Co-Extrusion (CoEx) for Cost Reduction of Advanced High-Energy-and-Power Battery Electrode Manufacturing

Corie L. Cobb, Ph.D. (Principal Investigator)
PARC, a Xerox Company
2016 Annual Merit Review
June 9, 2016

This presentation does not contain any proprietary, confidential, or otherwise restricted information
# Overview

## Timeline

- **Project start date:** December 17, 2015
- **Project end date:** December 16, 2018
- **Percent complete:** 5%

## Budget

- **Total project funding:**
  - DOE share: $2,999,115
  - PARC share: $787,478
- **FY 2015 Funding (DOE):** $0
- **FY 2016 Funding (DOE):** $1,476,420

## Barriers Addressed

- **Cost:** Current cost of Li-ion batteries is ~$250–$500/kWh, a factor of about two to three times too high on a $/kWh basis.
- **Performance:** High energy density battery systems to meet both volume and weight targets.

## Partners

**Project Lead**

- parc
  - A Xerox Company

**Project Partners**

- Oak Ridge National Laboratory
- Navitas Systems
- Ford
- Argonne National Laboratory

**Collaborations**
Relevance and Project Objectives

• Overall Project Objectives:
  • Demonstrate pilot scale, electric vehicle (EV)–relevant ≥14 Ampere hours (Ah) Co-extrusion (CoEx) pouch cells with:
    ➢ Cost Barrier: ≥30% reduction in $/kWh costs thru thick, structured high energy and power electrodes
    ➢ Performance Barrier: Gravimetric energy density improvement of ≥ 20% relative to conventional electrodes of the same chemistry

• FY2015/2016 Objectives:
  • Fabricate a demonstrator CoEx coin cell with ≥ 20% gravimetric energy improvement over a conventional baseline cell
  • Optimize the thick CoEx cathode design and matching graphite anode for EV applications with guidance from Ford
  • Conduct a technology evaluation & predictive scaling analysis on CoEx
## Milestones: FY 2015/2016

<table>
<thead>
<tr>
<th>Milestone</th>
<th>Type</th>
<th>Description</th>
<th>Due Date</th>
<th>Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specifications developed</td>
<td>Technical</td>
<td>Recommended cell targets for a Nickel Manganese Cobalt (NMC)-graphite materials set are identified.</td>
<td>2/3/2016</td>
<td>In Progress.</td>
</tr>
<tr>
<td>Geometries Identified</td>
<td>Technical</td>
<td>Modeling results in a subset of optimal geometries for the CoEx cathode, which show a 10-30% improvement over the selected baseline case.</td>
<td>4/13/2016</td>
<td>In Progress.</td>
</tr>
<tr>
<td>Cathode Films Demonstrated</td>
<td>Technical</td>
<td>Single-layer CoEx cathode films demonstrate a minimum crack-free thickness and half-cell measurements demonstrate &gt;142 mAh/g at C/2 discharge rate, tested at 4.2V.</td>
<td>9/16/2016</td>
<td>In Progress. Materials selected, ink formulations under investigation.</td>
</tr>
<tr>
<td>Baseline Validated</td>
<td>Technical</td>
<td>Baseline anode meets specifications.</td>
<td>7/28/2016</td>
<td>In Progress.</td>
</tr>
<tr>
<td>Capability Demonstrated</td>
<td>Go/No Go</td>
<td>A homogenous ≥120µm anode film demonstrates the capacity required to balance the CoEx cathode.</td>
<td>12/16/2016</td>
<td>Not started. Dependent on milestones above.</td>
</tr>
</tbody>
</table>
# Approach and Strategy: Timeline

<table>
<thead>
<tr>
<th>TASK</th>
<th>2015-2016</th>
<th>2016</th>
<th>2017</th>
<th>2018</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>12 1 2 3 4 5 6 7 8 9 10 11</td>
<td>1 2 3 4 5 6 7 8 9 10 11</td>
<td>1 2 3 4 5 6 7 8 9 10 11</td>
<td>1 2 3 4 5 6 7 8 9 10 11</td>
</tr>
<tr>
<td><strong>Project Management &amp; Planning</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Material &amp; Battery Design</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>CoEx Cathode Design, Fabrication, &amp; Testing</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Anode Design, Fabrication, &amp; Testing</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>CoEx Printhead Design &amp; Development</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>CoEx Process Development &amp; BMF Integration</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Pouch Cell Production &amp; Testing</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Pouch Cell Characterization &amp; Validation</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

- **PH** = Printhead
- **BMF** = Battery Manufacturing Facility

- Specifications Developed
- CoEx Geometries Modeled
- CoEx Films Tested
- Baseline Anode
- CoEx Matching Anode Tested
- CoEx 14Ah PH Designed
- 1st PH Fabricated
- 2nd PH Fabricated Qualified at ORNL
- CoEx Speed Demonstration
- CoEx Cathode Demonstration
- 14Ah Pouch Cells Fabrication
- 1-6 Ah Pouch Cells Meet Targets
- 14 Ah Cell Testing
- 14 Ah Cell Automotive Characterization
Approach and Strategy: Co-extrusion (CoEx)

Co-extrusion Printhead**

Using conventional battery materials, **thick CoEx cathodes** can change conduction pathways in lithium-ion batteries, decoupling power and energy trade-offs for a **30% reduction in $/kWh costs** and a **≥20% improvement in energy density**

**Funded in part by the Advanced Research Projects Agency-Energy (ARPA-E), U.S. Department of Energy, under Award Number DE-AR0000324**

High Conductivity Region

High Density Lithium Storage Region

Top View
Dried CoEx Cathode Sample

Current Collector

Li-ion flow
**Funded in part by the Advanced Research Projects Agency-Energy (ARPA-E), U.S. Department of Energy, under Award Number DE-AR0000324**

**Approach and Strategy: Co-extrusion (CoEx)**

- This project will leverage ARPA-E investment and optimize the CoEx cathode for EV applications
- The separator will not be printed for this project

**Co-extrusion Printhead**

**Past Project**

Printable Separator

Current Collector

Past ARPA-E Project: Separator & CoEx Cathode

Uncalendered sample cross-section

100µm

separator
cathode
current collector
Approach and Strategy: Co-extrusion (CoEx)

CoEx has been applied to solar cell metallization and integrated into high speed, high volume production.

Approach and Strategy: High Capacity Anode

- Develop and refine graphite-based anode slurry for coating adhesion, agglomerate cohesion, and high ionic and electronic conductivity by modifying binder and conductive additive.

  - **Method**: Anode slurries will be prepared with a NMP/PVDF solvent/binder system and slot-die coated to a sufficient thickness to balance CoEx cathodes. Anode formulations will be adjusted as needed to maintain sufficient anode coating integrity after calendering.

  - **Baseline Anode**: Electrochemical testing of baseline anodes developed at ORNL to quantify electrochemical performance. (Targets: 50-80 μm thick (2.5-3.0 mAh/cm²) after calendering and deliver >350 mAh/g)

  - **Thick Anode for CoEx**: Demonstrate a 125-200 μm uncracked anode (5-6 mAh/cm²) with a NMP/PVDF solvent/binder system to match CoEx cathode capacity; Show capability to maintain thick anode coating integrity after calendering to 30-40% porosity.
Approach and Strategy: High Capacity Anode

• Slot-die coating methodology:
  • Single-pass thick anodes with increased binder content
  • Double-pass thick anodes
  • Dual slot-die coated anodes

• Preferred materials for good thick coating integrity:
  • Showa Denko America SCMG-BH
  • Canada Carbon
  • Ontario Graphite

• Evaluated materials with non-optimized thick coating integrity:
  • ConocoPhillips A12 natural graphite and G8 synthetic graphite
  • Superior Graphite SLC 1520P, 1512P, and 1506T
  • GrafTech
**Approach and Strategy: High Capacity Anode**

ABR VTO Program historic baseline data from ORNL BMF. Data will be used as a guide for Task 3 and 6 as we progress to higher loading and thicker electrodes.

<table>
<thead>
<tr>
<th>Electrode</th>
<th>Composition</th>
<th>Areal Loading (mAh/cm²)</th>
<th>Porosity (%)</th>
<th>Electrode Thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cathode</td>
<td>NMC532/PVDF/CB = 90/5/5 wt%</td>
<td>1.9</td>
<td>36.8</td>
<td>52</td>
</tr>
<tr>
<td>Anode</td>
<td>Graphite/PVDF/CB = 92/6/2 wt%</td>
<td>2.4</td>
<td>32.5</td>
<td>55</td>
</tr>
</tbody>
</table>
Approach and Strategy: ORNL BMF

• End of Project Goals:
  • Integration of pouch cell scale CoEx printhead equipment at ORNL Battery Manufacturing Facility (BMF)
  • Production and characterization of 14 Ah pouch cells
  • Develop a plan for commercialization of the CoEx technology with potential end-users and suppliers

Slot-Die Coating Line at ORNL

PARC CoEx printhead

Slot-Die will be replaced with CoEx printhead & high pressure slurry dispensers
Technical Accomplishments and Progress

CoEx Cathode Print Feasibility Test

CoEx 1:
CoEx 2:
Print feasibility demonstrated previously on ARPA-E funding

- Developed a set of NMC 532 ink formulations to test the print feasibility of 2 different CoEx cathode structures
- With guidance from Ford and ORNL, electrochemical modeling will focus on optimizing the final geometry
Responses to Reviewer Comments

• This project is a new start.
Collaboration and Coordination

**Oak Ridge National Lab (Project Partner)**
Developing the matching high capacity anode, providing materials guidance, 1-6 Ah pouch cell assembly, and BMF integration assistance for CoEx hardware

**Ford Motor Company (Project Partner)**
Providing automotive guidance and recommendations on baseline electrode design, testing and cycling protocols, and market evaluation of CoEx technology

**Navitas Systems (Collaboration)**
Providing use of pouch cell assembly equipment for 14 Ah pouch cells in FY 2018

**Argonne National Labs (Collaboration)**
Providing guidance on best practices for coin cell assembly and half cell testing protocols
Remaining Challenges and Barriers

• CoEx Cathode (NMC 532)
  • Developing inks which enable thick (> 125 µm) dried cathode electrodes
  • Developing 2 inks with enough porosity/conductivity difference to enhance lithium ion pathways in thick electrodes
  • Scaling PARC’s existing CoEx printhead to fabricate the optimal structures determined from modeling to enable 1-14 Ah pouch cell production

• Anode (Graphite)
  • Developing the matching high capacity anode for the CoEx cathode and maintaining good electrode integrity
  • Designing an anode architecture with desirable power performance
Proposed Future Work: CoEx Modeling

These 3 examples have the same capacity (mAh/cm²) and we will use this as a guide for electrochemical modeling.

### Monolithic Comparison

**h** = 120 µm

<table>
<thead>
<tr>
<th>Description</th>
<th>Stripe Width Ratio</th>
<th>Stripe 1 Porosity (%)</th>
<th>Stripe 2 Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Monolithic case, assuming an electrode density of 2.8 g/cm³</td>
<td>N/A</td>
<td>34</td>
<td>N/A</td>
</tr>
</tbody>
</table>

### CoEx 1: Corrugated Design

**h** = 144 - 180 µm

<table>
<thead>
<tr>
<th>Description</th>
<th>Stripe Width Ratio</th>
<th>Stripe 1 Porosity (%)</th>
<th>Stripe 2 Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Corrugated design with open channels for electrolyte</td>
<td>2:1 to 5:1</td>
<td>34</td>
<td>100</td>
</tr>
</tbody>
</table>

### CoEx 2: Two-material Design

**h** = 130 - 140 µm

<table>
<thead>
<tr>
<th>Description</th>
<th>Stripe Width Ratio</th>
<th>Stripe 1 Porosity (%)</th>
<th>Stripe 2 Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Two-material design with a 20% porosity differential between the stripes</td>
<td>1:1 to 2:1</td>
<td>30</td>
<td>50</td>
</tr>
</tbody>
</table>
Proposed Future Work

• Ongoing for FY 2016:
  • With guidance from Ford and ORNL, PARC is modeling a series of CoEx geometries to determine an optimal subset of designs for initial printhead design work and coin cell fabrication
  • Increase crack resistance of dried, uncalendered thick CoEx electrodes and demonstrate >142 mAh/g at a C/2 discharge rate
  • Design, formulate and fabricate the necessary matching graphite anode for the CoEx cathode

• Proposed for FY 2016:
  • Conduct an EV pack scaling analysis with CoEx experimental and modeling data to estimate potential energy and power benefits
  • Modify the existing CoEx printhead and print CoEx cathodes for 1Ah pouch cell fabrication in Budget Year 2
Summary

• **Relevance**
  • Demonstrate pilot scale, electric vehicle (EV)–relevant Co-extrusion (CoEx) ≥14 Ampere hours (Ah) pouch cells with a 30% reduction in cost and a gravimetric energy density improvement of ≥ 20%

• **Approach**
  • Develop thick structured cathodes with CoEx to mitigate power and energy trade-offs in cathode electrodes
  • Modify, binder and conductive additive; dual slot die and/or double pass coating for a thick matching capacity anode

• **Technical Accomplishments**
  • Initial print feasibility for CoEx cathode structures has been demonstrated

• **Future Work**
  • Optimize CoEx cathode and anode for overall capacity improvement
  • Develop the necessary process for fabricating crack-free thick electrodes