A solid electrolyte interphase (SEI) forms on lithium-ion battery anode surfaces during cycling. The SEI results from the decomposition of the electrolyte\(^1,2\). On the one hand, SEI passivates the graphite anode and slows down electrolyte reduction; on the other hand, it results in continuous capacity fade. Furthermore, the SEI is important for critical degradation mechanisms in lithium batteries, e.g., thermal runaway, lithium plating, and dendritic growth. Therefore, we are studying the chemical composition and structural morphology of the SEI via continuum modeling.

Despite numerous experimental and theoretical works on SEI growth, its structure is not yet well understood\(^3\)\textsuperscript{–}\textsuperscript{5}. Various models predict the correct capacity fade, but no consensus is reached on the limiting process for SEI growth. Microscopic experiments reveal that the SEI is composed of a thin inner compact layer and a thick outer porous layer with different chemical composition and structural morphology\(^2\). Similarly, the role of the various solvents and co-solvents in its formation, is very complex and topic of ongoing research\(^6\)\textsuperscript{–}\textsuperscript{8}.

We present here, for the first time, a SEI growth model with an evolving porous structure. We model the growth of a porous SEI on top of the electrode surface in one dimension applying a new approach for quasi-diffusional growth inspired by phase-field theory (see Fig. 1). Focusing on the evolution of SEI morphology, we assume a single component SEI of Li\(_2\)CO\(_3\) and the reduction of EC with an inert co-solvent\(^2,6\).

Our simulations result in a porous SEI whose thickness grows with the square root of time as observed experimentally (see Fig. 2)\(^5\). The transport of charges through the porous SEI is limiting this SEI thickness and the resulting capacity fade. The transport of electrolyte, instead, is determining the SEI porosity. We highlight that we can analytically understand this behavior and give expressions for SEI thickness and porosity as a function of time and position.

![Figure 1: Modeling scheme for porous SEI growth](image)

**Figure 1: Modeling scheme for porous SEI growth**

A transport-limited SEI growth model is developed\(^9\). On the one hand, charges move from the electrode surface to the reaction sites within the SEI. Here we assume that neutral lithium diffuses through the SEI and move the charges, consistent with DFT calculations for Li\(_2\)CO\(_3\)\(^2\). On the other hand, electrolyte convection and EC diffusion transports the solvent through the porous SEI to these reaction sites.

![Figure 2: Evolution of SEI thickness](image)

**Figure 2: Evolution of SEI thickness**

We compare our approach to experiments and find SEI charge transport coefficients and SEI porosities in agreement with observations. However, further comparison with in-situ microscopic studies is necessary in order to validate this SEI growth model.

**Literature:**