

Electrochemical and material characterization of Li-ion cells for aging degradation modelling

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Lithium-ion batteries (LIBs) play a pivotal role in sustainable energy solutions, powering electric vehicles and storing renewable energy. However, degradation during aging causes capacity fade, increased internal resistance, and safety risks, limiting lifespan and reliability.

Among battery components, the positive electrode (PE) material is one of the most critical, as it governs key performance metrics: energy density, power density, cycle life, safety, cost, and sustainability. Widely used PEs include nickel manganese cobalt oxide (NMC) and lithium iron phosphate (LFP). NMC delivers high energy density (~170 mAh/g for NMC811 associated with a nominal voltage of ~3.7 V) but suffers from high cost, resource scarcity (cobalt/nickel), moderate thermal stability, and limited cycle life due to issues like particle cracking and transition metal dissolution. In contrast, LFP excels in safety, thermal stability (thanks to its 3D olivine structure), long cycle life (>2000 cycles), and low cost using abundant elements, though it offers lower energy density (with a nominal voltage of ~3.2 V vs. Li+/Li) and a reduced voltage range. To bridge these trade-offs, lithium manganese iron phosphate (LMFP, $\text{LiMn}_x\text{Fe}_{1-x}\text{PO}_4$) has emerged. LMFP preserves LFP's safety, cost, and stability advantages while raising the nominal voltage (~3.8 V) and boosting energy density by ~15–20% via manganese redox activity, providing a cobalt/nickel-free alternative. It is also common to use blends of different materials as PEs in order to combine the characteristics of the different materials.

This PhD project investigates degradation mechanisms in a commercial cylindrical cell (26700 format) with an LMFP:NMC blended PE and graphite negative electrode (NE). Accelerated aging tests were performed under varied protocols: calendar aging (at elevated temperatures) and power cycling (at different C-rates and depths of discharge). By adjusting conditions, dominant degradation pathways could be isolated and shifted (e.g., toward SEI growth, lithium loss, transition metal dissolution, or particle cracking).



Fig 1. Li-ion cell.

Degradation is currently quantified from electrochemical perspectives (capacity tracking, electrochemical impedance spectroscopy (EIS), incremental capacity analysis, and internal resistance evolution) and going to be modeled empirically to predict capacity fade and resistance increase. Then, Post-mortem material characterizations on pristine and aged cells—including scanning electron microscopy (SEM), X-ray diffraction (XRD), inductively coupled plasma (ICP) analysis, half-cell electrochemistry, and thermogravimetric analysis (TGA)—will be conducted to validate the proposed mechanisms and confirm the model accuracy.

This multi-faceted approach gives an insight aging in LMFP:NMC-graphite systems, provides a validated predictive tool for battery management, and supports strategies to enhance durability of next-generation sustainable LIBs.

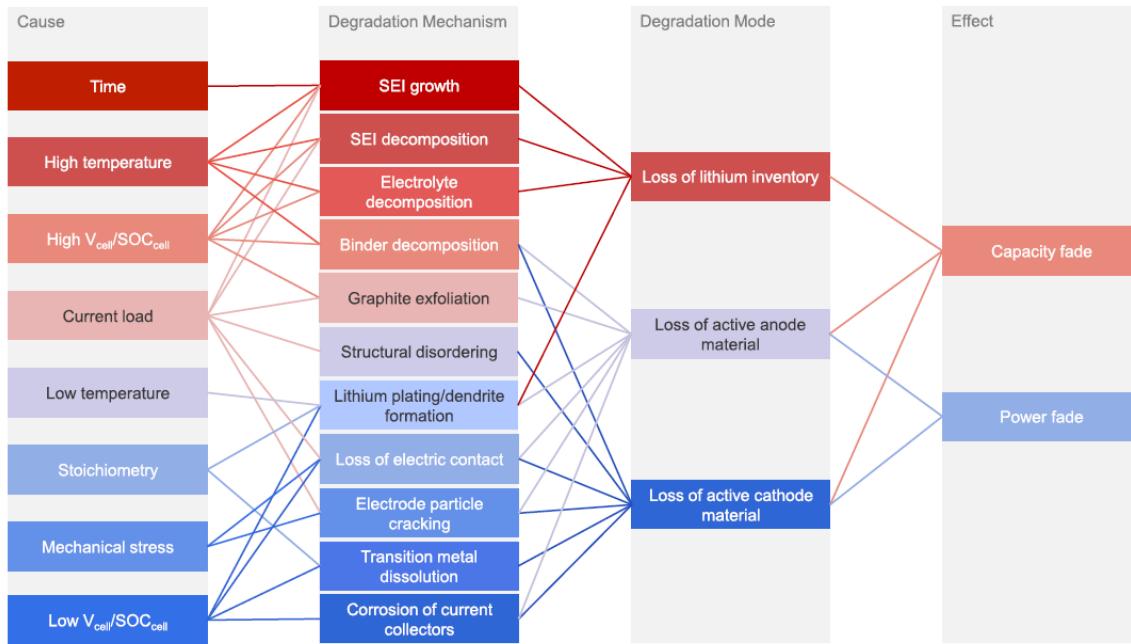


Fig 2. Cause and effect of degradation mechanisms and associated degradation modes.