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Understanding the Lithium Depth Profile Upon Lithiation of Extracted Silicon-Based Anodes

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A prominent strategy to increase the capacity of Lithium Ion Batteries is the use of silicon as anode material. However, the volumetric increase of silicon upon lithiation regularly results in a low cycling stability of the material. Our strategy is based on a partial lithiation of silicon to ~30%, which leads to a significant gain in cycling stability while maintaining a high capacity. The knowledge of the lithium distribution across such silicon electrodes is crucial to assess their behavior in working cells. In our study, we use Neutron Depth Profiling (NDP) on extracted silicon-based anodes to reveal the lithium depth distribution after different formation and lithiation steps. Thereby, we investigated three states of charge (SOC) and the formation with and without LiNO_3 as electrolyte additive, which can significantly increase the cycling stability of silicon. Our results show that lithium is evenly distributed in depth across all studied electrodes. The formation already leads to a lithium concentration of $\sim 9 \text{ mol/cm}^2$, which is inferred to be a consequence of lithium which is irreversibly bound in the as-formed solid-electrolyte-interface (SEI). With increasing SOC (15%, 30%) the lithium concentration consistently increases. Also, we observe a significant swelling of the electrode during the lithiation process. Notably, the LiNO_3 electrolyte additive leads to a higher lithium content in the SEI, which is a first indicator of differences in the formation process.

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