Dienstag, 4. Juni 2013 | Tuesday, 4th June 2013
09:00 – 09:45 Uhr

Cathodes – Technology Review

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Abstract. Components in batteries cannot be optimized individually. Very pronounced materials interactions require a comprehensive consideration at least on cell level. Therefore, this contribution reports on *in operando* characterization of Li-ion batteries by neutron radiation [1,2] with a focus on the positive electrodes (cathodes) and operation temperature. $LiCoO_2$ was already used in the first commercialized Li-ion battery by SONY in 1990, but is still the most frequently used cathode material. $LiCoO_2$ is intrinsically unstable in the charged state, especially at elevated temperature and in the overcharged state. Several advantages like better high-temperature stability, less sensitivity against slight overcharges and the applicability of cheaper raw materials are reached by replacing Co partially by Mn, Ni and Al. A very important point addresses the composite formation with binders and electronically conducting additives. These cathode mixtures need to ensure sufficient electronic contact between the particles and the current collector. Volume changes and distortions in dependence on the state of charge are a challenge for the mechanical integrity of the electrode and contribute to lifetime limitations by crack formation and contact losses. Such mechanical aspects are sometimes underestimated, but play an important role for most of the cathode materials like layered oxides $LiMO_2$ (M = Ni, Co, Mn, Al), but also for the cubic spinels, derived from the parent material $LiMn_2O_4$. Both volume changes and transport behaviour require nanostructured electrode concepts. This is a considerable challenge for process technology with respect to both surface coating and handling of nanoparticles. Electrode coating is established for micrometer-sized particles. Different compositions of electrode mixtures and processing conditions are needed for "nano powder", which are difficult to find and optimize individually for each new electrode material. The most promising strategy is an additional transformation of nanosized primary particles into secondary particles with micrometer sizes, preserving the intrinsic nano-composite structure and properties. Carbon-coated $LiFePO_4$ nanoparticles are an example, where the transformation into much larger secondary particles was successful. This "conditioning" enables the application of process steps, established for electrode mixtures with micrometer-sized particles. The highest economical impact might have an increase of the specific energy density of Li-ion batteries, which are mainly limited by the positive electrode materials. Besides new cell chemistries, which require major changes on a long-time scale, the reduction of inactive components could contribute significantly. Much thicker electrodes (above 1 mm) than the layers used at present (< 200 µm) have the potential for a real breakthrough. However, novel composite or hybrid concepts are first needed, which overcome the transport limitations for high power in such "thick electrodes". In addition to the expected gain in energy density a route to lower-cost production is offered by this approach.