

***In situ* proving reaction heterogeneity during solid-state synthesis of Ni-rich layered oxide**

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Ni-rich layered oxide has been received tremendous attention as promising cathode materials in Li-ion batteries due to its high discharge capacity. However, it suffers from poor cycle life attributed to various microstructural defects. To achieve structure integrity, rational tailoring synthetic pathway based on clear mechanistic understanding is essential. Herein, we investigate synthesis process of $\text{LiNi}_{0.6}\text{Co}_{0.2}\text{Mn}_{0.2}\text{O}_2$ by transmission electron microscopy combined with *in situ* X-ray diffraction and thermal analysis. It is revealed that overall synthesis reaction is governed by competitive reactions at low temperature between intrinsic thermal oxidation at bulk and topotactic lithiation near interface, which leads to spatially heterogeneous intermediates. Moreover, we found that thermal decomposition leads to formation of internal void and nanopore which are precursors for cell degradation. We proposed quasi-equilibrium pathway to promote topotactic lithiation which demonstrates enhancement of electrochemical stability by suppression of microstructural defects. This work provides new insight for material synthesis in terms of defect engineering based on mechanistic study.