**Multiscale imaging and computational modeling for understanding thick cathode degradation mechanisms.**

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Lithium-ion batteries operation involves coupled electrochemical-mechanical phenomena spanning multiple length scales. An in-depth understanding and quantification of the battery function and degradation mechanisms requires imaging capabilities able to provide information at a fine resolution over statistically relevant volume.

Plasma focused ion beam-scanning electron microscope (PFIB-SEM) enables statistical and rapid analyses of the microstructural transformation of large sub-volumes of composite electrodes at different cycling states. Here, by using a thick NMC811 (LiNi₀.₈Mn₀.₁Co₀.₁O₂) electrode as an example, a macro- to nanoscale 2D and 3D imaging analysis approach coupled with 4D (space + time) computational modeling to probe its degradation mechanism in a lithium-ion battery cell [¹]. Cracking of the NMC active particles increases and the contact area between Carbon/Binder and NMC decreases, correlating with the thick electrode degradation. Computational modeling indicates the CEI growth results in roughly 10% of the capacity loss.

This study unravels that the reaction heterogeneity within the thick cathode caused by the unbalanced electron conduction is the main cause of the battery degradation over cycling. The findings shed light on the crucial role of the electronic and ionic transportation networks in the performance deterioration of the thick cathode, which provides guidance for cathode architecture optimization and performance improvement during thick cathode system design process.

Reference:

1. Zhang et al., Coupling of multiscale imaging analysis and computational modeling for understanding thick cathode degradation mechanisms, Joule (2022), https://doi.org/10.1016/j.joule.2022.12.001