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

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PARC, a Xerox Company

2016 Annual Merit Review

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Project ID # ES266

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Overview

Timeline

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

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.

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

Partners

Project Lead

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Project Partners

Oak Ridge National Laboratory

Ford

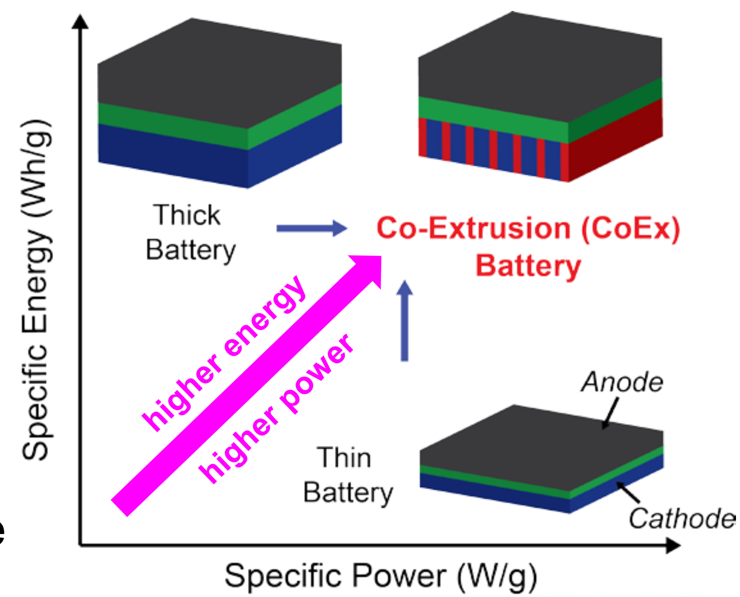
Collaborations

Navitas Systems

Argonne National Laboratory

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



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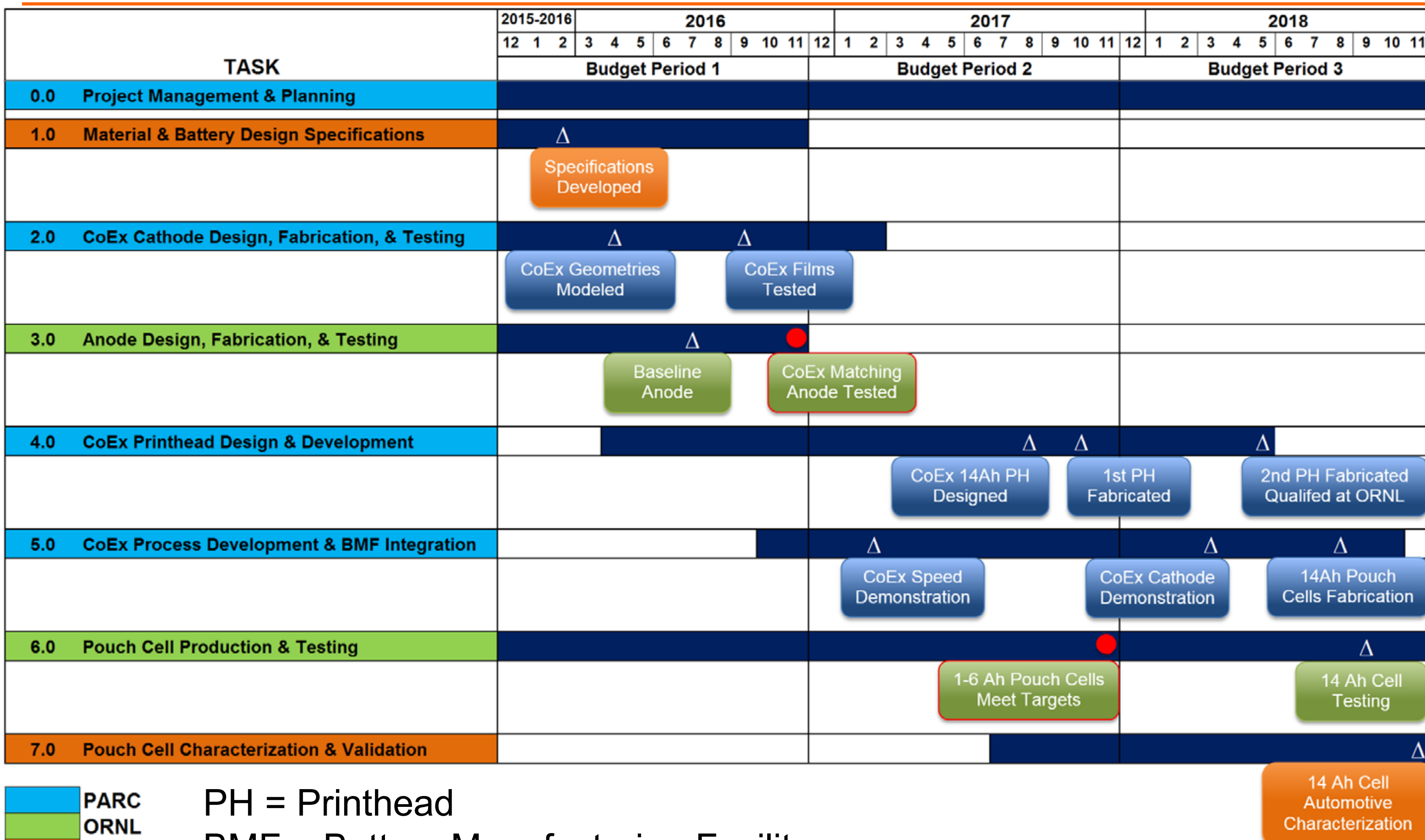
Milestones: FY 2015/2016

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

Approach and Strategy: Timeline

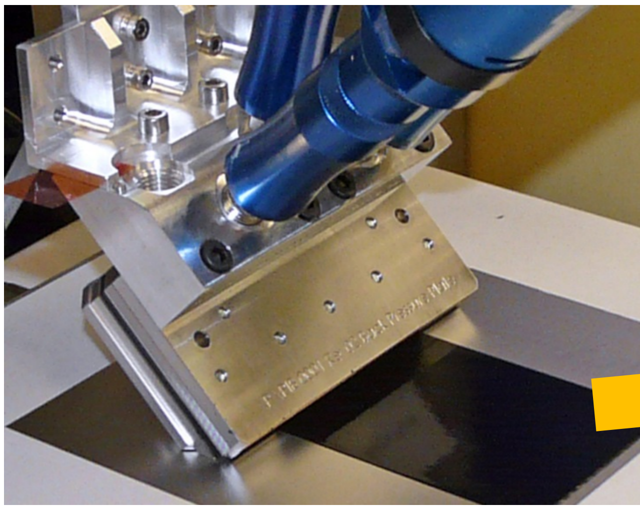


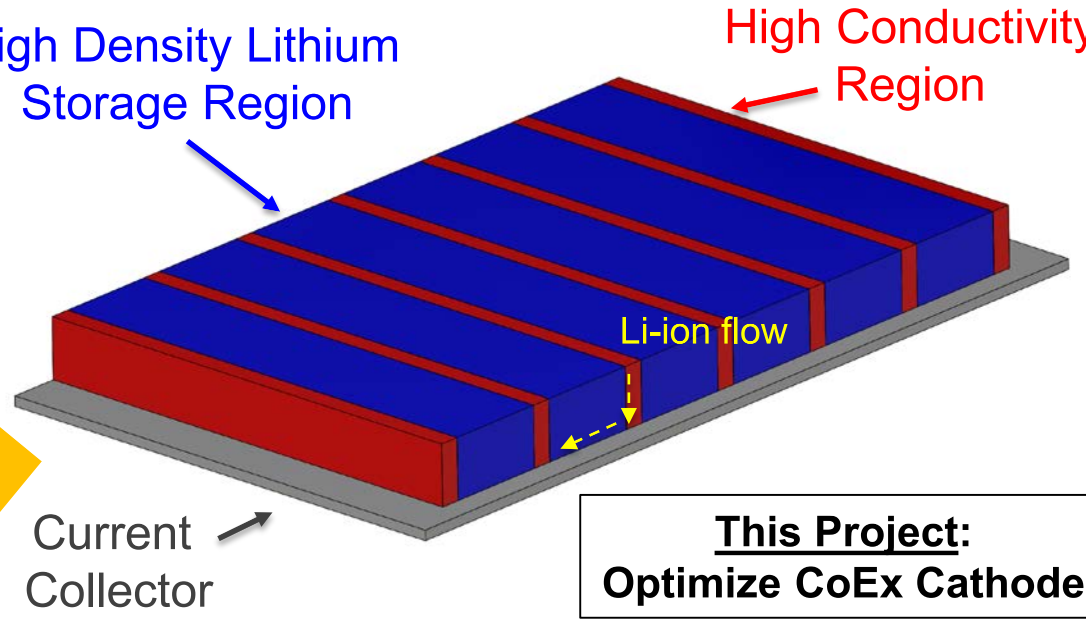
PARC
ORNL
Ford

PH = Printhead
BMF = Battery Manufacturing Facility

Approach and Strategy: Co-extrusion (CoEx)

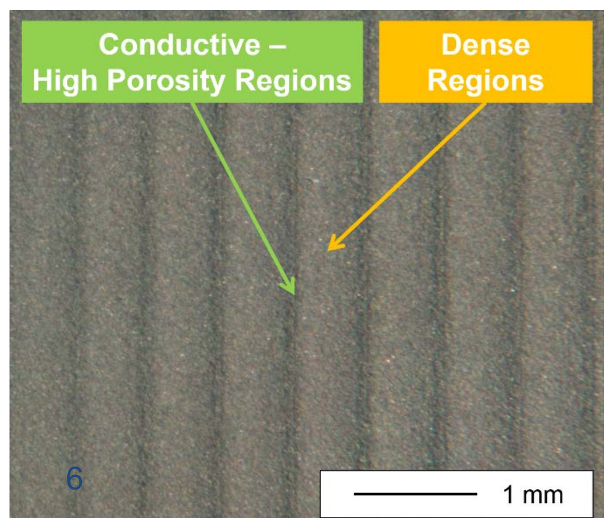
Co-extrusion Printhead**





This Project:
Optimize CoEx Cathode

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**



Top View
Dried CoEx Cathode Sample

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

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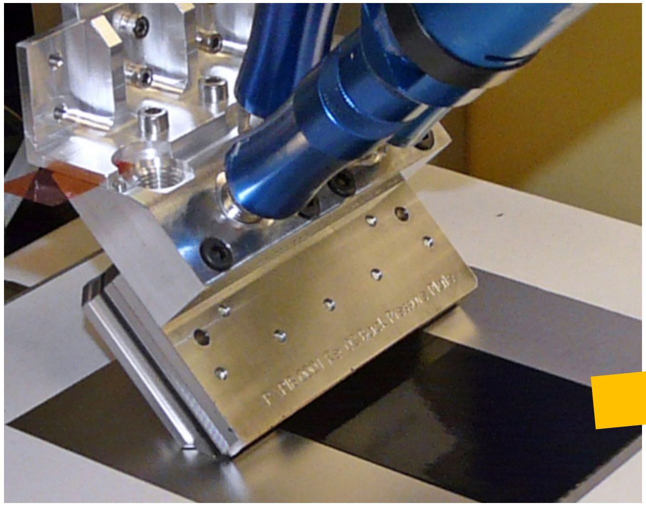
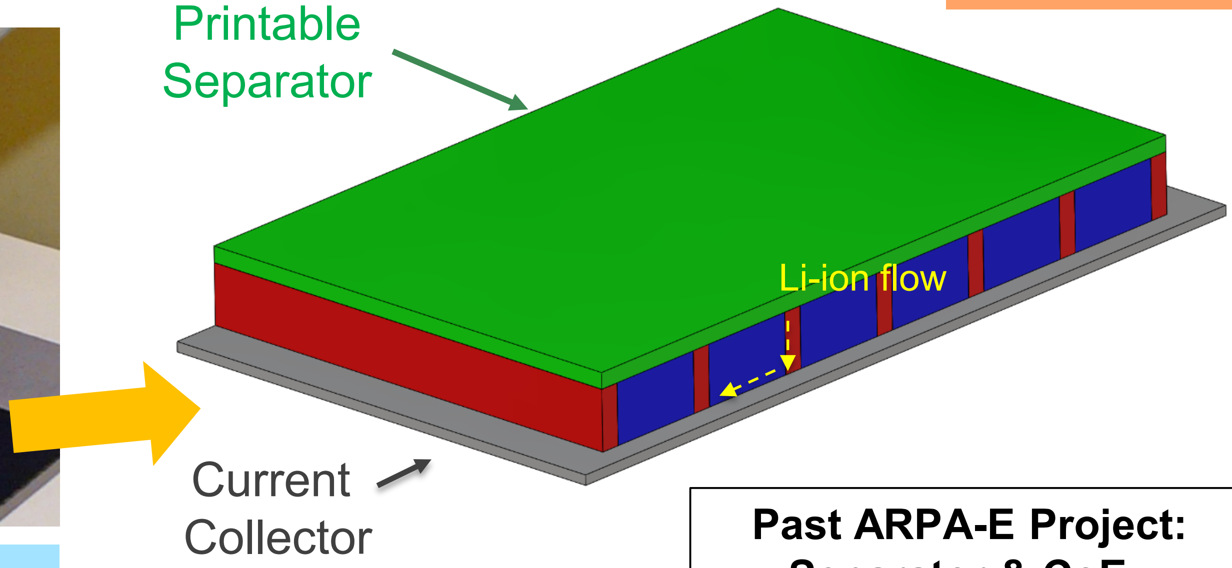
Approach and Strategy: Co-extrusion (CoEx)

Past Project

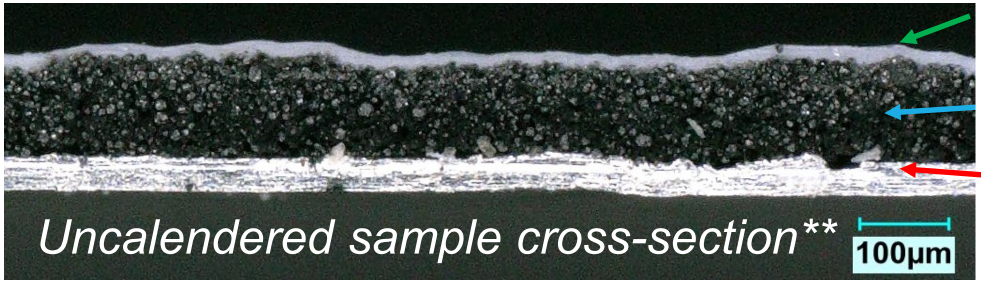
Co-extrusion Printhead**





Past ARPA-E Project: Separator & CoEx Cathode

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



*Uncalendered sample cross-section***

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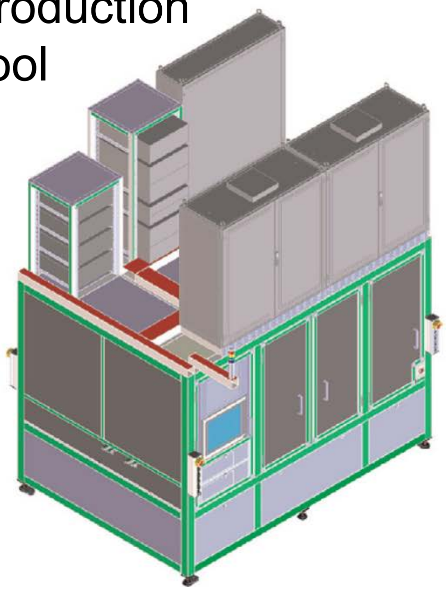
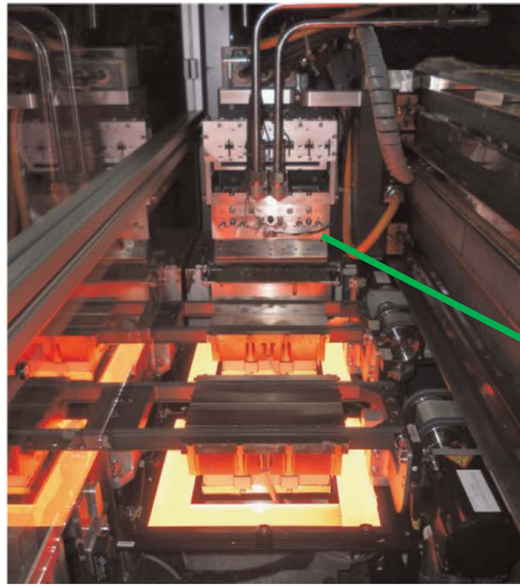
Approach and Strategy: Co-extrusion (CoEx)

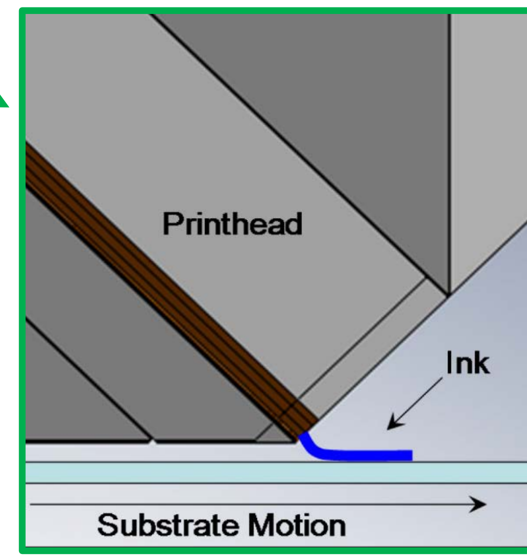
Past Project

Production Tool



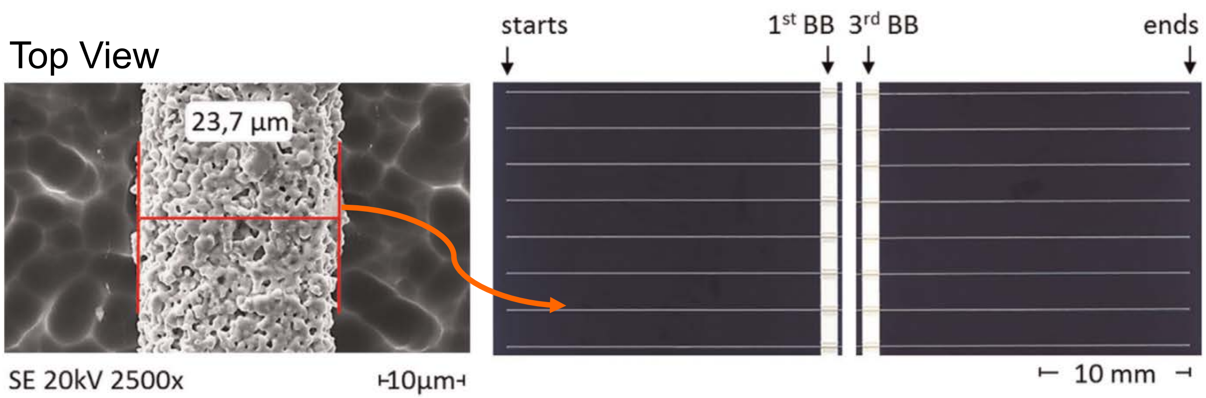


CoEx has been applied to solar cell metallization and integrated into high speed, high volume production



Side View of CoEx process

Top View



L.P. Richter, G. Fischer, L. Sylla, M. Hentsche, S. Steckemetz, M. Müller, **C.L. Cobb**, S.E. Solberg, R. Rao, S. Elrod, P. Palinginis, E. Schneiderlöchner, H. Neuhaus, "Progress in Fine Line Metallization by Co-extrusion Printing on Cast Mono Silicon PERC Solar Cells," *Solar Energy Materials and Solar Cells,* Vol. 142, pp. 18-23, 2015. doi:10.1016/j.solmat.2015.05.023

Approach and Strategy: High Capacity Anode

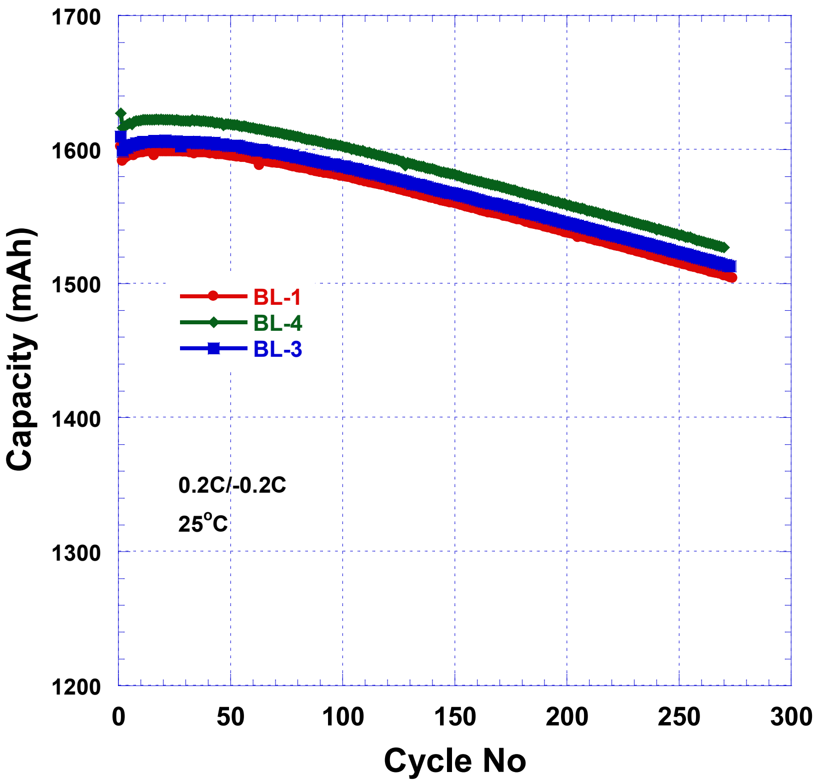
- Develop and refine graphite-based anode slurry for coating adhesion, agglomerate cohesion, and high ionic and electronic conductivity by modifying binder and conductive additive.
  - **Method**: Anode slurries will be prepared with a NMP/PVDF solvent/binder system and slot-die coated to a sufficient 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/cm^2) after calendering and deliver >350 mAh/g)
  - **Thick Anode for CoEx**: Demonstrate a 125-200 μm uncracked anode (5-6 mAh/cm^2) 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.

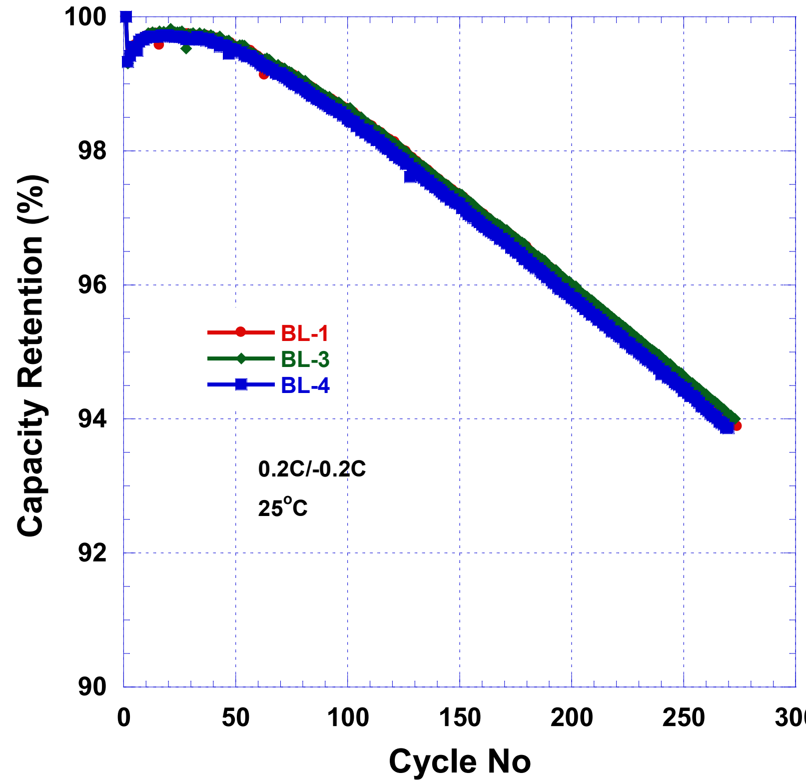
Approach and Strategy: High Capacity Anode

- 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.

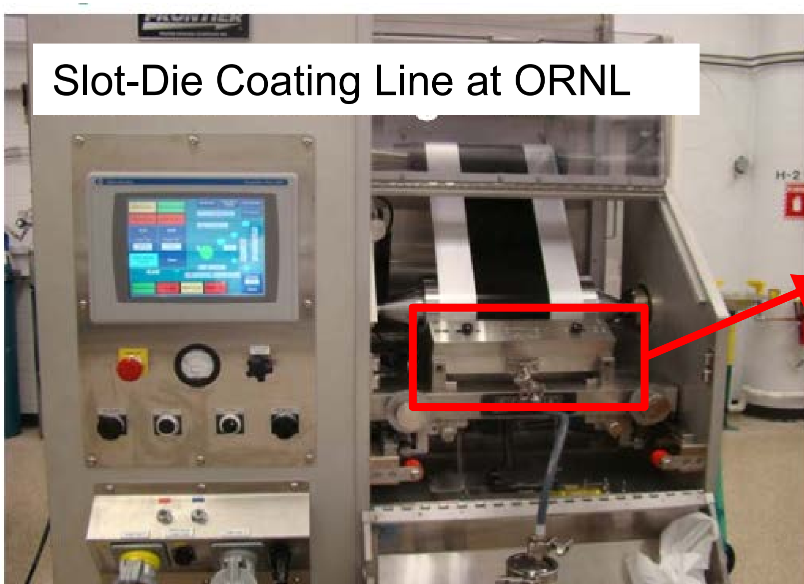


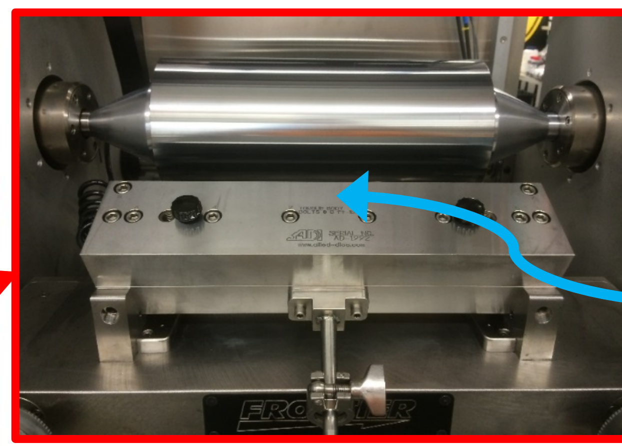


Electrode	Composition	Areal Loading (mAh/cm^2)	Porosity (%)	Electrode Thickness (mm)
Cathode	NMC532/PVDF/CB = 90/5/5 wt%	1.9	36.8	52
Anode	Graphite/PVDF/CB = 92/6/2 wt%	2.4	32.5	55

Approach and Strategy: ORNL BMF

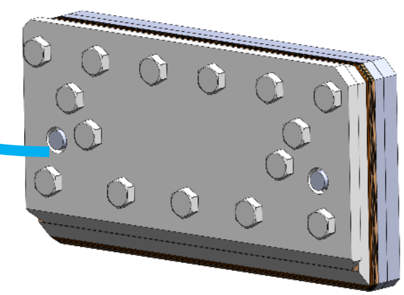
- End of Project Goals:
  - Integration of pouch cell scale CoEx printhead equipment at ORNL Battery Manufacturing Facility (BMF)
  - Production and characterization of 14 Ah pouch cells
  - Develop a plan for commercialization of the CoEx technology with potential end-users and suppliers





Slot-Die will be replaced with CoEx printhead & high pressure slurry dispensers

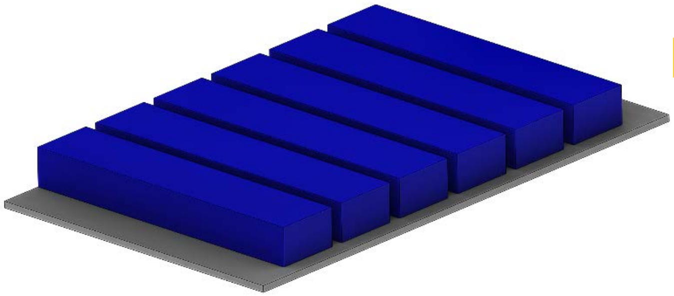
PARC CoEx printhead



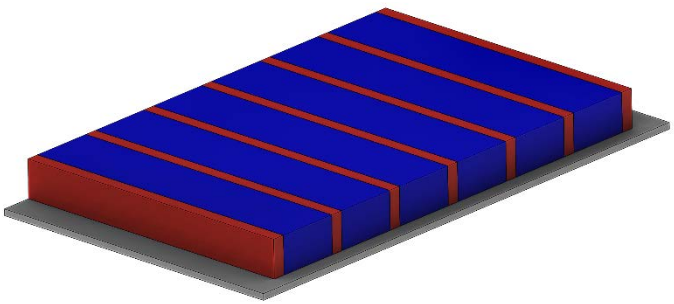
Technical Accomplishments and Progress

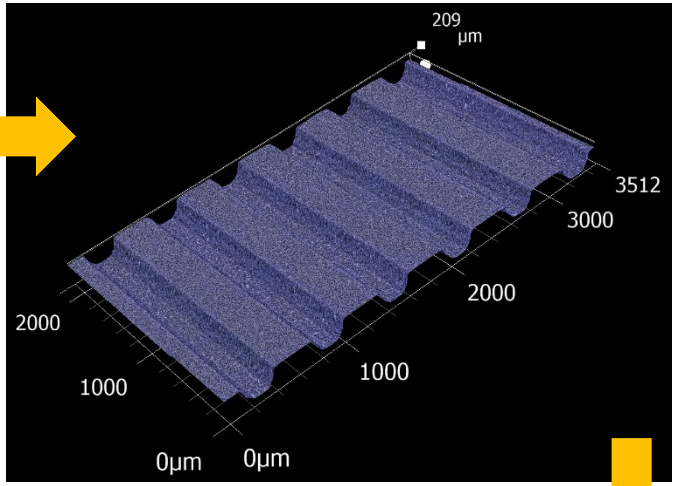
CoEx Cathode Print Feasibility Test

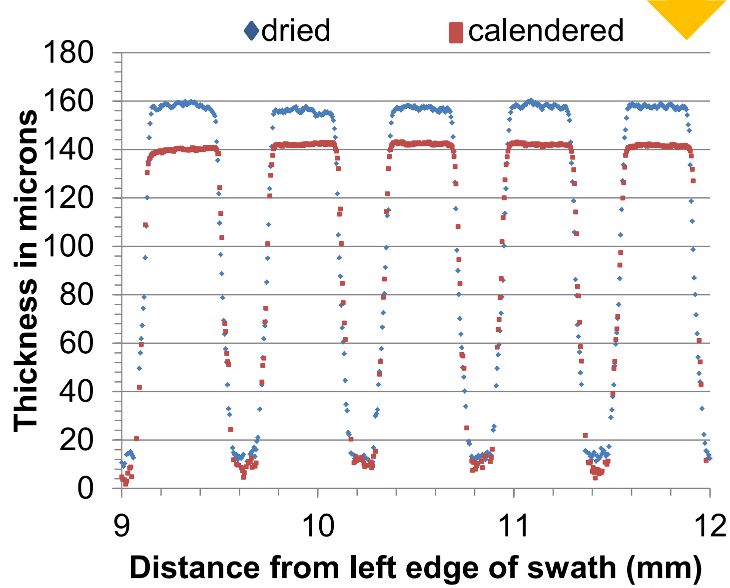
CoEx 1:



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







Print Profile

- 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.

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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

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Proposed Future Work: CoEx Modeling

These 3 examples have the same capacity (mAh/cm^2) and we will use this as a guide for electrochemical modeling.

Monolithic Comparison

h = 120 µm

CoEx 1: Corrugated Design

h = 144 - 180 µm

CoEx 2: Two-material Design

h = 130 - 140 µm

Description	Stripe Width Ratio	Stripe 1 Porosity (%)	Stripe 2 Porosity (%)
Monolithic case, assuming an electrode density of 2.8 g/cm^3	N/A	34	N/A
Corrugated design with open channels for electrolyte	2:1 to 5:1	34	100
Two-material design with a 20% porosity differential between the stripes	1:1 to 2:1	30	50

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

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