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Investigating the Formation and Lithiation of Silicon-based Anodes by Neutron Depth Profiling

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Silicon anodes are prominent candidates to enhance the capacity of Lithium Ion Batteries. However, the volumetric changes of silicon upon (de-)lithiation regularly lead to a low life time of such electrodes. Our strategy to prolong the life time is based on a partial lithiation of silicon to ~30%, which significantly increases the cycling stability while maintaining a three times higher gravimetric capacity compared to common graphite anodes. In our study, we use Neutron Depth Profiling (NDP) on extracted silicon-based anodes to acquire quantitative lithium depth distributions after different formation and lithiation steps. This can help to understand the performance of the electrodes and to identify lithium-containing side reactions which are invisible for electrochemical methods. We investigated three states of charge (SOC) and the formation with and without $LiNO_3$ as electrolyte additive, which was found to increase the cycling stability of silicon. Our results show that lithium is evenly distributed in depth across all studied anodes. The formation already leads to a moderate lithium concentration, which originates from irreversibly bound lithium in the as-formed solid-electrolyte-interface (SEI). With rising SOC (15%, 30%) the lithium concentration consistently increases and a sizable swelling of the electrodes is observed. Notably, the $LiNO_3$ electrolyte additive elevates the lithium content in the SEI, which indicates additional reactions in the formation process.

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