Understanding Coulombic Efficiency of Lithium Anodes through Operando XRD

N R. Geise[1], R. M. Kasse[2], H-G Steinruck[3], M. F. Toney[3]

[1] Department of Chemistry, Stanford University, USA
[2] Materials Science and Engineering, Stanford University, USA
[3] Stanford Synchrotron Radiation Lightsource, Menlo Park, USA

Next generation batteries will require higher energy densities, long cycle lives, safety, and affordability. Li-metal is a promising, high-capacity anode that can be incorporated into Li-ion, Li-S, and Li-air systems. However, in the process of lithium plating and stripping, there are large volume and morphological changes that cause uneven plating, growth of mossy and dendritic Li, as well as cracking and reforming of the solid electrolyte interphase (SEI). The loss of Li into the SEI and into Li deposits that become electrically disconnected ('dead' Li) are major, interrelated contributors to Coulombic efficiency losses in the Li-metal system. However, fundamentals of these issues are not well-understood, and current methods of characterization provide a limited picture of how Li nucleation and growth occurs.

X-ray diffraction (XRD) provides an in-situ probe to detect metallic Li and crystalline SEI components in a nondestructive and quantitative fashion. We have developed an operando electrochemical cell to measure the XRD during plating and stripping of Li onto an inert copper substrate. Our results confirm the ability to quantify Li plating throughout cycling. We are developing model, highly crystalline inorganic SEIs to better control SEI formation and develop this method. By tracking the development of XRD peaks of the crystalline SEI and Li phases, we can obtain quantitative results on the SEI and dead Li development through cycling and correlate these results with electrochemical data.