## A 4D Framework for Probing Structure-Property Relationships in Lithium Ion Batteries

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The modern world has an increasing reliance on portable energy storage solutions, such as lithium ion batteries (LIBs). With applications ranging from portable electronic devices to hybrid- and fully-electric vehicles, understanding the performance characteristics and failure conditions of commercial LIBs is paramount toward establishing their roles in modern society. Ensuring the safe and reliable operation of commercial batteries is of high importance in LIB commercialization, to make certain that a) the batteries will perform as expected, and b) that any failures will occur safely, with minimized risk to the consumer.

In spite of the vast role that LIBs play in a variety of applications, there remains a relatively poor understanding of when, why, and how batteries degrade and, ultimately, fail. It has been recently demonstrated that inhomogeneities in the microstructures – particularly tortuosity – can lead to unexpected behaviours [1-3]. Small anisotropies can have far-reaching effects on the performance of a battery [1], which may enhance cell degradation and even lead to failure [2]. Thus, it has been shown that studies of microstructure – and, in particular, microstructure evolution – are of critical importance toward understanding a battery's behaviour, and that 3D imaging may provide a unique pathway toward gaining this insight [1-4]. One of the more popular approaches for 3D characterization of batteries is X-ray microscopy (XRM), due to its unique abilities to achieve high spatial resolutions inside packaged structures without disturbing the packaging [5], providing the unique capability to track the fine details of the microstructure over a length of time. XRM is, thus, a non-destructive 3D imaging technique, which enables studies of microstructure evolution (so-called "4D" microscopy).

The present study sought to better understand the evolution of a packaged LIB by developing a research methodology for probing the 3D microstructure of a commercial battery at various stages of its life. A sample of six 18650 batteries was commercially sourced and the batteries were split into two experimental groups. Group A batteries were imaged as received using XRM (~2 µm voxel size), cycled until failure at a rate of 1.5C (40 minute charge/discharge), and then imaged again. Group B batteries were imaged as received, placed near to the cycling apparatus (but not cycled) in order to serve as a control group, and then imaged again. The 3D X-ray micrographs in the "fresh" and "aged" states were then aligned to each other and visually examined for signs of defects. It was observed that a) the primary failure mechanism in this case was engagement of the current interrupt device (CID), and b) that the primary microstructure evolution process at this length scale was the apparent closure of large-scale cracks after aging.

These results represent an exciting advancement in the field of battery characterization and present a unique 4D workflow for future studies. This presentation will detail the steps involved and discuss the details of what was observed in a real-world battery subjected to real-world loading conditions.

## References:

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PRISTINE CELL (CONTROL GROUP)

FAILED CELL (EXPERIMENTAL GROUP)



Figure 1. Engagement of the current interrupt device (CID) was determined to be the primary failure mechanism in the batteries under investigation.



Figure 2. 3D X-ray micrograph of a fresh battery (left) compared to the same region of the same battery after cycling to failure (right). In this case, many cracks that were observed in the fresh condition were not observed in the aged condition, suggesting a crack closure evolution process as a function of aging.