Spatial lithium distribution in Li-ion batteries revealed *in situ* by neutron scattering

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Energy storage media based on different technologies have gained in importance for a wide field of applications ranging from supplying portable devices to large electric vehicles. In recent decades the energy storage technology, based on lithium ions, dominates to the market due to the best compromise between energy and power densities (both gravimetric and volumetric), low self-discharge when not in use, tiny memory effect *etc*. Despite the overall success of Li-ion technology, a further progress permanently demands for lower-cost, longer-life, higher energy/power density batteries resulting in active development and research to the field. Nowadays modern Li-ion batteries are sophisticated electrochemical devices, possessing numerous degrees of freedom (chemical, morphological, transport) along with complicated geometries of the electrode integration. This along with the need to minimize the risks for possible materials oxidation, electrolyte evaporation, cell charge changes *etc*. requires new dedicated experimental techniques capable to reveal "live" information about processes occurring inside the cell. In such instance neutron scattering is already a well-established experimental technique for the characterization of Li-ion batteries[1]. Neutron scattering methods when combined with electrochemical characterization undergo an increasing relevance for studies of lithium-ion based electrochemical energy storage systems on different length scales, e.g. neutron imaging, reflectometry, small-angle neutron scattering, quasielastic neutron scattering and powder diffraction. Nowadays *in situ* experiments with neutrons are performed on different self-developed/special test cells and commercial Li-ion cells of diverse designs depending on the research targets and needs.
Simple in principle, but complicated in practice, designs of modern Li-ion batteries may result in spatial inhomogeneity of current, lithium or electrolyte distribution, which are often difficult to quantify, but they will surely affect performance, cycling stability and/or safety. Despite the increasing popularity of neutron scattering studies of batteries at their operating conditions the problem of cell non-uniformity (indirectly pointed by electrochemical measurements) and its effect is often not properly accounted in literature. Here a combination of three neutron-based experimental techniques, namely computed neutron tomography, high-resolution neutron powder diffraction and spatially-resolved neutron powder diffraction, applied *in situ* for studies on commercial Li-ion cells of the 18650-type will be presented along with results of electrochemical studies. The details of the cell organization on different length scales and its evolution on various factors like state-of-charge, temperature and fatigue will be presented in light of 3D lithium distribution in cylinder-type Li-ion batteries[2-3].

References:

[1] H. Ehrenberg, A. Senyshyn, M. Hinterstein, H. Fuess, in: E.J. Mittermeijer & U. Welzel (Eds.), Modern Diffraction Methods, Weinheim: Wiley-VCH, 2012, pp. 491-518.
[2] A. Senyshyn, M. J. Mühlbauer, O. Dolotko, M. Hofmann, H. Ehrenberg, Sci. Rep. 5 (2015) 18380
[3] M.J.Mühlbauer, O. Dolotko, M.Hofmann, H.Ehrenberg, A.Senyshyn, J. Power Sources 348 (2017) 145-149