Transport of Ions, Electrons, and Molecules across the Solid Electrolyte Interphase (SEI) in Lithium-Ion Batteries – What is our Current Level of Understanding?

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The solid electrolyte interphase (SEI) on the graphite anode of lithium ion batteries plays a crucial role for the battery performance. The SEI blocks electrons and solvent molecules, while Li+ ions can easily migrate across the SEI. However, quantitative measurements of transport coefficients for these species in the SEI are problematic due to the complex structure of graphite composite anodes. Therefore, we have grown model-type SEIs on planar glassy carbon electrodes and have characterized them by a combination of FIB/SEM, AFM-based scratching experiments, impedance spectroscopy and redox probe experiments. FIB/SEM and AFM experiments reveal a dual-layer structure of the SEI and give indication that the inner layer is composed of agglomerated spherical particles. Impedance spectroscopy and redox probe experiment show that the effective diffusion coefficient of Li+ ions and redox molecules in the SEI are virtually identical and, in addition, show the show the same temporal evolution after SEI formation and the same activation energy. Our results suggest that the inner SEI layer exhibits a small porosity, which plays a very important role for the transport. Finally, by comparing redox molecule transport and SEI growth rates, we give strong indication that redox molecules are transported much faster than the electrolyte solvent molecules.