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Ph.D. DISSERTATION DEFENSE

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Degree: Doctor of Philosophy
School/Department: Charles V. Schaefer, Jr. School of Engineering & Science / Chemical Engineering and Materials Science
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Title: High-Voltage-Stable Complex Oxide Cathodes for Advanced Lithium-Ion Batteries

Chairperson: Dr. Jae Chul Kim, Department of Chemical Engineering and Materials Science

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ABSTRACT

Over the past decade, lithium-ion batteries have evolved into indispensable energy sources for portable devices, electric vehicles, and large-scale energy storage systems. Currently, commercially available batteries predominantly utilize a limited set of cathode types composed of transition metals (TMs) and lithium (Li) frameworks. However, even as researchers continually refine these layered frameworks, they often suffer from irreversible phase transitions, oxygen release, and lattice distortion under prolonged cycling and at extremely high voltage.

In this work, we introduces a novel structure that enables a structurally consistent and uniform disordered rock salt layer on the surface of primary layered particles in ultra-high nickel (Ni > 0.95) cathodes. Numerous published literatures have demonstrated the compelling benefit of disordered rock-salt (DRX) cathodes due to their innate resilience against phase collapse and diminished reliance on lithium content for maintaining structural integrity at higher voltages. Therefore, by integrating a disordered rock-salt shell onto a layered core, we can harness DRX's remarkable tolerance to deep lithium extraction at high voltages, effectively limiting the structural damage and side reactions that typically plague Ni-rich layered oxides in theory. This approach seamlessly integrates the electrochemical functionality of disordered rock-salt (DRX) phases with the structural coherence of ordered layered frameworks, addressing persistent limitations in conventional cathode designs.

These DRX nucleated layered cathode achieves unprecedented integration of high capacity, long-term durability, and operational safety. In detail, the modified material reveals the significantly enhanced electrochemical performance in delivered capacity (up to 7 %) and capacity retention (up to 13.3 %) during long-term rigorous CCCV testing at a high operational potential. This enhancement is accompanied by suppressed side reactions and minimized irreversible phase transitions, attributed to the superior electrochemical and structural resilience of the active DRX layer. In addition, utilizing atomic scale diffraction and imaging techniques, the structural evolution reveals minimized lattice parameter variation, reduced microcracking generation, and enhanced morphological integrity, particularly under high de-lithiation states. Beyond immediate performance gains, this design framework provides a new perspective on cathode material engineering, emphasizing the importance of structural harmony and chemical resistance and opening more possibilities for broader electric vehicle adoption and enhanced sustainability.