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Combined neutron depth-profiling with ion-beam analysis study of Li surface gradients in Li-garnet solid electrolytes from all-solid-state-batteries

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Knowledge of surface and interface interaction is crucial for future next-generation battery concepts like all-solid-state batteries (ASSB). Spatial and time dependent Li distribution plays a significant role in understanding performance, aging, and failure mechanisms. However, Li detection remains a challenging task and only few techniques enable measuring its spatial distribution in battery components or even full cells. Non-destructive techniques are even more scarce. To our knowledge only neutron depth-profiling (NDP) and Ion-Beam-Analysis (IBA) are capable of absolute Li quantification under the given sample conditions. Both techniques originate from the utilization of nuclear processes to gain information about the nuclei present in the sample. However, due to the nature of the two approaches they differ in their resolving capabilities. IBA has a high lateral resolution but is rather limited in depth resolution, while NDP is conversely having a high depth but low lateral resolution. Thus, a combination of both methods would make the best of both worlds. Our study combines both techniques on identical samples of Li-ASSBs and allows us benchmark for the first time the strength and drawbacks of the two methods. We derive depth dependent Li-concentrations and validate a microstructural model of charge, discharge, and relaxation of ASSBs together with an electrochemical analysis. The work shows the fundamental advantages of such a combined approach to optimize materials and battery cells for ASSBs.

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