# Uncovering Degradation Pathways in High-energy NMC811 Electrodes – A Physics-based Predictive Model for Tropical e-Mobility Applications

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***Highlights***

- Developed a degradation-integrated Single Particle Model (*d*SPM) incorporating structural reorganization, passivation layer growth, and gas evolution in NMC811 electrodes.
- Captured counterintuitive degradation behaviour, where slow cycling under high SoC leads to accelerated ageing and gas evolution, especially at tropical temperatures.
- Provided a physics-based framework for predicting and optimizing battery performance in tropical e-mobility scenarios.

**Abstract**

High-nickel (NMC811) high-energy lithium-ion batteries (LIBs) [1] are widely favoured for electric vehicle (EV) applications due to their high voltage and energy density. However, these systems are prone to capacity fade, particularly at high states of charge (SoC), stemming from complex degradation mechanisms. This study presents a physics-informed, continuum-scale degradation-integrated Single Particle Model (*d*SPM) that captures the evolution of degradation phenomena in NMC811 positive electrodes over multiple charge-discharge cycles [2,3]. Key mechanisms include structural reorganization, gas evolution, and the growth of passivation layers. A Shrinking Core mechanism is employed to represent the phase transition between ordered and disordered phases, wherein the active core shrinks and the passivation shell grows, impeding lithium-ion transport. The model simulates Constant Current – Constant Voltage (CCCV) charging protocols and accounts for both the loss of active material (LAM) and lithium inventory (LLI). Using finite-difference methods to solve the coupled mass transport and degradation kinetics equations, the model delineates two dominant ageing regimes: diffusion-limited and reaction-limited degradation. Counterintuitively, simulations reveal that slower charging at ambient temperature (25°C) accelerates degradation due to prolonged cycling within the high-voltage regime (3.8 – 4.2 V), leading to increased gas evolution and structural breakdown. These effects are further exacerbated under tropical conditions (>45°C) [4], highlighting significant challenges for EV adoption in hot climates. Intriguingly, fast charging is shown to mitigate passivation effects and gas evolution, offering critical insights for battery management in tropical regions. This framework provides a predictive platform for optimizing battery materials, charging protocols, and thermal management, with future integration into full-cell models for lifetime prediction.

**References**

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