Electrolyte motion induced solid-phase lithium inhomogeneity in lithium-ion batteries

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Electrolyte motion induced salt inhomogeneity (EMSI) has been linked to capacity fade mechanisms such as lithium plating in lithium-ion cells, yet the liquid-phase salt gradients represent only the initial consequence of electrolyte motion. How EMSI ultimately translates into performance loss remains poorly understood. Here, we extend a previously developed modelling framework, incorporating electrolyte flow and electrode mechanics in an 18650 cylindrical cell, to decouple kinetic, transport and thermodynamic overpotential contributions, and trace the causal chain from salt redistribution to capacity fade. We show that EMSI-driven salt gradients propagate into persistent solid-phase lithium inhomogeneity in both positive and negative electrodes, causing uneven utilisation along the cell axis that manifests as apparent capacity fade, an effect further accentuated by the anode overhang near the electrode edge. By mapping the onset of lithium plating across a range of operating conditions, we demonstrate that plating susceptibility is governed by the interplay of liquid- and solid-phase concentrations and is significantly amplified by the mechanical constraints in the wound jelly roll. We reveal that the strong concentration dependence of electrolyte viscosity introduces an intrinsic asymmetry in salt redistribution: protocols with unequal charge and discharge rates produce a direction-dependent inhomogeneity that vanishes when viscosity is treated as a constant. These findings establish design guidelines for mitigating EMSI and underscore the necessity of accounting for electrolyte motion when optimising cell architecture and cycling protocols.