NMR-Powered Direct Probing Tool for Solid-Electrolyte Interphase Lithium-Ion Permeability

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Lithium metal is a leading candidate for replacing the anode in next-generation batteries for grid storage and transport. Although desirable for its high energy density, lithium metal currently suffers from low cycle life as well as safety issues. Being highly reactive, lithium metal forms a passivation layer known as the Solid-Electrolyte Interphase (SEI) – a heterogeneous barrier made up of electrolyte decomposition products. The SEI prevents further reactivity by blocking electron transport from the metal to the electrolyte. Yet, the SEI also impedes lithium-ion transport and causes uneven deposition on the metal surface, which in turn promotes dendrite formation that leads to cell failure. Presently, there is a deficit in techniques that can directly probe lithium-ion transport across the SEI without interference from other processes. Here, we develop a method to directly probe the lithium-ion permeability of the SEI under open-circuit voltage conditions using ^{7}Li Nuclear Magnetic Resonance alongside a fitting procedure based on a simple model. We employ this approach to compare SEIs in different electrolyte chemistries under electrochemical conditions to their spontaneously formed counterparts. Finally, we delineate the inherent challenges associated with this method and propose prospective enhancements to optimize its implementation.