Controlling Ni^{2+} from the Surface to the Bulk by a New Cathode Electrolyte Interphase Formation on Ni-rich Layered Cathode in High-Safe and High-Energy-Density Lithium-Ion Batteries

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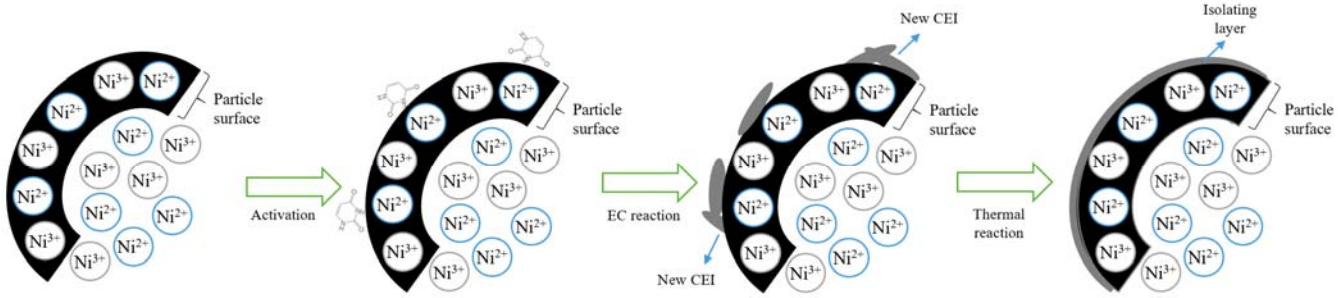
Abstract

Ni-rich high-energy-density lithium ion batteries pose great risks to safety due to internal short circuits and overcharging; they also have poor performance because of cation mixing and disordering problems. For Ni-rich layered cathodes, these factors cause gas evolution, the formation of side products, and life cycle decay. In this study, a new cathode electrolyte interphase (CEI) for Ni^{2+} self-oxidation is developed. By using a branched oligomer electrode additive, the new CEI is formed and prevents the reduction of Ni^{3+} to Ni^{2+} on the surface of Ni-rich layered cathode; this maintains the layered structure and decreases the cation mixing during cycling. In addition, this new CEI ensures the stability of Ni^{4+} that is formed at 100% state of charge in the crystal lattice at high temperature (660 K); this prevents the rock-salt formation and the overreduction of Ni^{4+} to Ni^{2+}. These findings are obtained using in situ X-ray absorption spectroscopy, operando X-ray diffraction, operando gas chromatography–mass spectroscopy, and X-ray photoelectron spectroscopy. Transmission electron microscopy reveals that the new CEI has an elliptical shape on the material surface, which is approximately 100 nm in length and 50 nm in width, and covers selected particle surfaces. After the new CEI was formed on the surface, the Ni^{2+} self-oxidation gradually affects from the surface to the bulk of the material. It found that the bond energy and bond length of the Ni-O are stabilized, which dramatically inhibits gas evolution. The new CEI is successfully applied in Ni-rich layered compound, and the 18650- and the punch-type full cells are fabricated. The energy density of the designed cells is up to 300 Wh/kg. Internal short circuit and overcharging safety tests are passed when using the standard regulations of commercial evaluation. This new CEI technology is ready and plans for future applications in electric vehicle and energy storage.

Keywords **-** cation mixing, cathode electrolyte interphase, synchrotron, internal short circuit, Ni-rich layered



Scheme 1 The proposed reaction mechanism of oligomer additive on NMC622.

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