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Dynamic lithium distribution in 18650-type Li-ion batteries on multiple length scales

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Lithium-ion batteries as energy sources in different kinds of portable electronics and electric vehicles play an important role in modern society. Permanent demand for improved energy/power densities and longevity results in the increasing engineered complexity of Li-ion batteries on different scale levels. Such complexity requires the application of non-destructive probes for characterization of batteries.

In-operando investigations of lithium-ion batteries (as closed electrochemical systems) are often not trivial as only a few methods exist to probe the conditions of the active materials locally in a non-destructive way. In such context diffraction-based methods are very popular in battery research (especially when combined with electrochemistry), either in *ex-situ* or *in-operando* modes.

In the current work the dynamic lithium distribution in a high-power 18650-type lithium-ion battery has been investigated on multiple length scales using both neutron and synchrotron based X-ray diffraction. The lithium distribution during dis-/charging has been investigated using spatially-resolved neutron powder diffraction on the graphite anode. Higher-resolution *in-situ* investigation in the fully charged state on both anode and cathode has been performed applying synchrotron-based X-Ray diffraction computed tomography. Finally, the dynamic lithium concentration in a single cathode layer has been investigated using synchrotron radiation with μm -sized beam. Non-uniform lithium distributions were observed on all investigated length scales, which was attributed to the complex character of current distribution in the studied high-power Li-ion battery. Such knowledge is highly relevant for further improvement of current state-of-the-art lithium-ion batteries in terms of effective use and understanding of issues related to enhanced power fade, fatigue and stable operation.

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