Understanding the Atomic Structure of Li-based Cathode Materials for Lithium-Ion Batteries by Advanced Transmission Electron Microscopyplayer

P.J. Ferreira

Materials Science and Engineering Program, The University of Texas at Austin, Austin, TX, USA

In order for Li-ion batteries to mature to a level useful for integration into the current or future energy infrastructure, basic problems such as cyclability, cost and rate capability must be overcome. The spinel cathode materials of the type $LiNi_xMn_{2-x}O_4$ (LNM) ($0<x\leq½$) have the advantage of being both cost-effective and high-rate capable materials, but they are plagued with cyclability problems. In the LNM system the main contributor to cycling degradation is the high operating voltage which leads to solid-electrolyte interphase (SEI) formation. To understand the passivation mechanism, it is crucial to determine the surface's atomic structure as it defines how reactive the cathode will be with the electrolyte during oxidation and reduction cycles. Hence, it is critical to understand the different phases that form in this system.

In this regard, aberration-corrected transmission electron microscopy was used to identify the surface and bulk structures in the LNM system. The analysis confirms the spinel structure and shows good agreement with computer simulations in the bulk. Near the surface however, other phases are observed. These include a rock-salt structure which is expected from x-ray diffraction results and a new phase, defined here as "ring-type structure", because of the characteristic rings that are formed within the first few atomic surface layers. All three phases are observed near the surface, however only the spinel is found within the bulk of the particles. This work enables us to develop a well-suited cathode material for future energy storage that will potentially spur the evolution of the future sustainable energy landscape.