QUANTITATIVE LITHIATION DEPTH PROFILING IN SILICON CONTAINING ANODES INVESTIGATED BY ION BEAM ANALYSIS

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The localization and quantitative analysis of lithium (Li) in battery materials, components, and full cells are scientifically highly relevant, yet challenging tasks. The methodical developments of MeV ion beam analysis (IBA) presented here open up new possibilities for simultaneous elemental quantification and localisation of light and heavy elements. It describes in detail the technical prerequisites and limitations of using IBA to analyse and solve current challenges in Li-ion and solid-state battery related research and development. Nuclear reaction analysis and Rutherford backscattering spectrometry can provide spatial resolutions down to 70 nm and 1% accuracy. To demonstrate the new insights gained by our technique, Silicon (SiO_x)-containing Carbon anodes are lithiated to six states-of-charge (SoC) between 0-50% in a 2032 coin-cell setup as shown in Fig. 1.

The quantitiative Li depth profiling of the anodes shows a linear increase of the Li concentration with SoC, as shown in Fig. 2, and a match of injected and detected Li-ions. This unambiguously proofs the electrochemical activity of Si. Already at 50% SoC, we derive C/Li=5.4 ($< LiC_6$) when neglecting Si, proving a relevant uptake of Li by the 8 atom % Si (C/Si≈9) in the anode with Li/Si≤1.8 in this case. Extrapolations to full lithiation show a maximum of Li/Si=1.04±0.05. The analysis reveals all element concentrations are constant over the anode thickness of 44 μm, except for a ~6 μm thick separator-side surface layer. Here, the Li and Si concentrations are a factor 1.23 higher compared to the bulk for all SoC, indicating preferential Li binding to SiO_x. These insights were so far not accessible with conventional analysis methods and are a first important step towards in-depth knowledge of quantitative Li distributions on the component level.
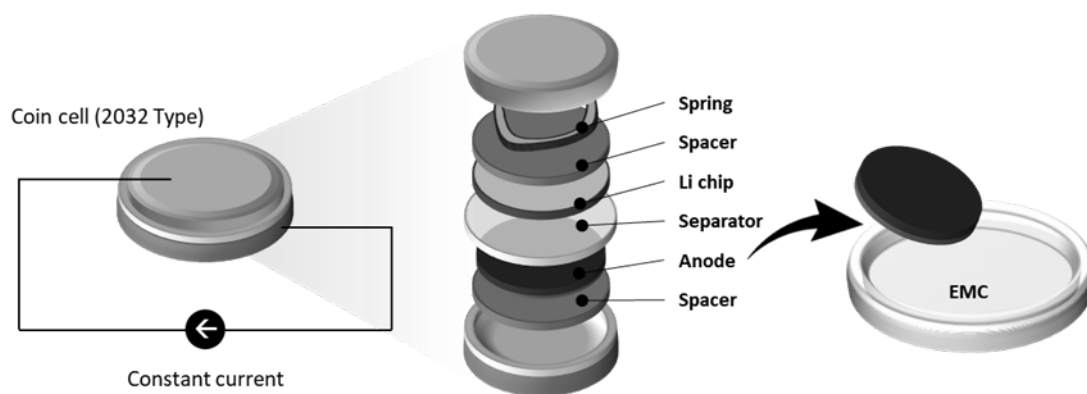
FIG. 1. Sample preparation/lithiation in a coin cell setup for later post-mortem analysis through IBA.
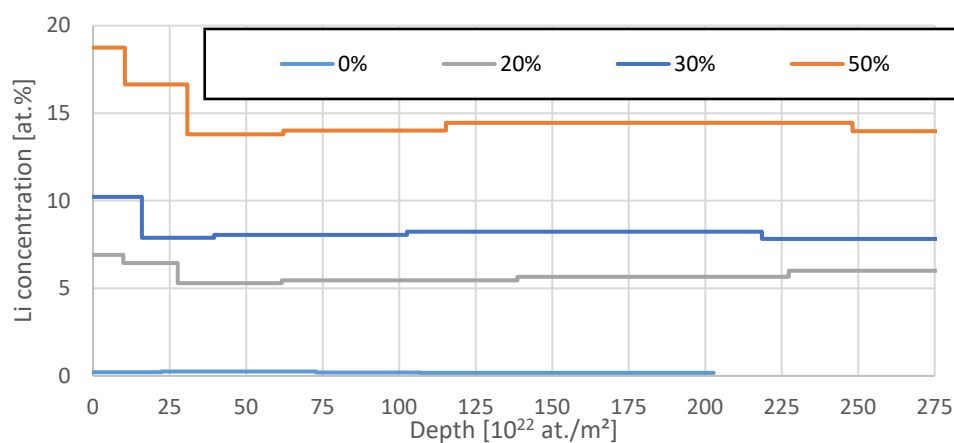

FIG 2: Comparison of the Li depth profiles. The first ~6 μm are enriched with Li. The region >200*10²² at./m² has to be considered with care due to the dominance of the pile-up signal in this deep region.