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Coupling Neutron Tomography with X-Ray Absorption and Scattering Tomography to Resolve Damage in Lithium Ion Batteries

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Neutron based techniques are uniquely interesting to study Lithium-Ion Batteries (LIBs). The direct sensitivity of neutrons towards lithium nuclei offer a plethora of opportunities to investigate the complex processes interacting to allow these devices to store electrical energy. Neutron computed tomography (NCT) in particular is interesting, as it allows one to directly map the distribution of lithium inside cells.

Thanks to the non-destructive nature of neutron methods, it further becomes possible to perform complementary techniques as well. By coupling NCT with X-Ray based Computed Tomography (NXCT) it becomes possible to segment different components by leveraging differences in the attenuation coefficients for elements with respect to neutrons and X-Rays. Further, diffraction/scattering based techniques can be employed as well, which offer a view into the processes occurring on the atomic/nano-scale. By utilising Small- and Wide-Angle X-Ray Scattering Computed Tomography (SWAXS-CT), which is capable of spatially resolving scattering properties in two dimensions, another source of insights can be gathered to aid in the study of LIBs.

In this work we measure aged industry-grade silicon-based LIBs with NXCT to investigate where lithium is located in degraded cells. This is combined with *operando* SWAXS-CT measurements performed on the same cell to map the crystallographic properties and match them with features observed in NXCT. Combining these techniques allowed us to probe peculiar zones inside the cells where the current collector has been massively deformed. We identify and characterise the resulting changes to the lithiation behaviour through localised *operando* scattering analysis and match the observations with insights gained from the neutron-based analysis. This correlated methodology ultimately enabled us to resolve the origins of these defects and offers valuable insights for manufacturers (1).

(1) E. Lübke et al, in review

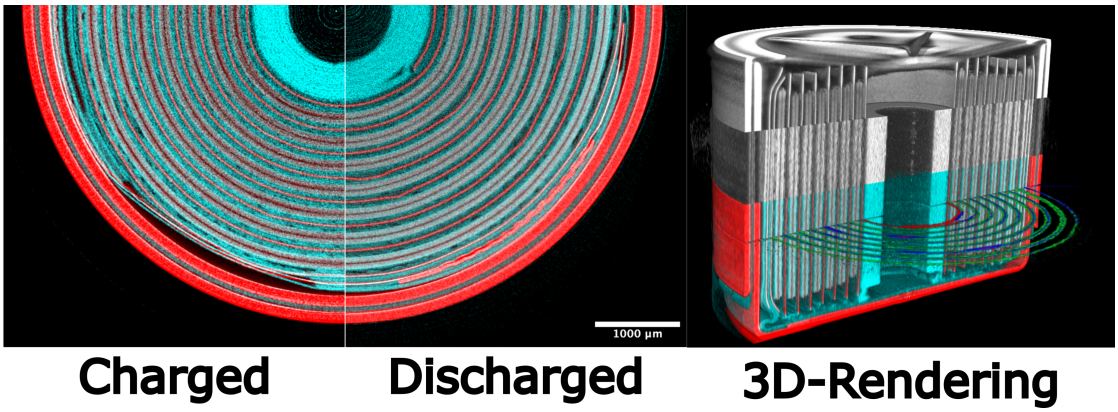

Figure 1: Left: Comparison of the same aged cell in the same position measured with NXCT in the discharged and charged state. Cyan = neutron attenuation component, red = X-Ray attenuation component. Right: 3D-rendering of NXCT data, with WAXS-CT data drawn in the measured position. Color-coded to reflect the lithiation state of graphite. Green = LiC6, Blue = LiC12, Red = Graphite with partially lithiated graphite phases.

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