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Electron Microscopy Investigation of Fading Mechanism of Electrodes

Introduction

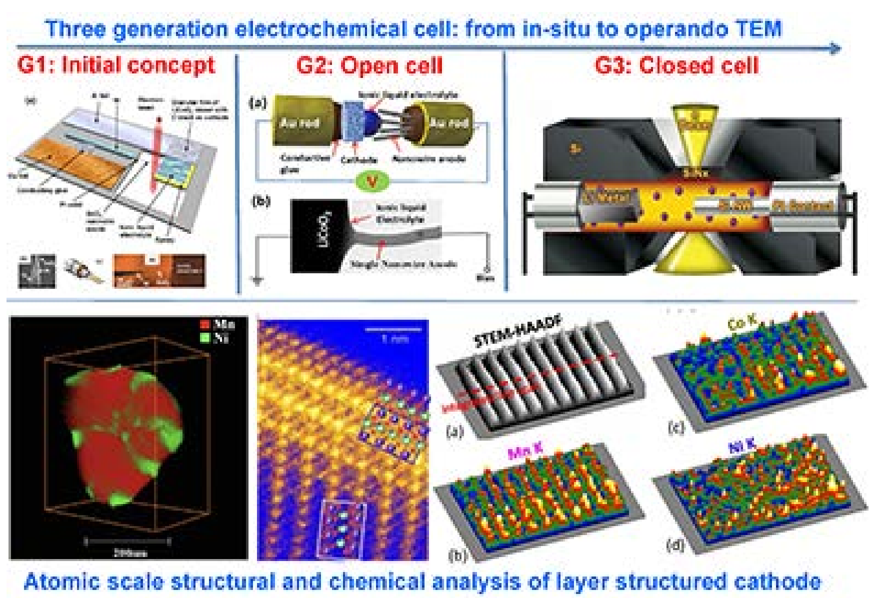
Both ex-situ and in-situ high-resolution transmission electron microscopy (TEM) are critical for probing the structure and chemistry of electrode and solid electrolyte interphase (SEI) layer, which leads to a direct correlation of microstructure/chemistry and their evolution with battery properties. The outcome of this work will provide key insight for designing of new electrode structure. Recently, we have developed new *operando* characterization tools to characterize SEI formation and electrode/electrolyte interaction using practical electrolyte that are critical for making new breakthroughs in the battery field.

Project Objective and Impact

The objective of this work is to use *ex-situ*, *in situ* and *operando* TEM and spectroscopy to probe the fading mechanism of electrode materials. The focus of the work will be on using *in situ* TEM under real battery operating conditions to probe the structural evolution of electrodes and interfaces between the electrode and electrolyte and correlate this structural and chemical evolution with battery performance. The success of this work will provide insight for designing of new electrode materials with increased the energy density of Li-ion batteries and accelerate market acceptance of electrical vehicles (EV), especially for plug-in hybrid electrical vehicles (PHEV) required by the EV Everywhere Grand Challenge proposed by DOE/EERE.

Key Achievement

- Demonstrated the first nano battery under operando TEM condition. A liquid cell based on closed window technique has been developed. This work paved the road for detailed study of SEI layer.
- Established a detailed mechanism on how the surface native oxide layer and artificial coating layer function in Si anode related to capacity fading and Coulombic efficiency.
- Investigated an artificial SEI coating layer on the lithiation kinetics of Si anode for lithium ion batteries, provide guiding information as what kind of coating layer can optimize the performance.



- Discovered that capacity/voltage fading of $Li_{1.2}Ni_{0.2}Mn_{0.6}O_2$ cathode is closely related to Ni segregation, dissolution, corrosion, fragmentation, and phase transformation of the materials.
- Stability of cathode depend on the crystallographic planes: plane with single ion species is more stable than mixed plane, providing guidance for designing of high performance cathode.
- Atomic level determination of mixing of Ni, Co, and Mn with Li layer in layered structure.
- Discovered phosphorus enrichment in the SEI layer of cathode, which depends on operating voltage and crystal structure.
- Discovered that coating layer on cathode surface can mitigate side reaction, prevent surface phase transformation, and mitigate the Mn reduction
- Established in-situ liquid SIMS and studied
- This project has leaded to 25 publications in the last five years.

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Selected Publications

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