

Modeling Solid Electrolyte Interphase Formation and Morphology

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When a lithium ion battery is fully charged, the potential of its negative electrode is outside the stability window of the electrolyte, typically a mixture of organic solvents (ethylene carbonate, dimethyl carbonate) with a lithium salt. Consequently solvent molecules are reduced at the anode surface and reduction products form a film thus passivating the electrode. This so called solid electrolyte interphase (SEI) slows down the reduction process to acceptable levels and is crucial for stable battery performance. Experiments indicate that the SEI has dual layer morphology [1], i.e. it consists of a dense inner layer (close to the electrode) and a porous outer layer (close to the electrolyte). So far, most theoretical studies describing SEI evolution homogenize the SEI structure, focusing solely on the layer growth [2,3,4].

We present a one dimensional model for porous SEI formation that additionally captures morphology properties of the SEI film. In our simulation the evolution of the SEI density and composition is traced along the axis perpendicular to the electrode surface. Solvent reductions reactions are “fueled” by electrons conducted through the solid SEI phase and solvent molecules which move within the pores of the porous structure via diffusion/convection (depicted in the figure below).

We can show – via simulation and theory – that these assumptions lead to the formation of a porous structure with constant, nonzero porosity. Additionally our model predicts the formation of dual layer morphology in a solvent mixture with two or more active species.

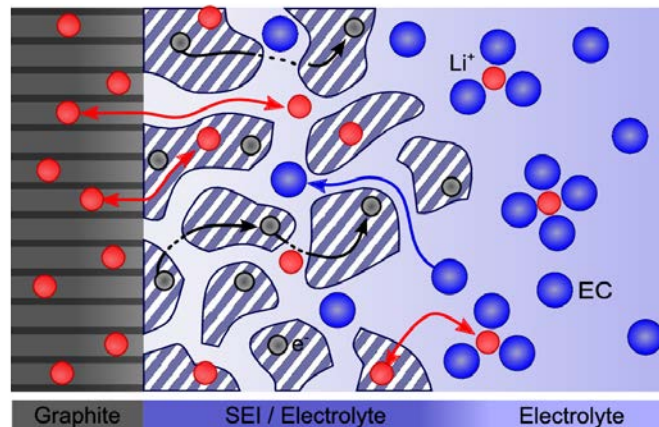


Figure: Cross section of graphite anode, porous SEI and electrolyte phase. Lithium can intercalate into the anode and move freely in all other phases. Solvent molecules and lithium salt anions move through the porous structure. Electron conduction through the SEI allows electrolyte reduction and SEI formation on any SEI/electrolyte interface.

References

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