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Dynamic structure evolution of extensively de-lithiated high voltage spinel Li1+xNi0.5Mn1.5O4 x<1.5

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Nickel manganese spinel LiNi0.5Mn1.5O4 is considered one of the most promising cathode materials for lithium-ion batteries. On the one hand, this is because of its high energy density due to its high voltage plateau at 4.7 V, and the high rate capability due to its three dimensional diffusion network in the cubic crystal system. On the other hand, this material is interesting because additional lithium can be incorporated into the crystal system by exploiting the redox chemistry of manganese, rather than using only the nickel. This feature can be used to compensate for losses in the initial cycles or to increase the overall capacity and energy density even further.
However, the extensive lithiation to Li2.5Ni0.5Mn1.5O4 in the complete voltage range from 4.9 V to 1.5 V leads to an aging of the material and capacity fading. In order to tackle this problem it is necessary to provide answers to two questions: Are there intermediate phases that can be identified and where is lithium intercalated to when all octahedral site in the spinel are occupied, at x>2 in LixNi0.5Mn1.5O4.
By applying potentiometric entropymetry and operando XRD, we were able to reveal a simultaneous formation of Li2Ni0.5Mn1.5O4 and Li2.5Ni0.5Mn1.5O4, both with tetragonal I41amd space group, but with large c and small a and b lattice parameters. Via neutron diffraction we where able to locate the additional lithium on octahedral 8a and tetrahedral 4a positions of the distorted tetragonal phase I41amd. Furthermore, the neutron diffraction revealed a partial site exchange of lithium and nickel ions. This observation could be identified as one aging mechanism for the material by perfoming post mortem XRD measurements on aged LiNi0.5Mn1.5O4 based electrodes.

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