Predicting and Understanding Novel Electrode Materials from First Principles

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Objective:

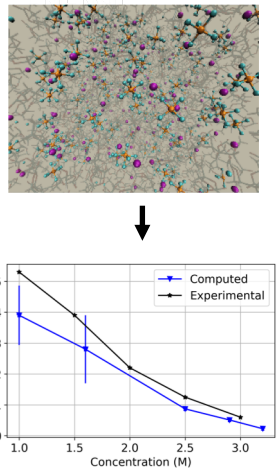
- Understand the Li-ion dynamics and stability (including oxygen retention, electrochemical window, etc.) in emerging Li-ion cell components; superconcentrated electrolytes, Si-Sn alloys (and their native oxides), and selected coatings of positive electrode via first principles and classical molecular dynamics studies.

Impact:

- Understanding transport mechanisms in superconcentrated systems will assist rational design and tailoring of electrolyte
- Improve stability of Si electrode through Sn alloying, while improving Li-ion dynamics
- Improve battery lifespan by reducing cathode degradation from oxygen loss

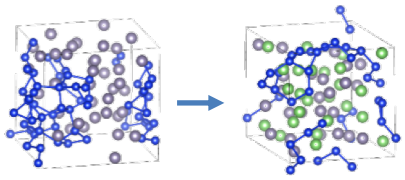
Transport and Stability in electrolyte/electrode

Conductivity of highly concentrated $LiPF_6$ in Propylene Carbonate

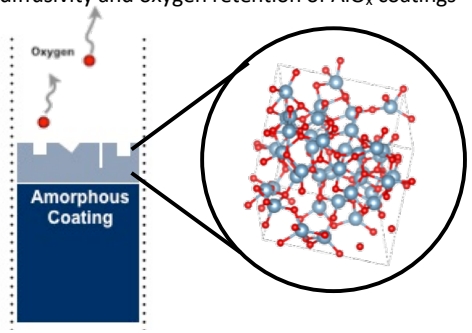


Experimental values from Takeuchi et al., 2009

Lithiation of Si-Sn and structure stability



Li diffusivity and oxygen retention of AlO_x coatings



Accomplishments:

- Discovery of the spontaneous oxygen evolution as a function of surface facet and state of charge for Li- excess Li_2MnO_3
- First computational framework to include dielectric constant contribution from contact-ion pairs, providing insight into speciation of salt in linear carbonate electrolytes
- Showed the contribution of the native SiO_2 layer on passivation and Li diffusion of Si
- **New Start** Quantification of transport mechanism change from standard to superconcentrated electrolyte
- **New Start** Evaluating the performance of Si-Sn alloys as Li-ion battery electrodes

FY19 Milestones:

- Benchmark algorithms for obtaining solvation structures and ion conductivity in highly concentrated liquid electrolytes (Q1)
- Identify Li conduction mechanisms in Si-Sn alloy anode (Q2)
- Evaluation of SiO_2 and SnO_2 as surface phases from the perspective of ionic conduction and reactivity (Q3)
- Evaluation of oxygen retention/Li diffusion in amorphous coatings using Materials Project infrastructure (Q4)

FY19 Deliverables:

Provide transport mechanism(s) of Li^+ in both superconcentrated electrolytes and Si-Sn alloys.

Funding:

— FY19: ?, FY18: $350k, FY17: $350k