Supplementary Information

Paradoxical Role of Rock-Salt Phases in High-Nickel Cathode Stabilization: Engineering Detrimental to Beneficial

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Supplementary Tables

Table S1. Comparative Performance Analysis of Surface Engineering Strategies for High-Nickel Layered Oxide Cathodes

Material System	Modification Strategy	Capacity Retention [cycles]	Initial C.E. (%)	Voltage Window (V *vs.* Li/Li^+)	Test Conditions	Ref.
I. Reference Materials						
$LiNiO_2$	Pristine	74 % [100]	82.5	2.7-4.3	30 °C, C/2	[1]
II. Surface Coating Strategies						
A. Metal Oxide Coatings						
PC-NCM811	Sucrose-derived carbon	76.7 % [200]	85.3	2.8-4.3	25 °C, C/2	[2]
SC-NCM811	Sucrose-derived carbon	84.8 % [200]	87.2	2.8-4.3	25 °C, C/2	[2]
PC-NCM83	Water-washed	52.8 % [200]	82.1	2.8-4.3	25 °C, 1C	[3]
PC-NCM83-AL	Al coating	68.3 % [200]	85.6	2.8-4.3	25 °C, 1C	[3]
PC-NCM83-Al-N	Al-N co-doping	96.5 % [200]	89.2	2.8-4.3	25 °C, 1C	[3]
III. Compositional Engineering						
A. High-Entropy Strategies	High-entropy doping					
HE-SC-N88		95.7 % [200]	82.1	2.8-4.3	25 °C, 1C	[4]
IV. Single-Crystal Architectures						
SC-Ni92-Pri	Single-crystal morphology	64.1 % [200]	[NA]	2.75-4.4	45 °C, 1C	[5]
SC-Ni93-Al	Al-coated single-crystal	72.1 % [200]	[NA]	2.75-4.4	45 °C, 1C	[5]
SC-Ni94-Zr	Zr-coated single-crystal	76.7 % [200]	[NA]	2.75-4.4	45 °C, 1C	[5]
SC-Ni95-ZA	ZA-coated single-crystal	84.7 % [200]	[NA]	2.75-4.4	45 °C, 1C	[5]

Table S2. Comparative Performance Summary of Cathode Engineering Strategies

Statistical Analysis of Performance Metrics by Modification Category

Strategy Type	Average Capacity Retention (%)[a]	Key Performance Advantages	Primary Limitations
Surface Coating	75.3 ± 8.2	• Suppressed TM dissolution • Enhanced CEI stability	• Initial rate capability reduction • Coating uniformity challenges
High-Entropy Doping	93.5 ± 2.1	• Superior structural stability • Entropy-driven stabilization	• Synthesis complexity • Optimization requirements
Single-Crystal Architecture	74.7 ± 7.6	• No grain boundaries • Mechanical robustness	• Reduced rate performance • Manufacturing scalability

[a]*Average capacity retention calculated from all reported values after 200 cycles at 1C rate, ±1σ standard deviation*

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