Nanoscale X-ray microscopy of High Voltage Lithium-Rich MNC Composite Cathodes with 2D/3D Chemical/Elemental sensitivity

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It is important to probe and to understand the correlation between morphological and chemical changes within battery electrodes under electrochemical cycling. This is specifically relevant for the new class of high voltage layered-layered lithium-rich composite cathode having generic composition, $Li_{1+x}M_{1-x}O_2$ (where M = Mn, Ni and Co), x = 0.2-0.4, where there are significant changes in form of transition metal (TM) rearrangement in the host structure leading to formation of a lower energy (voltage) phase that closely resembles to a spinel structure instead of the layered–layered structure. In this case it is important to quantitatively analyze the changes in cathode material with elemental/chemical sensitivity in addition to the 3D nano/micro structure retrieval, which could lead to better understanding of the structure-electrochemical property correlation.

We combined full-field Transmission X-ray Microscopy with X-ray Spectroscopy[1-2] to investigate single-pixel chemistry of lithium-rich composite cathode materials having nominal composition $Li_{1.2}Mn_{0.525}Ni_{0.175}Co_{0.1}O_2$. K-edge spectra of all three transition metal elements were acquired and analyzed with spatial resolution down to 30 nm over several fields of view, covering tens of microns.

The chemical maps (as shown in figure 1) generated from the TXM-XANES analysis[3] (linear combination fitting of every single pixel spectrum using bulk spectra of pristine and 200-cycle samples as principle components) of Mn k edge data clearly show degradation of the cathode material. Detailed interpretation of the 2D chemical distributions[4] and 3D elemental distributions[5] will be presented.

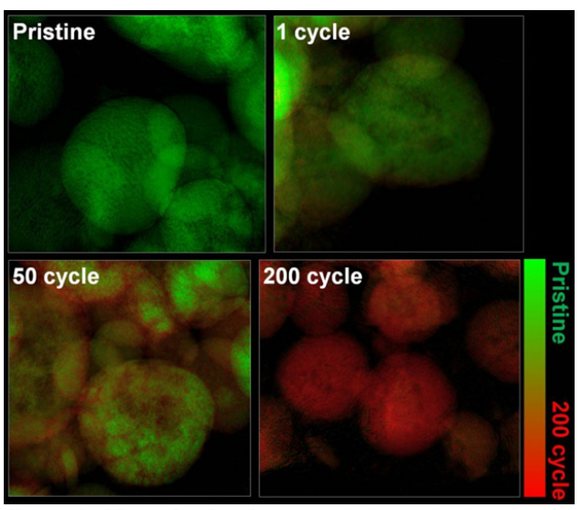

Figure 1. 2D projection images of the cathode material ($Li_{1.2}Mn_{0.525}Ni_{0.175}Co_{0.1}O_2$) with chemical sensitivity as a function of battery cycling. The size of each image is about 30×30 μm^2.

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