Development of High-Performance Lithium-Ion Cell Technology for Electric Vehicle Applications

P. I. Madhuri Thakur
Farasis Energy
June, 12th 2019

Project ID # bat355

FARASIS Empowering Innovation

Program Overview

Timeline

- Project Start Date: Feb. 2017
- Project End Date: Sept. 2020
- Percent complete : 66%

Budget

- $5.9M
- 50%/50% USABC/Farasis
- Subcontractors
- LBNL $300K
- ANL $400K

Barriers

- Achieving high energy density with stable chemistry to meet cycle life and calendar life goals.
- Meeting the cost target of ~ $0.10/Wh
- Manufacturing processes compatible with new materials

Partners

- BASF
- 3M/OneD material/SiO Supplier
- Argonne National Lab
- Lawrence Berkley Lab
- Solvay/ Daikin
- Entek/Celegrad

Relevance/Project Objectives

- <u>Project Objective</u>
  - Develop a EV cell technology capable of providing 280 Wh/kg after 1000 cycles at a cost target of $0.10/Wh.
  - To achieve this the BOL cell energy density target will be ~ 330Wh/Kg.

CELL LEVEL ATTRIBUTES	Units	Baseline (BOL)	Final Cells (BOL)- 2017	Final Cells (BOL)-2018
Cell Capacity (C/3 Rate discharge)	Ah	28.5	64	87
Cell Volume (without terminals/tabs)	L	0.222	0.24	0.3984
Cell Mass	kg	0.49	0.58	0.947
Vmin continuous, Vmax continuous (0 and 100% SOC)	V,V	3.0, 4.2	2.5, 4.5	2.75, 4.2
Vmin pulse, Vmax pulse (10 sec pulses)	V,V	2.5, 4.25	2.4, 4.5	2.5, 4.25
Vnominal (Wh/Ah)	V	3.68	3.55	3.6
Energy Density (volumetric)	Wh/L	470	947	786
Specific Energy	Wh/kg	215	392	331
Power Density (10 sec. HPPC power), 50% SOC	W/L	5440	2910	1778
Specific Power (10 sec. HPPC power), 50% SOC	W/kg	2460	1204	748
Target Cost / unit (>10 million cells/annum rate)	$	16 - 19	25 - 31	25 - 31
Cell format (cylindrical/prismatic)		Pouch	Pouch	Pouch
Cell dimensions: (height x width x thickness)	mm	230 x 160 x 6	230 x 160 x 6	294.4 x 100.4 x 14

Relevance/Project Objectives

- **Project Technical Target**
  - Year 1: Baseline deliverable (220Wh/Kg)
  - Year 2: Gen1 Deliverable (300Wh/Kg)
  - Year 3: Gen2 Deliverable (330Wh/Kg)
- **Cell Component R&D**
  - Comparative evaluation of electrode active materials.
  - Optimization of negative electrode formulation to achieve maximum energy density and cycle life.
  - Investigate effect of Si incorporation on negative electrode conductivity and mechanical stability relative to graphite-only active material electrodes.
  - Additive package development to optimize SEI formation on Si anodes.
  - Stability of Ni rich Cathode
- **Risk**
  - There are multiple interacting failure mechanisms at the materials and cell level that are barriers to achieving the system level battery performance goals.
  - Calendar life of the Gen1 and cycle life for Gen 2 Cells
  - Safety and cost targets

Project Milestones

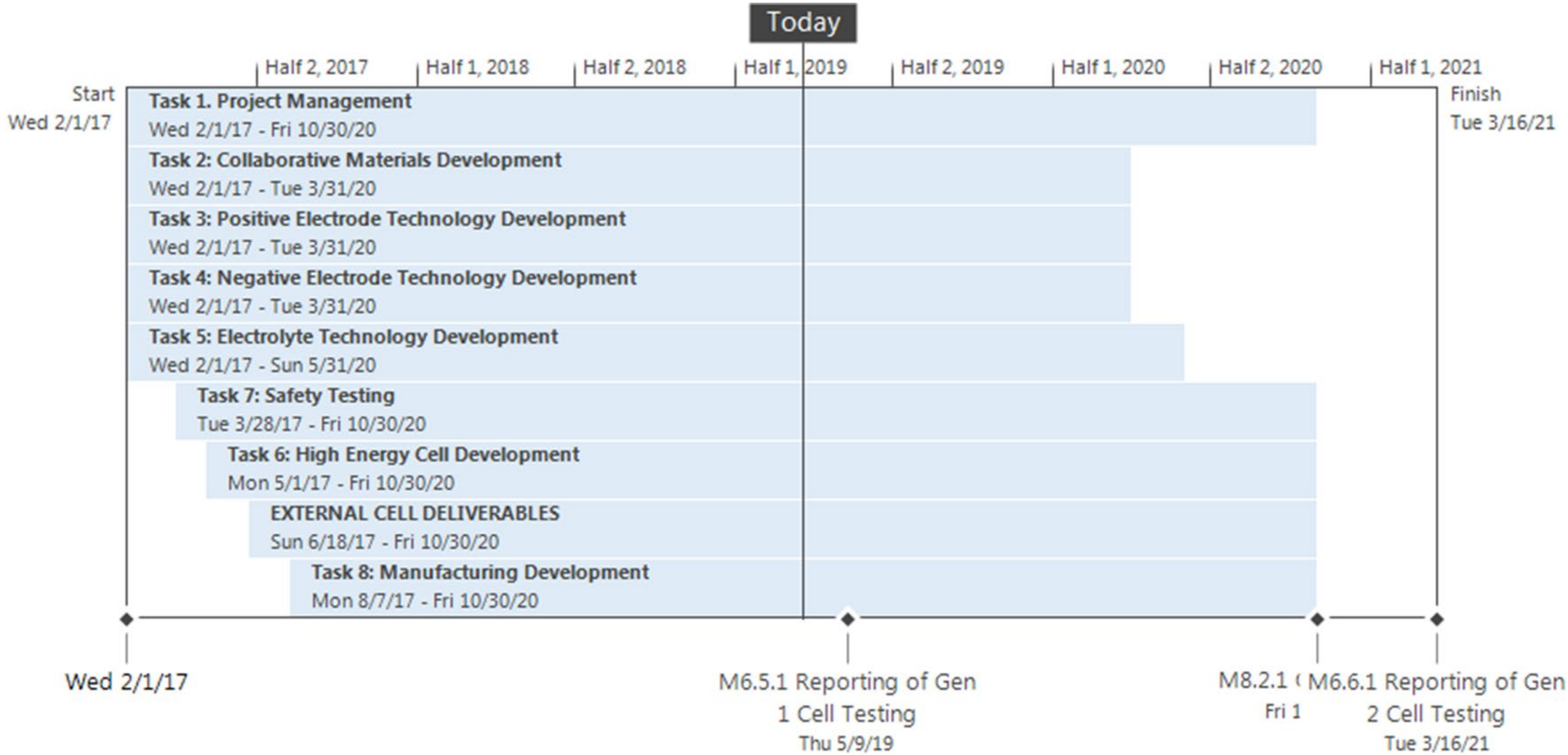
- The key Milestones are associated with demonstrating progress towards the goals of the project.

Milestone Summary Table

Task Number	Task Title	Milestone or Go/No-Go Decision Point	Milestone Number	Milestone Description (Go/No-Go Decision Criteria)	Planned Completion Dates	% Completed
3.3	Positive Electrode Developmental Cell Build	Milestone	M3.3.1	Production of pouch cells representing multiple positive electrode design variants	July, 2017	100%
4.3	Negative Electrode Developmental Cell Build	Milestone	M4.3.1	Production of pouch cells representing multiple negative electrode design variants	July, 2017	100%
6.2	Ongoing Cell Development	Go/No-go	G6.2.1	Demonstration of High-Energy Cells exceeding 300 Wh/kg after RPT2	April, 2018	100%
6.5	Generation 1 Cell Testing	Milestone	M6.5.1	Completed reporting of performance and safety testing results for Generation 1 cells	January, 2019	90%
8.2	Cost Model Development	Milestone	M8.2.1	Submission of detailed cost model based on new materials and processes used in Generation 2 cells.	October, 2020	20%
6.7	Generation 2 Cell Testing	Milestone	M6.7.1	Completed reporting of performance and safety testing results for Generation 2 cells	October, 2020	0%

Milestones Timing

- The key Milestones are associated with demonstrating progress towards the goals of the project.

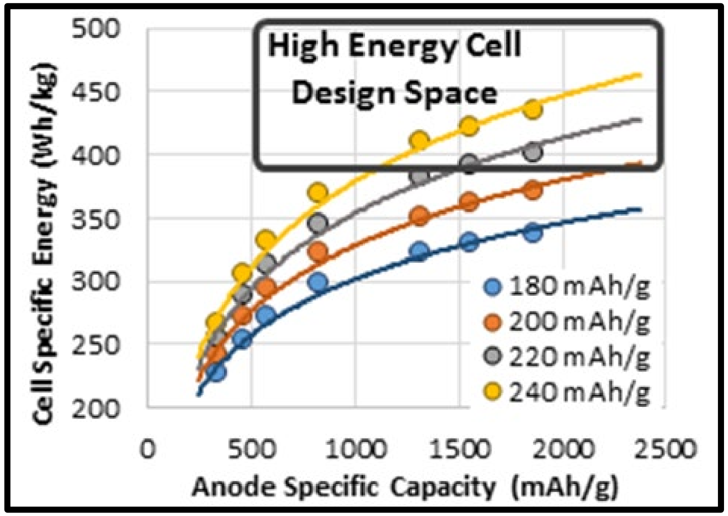


Technology Approach

- Development focused on addressing key current barriers to achieving high capacity, long cycle life and safer Li-ion cells
- **Electrode Chemistry:**
  - Stabilized Ni-rich Cathode: Capacity ~ 220-240 mAh/g
  - Silicon Composites: Capacity ~ 400-600 mAh/g

  Contributors: Argonne National Laboratory, OneD, 3M, SiO supplier , BASF etc.
- **Electrolyte Formulation:**
  - Fluorinated solvents
  - Stabilizing additives/salts.

  Contributors: Solvay, Daikin, 3M, BASF



- **Electrodes and Cell:**
  - Low reactivity conductive additives
  - Advanced binder formulations
  - Advanced separators: Coatings, high voltage stability

  Contributors: Lawrence Berkeley National Laboratory, Entek, Celgard

Strategy - Development Plan

Task Status: Colors	
Blue	Done
Green	Active with some delay
Red	Stopped
Black	Non-Active

Cell materials development

New materials sourcing and characterization
New materials improvement, synthesis, development (ANL, LBNL)
Form factor: Coin Cells
Metrics: Capacity, rate, stability

Positive electrode

C1: ~ 11 Materials Variations (~ 300 Wh/Kg)
C2: ~ 3 Materials Variations (shift to 400Wh/kg)
C3 : ~ 2 Materials Variation (Target ~ 330Wh/Kg)
Form Factor: Small Full cells (SLP - <2Ah), fixed anode, electrolyte, energy density
Metrics: Capacity, rate, cycling, safety

Negative electrode

A1: ~ 4 Materials Variations (~ 300Wh/Kg)
A2: ~ 4 Materials Composite Variations (shift to 400Wh/Kg
A3: ~ 2 Materials Variations (Target ~ 330Wh/Kg)
Form Factor: Small Full cells (SLP - <2Ah), fixed anode, electrolyte, energy density
Metrics: Capacity, rate, cycling, safety

Electrolyte

E1: ~ 10 Materials Variations (~ 300 Wh/Kg)
E2: ~ 5 Materials Variations (~ 320 Wh/Kg)
E3 : ~ 5 Materials Variations (~ 330 Wh/Kg)
Form Factor: 18650, <2Ah pouch, single cell design
Metrics: Capacity, rate, cycling, calendar life, safety

Cell

DOE1: ~ C1 & A1 (Optimized formulation for 3 C1 & 2 A1 with 5 E1)
DOE2: ~ C2 & A2 (Optimized formulation for 2 C1 & 2 A1 with 5 E1)
DOE3: ~ 7 Materials Variations
Form Factor: Full cells, fixed anode, electrolyte, energy density
Metrics: Capacity, rate, cycling, safety

Deliverables:
Baseline: Baseline (30Ah) NCM/Graphite
Gen1: Gen 1 Cells (40Ah) Chemistry from **DOE 1**
Gen 2: Gen 2 Cells (65Ah) Chemistry from **DOE2**

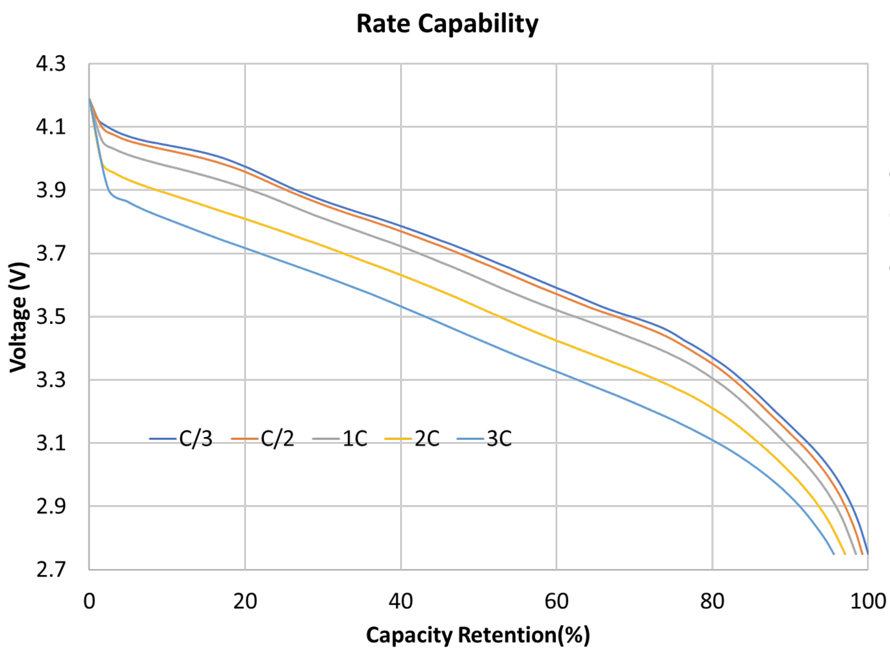
Technical Accomplishments
Gen1: Builds

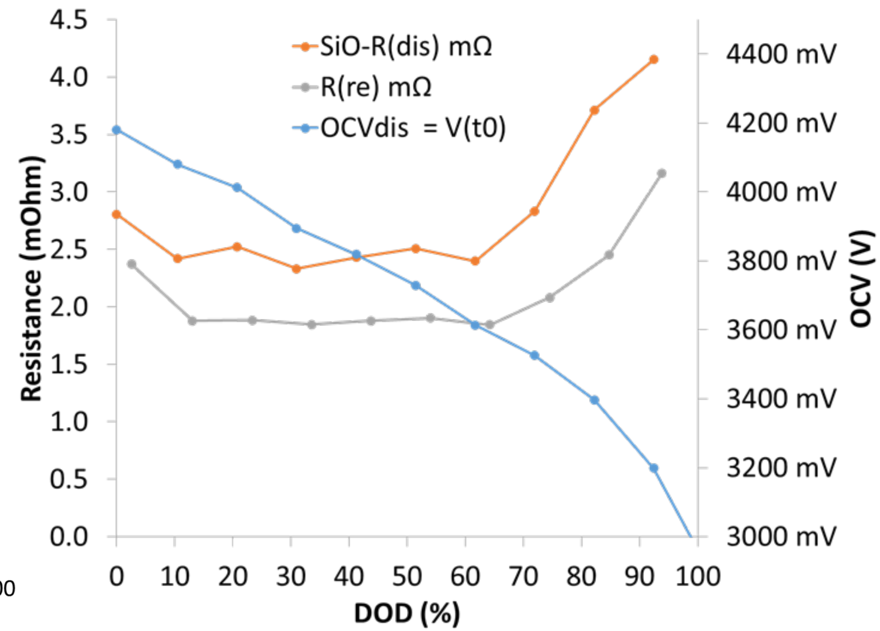
- **Number of Cell Builds**
  - Single layer pouch cell with 12 different cathode and 4 anodes
  - Single layer pouch cells for down-selected cathodes and anodes
  - 18650 Cells for the safety of materials and electrolyte optimization
  - Electrolyte optimization on 1.2 Ah pouch cells for down-selected cathode and anode
  - 35Ah Cell build with High voltage and Ni rich cathode and down selected anode
  - 2.5Ah/ 3Ah Cell build with Gen1 deliverable chemistry for cycle and storage life
  - Gen1 Delivered to National Labs
    - Energy Density: 600Wh/L
    - Specific Energy: 270Wh/Kg
    - Capacity: 41.5 Ah

Technical Accomplishments
Gen1 Build 1(35Ah) : DCR & Rate Capability

- Gen 1 Build 1 (35Ah):
  - Rate Capability
  - Cathode: Ni rich cathode
  - Anode: Si and carbon
  - Specific Energy: 265Wh/Kg

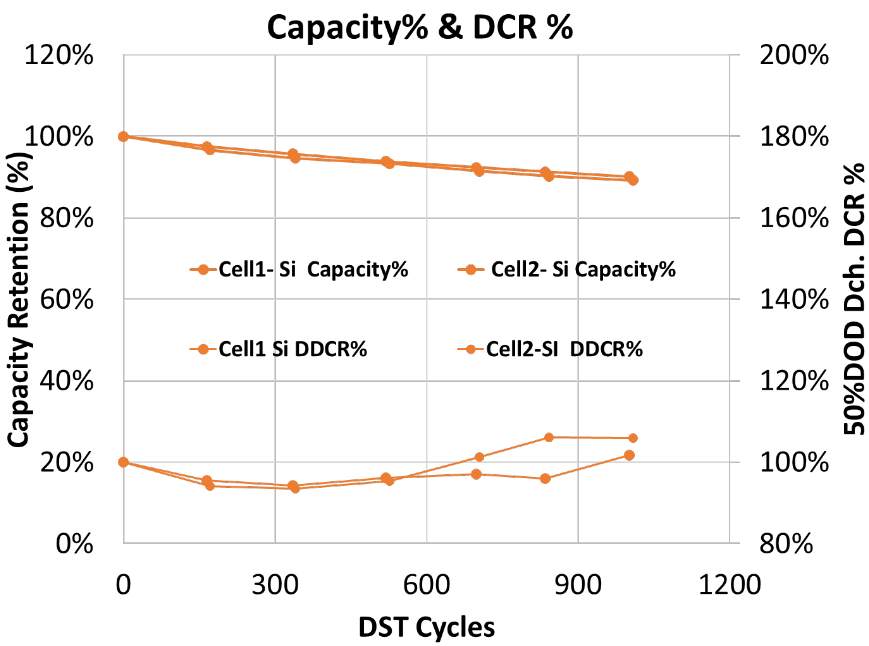
- Gen 1 Build 1 (35Ah):
  - HPPC: DCIR

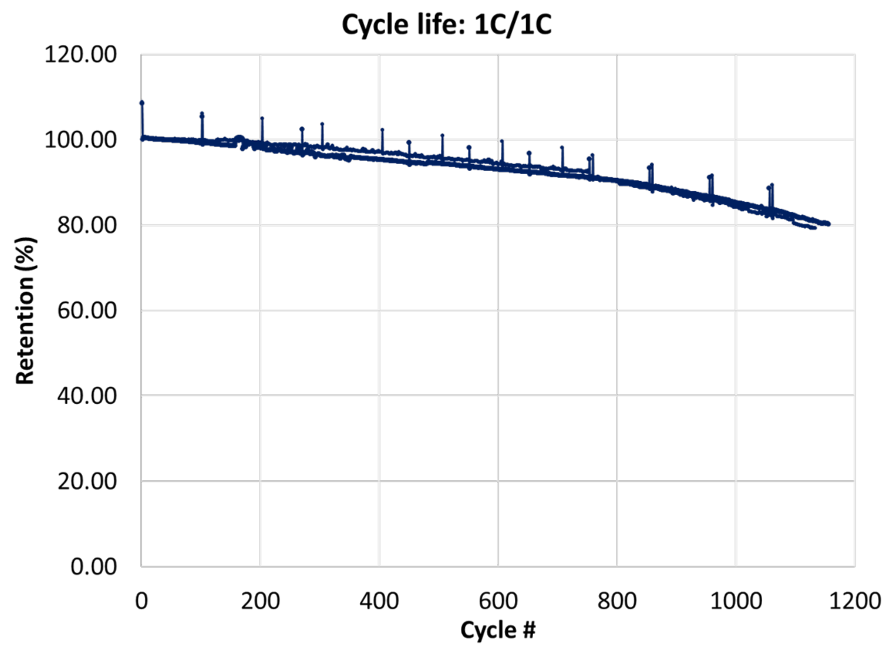




Technical Accomplishments Gen1 Build 1(35Ah) : Cycle life

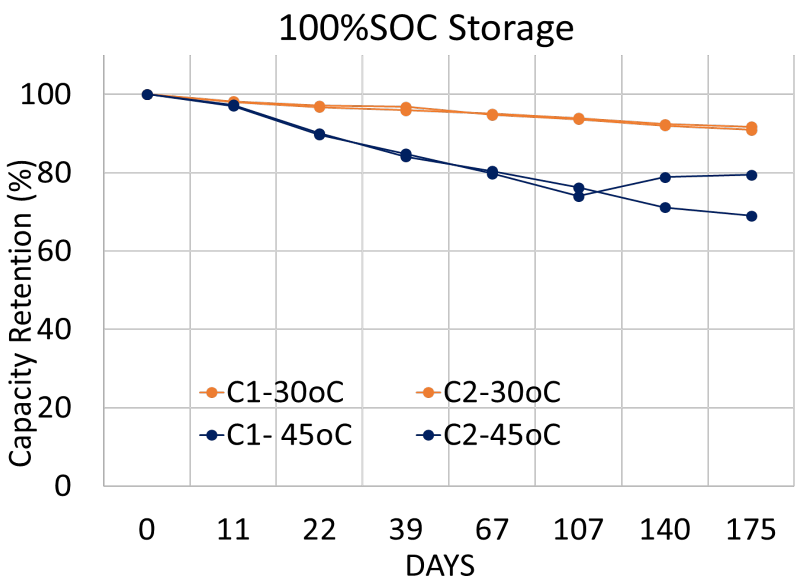
- DST Cycle life @ 30°C & Cycle life: 1C/1C
- Electrode : Ni rich cathode & Si carbon composite anode
- Specific Energy: 265Wh/Kg

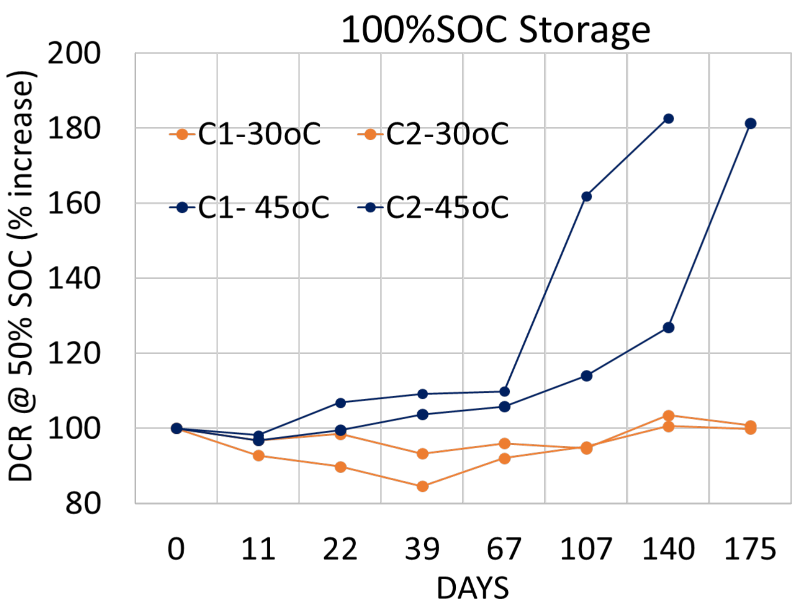




Technical Accomplishments
Gen1 Build 1(35Ah) : Calendar Life

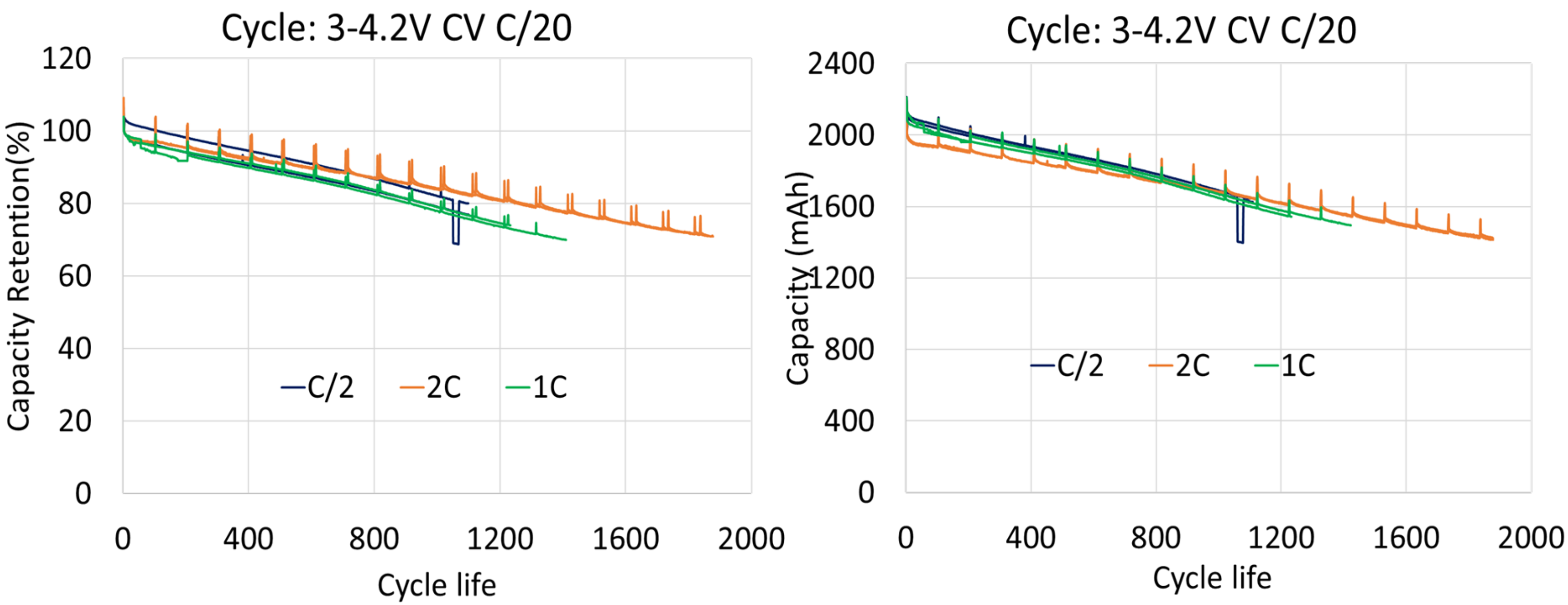
- Impedance, thickness and capacity





- Capacity retention and DCY at high temperature is not performing well.
- Optimizing the electrolyte additive for the high temperature storage performance

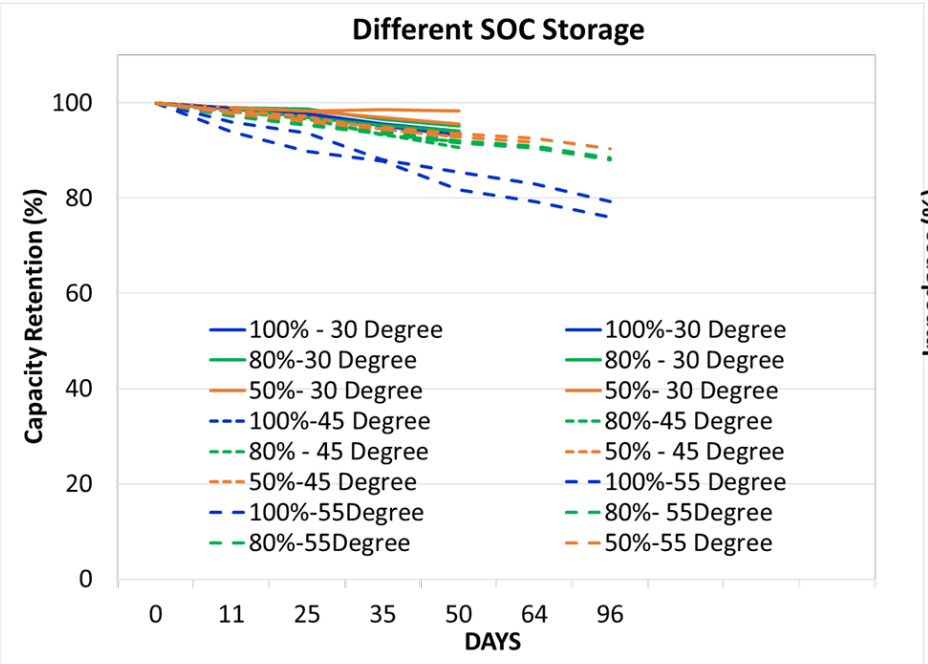
Technical Accomplishments
Gen1 Build 2(2.5Ah): Cycle Life

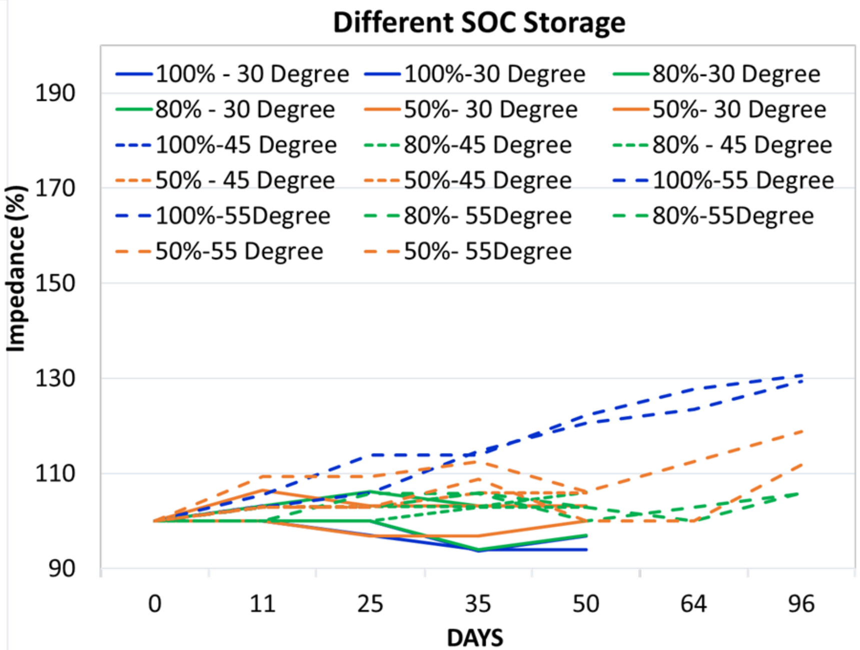


- Cell Charged @ 2C and 1C
- Dissected at the end of the cycle life to check for the Li plating- No Plating

Technical Accomplishments
Gen1 Build 2(2.5Ah): Calendar Life

- Impedance and capacity @ different SOC

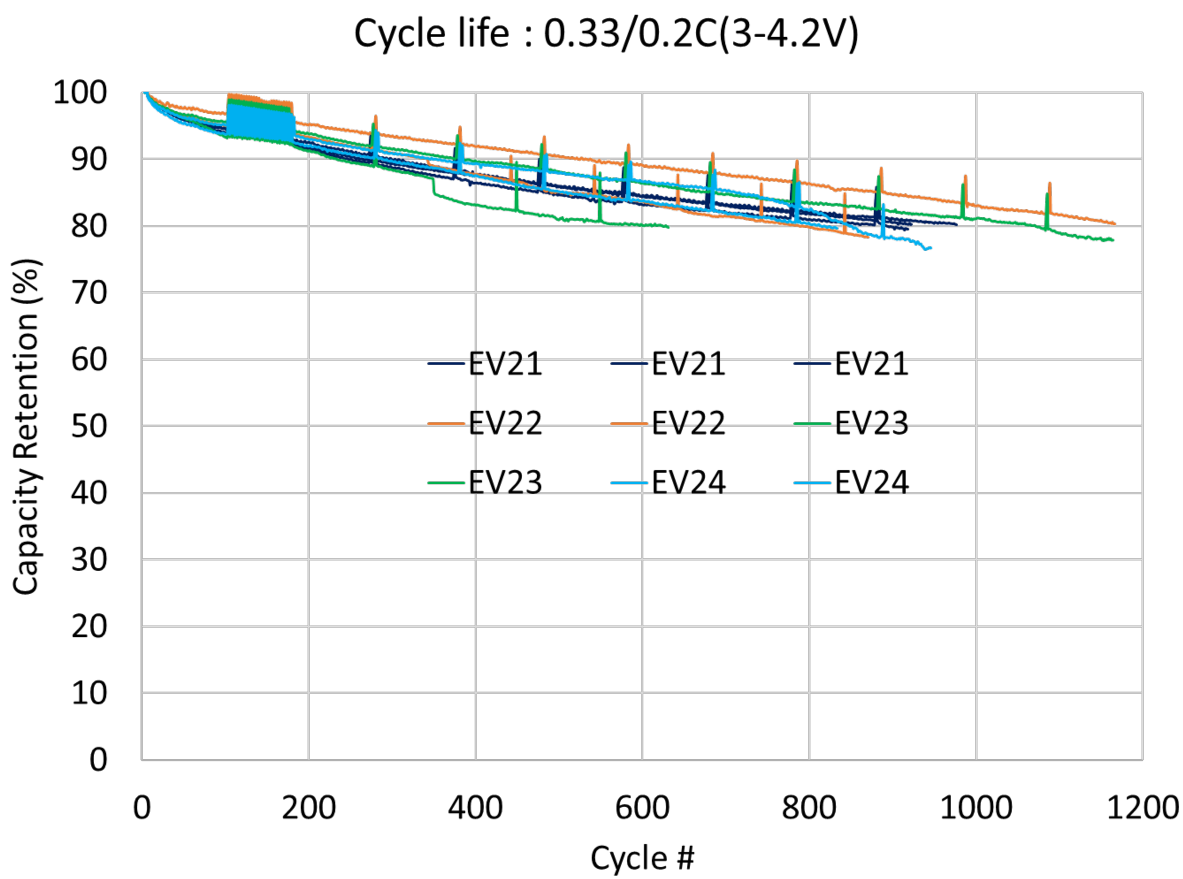




- Capacity retention and impedance for the cell stored for three months at 55° is poorer as compared to low temperature
- Storage of small cell is better as compared to the big cell, which indicates that the form factor impact the calendar life of the cells

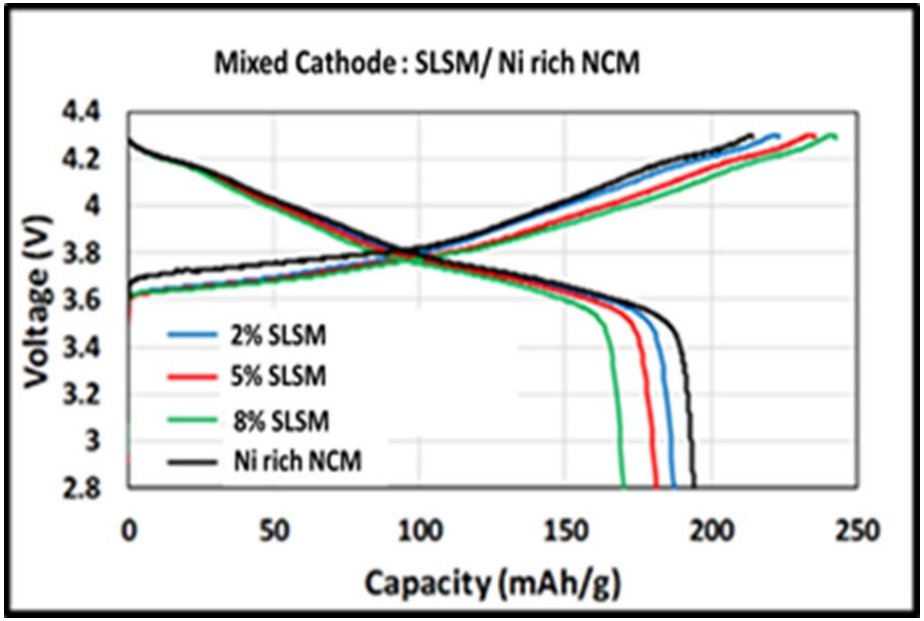
Technical Accomplishments
Electrolyte Technology Development

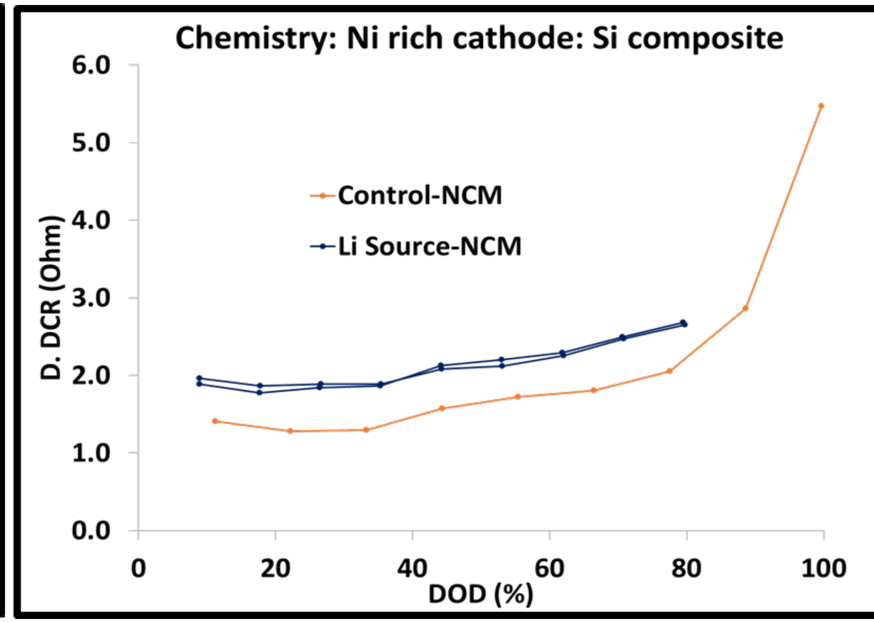
- Evaluated C1.1 (Ni rich NCM) with Si-graphite anode in a single layer pouch cells (at fixed anode capacity, same cathode & targeting 300 Wh/kg): Cycle life



Technical Accomplishments
Challenge with Li Source Development

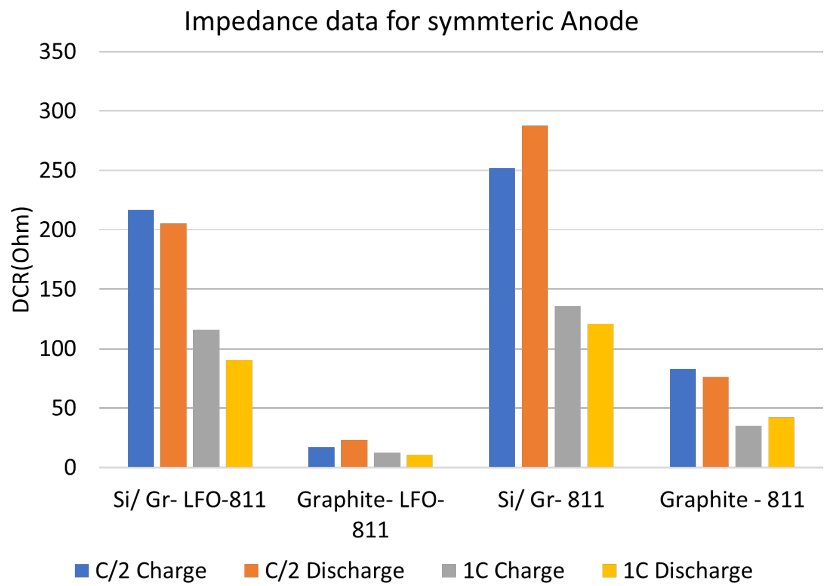
- Cathode: Ni rich NCM
- Anode: Silicon composite

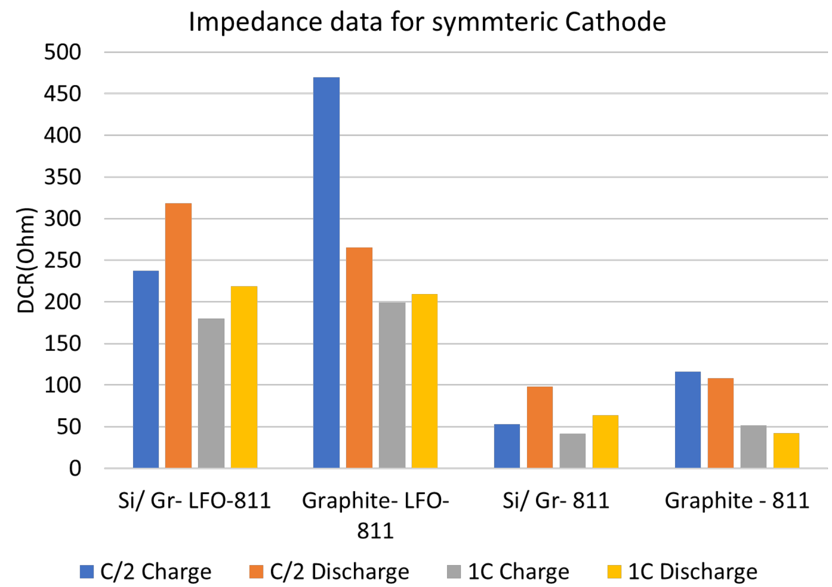




- Li source addition decrease the capacity of the cathode and to achieve the required energy Density need to use the anode with higher capacity or cathode with higher capacity
- Addition of Li source increase the impedance of the Cell

Technical Accomplishments
Challenge with Li Source Development





- Symmetric cells were assembled after washing the electrodes with EMC and left overnight in the glovebox before assembling.
- Some cells shorted and couldn't run the test protocol and are indicated above
- Anode with pure NCM have higher impedance as compared to the anode with LFO
- Cathode with LFO have higher impendence as compared to the pure NCM cathode.

Technical Accomplishments Summary Cell Development

- Baseline Cell Builds
  - Chemistry: NCM/Graphite
  - Delivered to National Labs
- Gen1 Cells Builds
  - Form Factor: Small pouch cells/ 40 Ah/ 80Ah
  - Chemistry: NCM/Si-Graphite
  - Specific Energy: 260-300Wh/Kg
  - Delivered to National Labs
- Internal Gen1 Cell Testing
  - Cycle life
  - DST Cycle life
  - Semi fast charge cycle life
  - Storage life at different form factor

Responses to Previous Year Reviewers' Comments

Comment: "significant amount of progress, especially for a new start, on all major technical categories. reviewer said that it would be good to see calendar life testing as part of the future work for this/these electrochemical couple(s)"

Response: Collected the calendar life data for the cell delivered to National Labs with Gen1 chemistry

Comment: "The single or double layer pouch cells are tested in flooded electrolytes. The reviewer asked if the author calculated how much electrolyte will be actually incorporated the in Gen 1 cell. If the single or double layer pouch cells are tested under such lean electrolyte conditions, the reviewer asked whether the cycling will last for the same few hundreds of cycling "

Response: Gen1 cell are tested in form factor starting from 500mAh till 42Ah and the amount of electrolyte needed per Ah is optimized and achieved more then 1000 cycle for 35 and 41.5Ah form factor cell which are build in the production facility.

Comment: "Electrode design parameters (i.e., areal capacity targets)"

Response: Capacity~ 300Wh/Kg, Cathode loading (4.39mAh/cm^2) and Anode loading (4.63mAh/cm^2)

Collaboration and Coordination with Other Institutions

- **Argonne National Laboratory (Chris Johnson):**

Federal Laboratory – Subcontractor helping to stabilize the materials and analytical work for project.

- Sacrificial Li-Source Development
    - Argonne National Laboratory and Farasis have develop the sacrificial Li- Source under a previous SBIR funded project. In collaboration with Argonne National Lab we will try to optimize the chemical composition of the materials for air stability.

- **Lawrence Berkeley National Laboratory (Robert Kostecki):**

Federal Laboratory – Subcontractor providing high voltage conductive additive for project.

- High Voltage Stable Conductive Additive
    - Robert Kostecki at Lawrence Berkeley National Laboratory has developed surface modified carbon blacks and nanofibers that can help to improved stability at high potentials (>4.4V)

Remaining Challenges and Barriers

- **Challenges with Si Anode**
  - Silicon volume expansion
  - Lithium consumption due to continual SEI growth
  - Irreversible capacity loss
  - Phase transition
  - Stability of Copper foil for higher amount of Si
  - Cycle life for higher amount the Si to achieve the targeted energy density of 330Wh/Kg of final delieverable

- **Challenges with Cathode**
  - Cathode > 200mAh/g
  - Stability of the cathode
  - Safety of Ni Rich cathode

- The sacrificial Li source will not be there on the final deliverable cell but we will continue the work to understand the impact of addition of the Li source into the cathode

Proposed Future Research

- Gen 1 Testing
  - Improving the storage life by optimization of electrolyte
- Optimization of Gen2 chemistry to hit the targeted capacity of 330Wh/Kg based on the new SOW
  - Cathode > 200mAh/g
  - Anode > 540mAh/g with coulombic efficiency of 88%
  - Optimization of electrolyte

Summary

- Gen 1 Cells deliver to National Lab (INL, SNL and IRNL) for independent testing
- Gen1 Cells are tested internally and achieved cycle life ≥ 1000
- Semi-fast cycling is done on the Gen1 Cells and shown that until 2C charging there is no Li plating on the cells
- Gen1 Cells shows good calendar life at room temperature but need further improvement at higher temperature and SOC.
- Understood the source of impedance increase in the cell with sacrificial Li source and needed more work to mitigate this so it will be not there in the final deliverable
- Final deliverable will be with high amount of Si and Ni rich cathode

Critical Assumption and Issue

- Final deliverable was design to specific energy of 400Wh/Kg (cathode capacity ≥200mAh/g and anode capacity ≥1500mAh/g).
- To achieve the target energy density 400Wh/Kg, we need pre-lithaition of Si anode. We selected Li-iron oxide source as a sacrificial Li source to compensate the irreversible loss of Si.
- The addition of the Li source to cathode increase the impedance of the cell and leads to lower cycle life and rate capability.
- Sacrificial Li source is air sensitivity, require processing to be done in the dry environment.
- Need more research and development to overcome all of the problems associated with the Li source and would be difficult to implement in the time frame required for the program builds.
- The final deliverables will be without the pre- lithiation and the specific energy will be 330Wh/Kg instead of 400Wh/Kg and work will be focus on the Si composite with higher columbic efficiency.