Electrodeposition of Metals in Nanostructured Electrolytes: Transport Phenomena and Stability

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Electrodeposition is used in various chemical manufacturing processes for creating metal, colloid, and polymer coatings on conductive electrode substrates. The process also plays an important role in electrochemical energy storage technologies based on batteries, where it must be carefully managed to facilitate stable and safe operations at low operating temperatures, high rates and over many cycles of charge and discharge. This talk considers the stability of electrodeposition of reactive metals such as lithium and sodium on planar electrodes with the goal of enabling next-generation secondary batteries based on lithium and sodium metal anodes. Such batteries promise substantial improvements in electrochemical energy storage over todays's state-of-the art lithium ion technology and are under active investigation worldwide.

Development of a practical rechargeable lithium metal battery (LMB) remains a challenge due to uneven lithium electrodeposition and formation of ramified denderitic electrodeposits during repeated cycles of charge and discharge. Known consequences of unstable electrodeposition in LMBs include accumulation of electrically disconnected regions of the anode or "dead lithium", thermal runaway of the cell, and internal short circuits, which limit cell lifetime and may pose serious hazards if a flammable, liquid electrolyte is used in a LMB. Lithium-ion batteries (LIBs) are designed to eliminate the most serious of these problems by hosting the lithium in a graphitic carbon substrate, but this configuration is not entirely immune from uneven lithium plating and dendrite formation. Specifically, the small potential difference separating lithium intercalation into versus lithium plating onto graphite, means that a too quickly charged or overcharged LIB may fail by similar mechanisms as a LMB.

Using a continuum transport analysis for electrodeposition in a structured electrolyte in which a fraction of the anions are fixed in space, I will show that electrodeposition at the lithium anode is intrinsically unstable at both low and high current densities. I will further show how the process can be stabilized through rational design of the electrolyte and salt. Building upon these ideas, I will explore structure and transport in novel nanoporous hybrid electrolyte configurations designed to stabilize metal anodes against dendritic electrodeposition and premature failure. Application of these electrolyte designs for LMBs will be used to evaluate stability conditions deduced from theory.