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Insights into the (de)lithiation mechanism of core-shell layered $\text{Li}(\text{Ni},\text{Co},\text{Mn})\text{O}_2$ cathode materials during cycling

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Layered $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ (NCM) oxides with core-shell morphology have been found to be prospective cathode candidates for advanced lithium-ion batteries. The electrochemical performances of NCM cathodes are tied to the transition metal relative ratios, thereby it is difficult to determine the real structure of core-shell NCM materials and to understand the synergistic effect of core and shell upon cycling. Herein, high-resolution neutron powder diffraction at the instrument SPODI was used to investigate the structure of synthesized NCM compound. The results show that the as-prepared NCM material consists of an inner Ni-rich core and a Mn-rich shell on a secondary particle level. Both core and shell possess a layered $\alpha\text{-NaFeO}_2$ -type structure with the same space group ($R\bar{3}m$) while a slight difference in lattice parameter. The (de)lithiation mechanism of core-shell NCM cathode materials was investigated by in situ synchrotron-based X-ray diffraction and absorption spectroscopy. These findings contribute to prepare layered Ni-based oxides with good electrochemical performances.

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