Commercialized lithium-ion batteries stand out among energy storage devices because they offer good energy and high power densities. On negative electrodes of lithium-ion batteries, a thin layer of electrolyte reduction products forms. Even though this solid-electrolyte interphase (SEI) suppresses the further electrolyte reduction, continued SEI growth is the main contributor to lithium-ion loss, capacity fade, and limited battery lifetime \cite{Keil2016}. Key questions about SEI chemistry and mechanisms are still unanswered. The experimentally observed square-root-of-time dependence of capacity fade suggests a transport limitation, but it does not distinguish between different transport mechanisms. We develop a series of models to predict additional dependencies, i.e., morphology of SEI and potential dependence of SEI growth \cite{Single2016, Single2017, Single2018}. Dedicated experiments can then identify the long-term growth mechanisms, e.g., battery storage at various state-of-charge \cite{Keil2016}, neutron reflectometry \cite{Steinhauer2017}, and SEI on model electrodes \cite{Kranz2017}.

Going beyond the standard SEI models, our model predicts the morphology of SEI by taking into account the interplay of structure, reaction kinetics and transport mechanisms \cite{Single2016, Single2017}. We consider all potentially rate-limiting transport mechanisms, e.g. electron conduction, electron tunneling, solvent diffusion and lithium interstitial diffusion. The emergence of a spatially structured SEI is the consequence of coupling these transport mechanism with different SEI formation reactions. Different SEI profiles generated in our model are studied with impedance simulations.

From the potential dependence of the SEI growth rate \cite{Keil2016}, we predict the mechanism behind its continued growth \cite{Single2018}. We present the first indirect experimental evidence that neutral radicals carry a negative charge and diffuse through the SEI.

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**Literature**


