Are Single Crystalline Ni-rich cathodes with full capacity utilization possible?

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Lithium ion batteries (LIBs) with high energy-density are in demand for portable electronic devices and electric vehicles (EVs). Since the energy density of lithium ion batteries relies on the cathode material, many researchers have focused on developing cathode materials with higher specific energy density and stable long term cyclability. In particular, the layered nickel-rich cathode materials are considered as promising cathode candidate for high energy density and low cost. High volumetric enegry density (≈ 3.4g cm^{-3}) is required to commercialize nickel-rich cathodes. However, these cathodes have been suffering from inherent structural and electrochemical degradation at high electrode density (>3.0g cm^{-3}). Single crystalline structure is essential factor to achieve high volumetric enegrgy denstity and stable cyclability.

For polycrystalline cathodes, the secondary particles are collapsed during electrode pressing process by high pressure, which increases the contact area between cathode and electrolyte. This result in the formation of resistant layer at the surface of cathode particles, for example SEI layer and cation mixing layer. Furthermore , microcracks are generated during electrochemical cycling and they hinder the lithium ion diffusion and cause capacity loss. However, single crystalline cathodes maintain the initial morpholgy after the electrode pressing process. The robustness of morphology prevents the formation of resistance layer, ensuring the stable electrochemical properties. In addition to this property, microcracks are suppressed in single crystalline cathode, resulting in the decrease in the gas evolution. The characteristics of single crystalline cathodes allow long-term cycle stability and the prevention of gas evolution.

In this work, we present single crystalline nickel-rich cathodes (≥ Ni 50%) which are developed in our group. These single crystalline cathodes exhibit long-term cycle stability and thermal stability at high electrode density (≥ 3.6g cm^{-3}). The electrochemical properties of single crystalline cathodes outperform polycrystalline cathodes. Our research will provide insights into the practical application of single crystalline nickel-rich cathodes.