# Towards ab initio models of the solid electrolyte interface and improved accuracy for energy related materials

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**Presented by…**


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**Abstract** Dr. Kent will discuss two distinct topics, the first relating to first principles modeling of electrolytes for lithium ion batteries, the second on quantum monte carlo methods, which offer significantly improved accuracy over the popular density functional approaches:

(1) The liquid electrolytes used in lithium ion batteries react on the battery electrodes, forming solid-electrolyte interphases (SEI). This results in capacity loss, an increase in cell resistance, and significantly alters the degradation mechanisms of the electrode materials. To help understand the SEI we have performed ab initio molecular-dynamics simulations of common carbonate electrolytes and lithium salts. We study the role of functionalization of graphite-anode edges on the reducibility of the electrolyte and the ease of Li-ion intercalation at the initial formation stages of the SEI. The molecular dynamics approach reveals, e.g., orientational ordering of the solvent molecules, and favored migration of inorganic (vs. organic) reductive components to the electrode.

(2) Despite recent improvements, practical density functional calculations lack accuracy when applied to many energy storage and catalytic materials. In the case of battery voltages, the challenge involves simultaneously obtaining accurate energies for an oxide and for a metal. Errors of 10% are common, suggesting also that diffusion barriers are suspect. Quantum Monte Carlo (QMC) is a method potentially offering much improved accuracy. However, QMC has only been broadly applied to molecules and select insulators. I will present very recent data for bulk aluminum and a variety of defects demonstrating excellent agreement with experiment, where such data is available. These developments, coupled with the availability of petascale computing, suggest QMC can serve as a benchmark for energy related materials.


**More info?**

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