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Uncovering Degradation Pathways in High-energy NMC811 Electrodes for Tropical e-Mobility Applications

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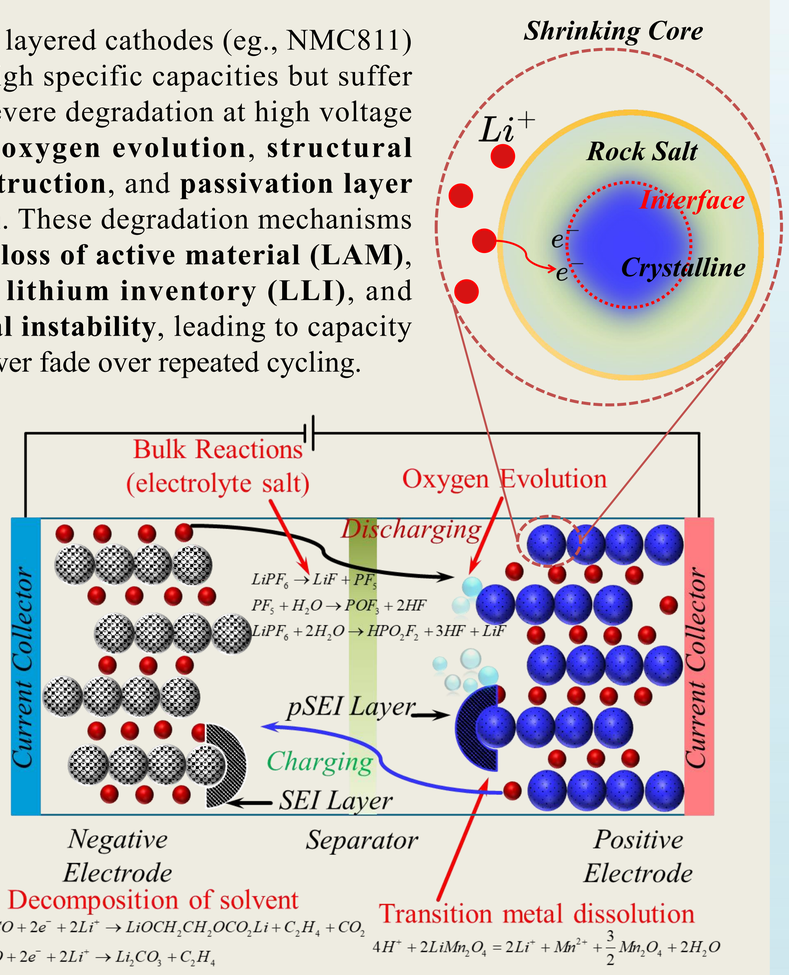
Introduction

Ni-rich layered cathodes (eg., NMC811) offer high specific capacities but suffer from severe degradation at high voltage due to **oxygen evolution**, **structural reconstruction**, and **passivation layer growth**. These degradation mechanisms induce **loss of active material (LAM)**, **loss of lithium inventory (LLI)**, and **thermal instability**, leading to capacity and power fade over repeated cycling.

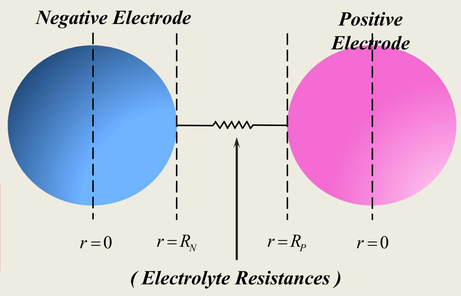


Theory & Simulations

Governing Equations

Fick's 2nd law of diffusion in spherical coordinate system:

$$\frac{\partial c_{s,j}}{\partial t} = \frac{D_{s,j}}{r_j^2}\frac{\partial}{\partial r_j}\left(r_j^2\frac{\partial c_{s,j}}{\partial r}\right)$$



Final Formulations

» Butler-Volmer Equation:-

$$J_j = \frac{I_j}{F} = k_j c_{s\max,j} c_e^{0.5}(1-X_{j,surface})^{0.5} X_{j,surface}^{0.5}\left[\exp\left(\frac{\alpha_p F}{RT}\eta_j\right) - \exp\left(-\frac{\alpha_n F}{RT}\eta_j\right)\right]$$

Overpotential

$$\eta_j = \phi_{1,j} - \phi_{2,j} - u_j$$

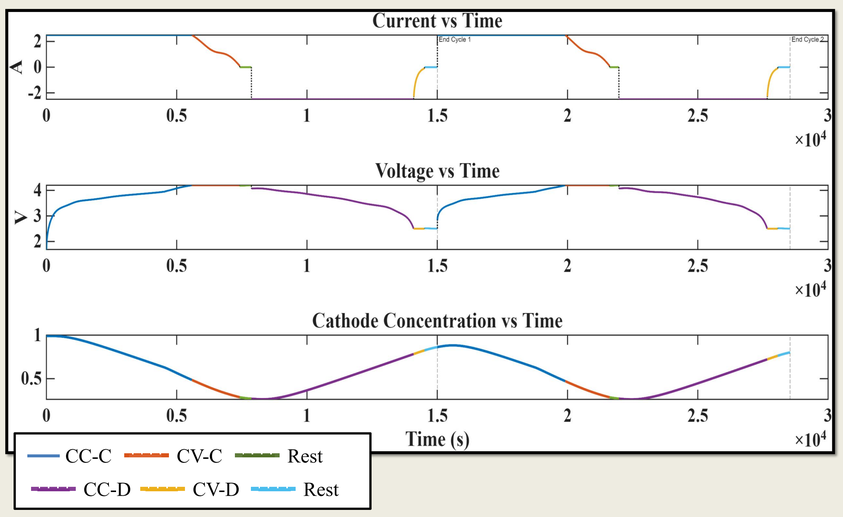
Solid phase potential; Solution phase potential

» $V_{cell} = U_p(x_p^{surf}, T_{ref}) - U_n(x_n^{surf}, T_{ref}) + \left(U_{p,T}\big|_{x_p^{surf},T_{ref}} - U_{n,T}\big|_{x_n^{surf},T_{ref}}\right)(T - T_{ref}) + \frac{2RT}{F}\ln\left(\sqrt{(m_p^2+4)} + m_p/2\right)$

$$+\frac{2RT}{F}\ln\left(\sqrt{(m_n^2+4)} + m_n/2\right) + I_{app}R_{cell}$$

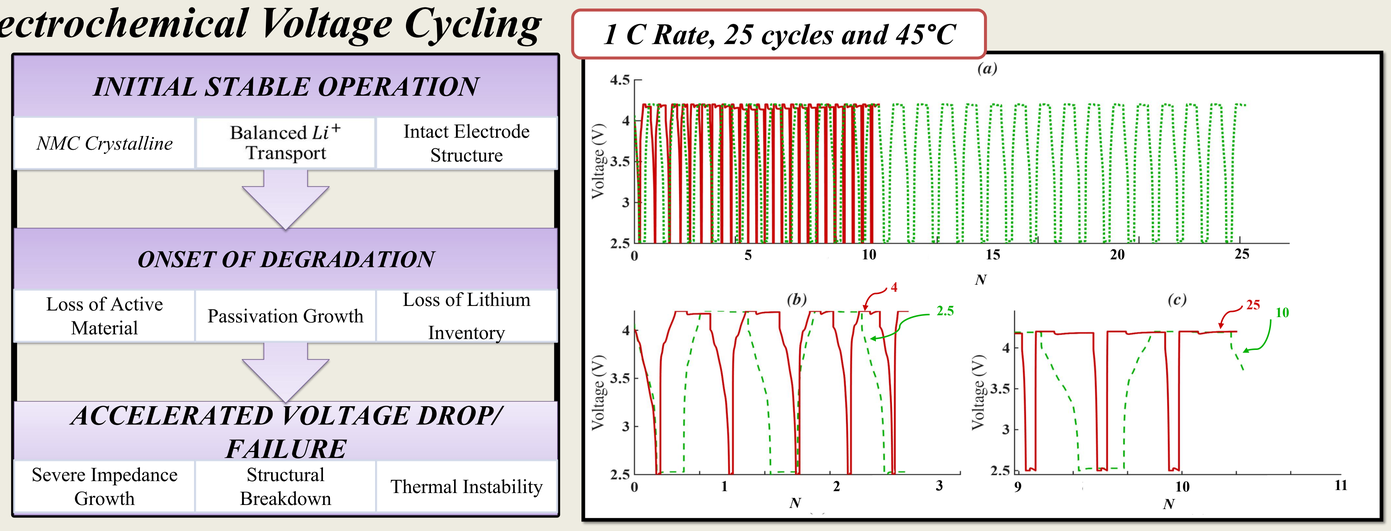
$R_{cell} = \Theta_1 + \Theta_2(T - T_{amb})$ — Parameterized using Experimental Data [4].

CC-CV T-dSPM Multi-Cycling

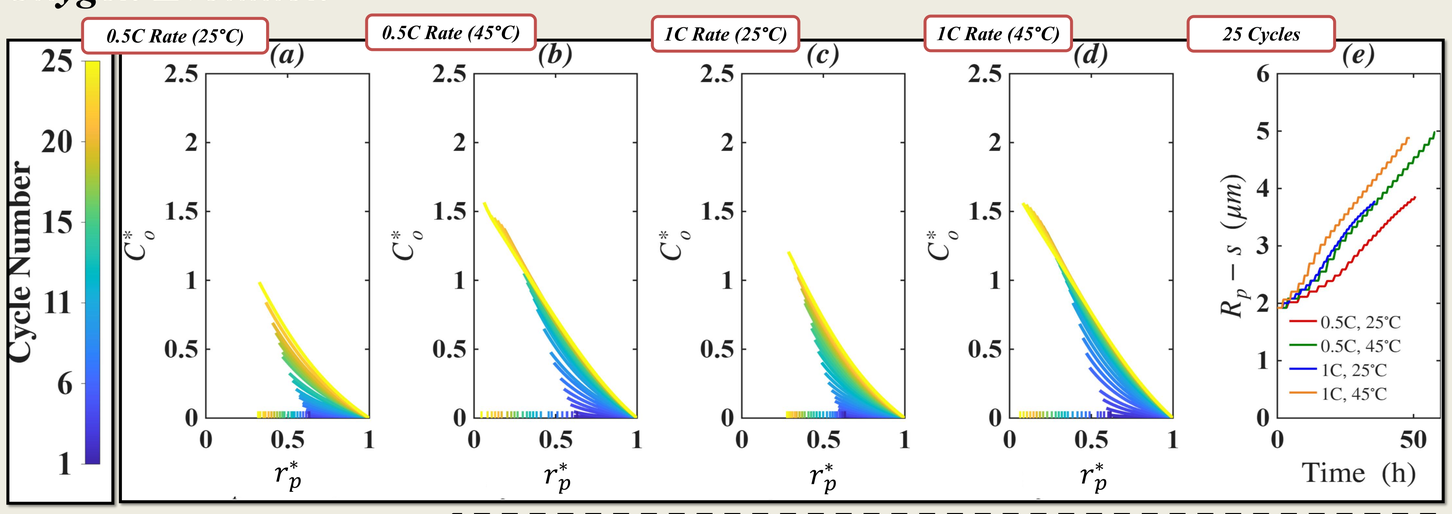


Results & Discussions

Electrochemical Voltage Cycling



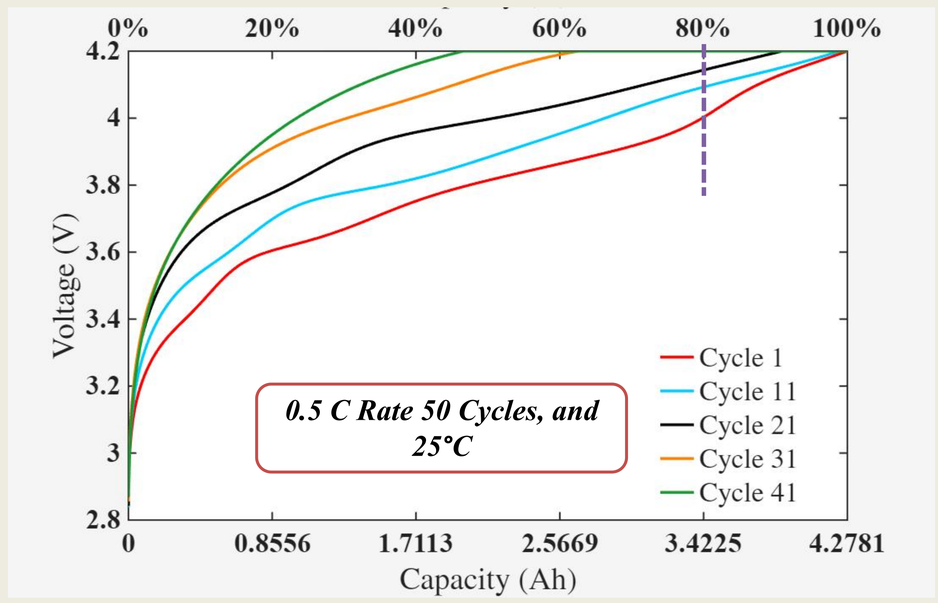
Oxygen Evolution



- The increasing oxygen concentration near the particle surface and the growing passivation layer with cycling confirm that **oxygen release from the core-shell interface accelerates at higher temperature and C-rate**, promoting faster rock-salt layer formation; Driving capacity fade.

Capacity Degradation Trajectory

- At 0.5C, ***passivation layer, active material Loss, lithium depletion, and increased diffusion resistance in Ni-rich cathodes*** elevate in cell with reduced capacity.



Conclusions

- Oxygen release at the core-shell interface drives the formation of a rock salt passivation layer, leading to LAM, Li-deposition, Capacity fade.
- Higher temperature and C-rate accelerate degradation through enhance O_2 evolution and shell growth
- The Thermally coupled *d*SPM successfully captures the coupled electrochemical-thermal degradation.

Impact Statement ***The insights enable optimized cathode design and operating strategies for longer-lasting , safer Li-ion batteries.***

References

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Acknowledgements

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