

## Boosting Reaction Homogeneity in High-Energy Lithium-Ion Battery Cathode Materials

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An intrinsic limitation of polycrystalline nickel-rich cathode materials in high-energy full-cells is discovered under industrial electrode fabrication conditions. Owing to their highly unstable chemo-mechanical properties, even after the first cycle, nickel-rich materials are degraded in the longitudinal direction of the high-energy electrode. This inhomogeneous degradation behavior of nickel-rich materials at the electrode level originates from the overutilization of active materials on the surface side, causing a severe non-uniform potential distribution during long-term cycling. In addition, this phenomenon continuously lowers the reversibility of lithium ions. Consequently, considering the degradation of polycrystalline nickel-rich materials, this study suggests the adoption of a robust single crystalline  $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$  as a feasible alternative, to effectively suppress the localized overutilization of active materials. Such an adoption, can stabilize the electrochemical performance of high-energy lithium-ion cells, which demonstrates superior capacity retention above ~80% after 1,000 cycles at 45°C.