Overview

Timeline
- Project start date: Nov. 15, 2012
- Project end date: Dec. 22, 2015
- Percent complete: 51%

Barriers
- Barriers addressed
  - Specific Energy
  - Life
  - Cost

Budget
- Total project funding
  - DOE share $1,147,684
  - Contractor share $123,042
- Funding received in FY14
  - $499,990
- Funding for FY15
  - $499,909

Partners
- XG Sciences - Project lead
- A123 System
- Georgia Institute of Technology
- Collaborators
  - Argonne National Laboratory
  - Ashland Specialty Ingredients
  - Daikin America
  - Lawrence Berkeley National Laboratory
  - Sandia National Laboratory
Overall Objectives:

• Demonstrate XG SiG™ Si-graphene nano composite next generation Li-ion anode:
  – 600 mAh/g (Specific Energy Barrier),
  – 85% 1st cycle efficiency and
  – 1000 cycles with 75% capacity retention (Life Barrier)
• Stabilize and optimize the XG SiG™ anode pilot production (Cost Barrier)
• Develop a scalable dispersion and coating process with desired electrode properties
• Validate the XG SiG™ technology in commercial grade 2 Ah prototype Li-ion cells

Current Term Objectives:

• Demonstrate XG SiG™ anode performance:
  – 600mAh/g (Specific Energy Barrier),
  – 85% 1st cycle efficiency, and
  – 500 cycles with 70% retention (Life Barrier)
• Define XG SiG™ manufacturing product and process variable limits
• Demonstrate 2-3 L XG SiG™ slurry preparation and coating
• Demonstrate XG SiG™ performance in 2Ah cells
Impact on Barriers

EV Everywhere program defines a cell specific energy target of greater than 350 Wh/kg with 1000 cycles

Project Objective #1: Improve XG SiG™ anode performance

a. 600 mAh/g (Energy Barrier),
b. 85% 1st cycle efficiency, and
c. 1000 cycles with 75% capacity retention (Life Barrier)
Impact on Barriers

EV Everywhere targets cutting battery costs to $125/kWh

- This has been one of the biggest challenges for Si-based anodes due to poor scalability and prohibitive process cost.
- XGS’ XG SiG™ manufacturing process specifically addresses the cost issue in three ways:
  - Use of a low cost Si precursor,
  - Incorporating XG SiG™ formation into an existing manufacturing process,
  - Automation and modular design of the production system making the XG SiG™ process less labor intensive
- Cost models show that XG SiG™ can achieve a competitive price as compared with graphite which is required for the commercial acceptance for PHEVs and EVs.
## Milestones

<table>
<thead>
<tr>
<th>Tasks</th>
<th>2014</th>
<th>2015</th>
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<tbody>
<tr>
<td></td>
<td>Q1</td>
<td>Q2</td>
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<tr>
<td>1. Improve EC performance of XG SiG™</td>
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<td>2. Optimize pilot production</td>
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<td>3. Characterize materials/electrodes/cells</td>
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<td>4. Optimize dispersion</td>
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<td>5. Optimize electrolyte/additives</td>
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<td>6. Design/build 2Ah prototype cells</td>
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</table>

**Milestones**

MS 1: Demonstrate XG SiG™ silicon anode material in full cells:

- 600mAh/g, 85% 1st cycle efficiency, 500 cycles with 70% retention

MS 2: Demonstrate 600mAh/g, 85% 1st cycle efficiency, 1000 cycles with 70% retention

MS 3: Demonstrate XG SiG™ manufacturing process readiness

MS 4: Demonstrate electrode coating ready for prototype cell builds 2~3 L slurry preparation

MS 5: Select final electrolyte / additive

MS 6 & 7: Demonstrate XG SiG™ performance in 2Ah cells
# Status of Current Term Milestones

<table>
<thead>
<tr>
<th>Milestone ID</th>
<th>Description</th>
<th>Status</th>
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<tbody>
<tr>
<td>1</td>
<td>Demonstrate 600mAh/g, 85% FCE, 500 cycles with 70% retention</td>
<td>Demonstrated XG SiG™ material 600 mAh/g, 85% FCE and 70% capacity retention at 500 cycles targets in small format cells</td>
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<td>3</td>
<td>Demonstrate XG SiG™ manufacturing product and process variable limits</td>
<td>XG SiG™ manufacturing product and process variable limits were defined and implemented resulting in improved material quality</td>
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<tr>
<td>4</td>
<td>Demonstrate 2-3 L XG SiG™ slurry preparation and coating</td>
<td>Electrode coating of XG SiG™ material was successfully demonstrated by A123 using their commercial coater (&gt;6 L) and Argonne National Lab.</td>
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<td>6</td>
<td>Demonstrate XG SiG™ performance in 2Ah cells</td>
<td>2 Ah pouch and cylindrical cells were produced and performance demonstrated. Performance gaps were defined that are being addressed in year 2.</td>
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</table>
Milestone: 600mAh/g, 85% FCE, 70% capacity @ 500 cycles
Status: Demonstrated XG SiG™ material 600 mAh/g, 85% FCE and 70% capacity retention at 500 cycles targets in small format cells

• Employ material modification to target more stable SEI layer
  – Si precursor
  – Composite formulation
  – Composite manufacturing process

• Reduced Si fracture
• Reduced Li⁺ reaction
• Optimized graphene nanoplatelet support
Approach to Manufacturing Process Improvement

**Milestone:** Define XG SiG™ manufacturing product & process limits

**Status:** XG SiG™ manufacturing product and process variable limits have been defined and implemented resulting in improved quality

- Define Measurement Capability
- Define XG SiG™ Product Specifications
  - Relate key performance to manufacturing metrics
- Develop Manufacturing Process Control
  - Define material – process parameter sensitivity
- Define Manufacturing Process Capability
  - Optimize process targeting manufacturing metrics
Approach to Dispersion and Coating Improvement

**Milestone:** Demonstrate XG SiG™ slurry preparation and coating

**Status:** Electrode coating was successfully demonstrated by A123 using their commercial coater (>6 L) and at Argonne National Lab

- Identify:
  - Industrial mixer types,
  - Preferred solvents,
  - Material and slurry limitations (e.g. particle size, viscosity, etc.) used by XG Sciences customers
- Characterize slurry shear conditions in successful XG SiG™ laboratory dispersion and coating (magnetic stirrer)
- Scale-up slurry conditions to customer equipment preferences
- Transfer dispersion recommendations to customer
Technical Progress

XG SiG™ met MS 1 target in 63 mAh pouch cells

Improvement tied to: (a) modified Si precursor, (b) optimized formulation, and (c) manufacturing process modifications

- Phase II (Year 1) – 63 mAh pouch cell, A: 734 mAh/g, 93% active
- Phase II (Year 1) - 1.7mAh coin cell, A: 707 mAh/g, 93% active
- Phase I – 35 mAh pouch cell, A: 600 mAh/g, 93% active

MS 1: 70% retention @ 500 cycles

Anode: PAA binder
Cathode: NCA
Protocol: CCCV/ CC, C/2
Voltage: 3.2 – 4.2V
Technical Progress

New Gen 3 Silicon anode stable over wide operating voltage
Improvement tied to improved formulation

Capacity retention (%)
Cycle number

Gen 2 (C/2, 3 - 4.2V)
Gen 3 (C/2, 3 - 4.2V)
Gen 3 (1C, 2.7 - 4.2V)

Anode FCC : 600 mAh/g
94% : 6% PAA
Anode loading :
  2.2 mAh / cm²
Anode density : 1.5 g/cc
Cathode : NCA
Protocol: CCCV/ CC

Anode FCC : 600 mAh/g
94% : 6% PAA
Anode loading :
  2.2 mAh / cm²
Anode density : 1.5 g/cc
Cathode : NCA
Protocol: CCCV/ CC
Pouch cell demonstration with new Gen 3 Silicon anode

Anode FCC : 600 mAh/g
94% : 6% PAA
Anode loading :
  2.2 mAh / cm² – Coin
  3.0 mAh / cm² - Pouch
Anode density : 1.5 g/cc
Cathode : NCA
Protocol: CCCV/ CC

Cycle number
Capacity retention(%)
Technical Progress

Industrial-type mixer used to develop slurry protocol in support of electrode coating readiness demonstration (MS 4)

Double planetary mixer (commonly used by battery manufacturers)

11 Liter mixing vessel

Mixing vessel open 5 kg batch size (typ.)
Technical Progress

Slurry protocol successfully transferred to A123 Systems in support of electrode coating readiness demonstration (MS 4)

Recommended procedure for preparation of anode slurry using XG Sciences’ AN-SH silicon-graphene
Technical Progress

Slurry coat ability demonstrated on pilot-scale equipment in support of electrode coating readiness demonstration (MS 4)

XGS benchtop coating trial

Machine coating trial at ANL

50 µm Coating
75 µm Coating

Coating on Cu compressed to 1.8 g/cc

5 mAh/cm² and 7.2 mAh/cm²
No cracks were observed
Technical Progress

Met MS 4 with successful coating demonstrated on commercial machine

- Aqueous slurry made with polyacrylic acid binder
- Double-sided coating
- Coat weight 4.0 mg/cm² per side
- High quality coating confirmed
- XG SiG anode exhibited much better internal resistance than typical graphite anode
Technical Progress

Production material shows good consistency in 1st Cycle Efficiency and capacity retention in support of MS 3 - Manufacturing process readiness

- Anode FCC: 600 mAh/g
- 97%: 3% CMC/SBR
- Anode loading: 3.3 mg/cm²
- Anode density: 1.62 g/cc
- Protocol: CCCV/ CC, 1C
Reviewer Comments, Collaboration & Remaining Challenges

• Project was not reviewed last year

• Collaborations and Coordination
  • XG Sciences – Prime
  • A123 Systems – Subcontractor
  • Georgia Institute of Technology – Subcontractor
  • Argonne National Laboratory (A. Jansen)
  • Ashland Specialty Chemicals
  • Daikin America
  • Lawrence Berkeley National Laboratory (G. Liu)
  • Sandia National Laboratory (C. Orendorff)

• Remaining Challenges
  • Further improve capacity retention to meet 75% capacity at 1000 cycles
  • Maintain manufacturing process control with new material formulations
  • Maintain dispersibility and coatability new potential new formulations
## Proposed Future Work

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<th>Barrier</th>
<th>Proposed work</th>
<th>Objective (MS)</th>
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<td>• Silicon composite material modification including</td>
<td>Demonstrate: 600 mAh/g, 85% FCE, 70% Cap retention @1000 cycles</td>
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<td>- Si precursor physical characteristics</td>
<td>(MS2)</td>
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<td>- Additives/ coatings to reduce electrolyte reactions</td>
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<td>• Graphene conductive additive</td>
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<td>- Modified functionalization</td>
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<td>• Binder and electrolyte</td>
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<td>- Continue supplier collaboration with focus on improved cycling stability,</td>
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<td>gas generation, reduced inactive material</td>
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<td>Cost</td>
<td>• Translate future material developments to plant production</td>
<td>Demonstrate: Manufacturing readiness (MS3)</td>
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<td>Specific Energy, Life</td>
<td>• Transfer future non-active materials (composite, conductive additive,</td>
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<td>binder, etc.) to A123 mixing and coating equipment</td>
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<tr>
<td>Specific Energy, Life</td>
<td>• Design, fabricate, assemble and demonstrate cells</td>
<td>Demonstrate 2Ah cells (MS 7)</td>
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Summary

• XG Sciences is on target
  – Goals met for Si anode performance based on full cell cycling in small format cells.
• All material developments
  – Transferred to high capacity plant production, material at numerous cell customers.
• Slurry and coating developments
  – Transferred to A123 Systems and numerous other cell manufacturers.

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