

TOPIC: Cell level – any technology

Effect of silicon fraction and polymer molecular structure on the electrochemical performance of high-capacity lithium-ion cells

K. Pesta¹, R. Gordon², A. Smith², N. Willenbacher¹

¹Karlsruhe Institute of Technology (KIT) – Institute for Mechanical Process Engineering and Mechanics – Applied Mechanics (MVM-AME), Gotthard-Franz-Str. 3, D-76131 Karlsruhe

²Karlsruhe Institute of Technology (KIT) – Institute for Applied Materials – Energy Storage Systems (IAM-ESS), Hermann-von-Helmholtz-Platz 1, D-76344 Eggenstein-Leopoldshafen

Silicon – with its low operating potential and much higher theoretical capacity than graphite – is assumed to be the anode material of choice for next generation lithium-ion batteries. However, silicon undergoes huge volume changes during lithium intercalation, deteriorating the mechanical integrity of the electrode and resulting in a significant capacity loss during cell cycling. These issues render the introduction of silicon-based anodes as state of the art particularly challenging. Several research activities focused on the compensation of these volume fluctuations using different binder types and concentrations. However, these efforts have proven only partially successful. Therefore, we aim at a proper selection of binder system and silicon concentration offering a fair trade-off between high capacity and low electrode degradation.

To this end, water-based silicon/graphite composite anodes with various mass ratios of silicon to graphite were prepared. Sodium carboxymethyl-cellulose (CMC) and styrene-butadiene rubber (SBR) were chosen as binder system, whereby CMC of differing molecular weight (250 kDa, 400 kDa, 700 kDa) were used. The adhesive and cohesive strength of the electrode layers was determined using 90°-peel tests and compression tests, respectively. In order to evaluate the electrochemical behavior at electrode level, the prepared anodes were paired with lithium metal, assembled to half-coin cells and cycled. Long-term cycling tests were performed using pouch cells comprising a commercial NMC811-based cathode. Additionally, the degradation effect of electrodes was investigated using post-mortem analysis.

Based on the mechanical tests, the adhesive strength of the electrode layer to the copper current collector shows no dependence on CMC molecular weight, indicating that solely SBR contributes to the bond strength at the interface. Interestingly, the adhesive strength increases upon addition of silicon. In contrast, higher CMC molecular weight and decreasing silicon content enhance the cohesive strength of the electrode layer. Results at half-cell level yield a more pronounced capacity loss for anodes including higher silicon fractions as well as anodes based on CMC of lower molecular weight. Cycling test results with selected anodes paired with high capacity cathodes exhibit remarkable specific capacity values and a stable cycling performance. This demonstrates that the mechanical properties of electrode layers directly influence the long-term electrochemical performance. Besides, the post-mortem analysis of the cells confirmed the stronger degradation of the composite electrodes based on CMC with lower molecular weight. This indicates that higher CMC molecular weight is indeed beneficial for electrochemical performance of silicon/graphite composite anodes.