Quantitative Observation of Interface Dynamics in Next Generation Batteries

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Reactions at the two interfaces between the main components of a battery – anode/electrolyte and cathode/electrolyte – determine the overall energy density and coulombic efficiency of the entire system. In many of the newly developed battery nanomaterials and nonaqueous electrolytes, the biggest challenge is the transition from promising performance of the half-cell to a fully operating battery. There are many factors preventing the successful commercialization of these next generation battery systems, which are usually triggered by side reactions leading to electrolyte breakdown, passivation or corrosion and the formation of a solid-electrolyte-interphase (SEI) layer. In addition, deposition of an excess of metal ions during charging leads to dendrite formation during cycling. Each of these effects is directly linked to the local chemistry/field at the electrode/electrolyte interface and a full understanding of the materials parameters that can lead to improved battery performance can be achieved only if we can measure this directly. Here we show the use of an *operando* electrochemical cell (*ec*-cell) in the scanning transmission electron microscope (STEM) to investigate the role and mechanism of electrolyte additives, the initial stages of deposition/dissolution of Li dendrite and formation of the SEI layer in Li-ion, Li-sulfur and Li-Air batteries. [1-4]. For all these measurements, understanding and subsequently controlling the electrochemical process requires the ability to directly observe the transients as they happen. STEM routinely has the spatial resolution to directly visualize these transient processes on the atomic scale. However, the typical current densities used in modern instruments have made beam damage prevalent and the limitation to imaging is now the sample rather than microscope. Our aim in performing *operando* studies is therefore to efficiently use the dose that is supplied to the sample and to extract the most information from each image. Optimizing the dose/data content in non-traditional ways involves two main strategies to achieve dose fractionation – reducing the number of pixels being sampled in STEM mode or increasing the acquisition speed of the images in TEM mode. For STEM, inpainting methods allow a dose reduction of an order of magnitude or more, allowing data to be automatically recorded in a compressed form [5]. For the TEM mode of operation, an increase in speed increases the number of images and means that compressive sensing (CS) and automated methods of tracking changes in the structure need to be developed so that only the important changes are automatically detected and recorded. In this presentation, the use of inpainting/compressive sensing and the potential advantages for higher precision *operando* measurements in the future will be discussed.

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