

## Electrochemical Energy Storage - Insights from *in operando* Measurements

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Intermediate energy storage is one of the most important challenges for a successful transition into a sustainable energy technology based on fluctuating primary sources. Electrochemical energy storage is of high relevance for this purpose, because the high values for the energy efficiencies of the one-step transformations between electrical and chemical energy forms cause only low losses. The key performance indicators of electrochemical energy storage devices depend mainly on the properties of the battery materials used and their specific behavior in a battery cell. Therefore, a systematic optimization of batteries requires a solid knowledge of the working and degradation mechanisms of the involved materials during regular operation conditions. The highly reactive environments inside a cell and the pronounced interactions between the individual components require investigations by dedicated comprehensive and complementary *in operando* methods, supported by *post mortem* studies. This contribution presents and discusses *in operando* techniques using X-ray, synchrotron and neutron radiation with a focus on diffraction and their role in the development of battery materials. Selected examples are shown for Li-ion batteries but also for electrochemical energy storage “beyond lithium”, like Na- and Mg-batteries. During charge and discharge ions are alternately inserted and extracted from host structures accompanied with oxidation and reduction reactions. The underlying crystal structure is essential for the ionic transport properties, and the structural response during cycling reflects the working mechanism and changes during ageing and fatigue. Most reliable data are obtained from high-quality commercial cells, but sometimes specially designed test cells are used, especially at low technology readiness levels. Therefore, different types of *in operando* cells are also compared.

Capabilities and challenges of *in operando* measurements are compiled and discussed.

This work contributes to the research performed at CELEST (Center for Electrochemical Energy Storage Ulm-Karlsruhe) and was funded by the German Research Foundation (DFG) under Project ID 390874152 (POLiS Cluster of Excellence).