Degradation Phenomena in Silicon-Carbon Composite Anodes from Industrial Battery Grade Silicon

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Silicon is a remarkable material - even in lithium ion battery technology it seems to find its utilisation. Silicon can potentially store ten times more lithium (3579 mAh/g) than the conventionally used graphitic carbons (372 mAh/g) [1]. But, as is often the case, great performance in one parameter comes with great costs in other parameters. The large lithiation capacity of silicon comes with extreme volume change during lithiation and delithiation, which severely impacts the materials cyclability. Typical mitigation methods for this involve using nanostructured silicon [2-3] , optimisation of binders [4] and adding appropriate SEI forming electrolyte additives [5].

Silicon as anode material has now grown to a mature field, and many degradation phenomena has been explored in detail, in particular on nanostructured silicon using powerful in-situ TEM studies [6]. For current Li-ion batteries, improvement of the anode capacity beyond approximately 1200 mAh/g has negligible impact on the overall cell capacity. Thus, composite anodes containing both silicon and a conventional anode material such as graphite, are sufficient for most batteries. While degradation mechanisms of pure nano-silicon structures have been studied in detail, similar phenomena specific to composite silicon-carbon electrodes have not. In our work we have performed post-mortem FIB-SEM and TEM studies to investigate degradation occurring over a large number of cycles, examining the effect and interplay between the graphite and the silicon in composite anodes. In this work, we have explored different degradation phenomena observed in silicon-carbon composite anodes, including electrode thickening, silicon migration, electrochemical sintering, dentritic surface formation, inhomogeneous lithiation and dependence of electrode thickness on lithiation level.

The composite silicon-carbon anodes are based on industrial battery grade silicon produced by Elkem AS, a world leading company for environment-friendly production of silicon. The electrochemical performance was determined by cycling of half cells where the working electrode was made by mixing a silicon-carbon composite powder with an organic binder in an aqueous slurry and coated on a dendritic Cu-foil, and the counter electrode was lithium metal. Structural properties and degradation mechanisms were examined by electron microscopy (SEM, FIB-SEM, TEM) and XRD. Support for this work was provided through the ENERGIX program of the Research Council of Norway.

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