In Situ AFM Investigation of SEI Formation on Silicon Electrodes

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The stability of the Solid Electrolyte Interphase (SEI) is critical in rechargeable Li-ion batteries. This issue is particularly challenging in silicon, which has a high Li capacity and a correspondingly large volume expansion. Our work employs in situ AFM to investigate SEI formation on amorphous Si, using both continuous thin films lithographically patterned islands. The patterned films enabled direct in situ comparison between the Si and the copper current collector. These experiments were conducted with a Dimension Icon AFM in a glove box using Peak Force Tapping mode, where electrochemical cycling was conducted in a closed electrochemical cell. In addition to monitoring volume and surface morphology evolution, the mechanical properties of the SEI relative were also probed at fixed voltages. These experiments allowed us to investigate SEI behavior in different electrolytes and with different cycling conditions. Both the electrolyte composition and the formation potential had significant effects on the SEI formation. An irreversible Si volume expansion was measured during the first cycle and is likely the product of lithiation induced changes to the silicon structure. Complimentary in situ stress measurements were performed to provide additional information. The cycled films were also examined with detailed TEM to characterize the SEI thickness and structure. The results from this full range of experiments were used to develop a detailed model of SEI formation, which was then employed to develop strategies for designing more stable SEI layers.