Evaluating Multi-salt electrolytes and Textured Supports for Zero-excess Lithium Metal Batteries

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Nowadays, secondary lithium-ion batteries (LIBs) play a crucial role in the green energy transition. However, their performance is approaching theoretical limits, and current manufacturing processes remain highly energy-intensive[1]. A possible short-term solution to maximize LIB energy density while reducing manufacturing costs is to assemble conventional LIB cathode materials in a "zero-excess"[2] configuration. This involves removing the anode compartment from the initial cell setup, as all the lithium required for the electrochemical processes is provided by the cathode material itself. During cycling, lithium is plated onto and stripped from a dedicated current collector during charging and discharging, respectively[3]. Despite its advantages, the commercial development of zero-excess Li-metal batteries is hindered by capacity losses during the initial lithium deposition and dendritic growth in subsequent cycles[4]. To address these challenges, we propose the use of a multi-salt electrolyte and a textured current collector as a viable and more sustainable alternative to pure lithium metal. The presence of different salts in the electrolyte composition alters the lithium solvation structure, increasing the proportion of inorganic species over solvent molecules within the solvation shell[5]. Upon lithium deposition during the first cycle, the preferential decomposition of these inorganic-rich species leads to the formation of a more robust and ionically conductive solid electrolyte interphase (SEI). Additionally, employing the Laser-Induced Periodic Surface Structure (LIPSS) technique [6] enables the fabrication of periodic nanostructures on the current collector surface, guiding uniform lithium nucleation and promoting the formation of a homogeneous lithium metal electrode. The application of these components in lithium metal half-cells was investigated through a combination of ex situ Raman spectroscopy and optical/electron microscopy, coupled with galvanostatic and chronoamperometric measurements. These results highlight a promising strategy to improve the stability and performance of zero-excess lithium metal batteries, paving the way for more efficient and sustainable next-generation energy storage systems.