A combined SIMS and XPS Study on the Mechanism of Amorphous Silicon Electrode Lithiation in Li-Ion Batteries

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Amorphous silicon is a promising high-capacity anode material (4200 mAh/g) for the next generation of lithium-ion batteries. Kinetic processes at the electrodes and the mechanism of lithium incorporation/removal during charging and discharging cycles play a key role for an understanding and optimization of these batteries.

In the present study we measured the modification of lithium distribution taking place during galvanostatic lithiation of about 600 nm thick amorphous silicon film electrodes at low current densities of about 30 μA/cm^2 (C/13) by Secondary Ion Mass Spectrometry (SIMS) and X-ray Photo Electron Spectroscopy (XPS). The silicon electrodes were produced by ion-beam sputter deposition. The results indicate a two-step lithiation procedure, where first the electrode is transformed into a homogeneously lithiated phase with a low Li content of about $Li_{0.25}Si$. During later stages of the lithiation process, the results indicate an inhomogeneous penetration of a highly lithiated phase with a moving phase boundary. The existence of such an inhomogeneous lithiation mechanism may lead, due to the high volume expansion present in these materials, to high stresses. This is especially true at the sharp interface between regions with different Li concentration. High stresses are detrimental to a stable cycling behaviour and are hold responsible for capacity fading during cycling. These initial results should be confirmed in future by detailed SIMS measurements as a function of current density and by *in-operando* methods like neutron reflectometry.