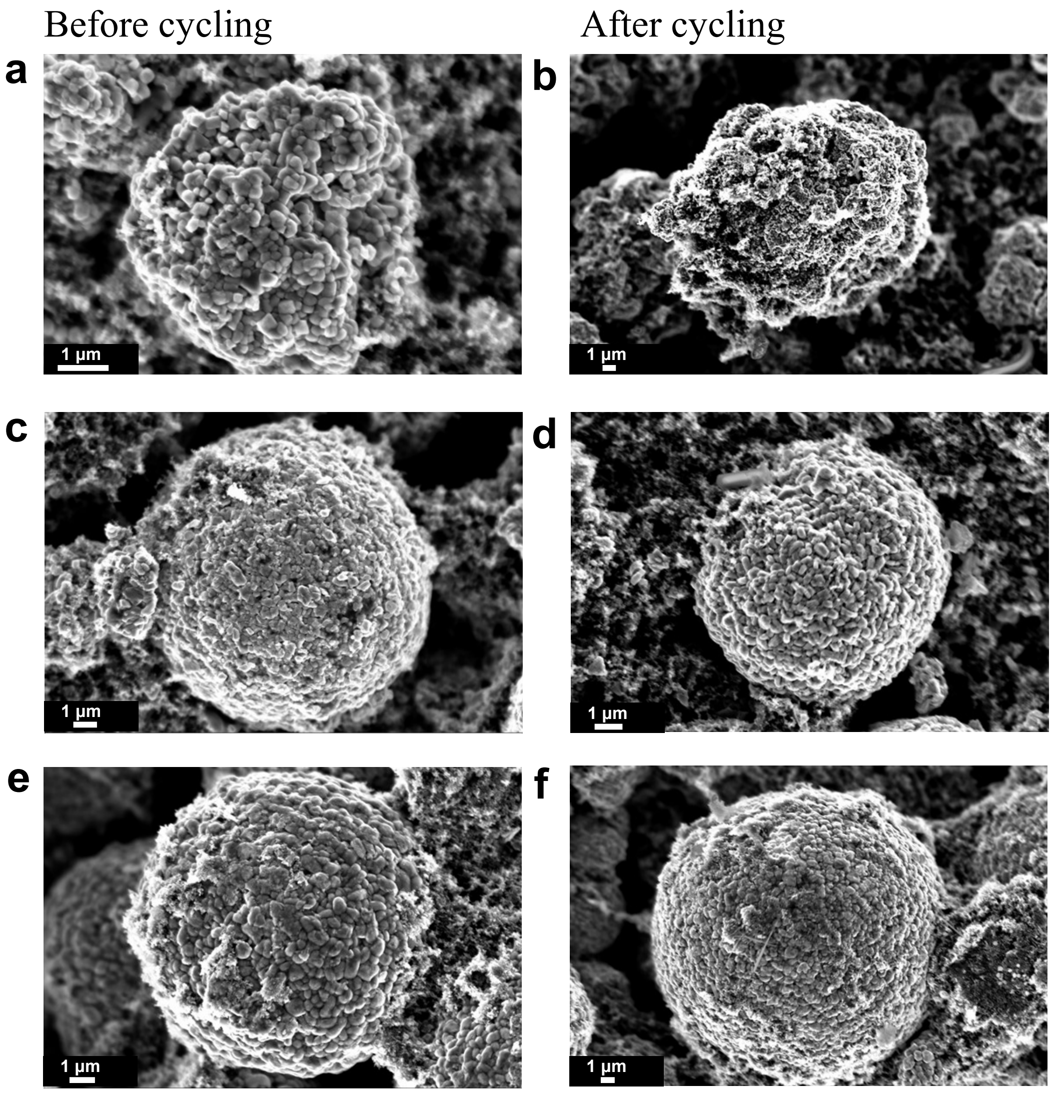

Figure S3. SEM images of NMC811 particles before and after 50 cycles. (a-b) NMC-R2. (c-d) NMC-CS. (e-f) NMC-BASF.

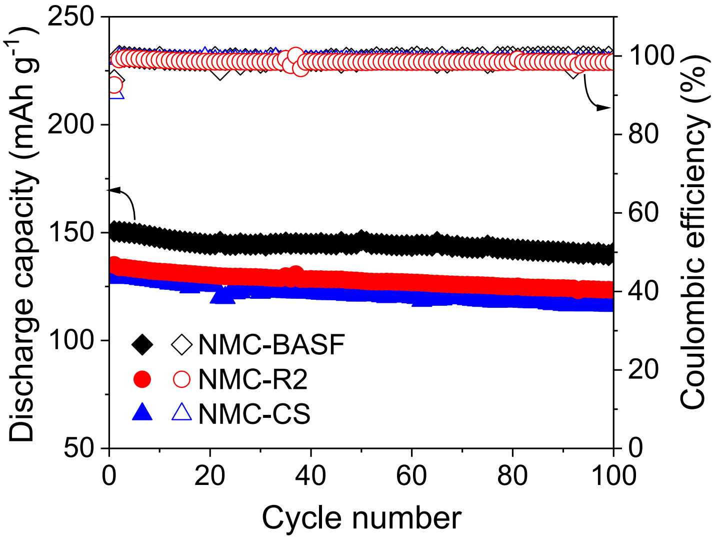

Figure S4. Cycling performance of NMC811||Li half-cells at 1C.

Table S6. Electrochemical performance of NMC811||Li half-cells: capacity, initial Coulombic efficiency, and capacity retention

Sample	Initial discharge specific capacities at 0.05C (mAh g⁻¹)	Coulombic efficiency	Capacity retention at 0.33C after 50 cycles	Capacity retention at 1C after 50 cycles	Capacity retention at 1C after 100 cycles
NMC-R2	173.8	91.1%	94.1%	95.0%	91.5%
NMC-CS	175.6	87.8%	93.2%	94.1%	89.1%
NMC-BASF	189.7	87.6%	93.6%	95.9%	92.6%

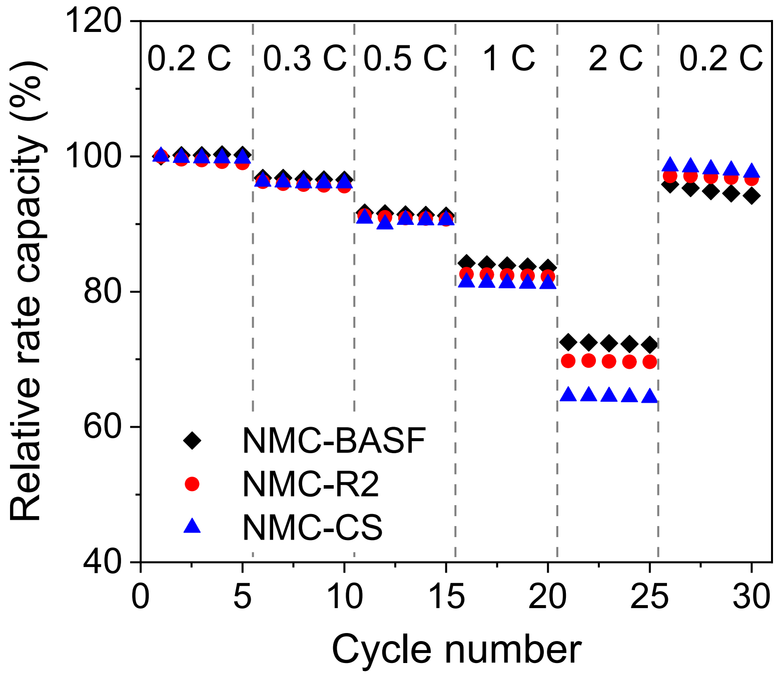

Figure S5. Relative rate performance as a function of cycle number, where the capacity at each C-rate is normalised to the initial capacity at 0.2C.

NMC-R2 NMC-CS NMC-BASF

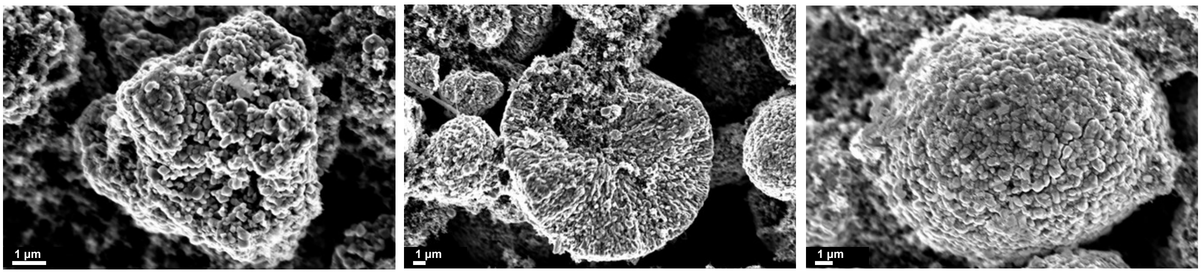

Figure S6. SEM images of NMC811 particles after 50 cycles.

Table S7. Comparative CO_2 emissions for Ni-rich NMC cathode production (NMC622 to NMC9.5.5) under different supply chain scenarios.

Comparison Scenario	CO_2 Emissions (kg CO_2 per kg Ni-rich NMC cathodes from NMC622 to NMC9.5.5)	Reduction ratio (%) vs. Altilium recycled supply chain
Primary Production – China	34.8–36.5	59-61
Hydrometallurgical Recycling – UK	16.7–18.7	15-24
Pyrometallurgical Recycling – UK	21.9–24.8	35-43
Altilium Recycled Supply Chain	12.0–16.7 (mean 14.2)	-

Table S8. Comparative CO_2 emissions reduction ratios for NMX cathode under the EU Battery Scenario.[8]

Comparison Scenario	Reduction ratio (%) vs. primary production	Reduction ratio (%) vs. primary production relying heavily on secondary supply
Hydrometallurgical Recycling	8-22	49-54
Pyrometallurgical Recycling	4-18	11-21
Direct recycling	36-41	55-63

Reference

1. M. Chen, Z. Zheng, Q. Wang, Y. Zhang, X. Ma, C. Shen, D. Xu, J. Liu, Y. Liu, P. Gionet, I. O'Connor, L. Pinnell, J. Wang, E. Gratz, R. Arsenault and Y. Wang, *Sci. Rep.*, 2019, **9**, 1654.
2. M. Beak, S. Park, S. Kim, J. Park, S. Jeong, B. Thirumalraj, G. Jeong, T. Kim and K. Kwon, *J. Alloys Compd.*, 2021, **873**, 159808.
3. R. Zhang, Y. Zheng, Z. Yao, P. Vanaphuti, X. Ma, S. Bong, M. Chen, Y. Liu, F. Cheng, Z. Yang and Y. Wang, *ACS Sustain. Chem. Eng.*, 2020, **8**, 9875-9884.
4. T. Wu, G. Wang, B. Liu, Q. Huang, Y. Su, F. Wu and R. M. Kelly, *J. Power Sources*, 2021, **494**, 229774.
5. P. Tammawat, T. Sesuk, P. Eiamlamai, P. Limthongkul and N. Kunanusont, *J. Power Source*s, 2025, **641**, 236827.
6. Z. Qin, T. Zhang, X. Gao, W. Luo, J. Han, B. Lu, J. Zhou and G. Chen, *Adv. Mater.*, 2024, **36**, e2307091.
7. H. Gong, Y. Cao, B. Zhang, J. Zhang, Y. Zhang, H. Wang, S. Zhang, X. Wang, Y. Mao, S. Liu, C. Han, Q. Xiang, C. Zhou and J. Sun, *Nat. Commun.*, 2024, **15**, 10243.
8. J. A. Llamas-Orozco, F. Meng, G. S. Walker, A. F. N. Abdul-Manan, H. L. MacLean, I. D. Posen and J. McKechnie, *PNAS Nexus*, 2023, **2**, pgad361.