Low Cost Manufacturing of Advanced Silicon-Based Anode Materials

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Avery J. Sakshaug, Abirami Dhanabalan, Chris Timmons, Aaron Feaver, Henry R. Costantino (PI)

Project ID: ES268

Overview

Timeline

- Start Date: January 2016
- End Date: September 2018
- Percent Complete: 80%

Budget

- Total Project Funding
  - DOE: $2.81M
  - G14: $1.23M
- Funding received in FY 2017
- Funding for FY 2018
  - No-cost extension

Barriers

- Cost: Anode materials that contribute towards the DOE target of $125/kWh
- Performance: Silicon based anodes to improve Li-ion energy density for vehicles
- Life: Maintain current cycle life of graphite anode Li-ion batteries

Partners

Relevance

- **Cost**: Current Li-ion battery cost structure will not enable widespread use of battery electric vehicles (BEV) or plug-in hybrid electric vehicles (PHEV)
  - Current technology trajectory will increase performance, but also increase cost
- **Performance**: BEV and PHEV range needs to be extended by increasing Wh/kg and Wh/L and maintaining power capability
- **Cycle Life**: Batteries with short life time i.e. 2-3 years can be tolerated in consumer electronics but not vehicles
- **Group14 targets**:
  - **Reduce cost** compared to current anodes for Li ion batteries
  - **Improve capacity** – increase EV range
  - **Maintain cycle life** of current batteries

Milestones

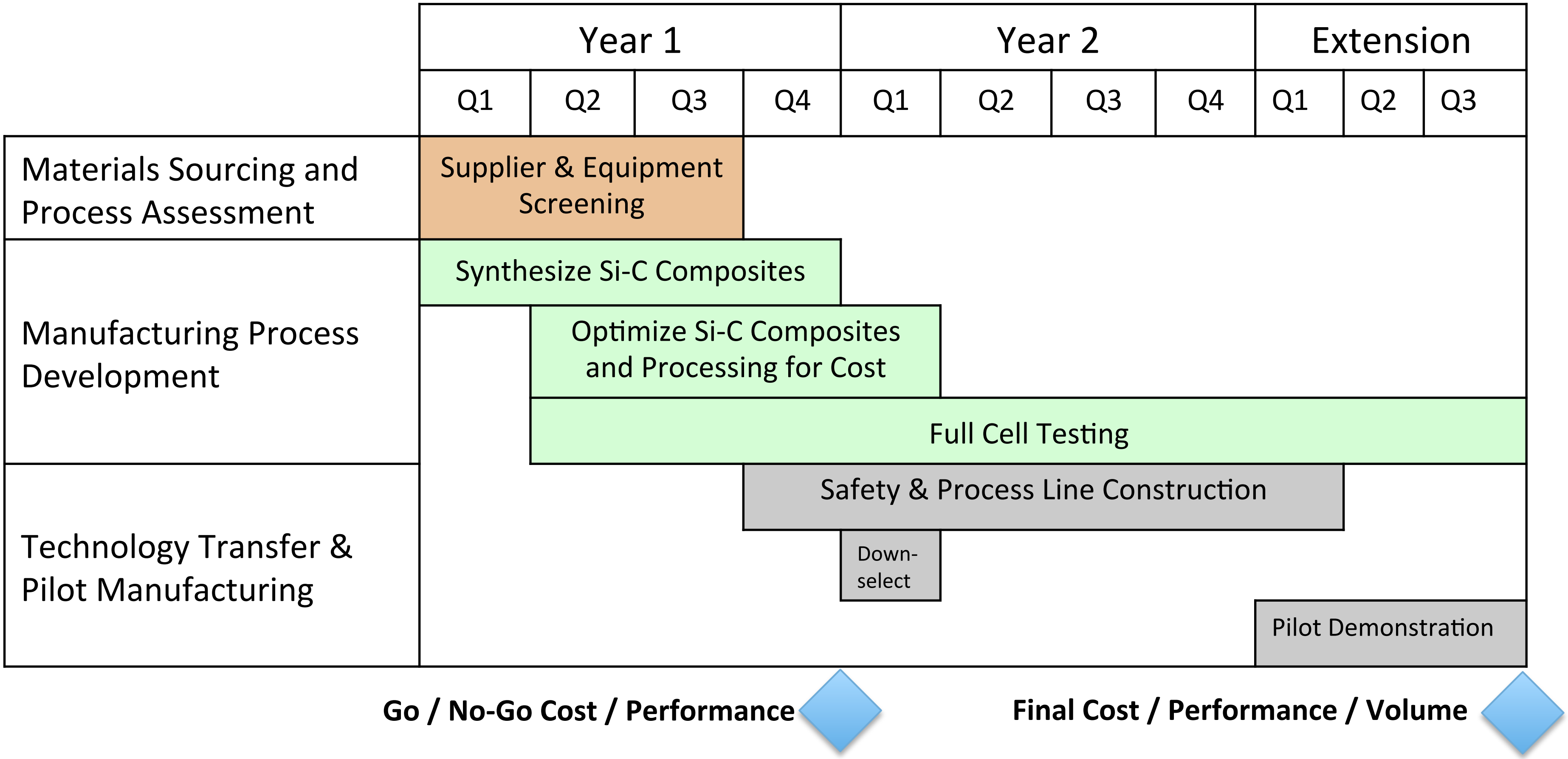
Year 1

Milestone	Type / Timing	Description
Supplier Identification	Technical / Q1	Identify minimum 2 suppliers for each new feedstock material required for Si-C composite. Materials must be available at full scale volume supporting < $125/kWh.
Sample Down-select	Technical / Q2	Down select to 3 lab-scale silicon samples for performance and cost
Synthesize Si-C	Technical / Q3	Synthesize 1x10g Si-C with 1000 mAh/g
Synthesize Si-C	Go/No-Go / Q4	Analysis indicates that the synthesized 1x1g Si-C with 1000 mAh/g is predicted to achieve 500 cycles at a projected cost of <$125/kWh

Year 2 / No-cost Extension Period

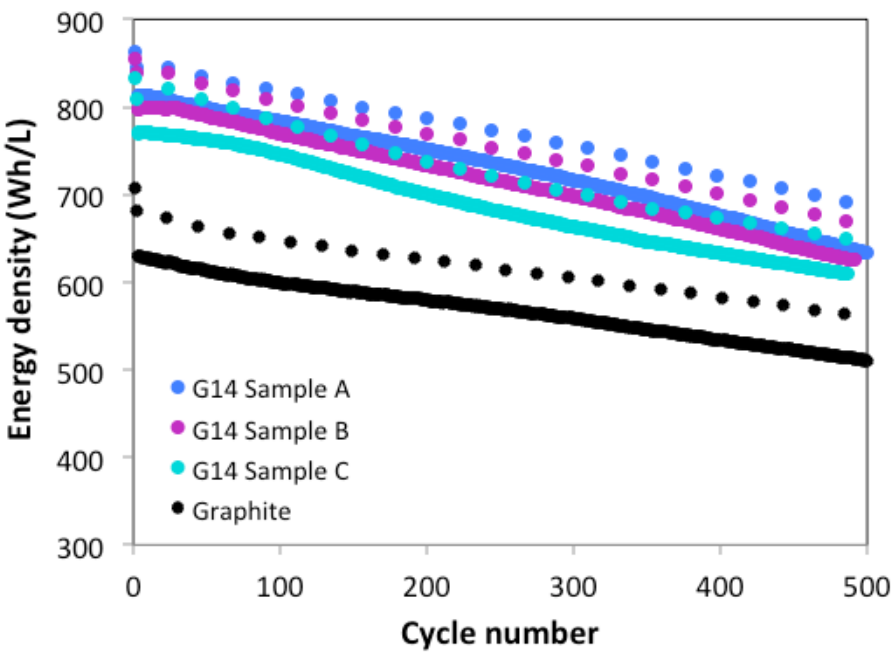
Milestone	Type	Description
Synthesize Si-C 1000 cycles	Technical / Q2	Synthesize 1x1g Si-C with 1000 mAh/g; predicted 1000 cycles; < $125/kWh projected cost
Performance Validation	Technical / Q3	Validate performance of at least one pilot-scale-synthesized material in the lab
Commission Equipment	Technical / No Cost Extension Period	Complete installation and commissioning of all new process equipment
Synthesis with Demo	Technical / No Cost Extension Period	The synthesis of 10 kg completes a demonstration 1000 mAh/g and predicted 1000 cycles at < $125/kWh at full scale volume

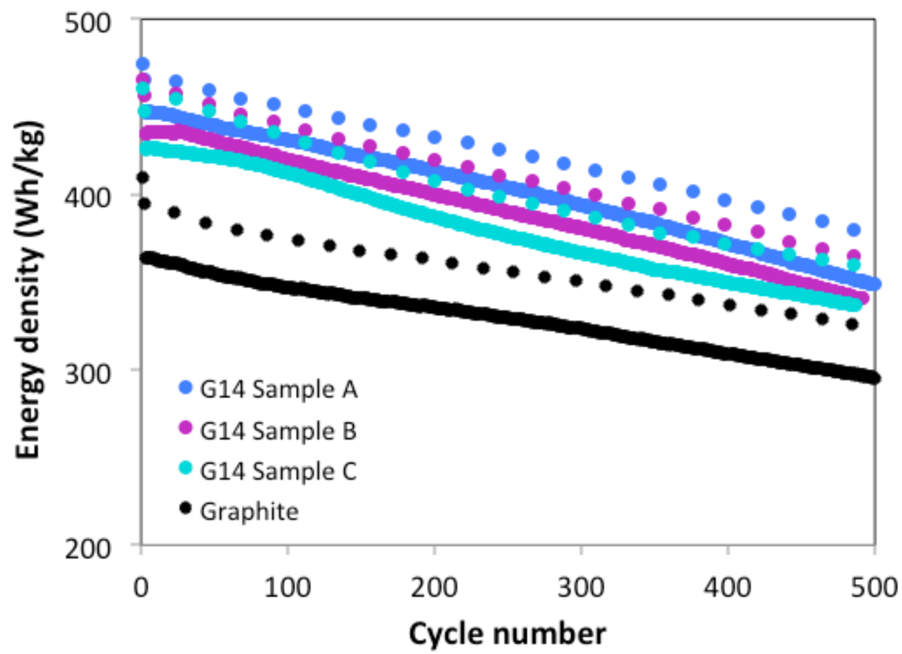
Approach / Strategy



- Leverage EnerG2 carbon expertise in carbon to create an ideal silicon support matrix material
- Develop and implement low cost silicon synthesis process compatible with the carbon platform
- Demonstrate success of the approach in full cell LIBs
- Manufacture at pilot scale for qualification with LIB customers using low cost process

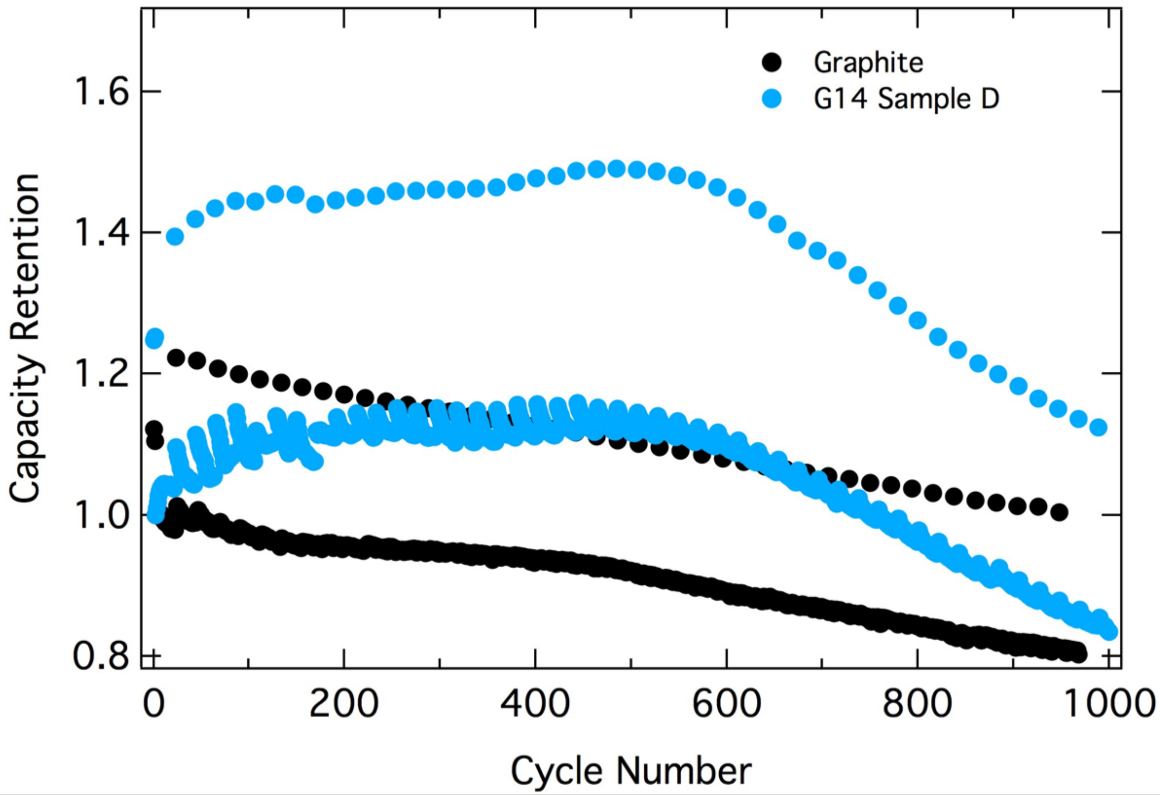
Technical Accomplishments:
Example of >500 Cycle Stability in Full Cells





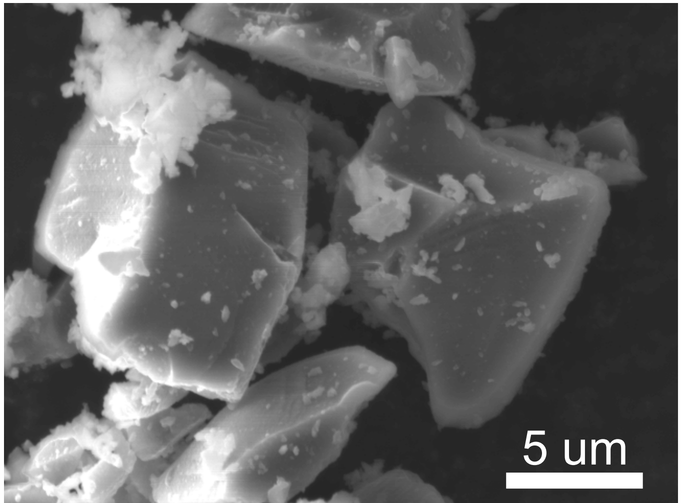
Volumetric energy (Wh/L) for three representative Si-C samples, cycled at C/2 rate, 2.5 – 4.2 V, with I/2 hold, cycled at C/10 every 20 cycles, 1 M LiPF6 in EC:DEC w/10% FEC, LiNiCoAlO cathode, 5-15% excess anode (anode=5% CMC-SBR, 5% Super-C45, ~16-19% Si-C, ~74-71% graphite).

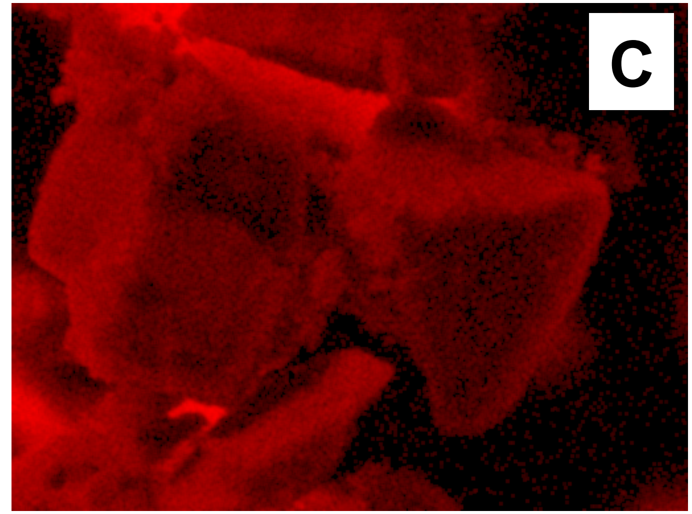
Technical Accomplishments: Example of Device-Level Enhancement

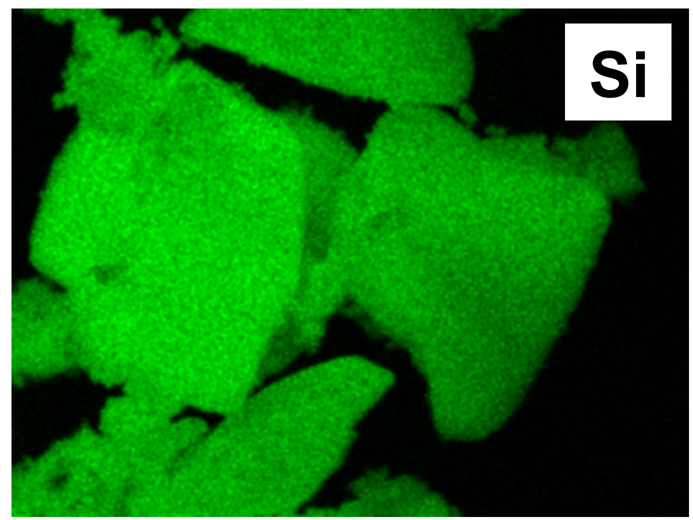


Capacity retention for pre-lithiated G14 Si-C, cycled at C/2 rate, 2.5 – 4.12 V, with I/2 hold, cycled at C/10 every 20 cycles, 1 M LiPF6 in EC:DEC w/10% FEC, LiNiCoAlO cathode, 5-15% excess anode (anode=5% CMC-SBR, 5% Super-C45, 31% Si-C, 79% graphite).

Technical Accomplishments: Homogeneity of Si in Composite





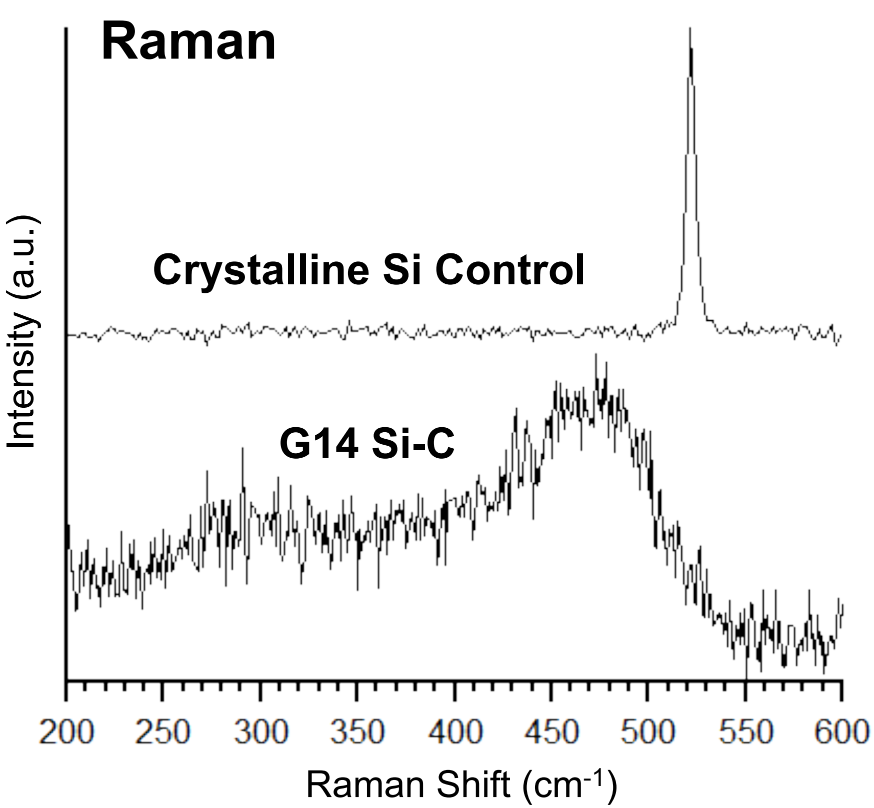


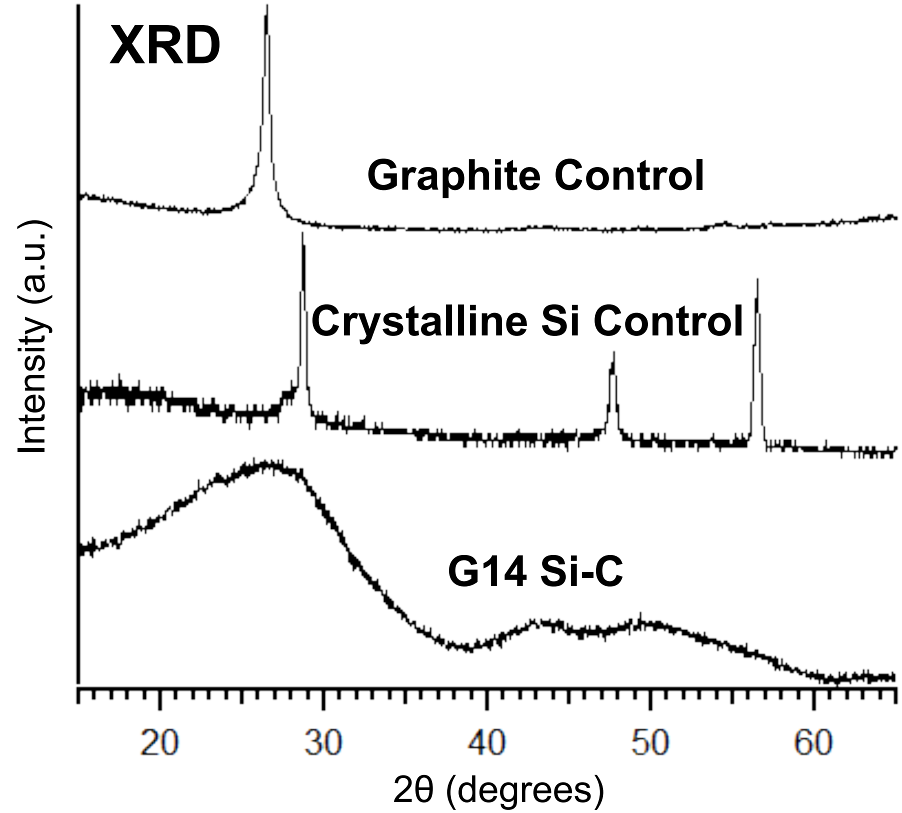
- Particles were examine be SEM in combination with energy-dispersive X-ray spectroscopy (EDS) to allows for elemental analysis and mapping
- Data demonstrates uniform distribution of carbon (C) and silicon (Si) throughout each particle

Acknowledgements:
Matt Lim & Peter Pauzauskie, UW

Molecular Analysis Facility

Technical Accomplishments: Ability to Achieve Amorphous Composite





- Raman and X-ray diffraction spectra consistent with amorphous Si and C

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Molecular Analysis Facility

Technical Accomplishments: Progress on Scale-up to Achieve Cost and Performance Requirements

- Specified, ordered, installed, and commissioned pilot scale equipment
- Identified, procured, outfitted, and obtained necessary operating licensure for MFG site in Woodinville, WA
- Secured pilot-scale quantities of raw materials to produce Si and C
- Pilot manufacturing in progress, experience to date support previous COGS projections commensurate with <$0.034/Ah as confirmed at lab scale
  - Consistent with project goal < $125/kWh at full scale volume

Responses to Previous Year Reviewers' Comments

Comment: *"…the data appear to show that all of the Si composite anodes are at the same energy density as a standard graphite anode after 500 cycles, and the trend line continues to point downward at a more rapid rate than for graphite only. The reviewer interpreted this to mean that the Si does not appear to be stabilizing in any way and the only stabilizing component is the graphite itself."*

> **Response**: G14 has shown multiple examples of cells with higher energy density relative to graphite @ cycle 500 (*vide infra*). Our capacity fade has similar trend as the graphite control we used in our device format and testing conditions. The increase in the energy density shown here comes from the addition of G14 Si-C composite.

Comment: *"…the development of Si anodes should be focused on electrolyte-anode pairing and stable SEI formation; it seems unlikely that this material will meet end of program 1,000 cycle goals using an off-the-shelf electrolyte formulation (and the reviewer understands this is likely outside the scope of this program)."*

> **Response**: We routinely test without any device or electrode level enhancements (except using FEC in electrolyte) to demonstrate G14's Si-C commercial relevance as a drop-in technology. With electrode level enhancement such as prelithiation, our material is capable of reaching up to 1000 cycles of stability (*vide infra*).

Responses to Previous Year Reviewers' Comments, Con't

Comment: *"…the coin cell data appear quite good: however, the reviewer would like to see more than "one or two" good cell before the team proceeds with process scale-up."*

> **Response**: We typically test 3 to 6 cells per sample to check the reproducibility of our results. We have shown in 2017 as well as in 2018 AMR presentations six different cells achieving ~500 cycles of stability.

Comment: *"…the energy density was very high, close to 800 Wh/L at 4.2V 2.5V at a C/10 rate. The reviewer said that the team needs to provide enough details on the cell modeling (e.g., loading, specific capacity of the Si/C composite, porosity, first cycle irreversible loss) to show how they achieved 800 Wh/L at only 4.2V."*

> **Response**: We calculate Wh/kg based on the active mass of anode and cathode; similarly, Wh/L is calculated based on the volume of anode and cathode laminates; Since we use coin cell format to test our materials, we do not include the weight or volume of the electrolyte, separator, current collector, and coin cell parts.

Comment: *"…the team needs to include a third-party to perform independent validation of the excellent density achieved at 4.2V…the reviewer would like to see more cycling and cell test data, perhaps by partnering with a group able to do that…"*

> **Response**: G14 has already obtained validation of excellent anode performance at lab scale with multiple third-parties, although said data are not sharable per partner agreements. Even so, and more importantly, G14 will obtain and present third-party validation of performance at pilot scale, commensurate with the stated goals on this DOE project.

Collaboration and Coordination with Other Institutions

- University of Washington Subcontract
- Pauzauskie Lab: Funded graduate student Matt Lim
  - Material modeling
  - Advanced characterization

- PNNL Subcontract
- Chongmin Wang Group: Funded post-doc Langli Luo
  - *In situ* TEM of Silicon Expansion
  - SAED
  - Advanced spectroscopy

Remaining Challenges and Barriers

- Improve Si-C full cell stability from 500 to 1000 cycles
- Produce pilot material and demonstrate performance in industrially relevant full cells
- Validate Si-C performance and cost targets at full scale volume according to DOE targets

Proposed Future Work

- Materials and process optimization at pilot scale (~kg)
- Pilot-scale validation of performance in full cells while maintaining projected cost target
  - Employ 3rd party for device construction
- Customer validation of Si-C performance at pilot scale

Summary

- Continued lab-scale validation of G14 Si-C performance
  - >500 cycle stability in Li ion full cells
  - Additional stability (e.g., 1000 cycles) achievable via device level enhancements
- Currently at pilot scale (~ kg)
  - Refinement of pilot process
- Progressing towards final project milestones
  - Pilot scale production meeting required COGS projected at full scale volume
  - Pilot material performance in industrially relevant full cells