Significant Cost Improvement of Li-ion Cells Through Non-NMP Electrode Coating, Direct Separator Coating, and Fast Formation Technologies

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Johnson Controls

Project ID #: ES133

2014 DOE Vehicle Technologies Program Review
June 17, 2014
Overview

Timeline
- Start: October 2011
- Finish: January 2015
- Final report to DOE: January 2015
- Percent complete 70% completed

Barriers
- Concentration polarization of dry electrode affects high current rate capability
- Dry cathode design related in process capability
- High self-discharge rate

Budget
- Total project funding
  - DOE: $3.67M
  - Johnson Controls and sub-recipients: $3.67M
- Funding received in 2013: $752k
- Funding for 2014: $2.02M

Partners
- Entek Membranes
- Maxwell Technologies
- University of Wisconsin – Milwaukee

(1) Tasks complete as of Mar 2014
Objectives - Relevance

Project scope
- Significant cost improvement of Li-ion manufacturing:
  - Non-NMP electrode coating process
  - Direct coating separator
  - Fast formation
  - Integrated cell design

Objectives
- Develop integrated cell with non-NMP electrodes with direct coated separator
- Develop dry electrode formulation and design: Complete investigation of effects of electrode design; loading weight and density and optimize electrode formulation
- Develop an additive and new formulation for process improvement for non-NMP electrode

Addresses Targets
- 50% manufacturing cost reduction
- Better than 90% performance of integrated cell compared to baseline’s performance

Impacts
- The performance of integrated cells have been improved so that it has achieved 37% lower impedance than baseline
- Detailed characterization of the microstructure of the electrodes helps to understand the performance of dry electrodes and the correlation of concentration polarization and morphology
### Milestones

#### Key milestones and decision points

<table>
<thead>
<tr>
<th>Year</th>
<th>Q1</th>
<th>Q2</th>
<th>Q3</th>
<th>Q4</th>
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<tbody>
<tr>
<td>2011</td>
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<tr>
<td>2014</td>
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</tbody>
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**Planning**

**Electrode Non-NMP Coating (Maxwell)**
- 2012: 80% perf of dry coating electrode to baseline
- 2013: 90% perf of dry coating electrode to baseline
- 2014: 95% perf of dry coating electrode to baseline

**Separator (Entek)**
- 2012: 80% perf of coating separator to baseline
- 2013: 90% perf of coating separator to baseline
- 2014: 95% perf of coating separator to baseline

**Water based cathode (JCI & UWM)**
- 2013: 100% perf compared to PVDF binder electrode

**Formation (JCI & UWM)**
- Wetting: 10% improvement
- Aging time: 50% reduction

**Cell Development (JCI)**
- Baseline cell
- 2013: 80% compared to baseline
- 2014: 90% compared to baseline
- 50% cost reduction

**Major Deliverables (JCI)**
- 2011: 18 x15 Ah baseline cells/ baseline cost model
- 2012: 18 x3 Ah advanced cells
- 2013: 24x15 Ah advanced cells/ final cost model
Approach

**Dry coated electrode**
- Optimize formulation and design to achieve electrode target specification and performance
- Improve the micro-structure of electrode for high rate capability performance
- Develop the process of automated pilot line for large cell build

**Water based cathode binder**
- Develop new cathode binder which is both electrochemically and chemically stable
- Develop an additive and new formulation for process improvement
- Investigate corrosion prevention methods and risk mitigation to allow for aqueous solvent manufacturing
- Perform trial run on production line

**Direct-coating of separator material on Li-ion electrodes**
- Formulation changes to improve porosity for Si/PVDF direct coated separator
- Laminate a free-standing separator on anode
- Develop roll-to-roll process for scale-up to large format cell build improving lamination strength and thickness variation

**Fast Formation**
- Develop new activation and detection process to improve cell uniformity, accelerate detection time, and minimize cell degradation
Technical Accomplishments
Dry Coating Electrodes

Dry Coating Electrodes

- The results for the optimized formulation and design of dry electrodes demonstrate improved performance
- 30% lower ASI compared to initial dry electrode, and 16% better ASI than the baseline
- The results of high current continuous discharge tests have shown lesser performance against the baseline due to concentration polarization
- The root cause of the concentration polarization is the smaller pore size and lower porosity in the electrode micro-structure, particularly in the cathode
Technical Accomplishments
Water Based Cathode

Water Based Cathode

- Cycle and calendar life performance of water based cathode are slightly better than the baseline due to superior adhesion strength.

- The rate capability performance has been improved through many trials with a pH balancer and conductive graphite to improve slurry dispersion and electrode quality.

- We have investigated corrosion prevention methods and risk mitigation to allow for aqueous cathode binder processing.

- We have initiated the transfer of the water based cathode into our production environment.

![Cycle Life Graph]

1C/1C 100% DOD

![Rate Capability Graph]

Baseline
- A binder with additive
- B binder with additive
- B binder without additive

Experimental table for better slurry property:

<table>
<thead>
<tr>
<th>Test</th>
<th>Binder</th>
<th>pH balancer</th>
<th>Conductive agents</th>
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<tbody>
<tr>
<td>1</td>
<td>A</td>
<td>None</td>
<td>CB</td>
</tr>
<tr>
<td>2</td>
<td>A</td>
<td>0.1% additive</td>
<td>CB</td>
</tr>
<tr>
<td>3</td>
<td>A</td>
<td>0.2% additive</td>
<td>CB</td>
</tr>
<tr>
<td>4</td>
<td>A</td>
<td>0.3% additive</td>
<td>CB</td>
</tr>
<tr>
<td>5</td>
<td>B</td>
<td>None</td>
<td>CB + CG</td>
</tr>
<tr>
<td>6</td>
<td>B</td>
<td>0.1% additive</td>
<td>CB + CG</td>
</tr>
<tr>
<td>7</td>
<td>B</td>
<td>0.2% additive</td>
<td>CB + CG</td>
</tr>
<tr>
<td>8</td>
<td>B</td>
<td>0.3% additive</td>
<td>CB + CG</td>
</tr>
</tbody>
</table>
Technical Accomplishments
Direct Coated Separator

Solvent Coating
- The solvent based separator meets all desired product parameters using a diverse co-solvent to polymer ratio and showed good performance similar to the baseline.
- This method was not selected due to difficulty in producing uniform thickness and porosity.

Lamination
- Lamination is the most promising of the three methods. The cells built with laminated separator show superior results in power performance.
- Investigating roll-to-roll lamination process to further improve thickness variation control.

Porosity control for solvent coating

HPPC

5C Discharge
Technical Accomplishments
Fast Formation

**Activation Process**
- We have conducted step-charge and step-aging to activate cells faster and more uniformly.
- The results of the new activation process show lesser variations for 1st cycle capacity, impedance, and dV due to improved cell uniformity.

**Detection Process**
- Formed at a low state of charge and various temperatures to maximize detectability and minimize cell degradation.
- We have conducted a design of experiment with three factors and three levels to optimize the best conditions for the detection process.
Technical Accomplishments

Cell Development

Integrated Cell Development

- We have optimized the cell and electrode design to integrate the new technologies and build them on our pilot line.

- 3Ah integrated cells with the dry electrode and laminated separator show 37% lower DC resistance than the baseline design.

- Rate capability with continuous high current shows 18% lower capacity retention than the baseline due to higher diffusion polarization.

### Integrated cell design for interim 3Ah cell

<table>
<thead>
<tr>
<th>Item</th>
<th>Unit</th>
<th>Baseline Cathode</th>
<th>Baseline Anode</th>
<th>Dry Electrode Cathode</th>
<th>Dry Electrode Anode</th>
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</thead>
<tbody>
<tr>
<td>Loading weight</td>
<td>mg/cm²</td>
<td>15.5</td>
<td>8.4</td>
<td>18.6</td>
<td>10.7</td>
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<tr>
<td>Density</td>
<td>g/cc</td>
<td>2.8</td>
<td>1.33</td>
<td>2.85</td>
<td>1.35</td>
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<tr>
<td>Stacks Number</td>
<td></td>
<td>8</td>
<td>9</td>
<td>8</td>
<td>9</td>
</tr>
<tr>
<td>Capacity</td>
<td>Ah</td>
<td>3.1</td>
<td></td>
<td>3.3</td>
<td></td>
</tr>
</tbody>
</table>

**HPPC**

![HPPC Graph]

**Rate capability**

![Rate capability Graph]
Technical Accomplishments
Process Development

- We have optimized the electrode notching process to accommodate the anode with the laminated separator
- By laminating the separator onto the anode, we were able to eliminate the zig-zag separator process and speed cell build time from 1 cell per minute to 3
- Eliminating process steps will allow future machines to be faster, smaller, simpler, and less expensive
Cost saving comparison for 15Ah

- Baseline cost as 100%
- New technology cost compared against baseline cost

Cost saving comparison for 15Ah

- Coat and mix, formation, and cell assembly have the highest cost improvement
- Slitting and calendering process costs are higher but not enough to have negative impact on overall process cost savings
- Dry coating has a cost advantage in the Coating and Mixing process step
Responses to Previous Year Reviewer’s Comments

Approach to performing the work

“The approach to lowering cost by greater than 50% is to make significant improvements in the Li ion manufacturing process by developing a non-NMP electrode coating process and a direct coated separator. This is a good approach and if successful has a very high chance of lowering battery costs. The reviewer criticized that no specifics or a pathway on how to accomplish this was provided.”

Response: This project has a higher goal to reduce barriers to vehicle electrification specifically by reducing Li-ion cell manufacturing costs. To achieve this goal, we have partnered to develop and integrate 3 technologies into a single cell that meets 80% of the performance of the baseline cells.

Technical accomplishments

“The reviewer noted that progress seemed slow since the project inception in 2011. The reviewer commented that they did not have sufficient data to evaluate the progress on shortened formation”

Response: The first year of the project was focused on creating the baseline and down-selecting technologies. This work was completed according to plan.

More technical details regarding the advancement in fast formation and improved detection are provided in this year’s poster presentation.
## Collaboration

<table>
<thead>
<tr>
<th>Entek Maxwell</th>
<th>Maxwell Technologies</th>
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<tbody>
<tr>
<td>Award sub-recipient</td>
<td>Award sub-recipient</td>
</tr>
<tr>
<td>Leader in micro-porous membranes</td>
<td>Leader in ultra-capacitor technology</td>
</tr>
<tr>
<td>Focus on direct coated separator</td>
<td>Focus on dry coating electrode research</td>
</tr>
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<tr>
<th>Johnson Controls</th>
<th>University of Wisconsin - Milwaukee</th>
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<tbody>
<tr>
<td>Award prime recipient</td>
<td>Partner in innovation</td>
</tr>
<tr>
<td>Leader in Lead acid and Li-ion batteries</td>
<td>Leading institute in material science and energy storage</td>
</tr>
<tr>
<td>Focus on cell design integrating new advanced technologies from our partners, water based cathode, and fast formation.</td>
<td>Focus on Al corrosion and wetting phenomenon of Li-ion cell, modeling, and cell characterization</td>
</tr>
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</table>
Future Works

Remainder of 2014
- Verify fast formation process against the formation process in our plant
- Complete process development for roll-to-roll dry electrodes, lamination and cell assembly
- Complete final optimization for dry electrode using pilot scale equipment
- Integrate technologies, scale up the process, and optimize to improve cell performance
- Finalize 15 Ah cell and electrode design
- Build and evaluate final deliverable 15Ah cells that integrate the technology advancements

FY15
- Deliverables to DOE:
  - Twenty-four 15Ah advanced cells
  - Final cost model
  - New formation process results
- Transfer advanced technologies into production

Remaining Challenges and Barriers
- Scale-up advanced technologies (3Ah → 15Ah, sheet → roll-to-roll)
- Concentration polarization of dry electrode affects continuous high current rate performance
- High self-discharge rate and dV variation
- Transfer new technologies to product practically
Summary

Relevance

- Develop integrated cell with dry electrode, direct coated separator, and fast formation to accomplish 50% cost savings while maintaining 90% performance compared to baseline design

Approach

- **Dry electrode**
  Improve the micro-structure and morphology of the electrode and develop the process of automated pilot line for large format cell builds

- **Water based cathode**
  Develop an additive and new formulation for mixing and coating process improvements

- **Direct coated separator**
  Develop roll-to-roll process for scale-up to improve lamination strength and reduce thickness variation

- **Fast formation**
  Develop new activation process to improve cell uniformity using step-charging and step-aging process and develop an improved detection process at low SOCs

Technical accomplishments

- **Dry electrode**
  The cells built with optimized dry electrode demonstrate 30% lower ASI and 10% higher rate capability than initial electrode design

- **Water based cathode**
  The results show 90% capacity at 2,500 cycles and similar performance compared to the baseline. The rate capability performance has been improved via an optimized pH balancer and conductive graphite.

- **Direct coated separator**
  The cells show 9% lower ASI and 27% better rate capability compared to the baseline

- **Fast formation**
  The results of the new activation process show lesser variation for 1st cycle capacity, impedance, dV and capacity retention after calendar life. The detection process at low SOC demonstrates improved detectability and lower cell degradation.

Proposed future research

- Deliver 24 15Ah final cells and final cost model to DOE
- Transfer technologies to production