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VEHICLE TECHNOLOGIES OFFICE (VTO)
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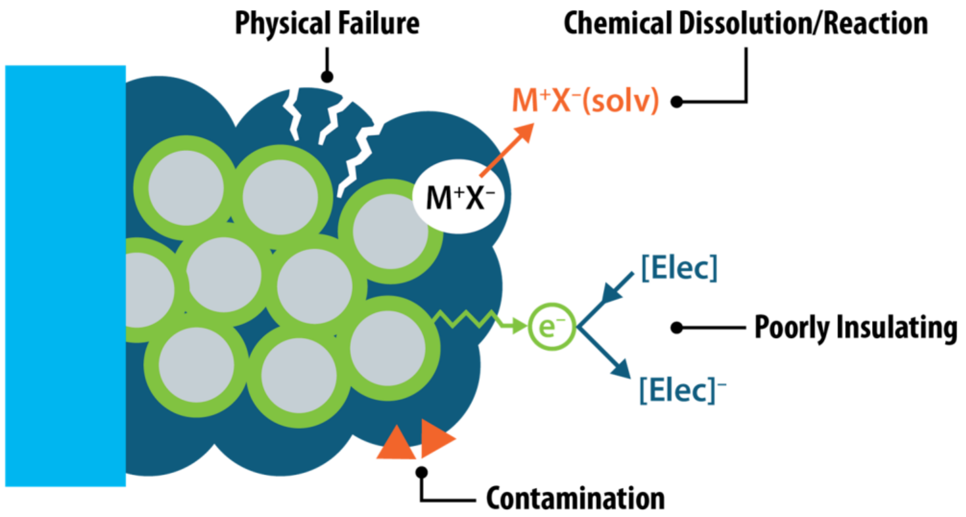
THE SILICON CONSORTIUM PROJECT: MECHANICAL PROPERTIES OF SILICON ANODES



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Poster ID: bat499
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SEI = solid electrolyte interphase, (PH)EV = (plug in) hybrid electric vehicles

OVERVIEW

Timeline

- October 1st 2020 - September 30th 2025.
- Percent complete: 10%

Budget

- Funding for FY21: $7500K

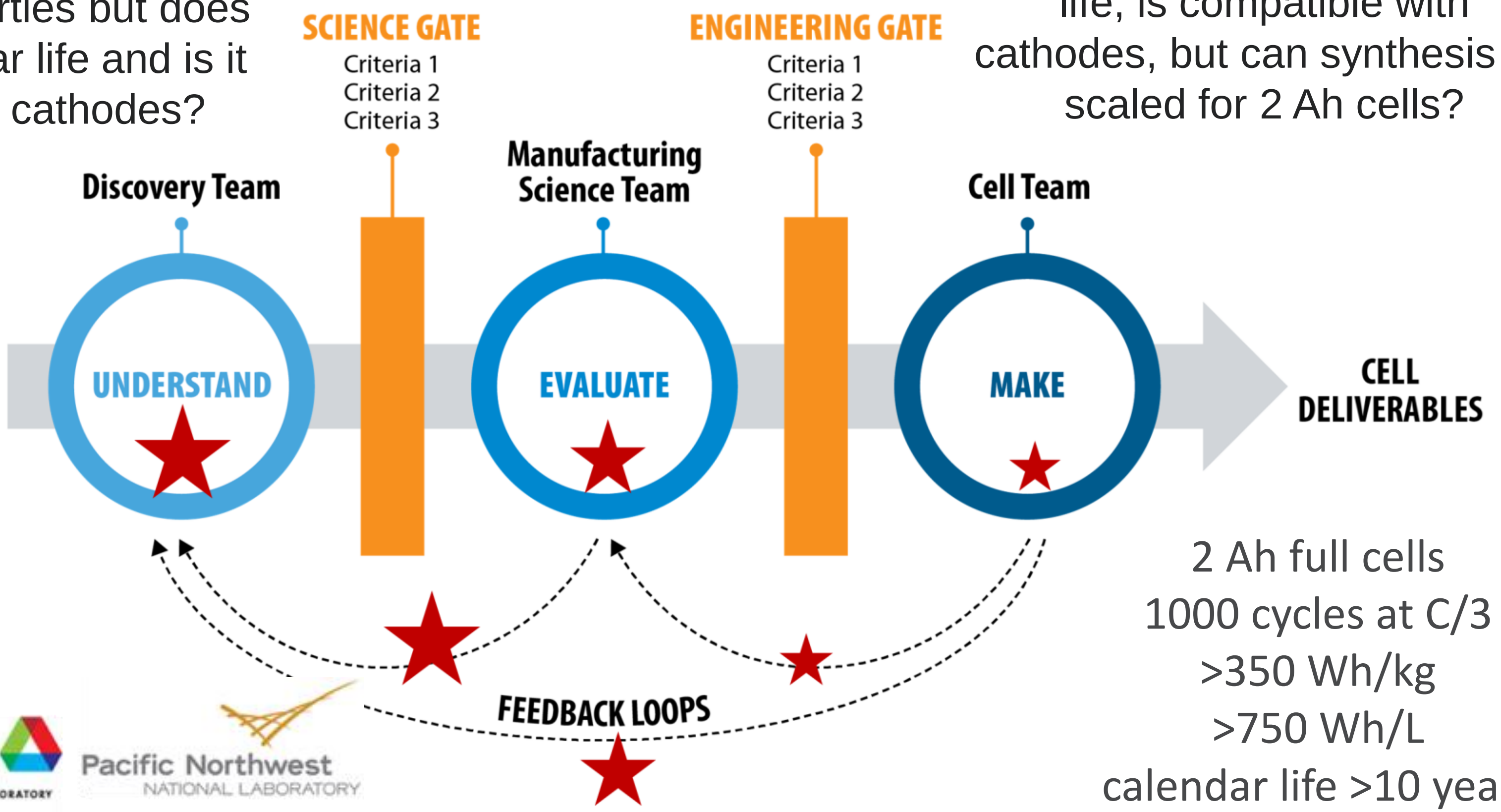
Barriers and Relevance

- Development of PHEV and EV batteries that meet or exceed the DOE and USABC goals. Specifically targeting the development of calendar life in silicon anode.
  - Cost, Performance and Safety

Mechanical Characterization Thrust Focus

Example gate criteria:
Additive enables better SEI mechanical properties but does it improve calendar life and is it compatible with cathodes?

Example gate criteria:
Additive enables favorable SEI mechanics, improves calendar life, is compatible with cathodes, but can synthesis be scaled for 2 Ah cells?



2 Ah full cells
1000 cycles at C/3
>350 Wh/kg
>750 Wh/L
calendar life >10 years

Partners

NREL, Sandia National Laboratories, Berkeley Lab, Oak Ridge National Laboratory, Argonne National Laboratory, Pacific Northwest National Laboratory

THRUST TASKS

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Tasks

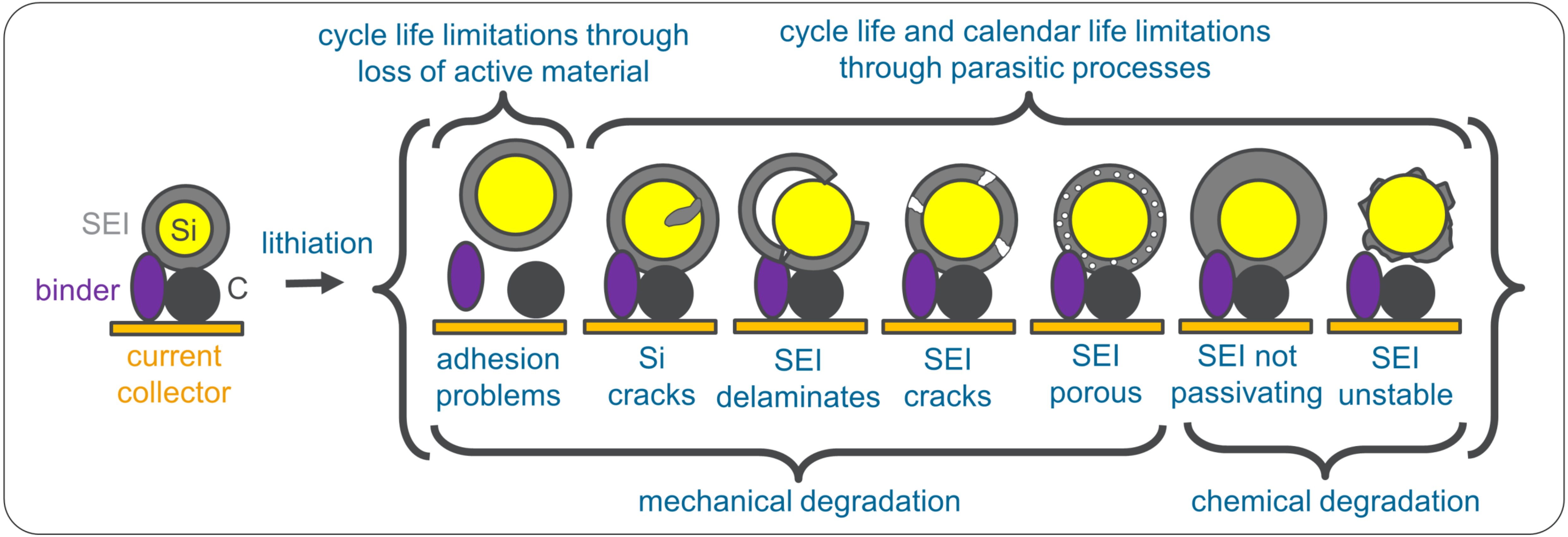
- Advanced Characterization of the Si/SEI/Electrolyte Interface Function
- Electrochemical Stability of the SEI
- **Mechanical Characterization of the SEI**
- Next-Generation Materials discovery and development
- The Science of Manufacturing
- Cell manufacturing

MILESTONES

- Establish a Pre-lithiation protocol that can be utilized by all partners Q1 (complete)
- Go/no-go on HF etching of Silicon Oxide-silicon as viable route to silicon Q2 (complete)
- **Go/No go on the Moire interferometry at as a method of probing the calendar life of the silicon SEI? Q3 (complete)**
- Produce 20 grams of next generation silicon's with at least two different coatings, at least one of which exhibits enhanced calendar life over the baseline commercial silicon (NREL-centric) Q4
- Advanced version of the calendar life protocols that quantifies calendar life in silicon-based anodes within 20% of the "real" calendar life predictions of calendar life. Q4
- Synthesis and testing of 5 different metallic glasses with theoretical capacities > 1000 mAh/g Q4
- **Identify active cell components and cell designs to achieve stable calendar life electrode performance with a cell build demonstrating 300 cycles with <20% capacity fade. Q4**

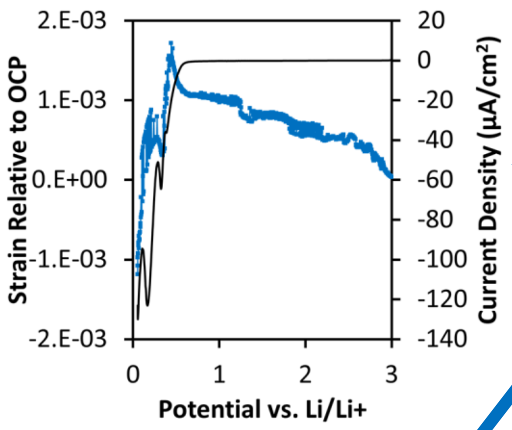
MECHANICAL CHARACTERIZATION THRUST OBJECTIVES

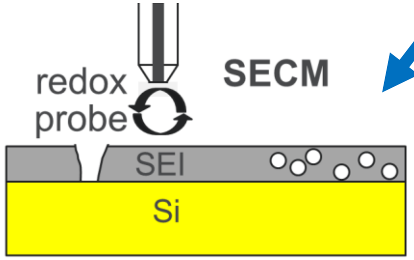
The thrust objective is to understand and mitigate the mechanical degradation mechanisms that lead to cycle and calendar life limitations.



- Si volume expansion leads to Si and SEI mechanical failure mechanisms → affects cycle and calendar life
- Calendar life is likely impacted by BOTH chemical and mechanical SEI degradation

MECHANICAL CHARACTERIZATION THRUST APPROACH





Apply Electrochemistry, Measure Forces

Moiré Interferometry FY21
SECM / Redox Probes FY21
Soft Substrate FY21

FY21 Scratch, Nanoindentation
FY21 Tensile/Adhesion Testing
FY21 CRFM, QNM
FY21 Varied Cycle Cal. Tests
Bragg Grating

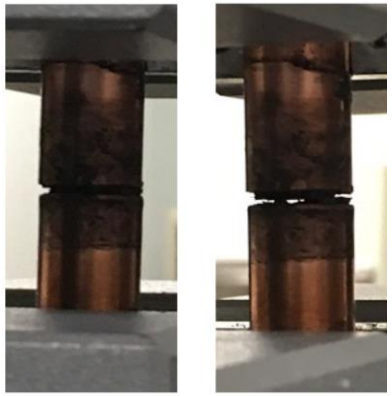
Thin Films — Simulations inform transition from model systems to electrodes — Electrodes

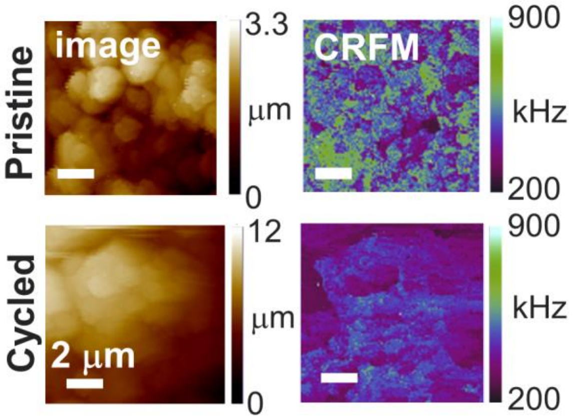
Stiff Substrate FY21
Compressed Electrodes

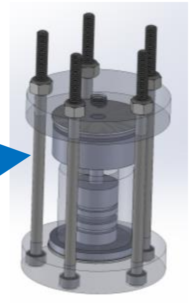
Scratch, Nanoindentation
Tensile/Adhesion Testing
CRFM, QNM
Bragg Grating
FY21 Compressed Electrodes
FY21 Microcalorimetry

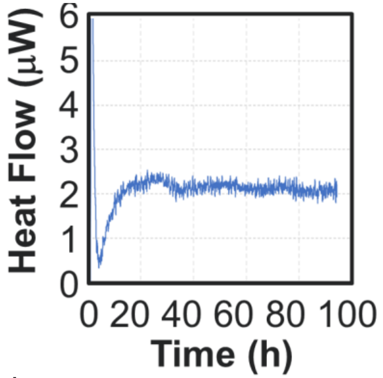
Any proposed future work is subject to change based on funding levels

Apply Forces, Measure Electrochemistry









SECM = scanning electrochemical microscopy, CRFM = contact resonance frequency microscopy, QNM = quantitative nanomechanics

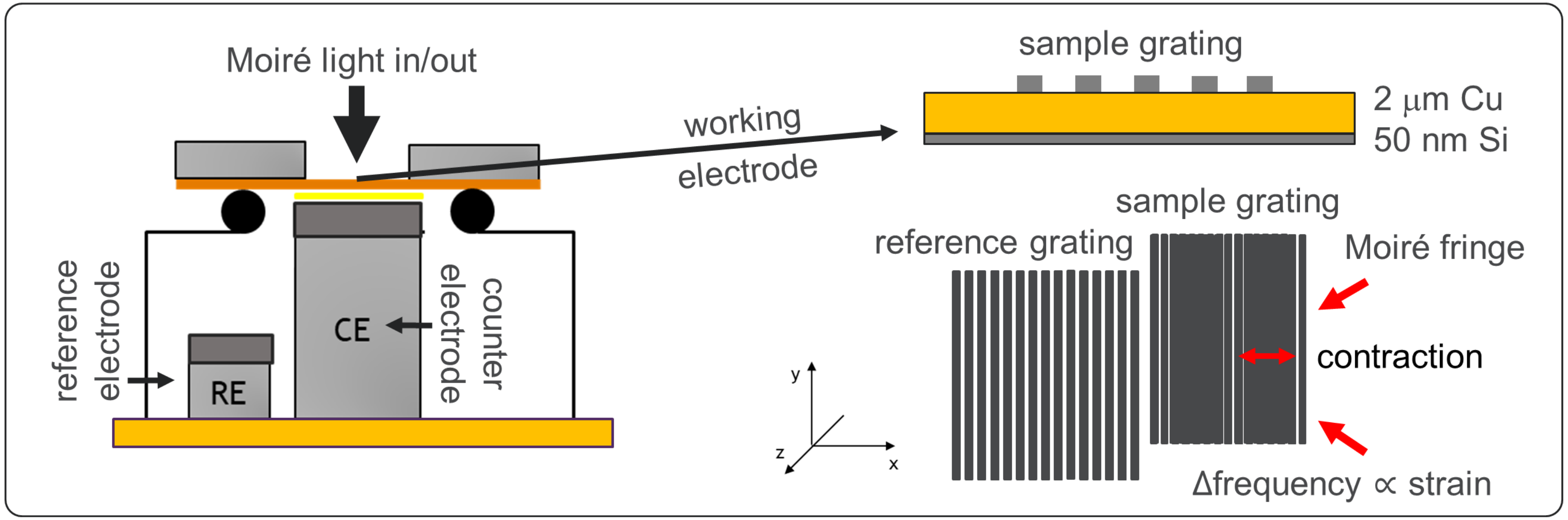
TECHNICAL ACCOMPLISHMENT AND PROGRESS: FY21 Q3 MILESTONE PROGRESS: NO-GO ON MOIRÉ INTERFEROMETRY

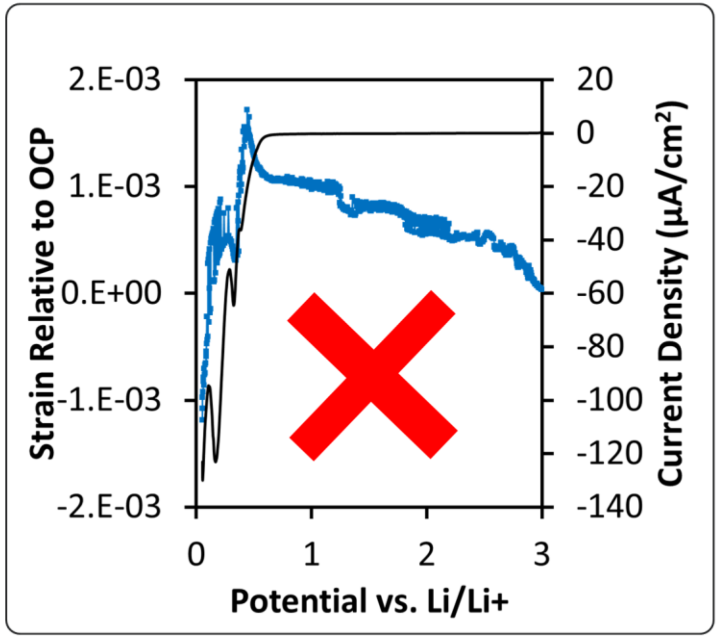
Motivation

Understanding of SEI mechanical failure mechanisms enables understanding that guides mitigation.

Approach

Built Moiré interferometer with in-situ electrochemical cell to measure SEI strain through interference between sample and reference gratings.





Results

Sample tilt is not controlled effectively for the in situ setup (confirmed by Gaussian beam simulations) and the data is not accurately showing strain.

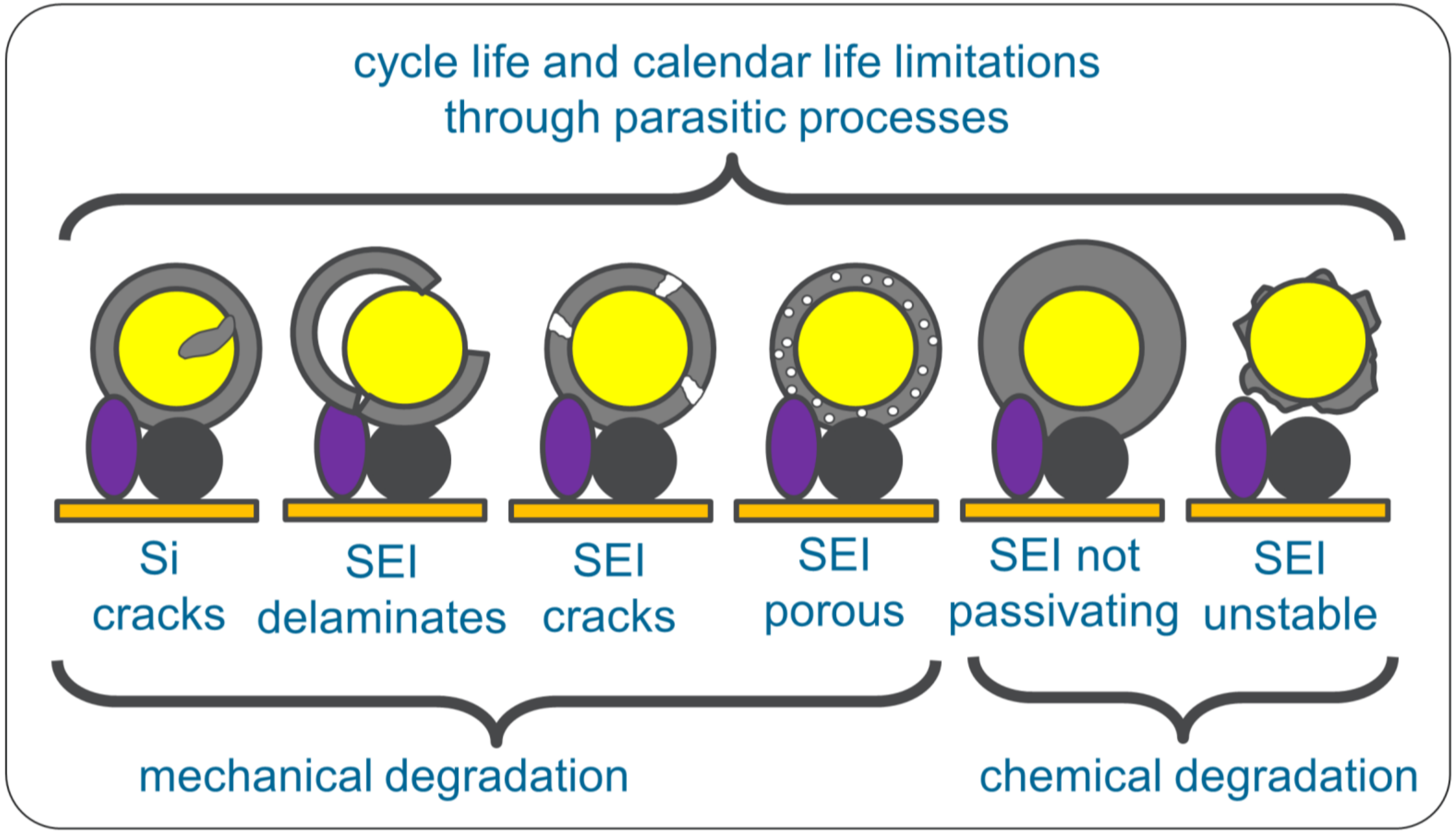
Impact

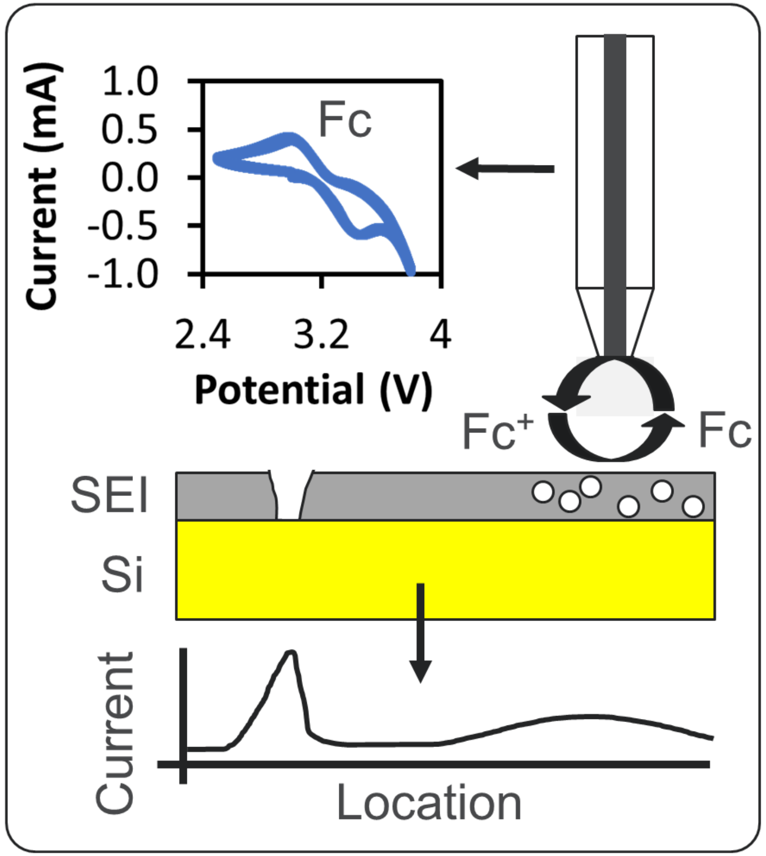
NO GO decision and need new approach.

NEW APPROACH: SECM AS ALTERNATIVE TO MOIRÉ INTERFEROMETRY

Question
Does passivation failure leading to poor calendar life occur due to local SEI mechanical failure or globally (cracking, delamination, or porosity)? How does this change with SEI composition (additives, alternative electrolytes)?

Updated Approach
Map passivation in a scanning electrochemical microscope (SECM) to understand SEI failure mechanisms using ferrocene (Fc) and other redox probes.





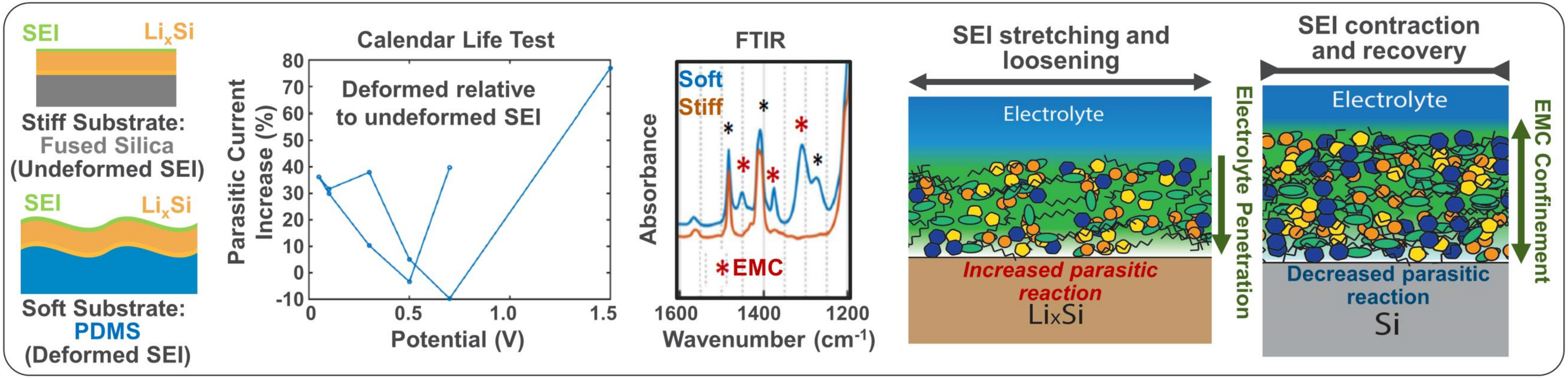
TECHNICAL ACCOMPLISHMENT AND PROGRESS: IMPACT OF MECHANICAL SEI DEFORMATION ON PASSIVATION

Motivation

Understand mechanics contribution to calendar life limitations.

Approach

Cycle and run calendar life tests on thin film Si on stiff substrates (rigid silica) to prevent SEI deformation and soft substrates (PDMS) to allow SEI strain.



Results

SEI deformation (soft substrate) leads to lower Coulombic efficiency, increasing parasitic current during calendar aging, and EMC penetration and trapping in the SEI (evidenced in FTIR).

Impact

SEI strain is caused by Si expansion during cycling and induces porosity that enables parasitic electrolyte reactions. Other electrolytes will next be studied.

FTIR = Fourier transfer infrared spectroscopy, EMC = ethyl methyl carbonate, PDMS = poly (dimethylsiloxane)

Any proposed future work is subject to change based on funding levels

SUMMARY, CHALLENGES, AND NEXT STEPS

- **This Mechanical Characterization thrust seeks to understand how mechanical properties of the SEI impact calendar life of Si anodes.**
  - Moiré interferometry milestone is a no-go decision due to challenges with the in-situ setup.
  - Soft and hard substrate experiments demonstrate that strain induces SEI porosity and increases parasitic reactions between Si and the electrolyte, leading to calendar life limitations.
  - Scanning electrochemical microscopy will help identify mechanisms of SEI failure under varied conditions.
  - **Impact:** Using these techniques developed in conjunction with the Advanced Characterization thrust helps the Next-Generation Materials and Electrochemical Stability thrusts understand SEI properties to design more passivating electrolytes and materials and meet calendar life goals.

- **This Mechanical Characterization thrust seeks to understand how mechanical properties and mechanical manipulation of porous electrodes impacts cycle and calendar life of Si anodes.**
  - Setting up variable pressure testing of cells in a microcalorimeter will help us understand how pressure impacts parasitic reactions that lead to calendar life limitations (and mitigate those reactions).
  - Carrying out mechanical characterization on porous electrodes (CRFM, QNM, nanoindention, tensile testing) will help us correlate mechanical properties with calendar and cycle life performances.
  - **Impact:** Using these techniques developed in conjunction with the Next-Generation Materials helps the Science of Manufacturing and Cell Manufacturing thrusts develop design rules to better mitigate cycle and calendar life limitations.

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