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Correlative *In Operando* Studies of Abusive Cycling Conditions for Li-ion Batteries

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Safety of lithium ion batteries (LIBs) has been a primary concern since their first appearance in commercialized products [1]. Abusive conditions such as over-charging/discharging, overheating, and internal short circuit lead to gas evolution and mechanical expansion in LIBs, and result in transformations to electrode morphology [2]. These changes potentially negatively impact electrochemical performance and even the safety characteristics of LIBs [3]. Here, we studied the use of an in-house transmission X-ray microscope (TXM) to visualize mechanical expansion and degradation of electrode stacks during the abusive cycling of LIBs [3, 4]. We also characterized the capacity fade of LIBs with Electrochemical-Acoustic Time-of-Flight (EAToF) [5] and electrochemical impedance spectroscopic (EIS) measurements.

Commercial lithium cobalt oxide (LCO) / graphite pouch cells (nominal capacity of 400 mAh and dimension of 27.0 × 27.0 × 4.8 mm) were cycled at various C rates (C/2, 1C, 2C, 3C, 4C) under constant current within a specified cut-off voltage range (upper limit: 4.38 V and lower limit: 3.0 V). EIS was performed at the top of the charge and bottom of the discharge cycles, using potentiostatic measurements with an amplitude of ± 20 mV about open circuit potential and a frequency range of 100 kHz to 0.01 Hz. A Carl Zeiss Xradia 520 Versa TXM was employed to obtain X-ray 2D radiography and 3D tomography of LIBs. Other camera settings were varied with the sample geometry. The false-color 3D rendering shows the jelly roll electrode assembly with current collectors (Fig. 1A). The high intensity regions in the TXM micrographs are dominated by materials that strongly absorb X-rays (LCO, Cu foil), and the weakly absorbing materials (graphite layer, Al current collector, separator) appear as dark regions between these strongly absorbing layers (Fig. 1B). Due to the cut-off voltage limits, the LIBs with higher cycling rates (i.e. 3C, 4C rate) do not exhibit distinctive mechanical deformations (Figs. 2A-B). Image analysis shows small displacements in the circular turn of the electrodes, and large displacements in the flat panel of jellyroll structure along the minor axis (Figs. 2C-D). Mechanical deformation of LIBs, resulting from the evolution and accumulation of gas between electrode layers, will be correlated to the capacity fade and EIS measurements during abusive cycling.

In summary, we used an in-house TXM as an efficient tool for imaging the internal structure of LIBs. We will examine multi-length scale 3D investigations of LIBs using *in operando* TXM/EAToF/EIS. X-ray microscopy can be useful to probe cell failure mechanisms for both *in situ* and *ex situ* analysis of LIB electrode architectures [6].

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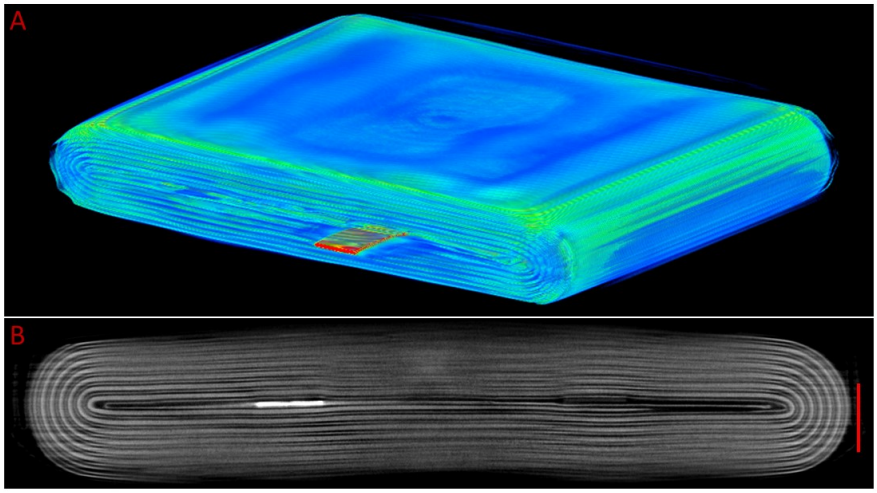

Figure 1. (A) Full 3D rendering, and **(B)** 2D cross-section of a fresh LIB pouch cell. A scale bar is 2000 µm.

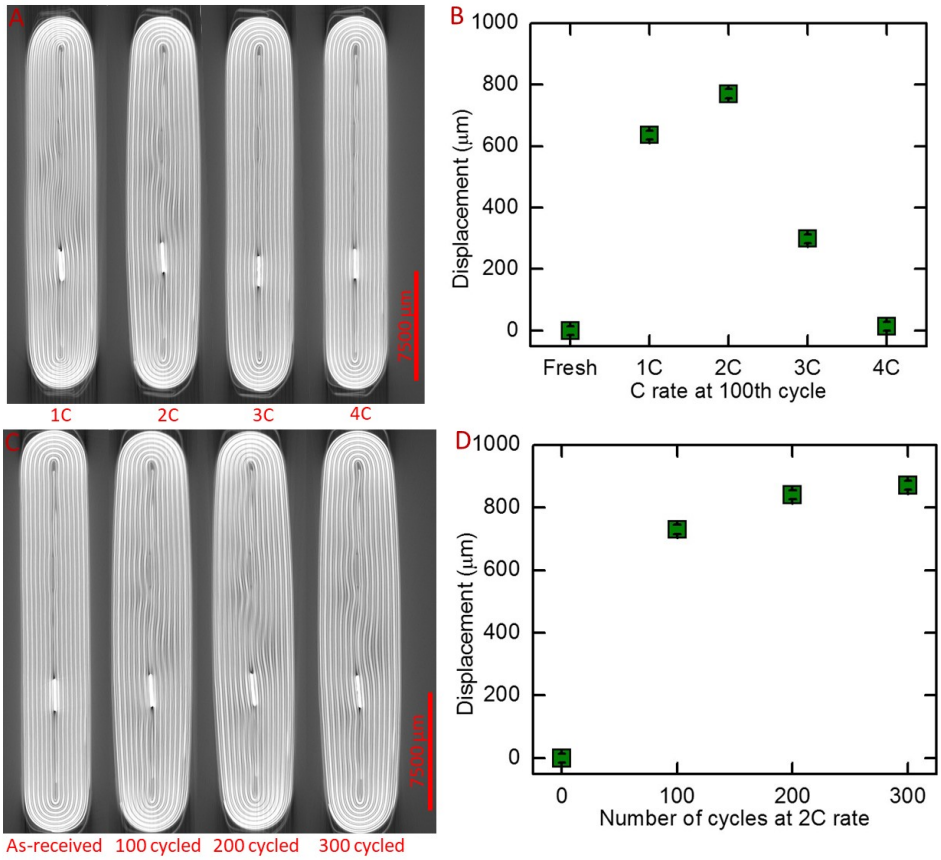

Figure 2. (A) 2D cross-sectional TXM micrographs, resolved at the same position of different LIBs, for LIBs cycled 100 times at 1C, 2C, 3C, and 4C rates. **(B)** Measured data for mechanical expansion of LIBs. **(C)** 2D cross-sectional TXM micrographs for a single LIB after 0, 100, 200, and 300 cycles at a 2C rate, resolved at the same position of the LIB. **(D)** Measured data for mechanical expansion of the LIB.

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