

High-Energy Nickel-Cobalt-Aluminium Oxide (NCA) Cells on Idle: Anode- versus Cathode-Driven Side Reactions



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Invited for this month's cover picture is the group of Prof. Harry Hoster at Lancaster University. The cover picture illustrates how state of charge (SoC) influences the capacity fade of a widely employed automotive Li-ion battery chemistry when idle, e.g. when EVs are parked. The chemical degradation is revealed to be aggravated when devices are kept at around 80% SoC. Read the full text of the Article at 10.1002/batt.202100046.

What is the central finding of this research and why is it so pertinent?

Li-ion batteries degrade even if not in use. Such type of silent degradation actually dominates ageing of batteries in most electric cars, given that they can be parked > 90% of the time (and possibly more so during the COVID-19 pandemic). To elucidate the underpinning chemical deterioration, we performed a systematic investigation of the effect of state-of-charge (SoC) and temperature on NCA/Gr-SiO_x 21700 cells—a commercial battery widely employed in electric cars. Capacity fade of those cells does not monotonically increase with SoC or cell voltage, contrary to common belief. Instead, we found a 'spoon-shaped' profile: cells stored at 100% SoC have better capacity retention than cells stored at 80 or 90% SoC. This non-monotonic relationship between capacity fade and SoC of NCA/Gr-Si batteries results from enhanced cross-talk between the cathode and anode.

A central element is a corrosion-style coupling of electrochemical reactions at the cathode: in the absence of an external current, parasitic CO₂ formation is coupled to Li⁺ reduction. The Li⁺ concentration in the electrolyte is maintained through



supply from the anode, which diverts lithium away from parasitic solid–electrolyte interphase (SEI) growth. Hence, higher irreversible self-discharge can alleviate reversible capacity loss. The ‘spoon-shaped’ profile arises because cathode-driven rates of self-discharge monotonically increase with the cell voltage, while the anode-governed supply of reactants reaches a plateau once the cell is slightly more than half-full. At $T > 40^\circ\text{C}$, self-discharge becomes a major problem of its own. Such temperatures are easily encountered by electric vehicles parked in cities and elsewhere, and users typically keep their devices at high levels of SoC. Finally, our findings are important for battery operation strategies in view of maximising their lifetime, ultimately reducing their carbon footprint.

How did this project start and where is it heading to?

The project started with an industry collaboration, led by AMTE Power and funded by Innovate UK. The Lancaster BatLab team was tasked with developing a framework for insurance-based warranty for new battery cells. Calendar ageing studies were part of the usual test matrix. The unusual spoon-shapes of degradation rates appeared after a few weeks, which led to further experiments and deeper investigation. In the Multi-

Scale-Modelling consortium (led by Gregory Offer at Imperial College and Funded by the UK Faraday Institution), our team works on physics-based degradation models. It was a logical step to couple both activities and started modelling the findings reported here, which initially were merely empirical.

Our team now looks in more detail at the electrochemical reaction mechanisms behind the coupled SEI growth and self-discharge processes. This includes a deeper study of the mechanisms behind the accelerated high-temperature self-discharge, which we believe involves accidental formation of electrocatalytically active sites at the cathode.

Altelium Ltd., a company specializing in battery-related warranty products in the insurance market, is the Lancaster spin-off that resulted from our work. Warranty products are in high demand at smaller companies who need external financial backing for the warranties they issue. Without such warranties, they cannot compete with established big players. Thus, our research will eventually enable more battery products from smaller companies to enter the market.