Amorphous Silicon Nitride - a Novel Anode Material for Li-ion Batteries

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Silicon has a great potential as anode material for lithium ion batteries due to its high theoretical capacity; however, there are several obstacles that need to be overcome in order to make it a commercially viable option. Two of the main issues stem from the fact that silicon undergoes a large volume change during lithiation and delithiation (1). This makes forming a stable solid electrolyte interphase (SEI) difficult, resulting in a continuous loss of electrolyte and lithium as SEI is formed and broken each cycle. Uneven expansion and contraction also causes the silicon to fracture, both exposing new surface on which more SEI might form, as well as electrically disconnecting material from the electrode, rendering it inactive.

One method to reduce these effects has been to use so-called in-situ convertible oxides, like tin oxide and silicon sub-oxide. In this work we investigate the use of amorphous silicon nitride as an alternative convertible alloy anode material. Silicon nitride is believed to form lithiated silicon and lithium nitride or one of several lithium silicon nitride ternary phases during initial lithiation (2, 3). The resulting material therefore believed to combine the high lithium storage capacity of silicon with the high lithium ion conductivity and structural stabilization of inactive lithium containing nitrides. This principle has been investigated in both a thin film electrode system, using PECVD deposited a-SiNx:H thin films, and particle based composite electrodes with amorphous SiNx nanoparticles made in an in-house developed synthesis reactor. By comparing materials with different nitrogen content, the effect of nitrogen content on the materials lithiation capacity, Coulombic efficiency, and cycling stability was explored.

Figure 1 shows the cycling performance of an a-SiNx thin film electrode compared to a pure Si reference, demonstrating that, at the cost of reduced initial capacity, improved cycling stability is gained. The same principle is shown in Figure 2 for more commercially relevant particle based electrodes, demonstrating that electrodes with capacity in excess of 1000 mAh/g can be made without compromising the cycling stability. Stoichiometric silicon nitride, on the other hand, exhibited negligible capacity, underlining the importance of understanding the material characteristics in order to fully utilize its potential.

References: