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New insights in battery electrolyte behavior during cycling and heating of batteries using dynamic micro-CT

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In this work, we present unique results of dynamic micro-CT to study the behavior of Li-ion batteries during two important events that determine battery quality and lifetime. First, we show the interactions between different battery components during the first few charge and discharge cycles of a battery, the so-called formation cycles. How these cycles are performed has a huge impact on the lifetime, capacity, and overall quality of Li-ion batteries. During these cycles the solid-electrolyte-interface (SEI), a thin layer on the surface of the electrode material, is formed. This SEI is formed by the reaction between the electrode and the electrolyte and serves as a layer where Li+ ions can be embedded and removed during further cycling of the battery. This makes optimizing the formation cycles one of the most important research topics in battery production. Since the SEI is very thin (100 nm), high-resolution methods such as (FIB)SEM are used to study it. Although dynamic micro-CT cannot visualize the actual SEI layer, it can be used to visualize 3D electrolyte movement in real time and study swelling and shrinking of the entire battery during cycling. For the experiment, 15 x 25 cm large pouch cells were pressurized and mounted in the TESCAN DynaTOM, a unique micro-CT system with a rotating gantry to allow for complex in-situ experiments. Using an externally controlled potentiostat, several charge and discharge cycles were programmed with specific charging speeds, and several high-quality static (60 minutes per scan) and fast dynamic (2 minutes temporal resolution) micro-CT scans were performed at 13 predetermined intervals during the 28-hours procedure. In this work, we show results on 4D electrolyte movement, gas formation and structural dynamics in these pouch cells.

In a second experiment, the behavior of cylindrical cells under increased temperatures was observed. Elevated temperatures as low as 60 $^{\circ}$ C already have a negative effect on lifetime and will result in degradation and irreversible damage. To observe the structural effect of increased temperatures, a 26650 cell was heated up to ~60 $^{\circ}$ C using Peltier elements powered through the slipring of the scanner's rotation stage. In a series of dynamic and time-lapse micro-CT scans, the movement of the electrolyte in and out of the electrode layers was observed in 3D. Novel machine learning protocols enable segmentation of the different battery components (cathode, anode, electrolyte, and gas) and follow their movement over time. In further experiments, the effect of higher temperatures, and faster or longer heating will be examined.

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