Diffraction-based studies of lithium distribution in 18650-type Li-ion cells at multiple length scales

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The problem of energy storage is a key challenge we are facing in 21th century. Our daily life has been changed by the development of electrochemical energy storage, ranging from Volta cells through lead-acid batteries to complex hybrid storage solutions explored nowadays. Lithium-ion batteries became an important part of this chain immediately after their commercialization in 1991 and since then they underwent a rapid development. Their outstanding energy/power density compared to other electrochemical energy storage systems available at the market make Li-ion batteries dominating in the segment of energy storage for portable electronics and electric drivetrains. Despite its overall popularity and widespread, the Li-ion technology has a high improvement potential, especially in the aspects concerning power and energy density, power fading, safety etc.

The safety of Li-ion cells depends on a variety of microscopic factors maintaining the integrity of the cell. Chemical, mechanical and morphological uniformity of the cell components is crucial for optimization, estimation and prediction of cell parameters and cell behavior during standard operation and misuse. “Real-life” cells are complex, closed systems exhibiting non-uniform distributions of lithium, electrolyte, salt or other components that highly depend on the state-of-charge (SOC) of the cells and properties like capacity, current, temperature, pressure, etc. Cell aging leads to the development of a heterogeneous distribution of battery parameters, which are often hard to predict by modeling and require experimental input based on complex characterizations.

Selected methodologies for probing the uniformity of cell components are briefly reviewed in Ref. [1]. A number of microscopy, spectroscopy or scattering-based techniques can be applied for *ex-situ* or *post mortem* analysis of cell components, mainly active electrode materials. If non-destructive methods have to be applied (due to closed system features), imaging and diffraction experiments using either neutrons or synchrotron radiation are the optimal choice to examine instrument-adopted test cells or commercially available large format Li-batteries. In recent years a number of methods to combine imaging and diffraction have been developed, either in the form of Bragg-imaging, spatially-resolved diffraction or computed tomography using an X-ray diffraction signal (XRD-CT) instead of attenuation (X-ray CT). The XRD-CT method has created a kind of new non-destructive insight with unprecedented sensitivity and contrast and is currently under active development.

In the current contribution a series of diffraction-based methods applied to probe inner structure of cylinder-type Li-ion batteries and lithium distribution will be reviewed in line with standard bulk high-resolution neutron powder diffraction. The methods will include applications using neutron scattering and high-energy photon beams, specifically spatially-resolved diffraction using radial oscillating collimators and conical slits along with XRD-CT and neutron diffraction –CT at multiple length scales.

[1] Mühlbauer M.J., Schökel A., Etter M., Baran V., Senyshyn A. Probing chemical heterogeneity of Li-ion batteries by in operando high energy X-ray diffraction radiography. *Journal of Power Sources*, *403*, 49-55 (2018).

[2] Petz D., Muhlbauer M.J., Baran V., Schokel A., Kochetov V., Hofmann M., Dyadkin V., Staron P., Vaughan G., Lienert U., Muller-Buschbaum P.,Senyshyn, A. Lithium distribution and transfer in high-power 18650-type Li-ion cells at multiple length scales. *Energy Storage Materials*, *41*, 546-553 (2021).