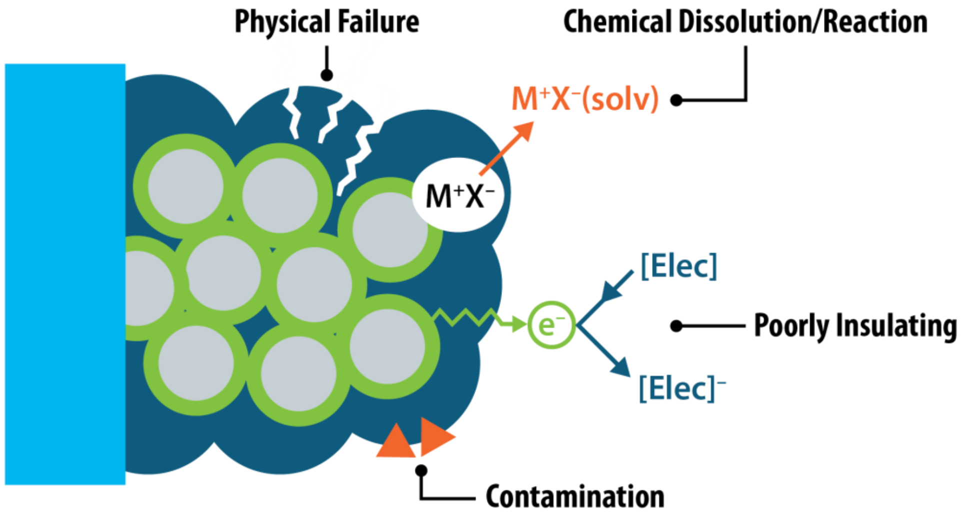
U.S. DEPARTMENT OF ENERGY'S (DOE)
VEHICLE TECHNOLOGIES OFFICE (VTO)
2020 ANNUAL MERIT REVIEW (AMR)

SILICON CONSORTIUM PROJECT: SCIENCE OF MANUFACTURING FOR SILICON ANODES



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Project ID: bat500
Poster presentation

NREL Transforming ENERGY · Sandia National Laboratories · Berkeley Lab · Oak Ridge National Laboratory · Argonne National Laboratory · Pacific Northwest National Laboratory

SCIENCE OF MANUFACTURING IS FOCUSED ON REDUCING DEVELOPMENT TIME AND BOTTLENECKS

Timeline

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

Barriers

- 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

Budget

- Funding for FY20: $7500K

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

OVERVIEW – SCIENCE OF MANUFACTURING

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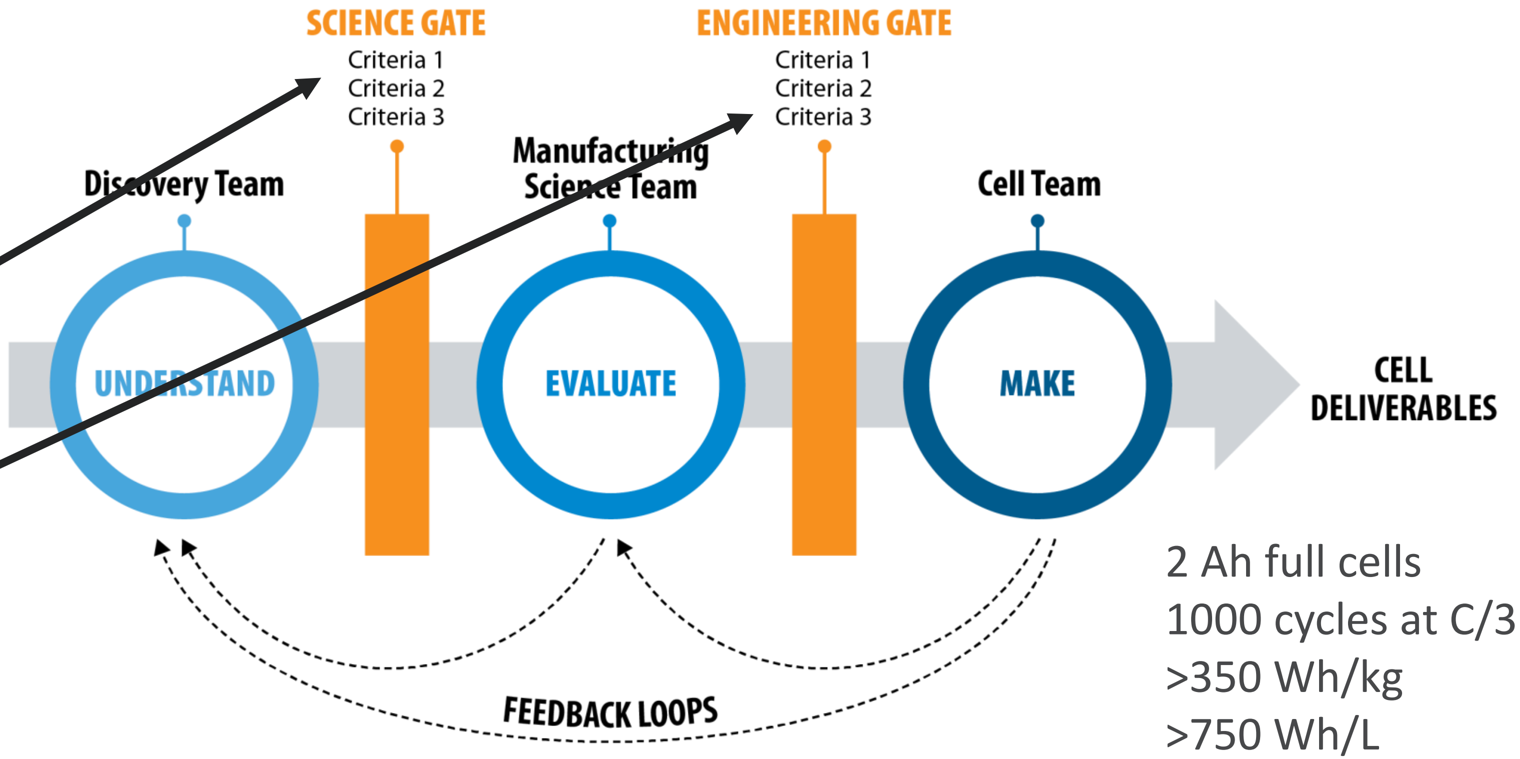
Example stage gate:
How do reduce the development time to implement a new material
- e.g., polymer

Can a new material be scaled to sufficient volumes?
- e.g., silicon and electrolyte



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

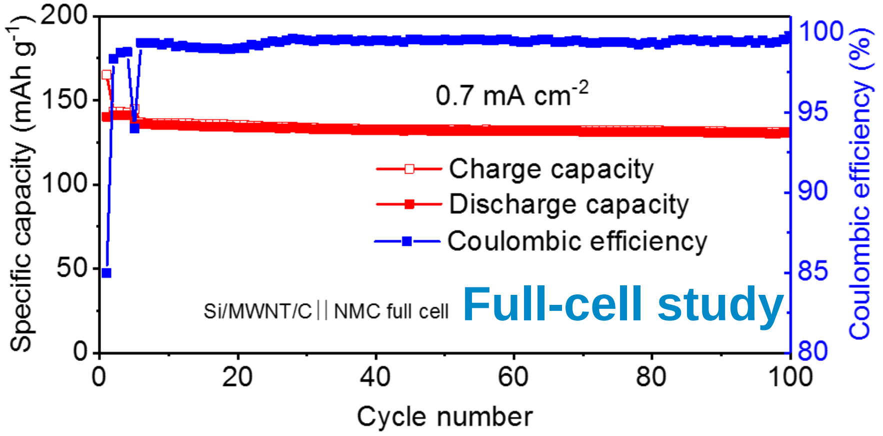
MILESTONES

- 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 spectroscopy 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

SCIENCE OF MANUFACTURING OBJECTIVE

SMT breaks down new cell chemistries to understand and predict how to scale up effectively

Materials discovery team identifies a promising material in small scale testing



Example of promising material from PNNL demonstrating excellent cycle life at a small scale

1. Process of prelithiation
- How do we scale up to 2 Ah cells?
- What procedures work best that can be applied to team?

2. Fabricate the new material at scale
- What happens when you make material at 50 gram scale?
- How reproducible is the synthesis?

3. New binder incorporation
- What are the protocols for processing electrode?
- How reproducible is the synthesis?

4. Scale to 4 mAh/cm^2
- How do you make the cells with the right energy density?

SCIENCE OF MANUFACTURING APPROACH

Engage all team members to rapidly understand key materials and processing issues and develop protocols for standardizing the development of components

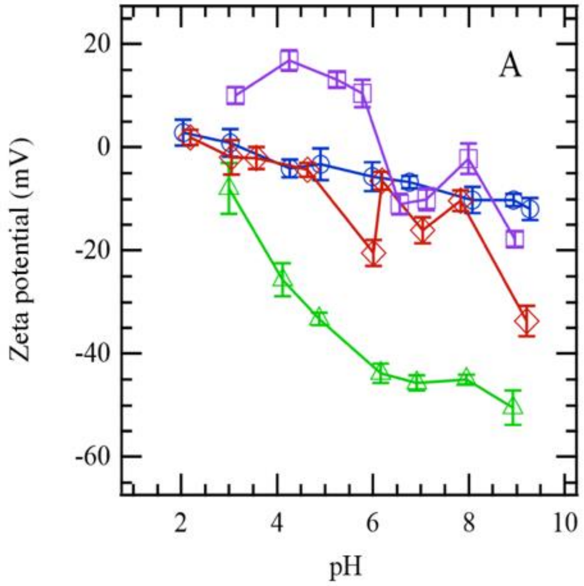
Protocol development

Prelithiate SiO_x anodes versus lithium metal

- *Punch out anodes to xx3450 size (14.1 cm²) and vacuum dry*
- *Cut out oversized lithium foil sheets*
- *Assemble half cell in large reusable SS cell fixture from ATD Program*
- *Use Gen2 electrolyte + 4 wt.% FEC*
- *Apply formation cycles:*
  1. *Lithiate to 10 mv*
  2. *Cycle 6x (700 – 50 mV)*
  3. *End on delithiation at 850 mV*
- *Harvest electrode in dry room, and rinse briefly (15 sec) with DMC*

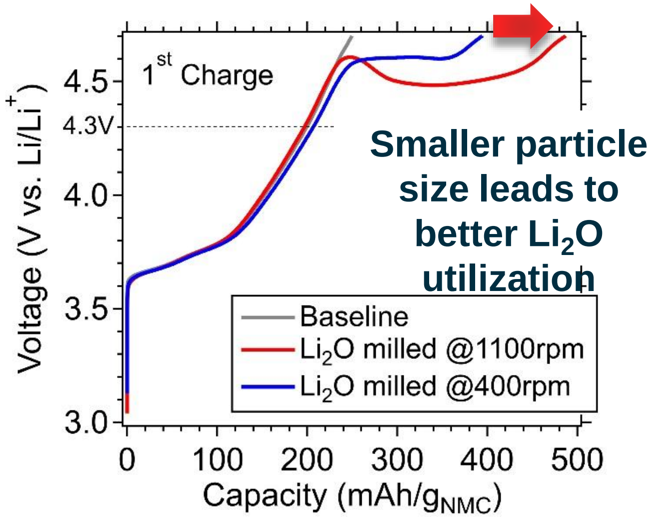
See Poster bat497

Colloidal processing methods



Measure zeta potential to predict processing regimes

Materials development



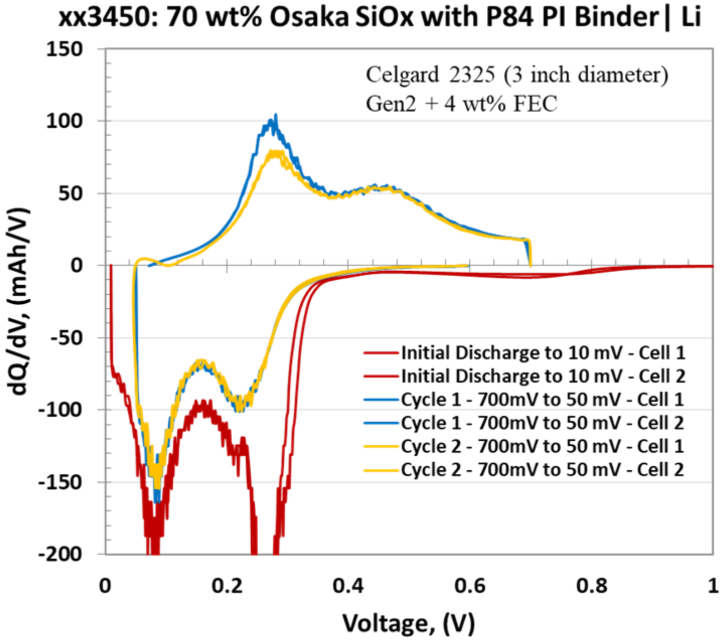
Evaluate processes to make electrodes at scale

See Poster bat499

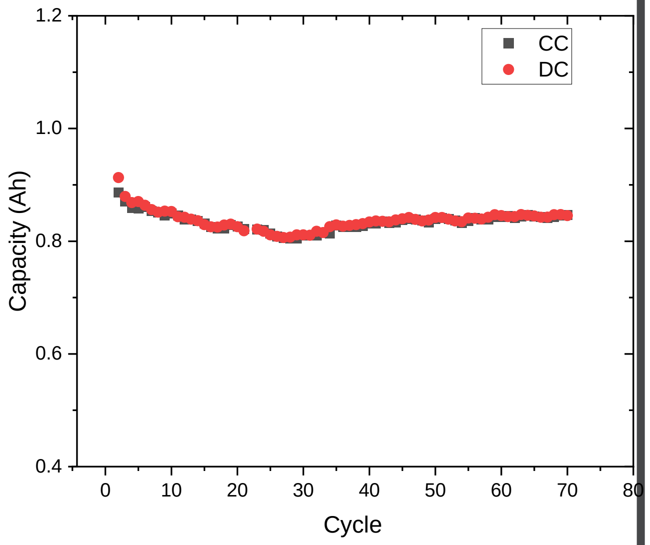
PRELITHIATION PROTOCOLS

Developed methods that could be uniformly applied from laboratory to laboratory

Electrochemical prelithiation compensates for initial lithium losses from SEI formation – good for all labs



Thermal evaporation lithiates silicon composite electrodes and can be readily incorporated within cell Highly effective at controlled addition of Li at scale.

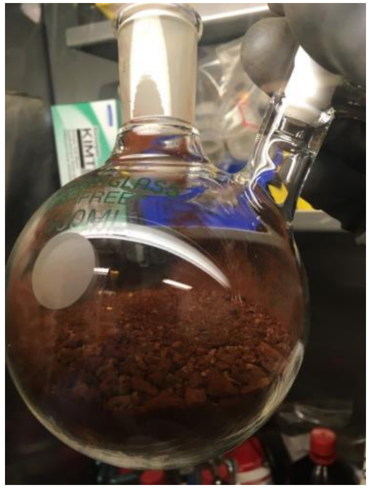


Also exploring prelithiation with SLMP materials and choice of solvents/salts to form SEI initially

Results and protocols extended to Cell Build and Materials Discovery Teams to speed development

FABRICATE NEW CARBON COATED MATERIALS

Scaled up experimental material and coating processes to 20 grams



~5.7 g of PECVD Si
(5.9 nm diameter)

1. Mix Si with petroleum pitch carbon/toluene sol'n (50/50 wt% Si/pitch)

2. Evaporate toluene from pitch carbon solution

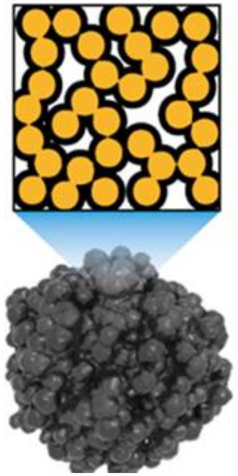
Si/pitch in toluene

Unannealed Si/pitch solid

3. Anneal Si/pitch under nitrogen, 700 ˚C, 1 h

4. Ball mill Si/pitch and sieve through 325 mesh screen

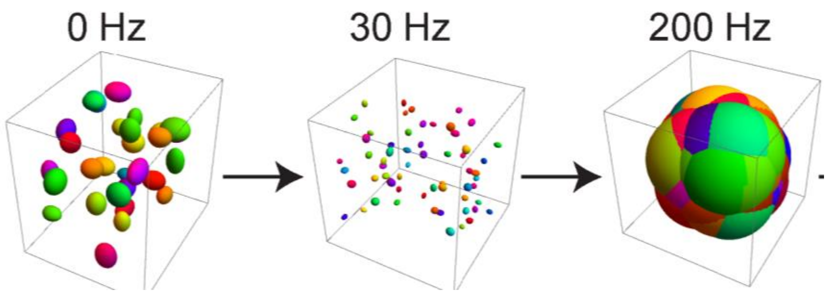
PECVD Si NPs produced and processed at



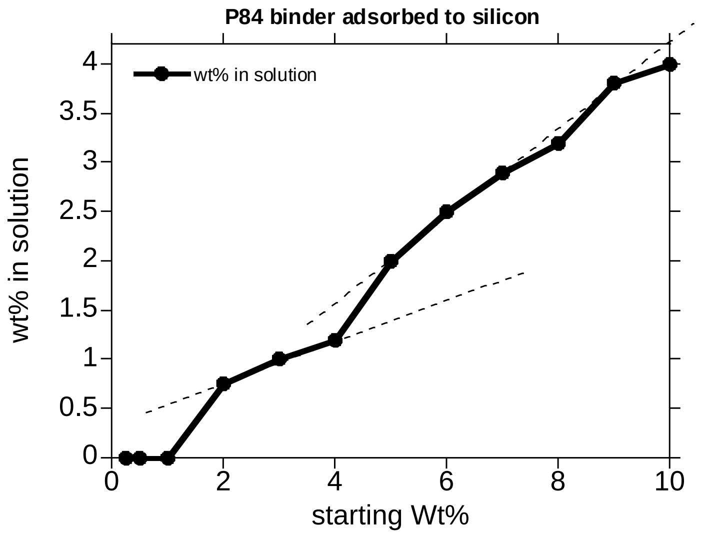
Explored pitch as a carbon source and evaluated carbon weight loading, colloidal properties, and how they related to electrochemistry and extended results to team to standardize processing.

UNDERSTAND AND INCORPORATE NEW POLYIMIDE BINDER

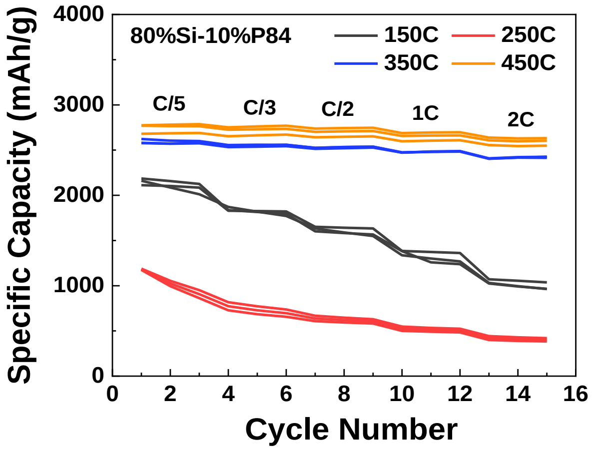
PI binder shows enhanced cycling stability over traditional materials – key is to rapidly utilize within program



Using neutron scattering able to identify casting regions with ideal polymer ordering ensuring rapid ion transport in electrode



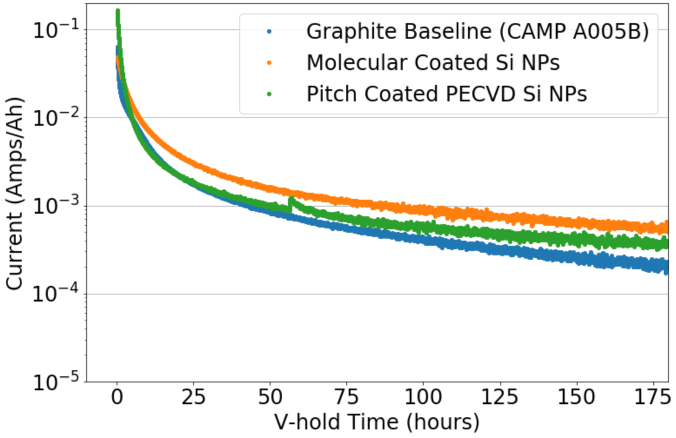
Polymer adsorption isotherms reveal regions of distinct polymer layering – provides guidance to rapidly optimize slurry processing



Explore drying protocols to produce best electrodes and distribute to consortium

Results and protocols extended to Cell Build and Materials Discovery Teams to facilitate processing and identify promising areas to explore

ADDRESSING CHALLENGE SCALING TO 4 AH CELLS

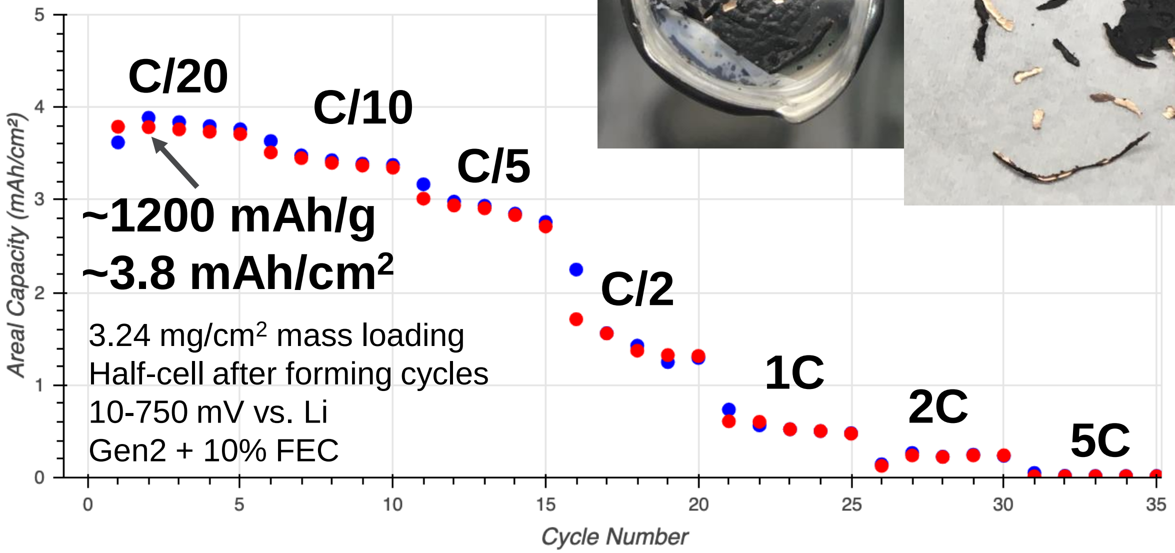


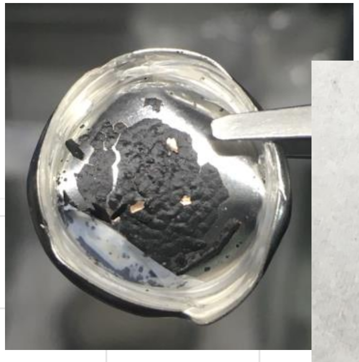
Identified pitch coated electrodes have outstanding calendar life in test cells.

However, at thicker loadings to reach cell metrics have problems with:

1. cycle performance
2. Mechanical forces destroying electrodes

Mechanical stresses shred current collector after cycling





Team is focusing on developing new electrode slurry protocols to facilitate ion transport and mechanical properties.

Results transferred to cell build team for scale up.

CONCLUSIONS AND NEXT STEPS

Conclusions:

- Achieved 1000 cycles with >80% capacity retention with new cell chemistries
- Eliminated HF etching of SiO_x as a viable process but able to pull out several new directions:
  - Polyimide binder shows promising mechanical properties and processing parameters
  - Identified pitch coatings as way to reduce corrosion current
- New, scalable, processes of prelithiation from evaporation
- Able to rapidly reduce the development time and Edisonian-like advances to cell development
- Standardizing protocols reduces development time and ensures transferability of results.

Next steps:

- Explore new processing regimes with reduced or mixed binder compositions
- Develop thermal evaporation to understand differences between and electrochemical and a chemically formed SEI
- Optimize carbon coatings beyond pitch → pyrolyzed carbons or specific functional coatings
- Work with mechanical team to understand properties of P84 electrodes under lithiation

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