Watching reactions take place at the atomic scale (and learning from them)

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Abstract

The majority of the research undertaken in my group focuses on making better batteries to meet the demands of emerging applications. A large proportion of the function of batteries arises from the electrodes, and these are in turn mediated by the atomic-scale perturbations or changes in the crystal structure during an electrochemical process (e.g. battery use). Therefore, a method to both understand battery function and improve their performance is to probe the crystal structure evolution *in operando*, i.e., while an electrochemical process is occurring inside a battery.

So, in my group we use *in operando* neutron powder diffraction, with its sensitivity towards lithium, to literally track the evolution of lithium in electrode materials used in rechargeable lithium-ion batteries. In addition, the ability to test smaller samples (e.g. in coin cells) with *in operando* X-ray powder diffraction has allowed us to probe other battery types, such as primary lithium and ambient temperature rechargeable sodium-ion batteries, and other configurations, such as thin film devices. With the information from these experiments we have directly related electrochemical properties such as capacity, battery lifetime and differences in charge/discharge to the content and distribution of lithium or sodium in the electrode crystal structures.

We are expanding our footprint in both the analytical techniques we use and the reactions we explore. Recent work has been directed towards realizing *in operando* neutron imaging, *in operando* X-ray absorption spectroscopy and *in situ* solid-state NMR allowing us to probe non-crystalline components in devices. We are also investigating formation reactions, i.e., literally watching synthesis of crystalline materials, and tracking the distribution of electrolytes during processes. The combination of these techniques and reactions provides more insight into the mechanism of device operation and the interactions at play.

Finally, materials discovery plays a large part in our synthetic work. We have two new research dimensions underway, the electrochemical tuning of the negative thermal expansion materials to obtain zero thermal expansion materials and the scaffolding of layer-structured electrode materials to increase electrochemical performance in rechargeable batteries.

This talk will provide a flavor of the work being undertaken in my group, emphasizing the highlights and our future directions.