

Analysis of Film Formation Chemistry on Silicon Anodes by Advanced *In Situ* and *In Operando* Vibrational Spectroscopy

PI: G.A. Somorjai (U.C. Berkeley)

Technical Approach:

- Determine formation/degradation mechanisms of the solid electrolyte interface (SEI) on the surfaces of Si anodes during charge/discharge cycles by applying advanced *in situ* vibrational spectroscopies.
- Determine how the properties of the SEI contribute to failure of Si anodes.
- Develop electrolyte additives and/or surface modification methods to improve Si anode capacity loss and cycling behavior.

Status:

- Modifying SFG apparatus for high-resolution spectra of Si particles and microstructures / electrolyte interface.

Initiated

- Performing SFG measurement under constant and dynamic potentials of 1 M LiPF₆ in EC w/o DEC in order to probe the SEI formed on amorphous silicon anodes.

Initiated

Technology:

SFG revealed that linear alkyl carbonate formed Si-alkoxide bonds only in Si(100)-H terminated anode. In Si(100)-O_x only soluble products leading to anode failure were detected.

Objectives:

- Chemical composition and structure of the SEI at the Si/electrolyte interface:
 - which electrolyte components are involved
 - how lithium ion solvation is involved
- SEI inhomogeneity across Si electrode surface
- Stability of the SEI with respect to:
 - the crystal size, structure, and orientation of Si
 - doped or undoped Si
 - electrolyte composition

Deliverables: Understanding of the composition, structure, and formation/degradation mechanisms of the solid electrolyte interface (SEI) on the surface of Si anodes during charge/discharge cycles.

Funding:

Duration: 4 yrs (Yr 4)
FY16 Budget: \$200K (DOE)

Milestones:

Q1: Modifying SFG apparatus in order to obtain high resolution spectra further allowing to research the electrolyte / Si interface.

Q2: Performing SFG measurement under constant potentials (-0.01 V, 0.5 V and 1.0 V vs Li/Li⁺) of 1 M LiPF₆ in ethylene carbonate (EC) w/o diethyl carbonate (DEC) in order to probe the SEI formed on amorphous silicon.

Q3: Performing ps-SFG measurements under dynamic potential of 1 M LiPF₆ in EC : DEC mixture. Go/No-Go: Can we distinguish between the various SEI products in the C-H stretch mode (2800 – 3200 cm⁻¹)? If not proceed to conventional C-O region (1700 – 1800 cm⁻¹)

Q4: Ring opening kinetics of EC in the presence of LiPF₆ and DEC) by fs-SFG measurement in tandem with cyclic voltammetry.