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Investigations of Silicon-Based Anodes for Li-Ion Batteries Using X-Ray and Neutron 3D/4D Imaging Techniques

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Silicon-based anode materials are one of the most promising approaches to further increase the energy density of lithium-ion batteries. However, Current materials are limited by poor cycling stability and rapid capacity fading, mainly caused by the massive volume expansion of Si during lithiation and subsequent strain on the material composite [1]. Furthermore, this electrode swelling also results in continuous solid electrolyte interface (SEI) growth, which hinders the migration of Lithium and leads to permanent capacity loss [2] [3].

To optimize these materials analytical techniques able to probe the local 3D morphology and Li content are necessary. By applying methods such as neutron computed tomography (NCT), X-Ray computed tomography (XCT) and synchrotron scattering micro-tomography (SMT) structural ageing and changes in the distribution of lithium in the various components can be characterized and quantified for different charging states and recharge cycle numbers.

This study investigates an anode material based on a dual phase alloy system of amorphous Silicon (a-Si) with crystalline iron silicide (c-FeSi2) and graphite [4] [5]. Li-ion battery coin cells containing this silicon-graphite composite anode material were industrially produced and aged by performing 1, 300, and 700 charge-discharge cycles, corresponding to remaining capacities of 100, 70 and 50 %, respectively. NCT and XCT scans were acquired at the NeXT instrument of the ILL in Grenoble (France) [6], and SMT measurements performed at ID31 of the neighboring ESRF. Figure 1 shows a schematic diagram of the XCT/NCT setup used to measure these complementary methods, as well as sample slices through the reconstructed volumes taken before any post processing steps.

These datasets were reconstructed and evaluated using specially developed data processing pipelines. Alignment of the datasets was achieved by following a modified digital volume correlation algorithm to handle image volumes acquired using different modalities [7]. Using this multi-modal registration process, the NCT and XCT scans were aligned and segmented to combine the complementary datasets and create a 4D model of the Li-ion battery coin cells.

Figure 2 shows a sample of the combined NCT (cyan) and XCT datasets (red). Cropped slices of the measured volumes, taken perpendicular to the beam, were averaged over 29 slices, and depicted side by side with the main components labelled. Comparing one of the highly cycled cells with 50% remaining capacity (top) with the reference cell without electrolyte (bottom), trapped Lithium can be observed in the anode layers, visible as cyan colored blotchy areas in the otherwise dark component.

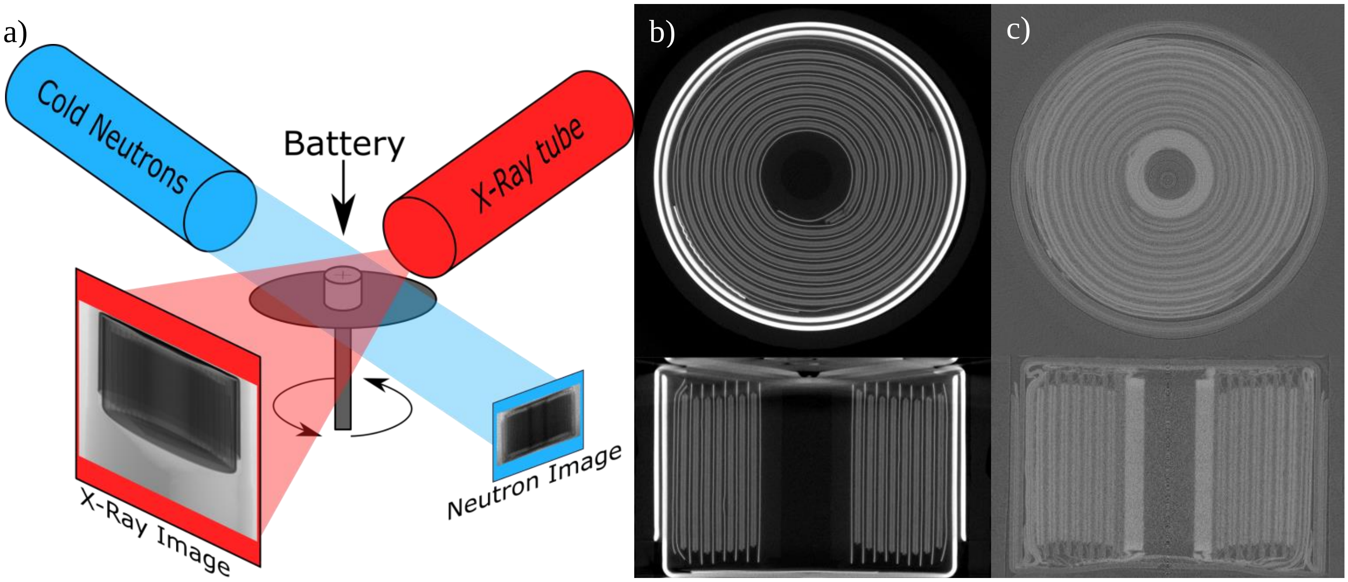

Figure 1. a) Schematic depiction of the measurement setup present at NeXT at ILL, Grenoble. The neutron guide provides a cold neutron beam exhibiting a flux of $1.4*10^{10}$ n/cm^2/s, with the peak flux at 2.8 Å. Images depicted on the detector screens represent samples of the raw Neutron / X-ray image data before corrections and reconstruction. b) / c) Reconstructed XCT / NCT volumes before post processing. Top figure shows a sample slice through the reconstructed volume, perpendicular to the beam direction. Bottom figure shows a sample slice of the volume's side view.

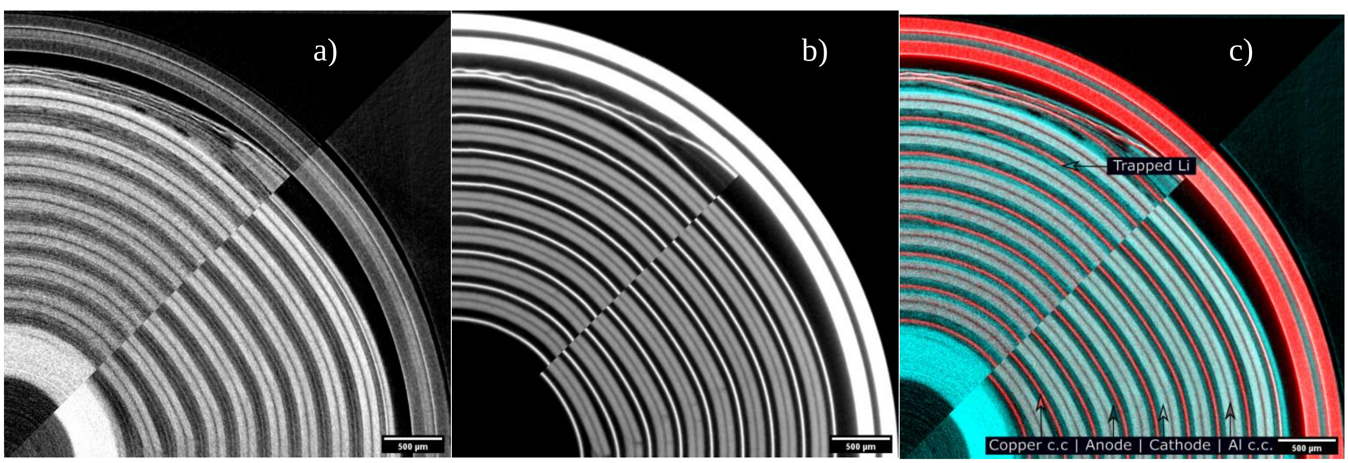

Figure 2. Averaged and cropped slices of Li-Ion battery coin cells sliced perpendicular to the beam direction. Pixel size of ~ 4.2 µm. Comparison of a highly cycled cell with 50% remaining capacity (top) with a reference cell without electrolyte (bottom). a) NCT-Slice. b) XCT-Slice of the same cell, aligned to the orientation of the NCT slice. c) Combined NCT (cyan) and XCT datasets (red) with labeled components.

The change of lithium distribution in the components between different cycle numbers and charge states in the attenuation-based tomography techniques of NCT / XCT is quantified by modelling expected attenuation parameters to the observed values. From this information, trapped Lithium can be identified and changes in the distribution are analyzed to point out possible degradation and failure mechanisms.

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