

Modelling SEI Growth and Function During Battery Operation

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The solid-electrolyte interphase (SEI) substantially influences the lifetime of lithium-ion batteries. Nevertheless, the mechanism responsible for the long-term growth of SEI during storage as well as operation remains controversial.

This talk discusses the reaction and transport processes relevant for SEI growth. Our thermodynamically consistent model for the SEI formation reaction shows that a transition from compact to porous is natural for self-passivating films [1]. This agrees with the observed dual-layer structure of SEI. However, long-term SEI growth is determined by charge transport through SEI. We show that electron diffusion can explain both the observed state-of-charge dependence and the time dependence during battery storage, in contrast to solvent diffusion [2]. Due to self-discharge, this dependence can even explain observed deviations from the typical square-root behavior in the time domain [2].

We extend and validate our model for electron diffusion through SEI to model batteries in operation [3,4]. In this case, we find a sequence of growth regimes, i.e., reaction-limited, diffusion-limited, migration-limited. In micro-structure resolved 3D simulations, our electron diffusion model predicts, for the first time, the experimentally observed heterogeneity in SEI thickness throughout the porous electrode [5]. Finally, we present our thermodynamically consistent model for the effect of mechanics on SEI structure and function during continued charge and discharge of silicon electrodes [6].

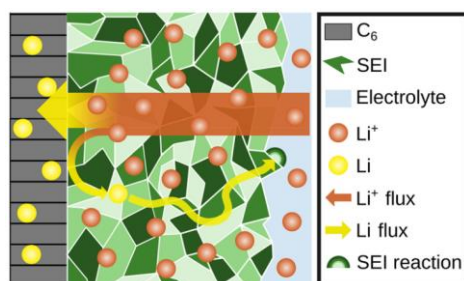


Figure 1: Scheme of electron diffusion through localized states [3].

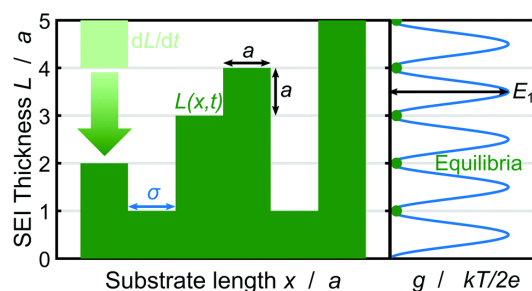


Figure 2: SEI reaction model

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