in operando Neutron Scattering Studies on Commercial Li-ion Batteries

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Energy-storage systems for the power train in electrical vehicles require both high specific power and energy densities simultaneously. Li-ion batteries appear today as the most promising concept for this application. The necessary high currents are limited by the Li-ion mobility, especially in the solids, and sufficiently high currents are very often only obtained for nanosized particles. However, the huge surface area of nanoparticles in combination with high cell voltages results in pronounced surface reactivity and degradation phenomena. Therefore, complex electrode systems are used, which enhance Li-ion transport by morphology and texture, stabilize the interface between solid electrode particles and electrolyte by coatings and ensure sufficient electronic conductivity by admixtures of carbon or other additives. The huge number of degrees of freedom and the pronounced interaction between different components make a purely empirical optimization of battery materials a not very promising approach. A key for new Li-ion batteries with enhanced performance is the understanding of the underlying processes and relevant fatigue mechanism. This knowledge will allow for an identification of the limitations of proposed materials and to develop strategies for improved materials concepts. The complexity of complete energy storage devices calls for new dedicated methods to reveal the relevant mechanisms and processes under real operation conditions. Neutron diffraction and tomography are non-destructive methods, very well suited for such *in operando* studies of commercial Li-ion batteries. The ability of neutron scattering to localize light atoms (e.g. Li, C, O) and the high penetration depth of neutrons are the most important aspects of this unique tool. In addition, the scattering contrast between transition metals with nearly the same non-resonant X-ray scattering factors allow for a distinction of their site-specific occupation numbers. The latter aspect is for example essential to reveal the Mn- and Ni-distribution in $LiNi_{0.5}Mn_{1.5}O_4$ and to distinguish the primitive and the face-centred cubic polymorphs with different electrochemical behaviours [1].

In this talk a combined neutron diffraction and tomography study on commercial 18650-type Li-ion batteries will be presented. The *in operando* neutron scattering experiments have been performed at the experimental stations SPODI and ANTARES, respectively, at the research reactor FRM-II in Garching near Munich, Germany. The investigation of different states of charge of identical batteries, a "fresh" and a "fatigued" one in comparison, does not only elucidate the underlying mechanisms during reversible extraction and insertion of Li-ions, but also their changes at high cycle numbers and, hereby, reveal the relevant degradation phenomena.

References

[1] A. Bhaskar, N. N. Bramnik, A. Senyshyn, H. Fuess, H. Ehrenberg, *Synthesis, characterization and comparison of electrochemical properties at room temperature and elevated temperature for $LiM_{0.5}Mn_{1.5}O_4$ (M = Fe, Co, Ni) as cathodes for high voltage lithium-ion batteries*, J. Electrochem. Soc. 157 (2010) A689-A695.