Chemical-mechanical modeling of SEI on silicon particles

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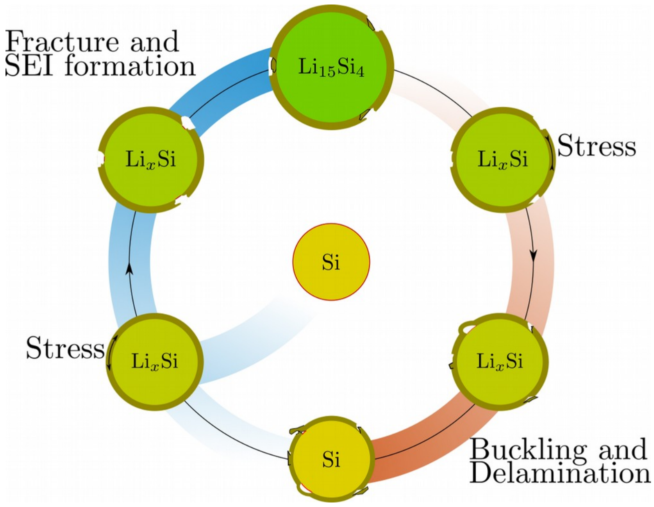
Lithium-ion batteries combine good long-term stability and performance. These properties have made Lithium-ion batteries the benchmark energy storage for hand-held electronics and electric vehicles. Nevertheless, there is an industrial urge to further push the technology towards higher capacities and longer battery life. Increasing the lifetime of Lithium-ion batteries is currently a fundamental challenge for battery research. One important mechanism contributing to the capacity loss is the growth of a solid-electrolyte interphase (SEI) during storage and cycling.
Silicon anodes are a promising approach for further increasing the capacity of Lithium-ion batteries, as they show the tenfold theoretical specific capacity of the currently used graphite anodes. However, the SEI growth is even more severe for silicon anodes: large volume expansions of ~300% exert high mechanical stresses and fracture the SEI. The resulting cracks subsequently expose the pristine electrode leading to SEI reformation and thereby continuous capacity decrease.



In order to further understand SEI growth, our group developed a model describing SEI growth on graphite during storage [1-4]. Thereby, the diffusion of neutral Li interstitials from the electrode to the electrolyte was found to cause the long-term growth of the SEI [1,2].
Additionally, a thermodynamical framework was developed to describe the coupling of chemical, electrical and thermal effects [5]. We extend this model for mechanical effects and investigate how the interplay of chemistry and mechanics impacts stability and growth of the SEI during battery operation.
Understanding these relationships identifies critical operating conditions and aid in the design of new electrodes. Thereby, batteries with higher capacity and long-term stability can be developed.

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