Structural Stability of Nickel-Rich Layered Cathode Materials

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To increase the energy density of Li batteries (including conventional Li ion batteries and solid state batteries), cathode materials with higher voltages and/or higher capacities are required, but safety and cycle life should not be compromised. Nickel-rich $LiNi_{1-x-y}Mn_xCo_yO_2$ (NMCs) can provide higher capacities and lower cost in comparison with low nickel content NMCs. In order to make nickel-rich NMCs viable for commercial lithium batteries, methods need to be developed to improve the structural stability (i.e., crystal structure, electronic structure) upon battery cycling and/or under abused thermal conditions.

In this study, NMCs with various chemical compositions were synthesized and cycled to different states of charge at various cycle numbers and voltage windows. *In situ* and *ex situ* characterizations (XRD, XAS, TXM and STEM-EELS) were performed to study the evolution of crystal and electronic structures at multiple length scales. It was found that in nickel-rich NMCs (e.g., NMC622) the structural change involved all three transition metals, in particular, the participation of nickel is much stronger than low nickel content NMCs. As the nickel content increases, the structural behavior becomes closer to $LiNiO_2$ materials, with several phase changes. The roles of cobalt and manganese are similar to those in low nickel content NMCs.