Initial Formation of the Solid Electrolyte Interphase in Li Metal Batteries

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Advances in energy storage technology beyond traditional lithium ion battery materials are needed to meet the demands of the future which include a growing number of electric vehicles and grid storage applications. Next generation technologies such as Li-S and Li-O_2 batteries employ metallic lithium as an anode material due to its high specific capacity and low electrochemical potential. However, lithium presents a number of challenges including uncontrolled electrolyte consumption, formation of dendrites and "dead" lithium, as well as formation of a non-passivating and inhomogeneous solid electrolyte interphase (SEI). While the composition, morphology, and electrical properties of the SEI are critical to overall battery performance and lifetime, there is a lack of fundamental understanding regarding its initial formation and how these early processes relate to cell performance after many cycles.

In this work, we are using soft X-ray absorption spectroscopy (XAS) to study the initial stages of SEI formation with half cycle resolution in Li-Cu cells employing an ether-based electrolyte composed of 1 M lithium bis(trifluoromethyl)sulfonimide (LiTFSI), 0.3 M lithium nitrate ($LiNO_3$) in a mixture of 1,3-dioxolane (DOL) and 1,2-dimethoxyethane (DME) (v/v = 1:1), which is a standard electrolyte used in Li-S batteries. Coin cells are galvanostatically or potentiostatically cycled under varying conditions to probe the various stages of SEI formation. Copper electrodes are then removed from disassembled cells and rinsed prior to X-ray characterization. The XAS intensity from the species composing the SEI increases with cycling, indicating continued decomposition of the electrolyte. Interestingly, the SEI present after a plating half cycle differs noticeably from the SEI present after a stripping half cycle. Carefully controlled formation of the SEI during the first several half cycles may lead to large effects on the long-term composition and structure of the SEI and determine overall battery performance and cycle life.