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

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PARC, a Xerox Company
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Project ID # BAT266

Overview

Timeline

- Project start date: December 17, 2015
- Project end date: Aug 17, 2019
- Percent complete: 50%

Barriers Addressed

- **Cost**: Current cost of Li-ion batteries is ~$200–$300/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 2017 Funding (DOE): $451,231
- FY 2018 Funding (DOE): $747,867

Partners

Project Lead

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Project Partners

Oak Ridge National Laboratory

Ford

Collaborations

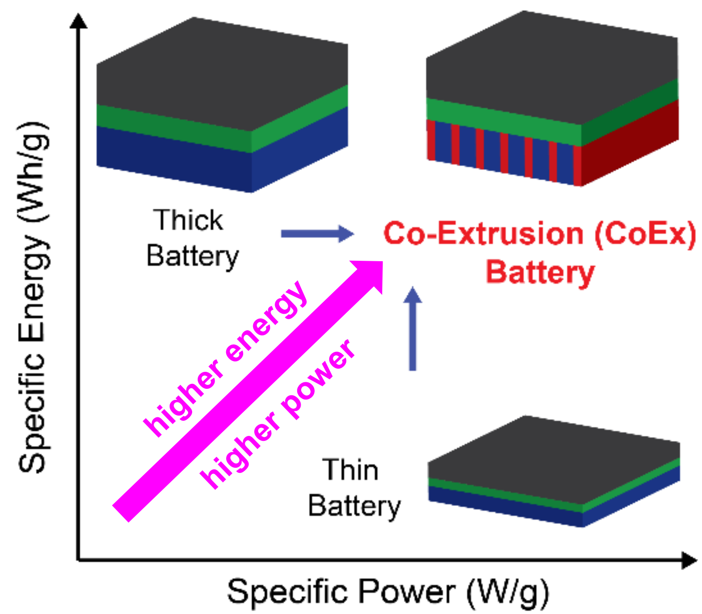
Navitas Systems

Argonne National Laboratory

SLAC National Accelerator Laboratory

Relevance and Project Objectives

- Overall Project Objectives:
  - Demonstrate pilot scale, electric vehicle (EV)–relevant 14 Ampere hours (Ah) pouch cells using Co-extrusion (CoEx), addressing:
    - Cost Barrier: ≥30% reduction in $/kWh costs through thick structured high energy and power electrodes
    - Performance Barrier: Gravimetric energy density improvement of ≥ 20% relative to conventional electrodes of the same chemistry
- March 2017–March 2018 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
  - Design and fabricate the printhead that will print CoEx cathode electrodes for small (1 Ah) pouch cells



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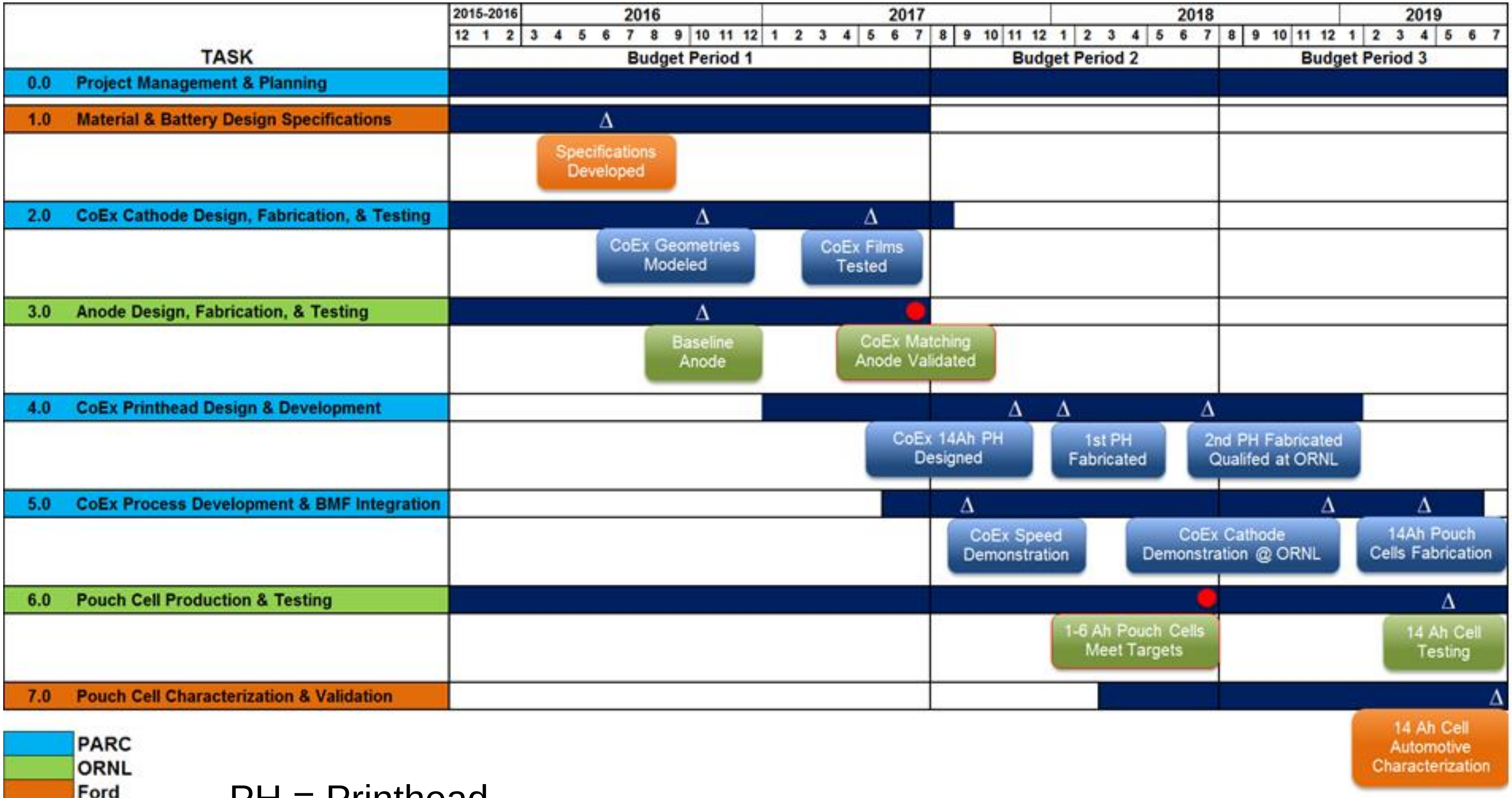
Milestones: FY 2018/2019

Milestone	Type	Description	Due Date	Status
Integrated Design Complete	Technical	Complete design of printhead and support hardware for CoEx printhead capable of pouch cell printing	11/24/2017	Completed
Fabrication Complete	Technical	Printhead and support hardware are fabricated	1/19/2018	Completed
Duplicate Hardware Qualified	Technical	A duplicate printhead system is fabricated and qualified for use on BMF at ORNL	8/16/2018	Not Complete
Pouch Cell Test Complete	Go/No-Go	Fabricate and test at least 5 full ≥ 1 Ah pouch cells which meet performance targets within 20%	8/16/2018	Not Complete
CoEx Printhead Demonstration	Technical	CoEx printhead is able to generate ≥ 125 µm cathodes on web coater with no visible print defects	12/5/2018	Not Complete
Large Pouch Cell Fabrication	Technical	CoEx hardware is able to fabricate CoEx electrodes for 14 Ah pouch cells	4/1/2019	Not Complete
Electrochemical Testing and Final Report	Technical	14 Ah pouch cells assembled and characterized	8/6/2019	Not Complete

Approach and Strategy: Timeline

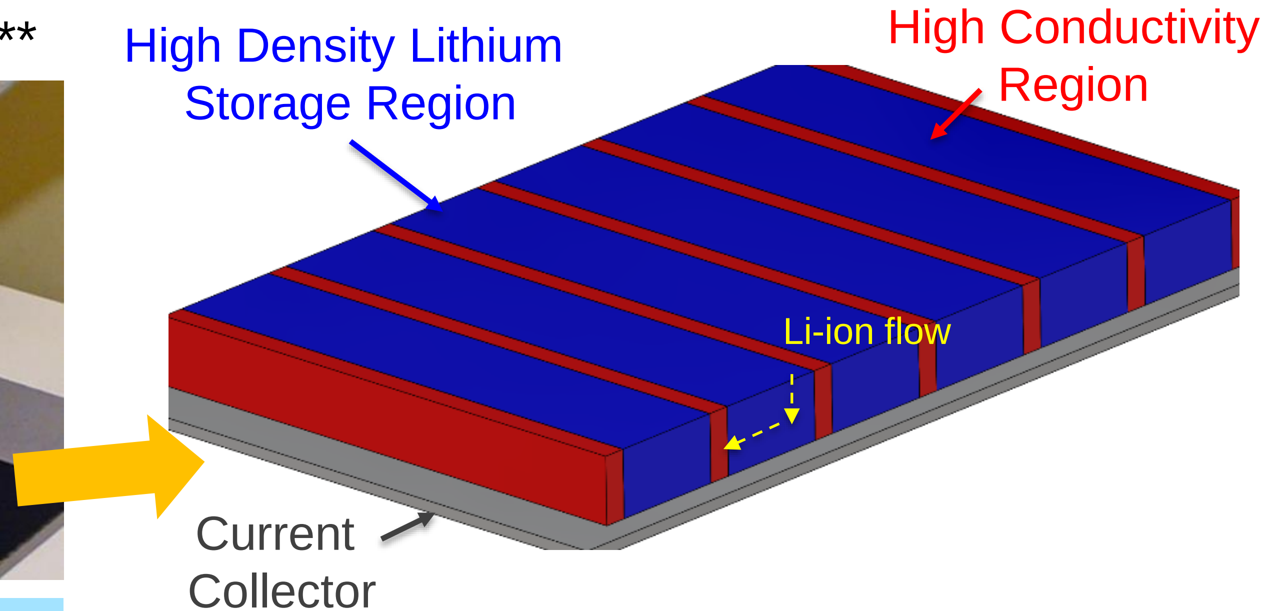


PH = Printhead

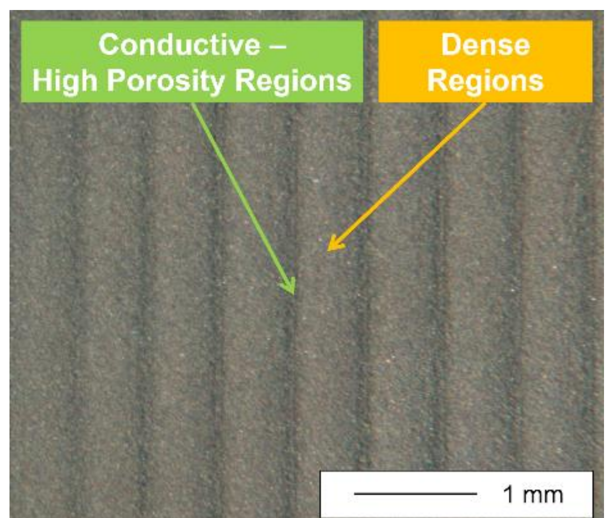
BMF = Battery Manufacturing Facility

Approach and Strategy: CoEx Cathodes

Co-extrusion Printhead**



The CoEx printhead enables the high-speed deposition of a cathode film with interdigitated regions of differential porosity. Thick, co-extruded cathodes can change conduction pathways in lithium-ion batteries, decoupling power and energy density trade-offs.



Top View
Dried CoEx Cathode Sample

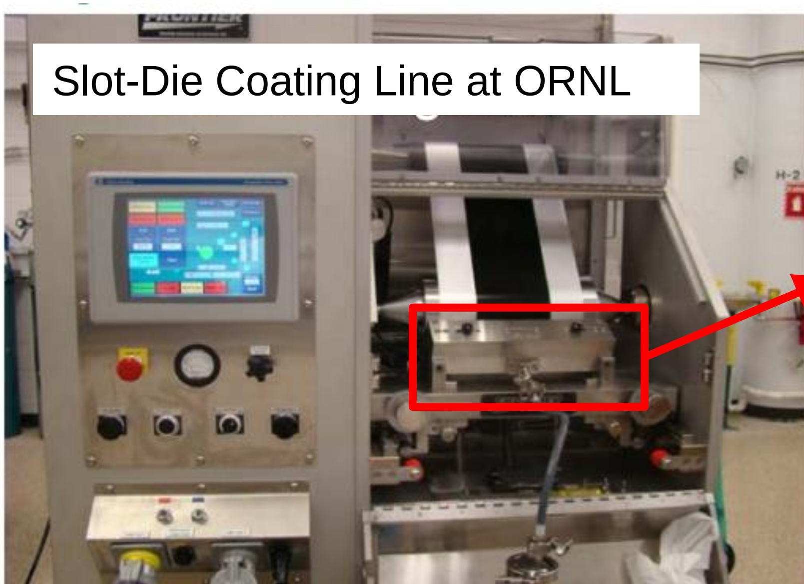
***Funded in part by the Advanced Research Projects Agency-Energy (ARPA-E), U.S. Department of Energy, under Award Number DE-AR0000324*

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 (thick) uncracked anode (5-6 mAh/cm^2) with NMP/PVDF solvent/binder system to match CoEx cathode capacity; Show capability to maintain thick anode coating integrity after calendering to 30-40% porosity.

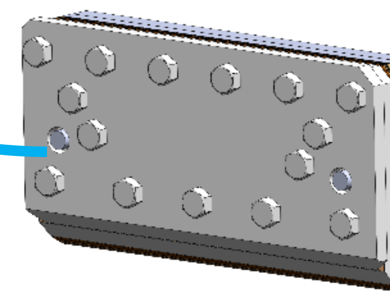
Approach and Strategy: ORNL BMF

- 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

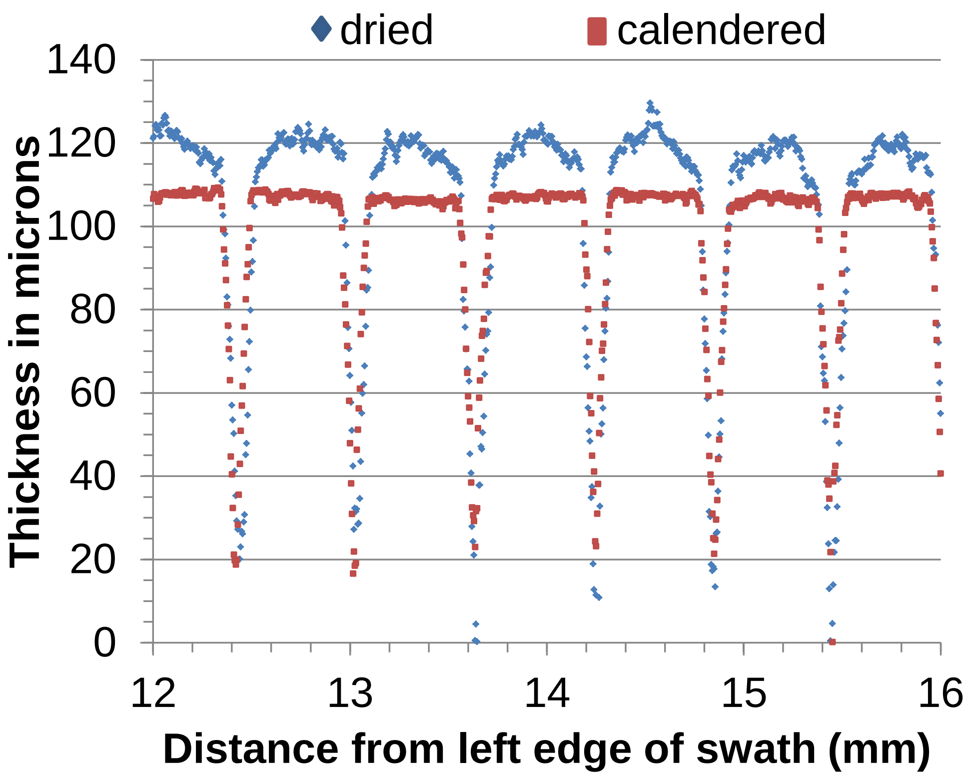


Slot-Die will be replaced with CoEx printhead & high pressure slurry dispensers

PARC CoEx printhead



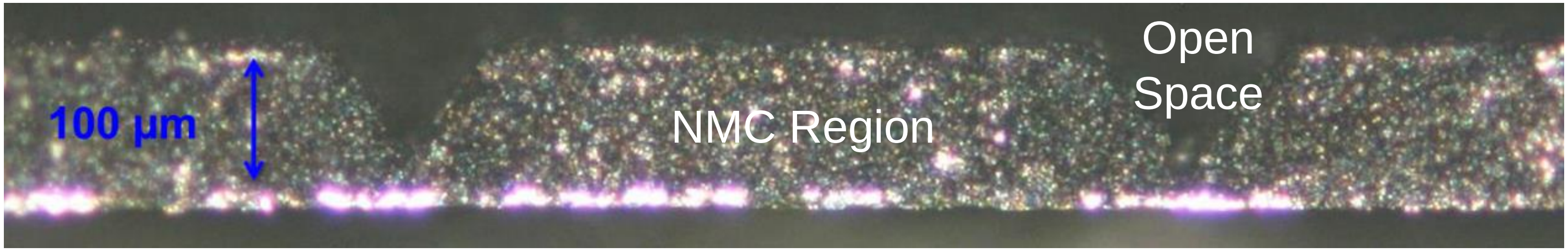
Printing of CoEx Structured Cathodes



Data Previously Presented

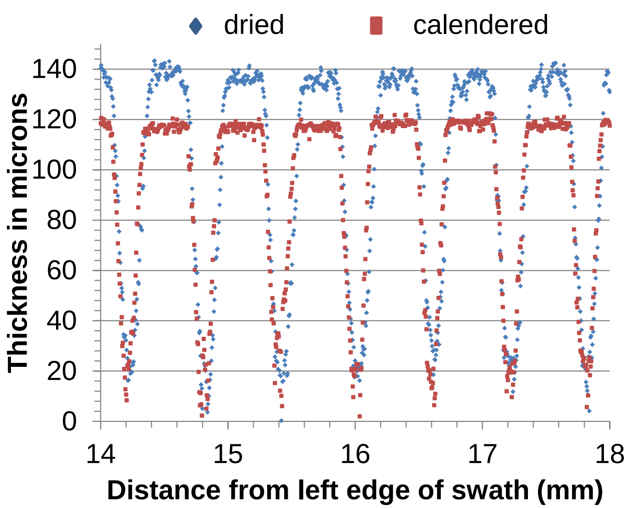
CoEx printheads and slurry formulations can be used to create corrugated electrode structures, with periodic grooves that fill with electrolyte and provide fast ionic transport

These grooves form during drying and maintain their shape during calendering process

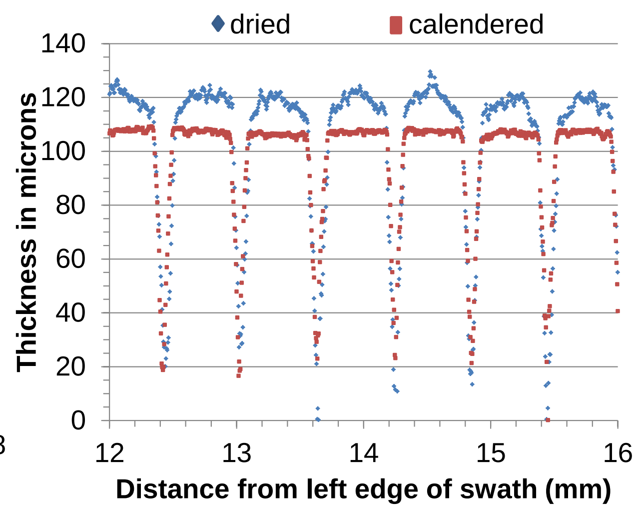


Current Collector

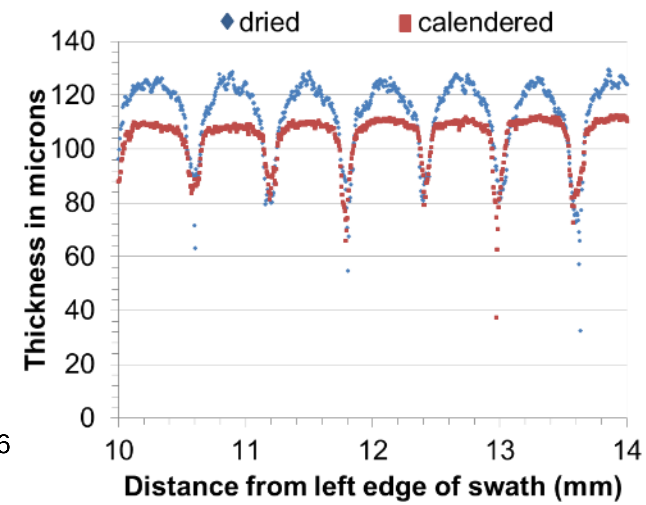
Tuning of CoEx Structures



Deep Grooves



Intermediate Grooves



Shallow Grooves

- By adjusting the composition of the CoEx paste formulations and print process parameters, we can tune the shape and periodicity of the structuring
- We performed electrochemical tests on a variety of CoEx structures with similar loading but varying geometry

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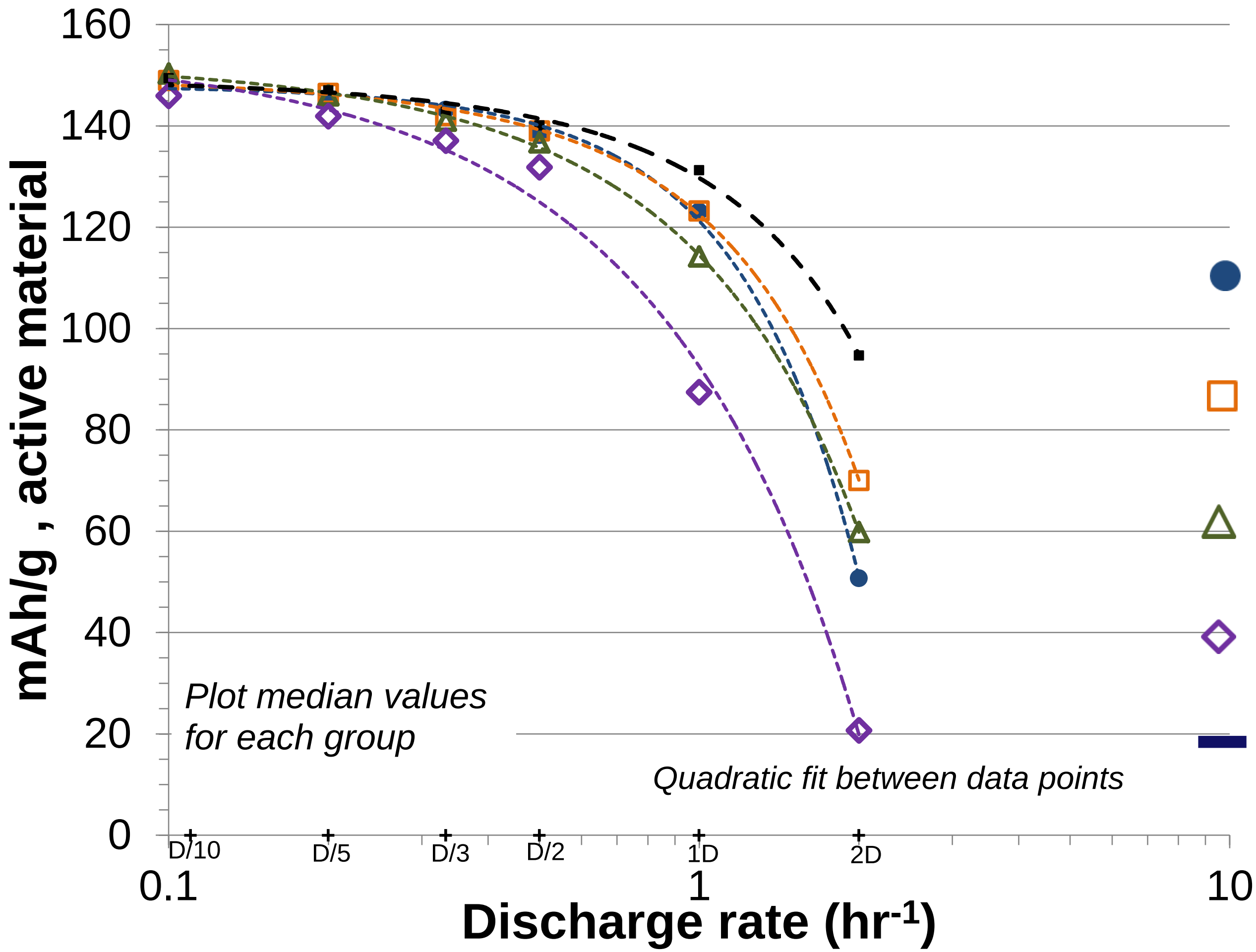
Technical Accomplishments

Comparing CoEx Structures – Discharge Rate



Half-cells:

Cathode	Thick µm	Active material mg/cm^2
CoEx Shallow Grooves	113	24.7
CoEx Medium Grooves	108	23.2
CoEx Deep Grooves	102	21.6
Thick Homogeneous	94	22.2
Baseline	69	15.8

Baseline cathodes assembled with Li-metal anodes in CR2032 format and tested at PARC.

CoEx electrodes show significant improvement over unstructured electrodes with similar loading

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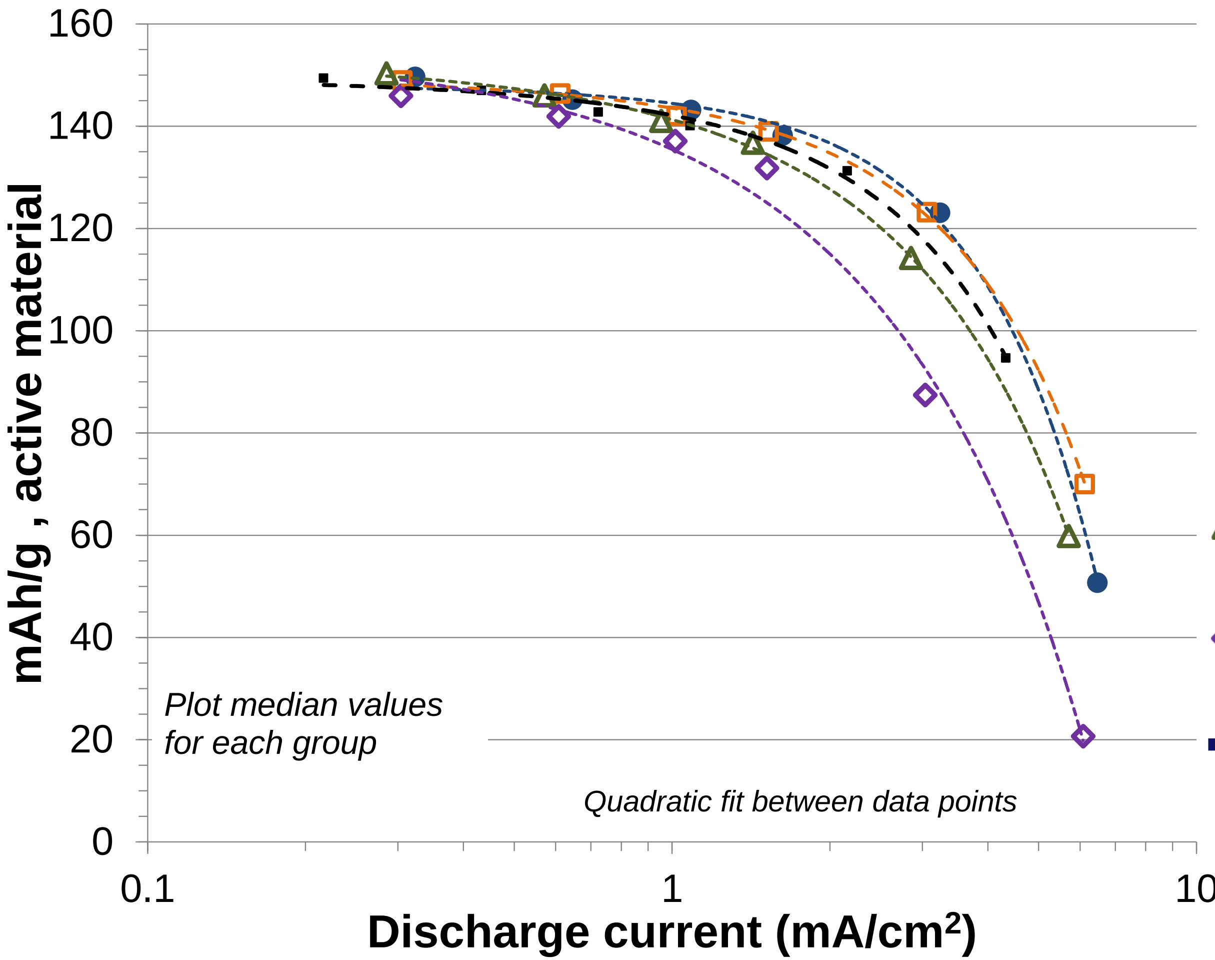
Technical Accomplishments

Areal Current Capability of CoEx Cathodes



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Baseline cathodes assembled with Li-metal anodes in CR2032 format and tested at PARC.

CoEx prints have increased discharge current capability compared to baseline

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Predicted CoEx Pouch Cell Performance

- Electrochemical results and geometric scaling arguments are used to predict the improvement at the pouch cell level due to CoEx structuring
- Results show moderate improvements at ≤ D/2 vs thinner baseline conventional cathodes and dramatic improvements at ≥ 1D vs unstructured, thicker cathodes

Best CoEx Cathode Vs Baseline Conventional Unstructured Cathode

D-rate	mAh/g	Wh/ kg	Wh/ L	W/kg
D/10	9%	10%	9%	10%
D/5	8%	9%	8%	9%
D/3	9%	9%	8%	9%
D/2	8%	6%	5%	6%
1D	2%	-3%	-4%	-3%
2D	**-46%**	**-44%**	**-44%**	**-44%**

Best CoEx Cathode Vs Thick Unstructured Cathode

D-rate	mAh/g	Wh/ kg	Wh/ L	W/kg
D/10	2%	1%	-1%	1%
D/5	1%	2%	-1%	2%
D/3	3%	3%	0%	3%
D/2	3%	4%	1%	4%
1D	**35%**	**36%**	**33%**	**36%**
2D	**119%**	**136%**	**130%**	**136%**

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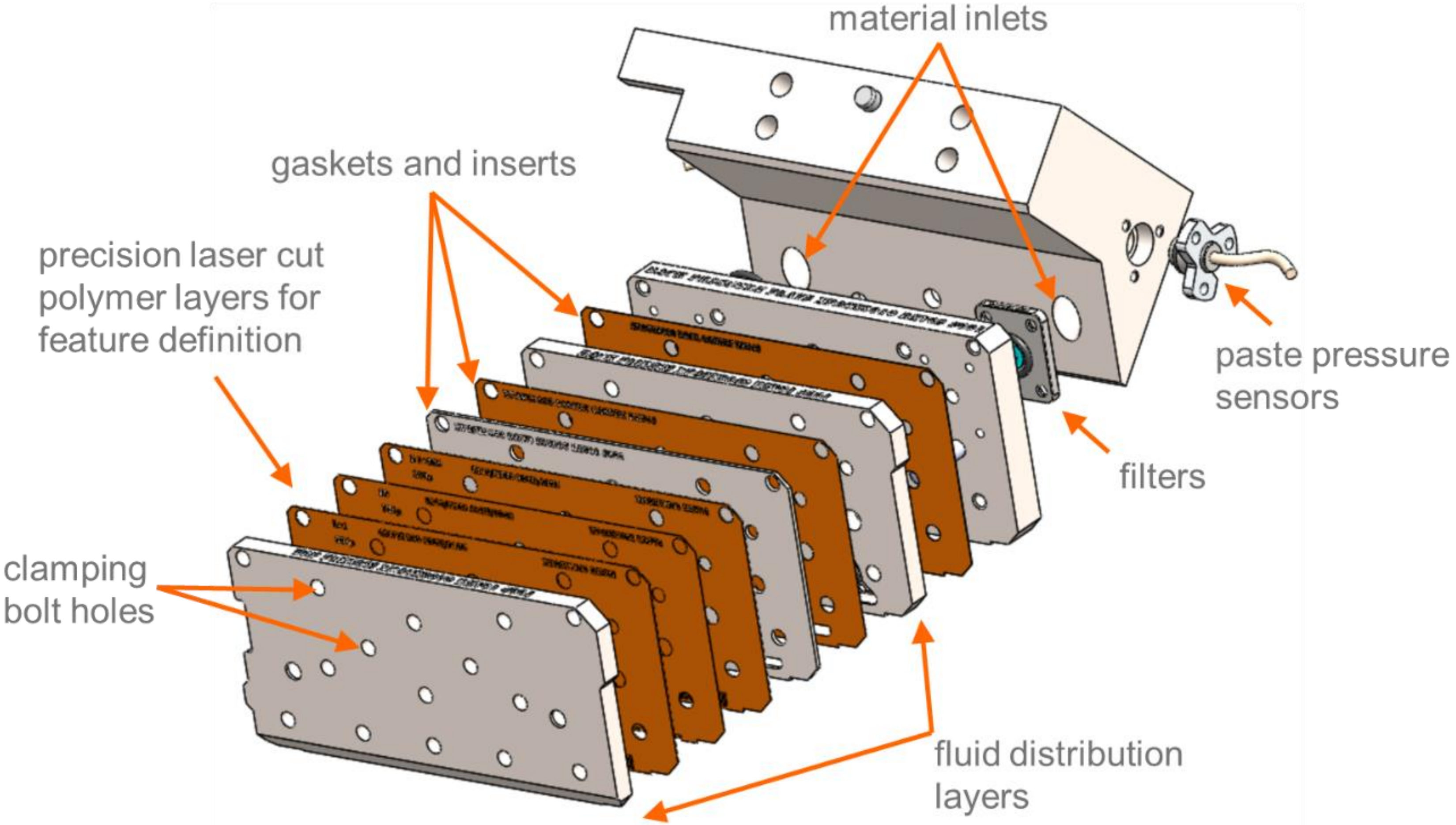
Printhead Development

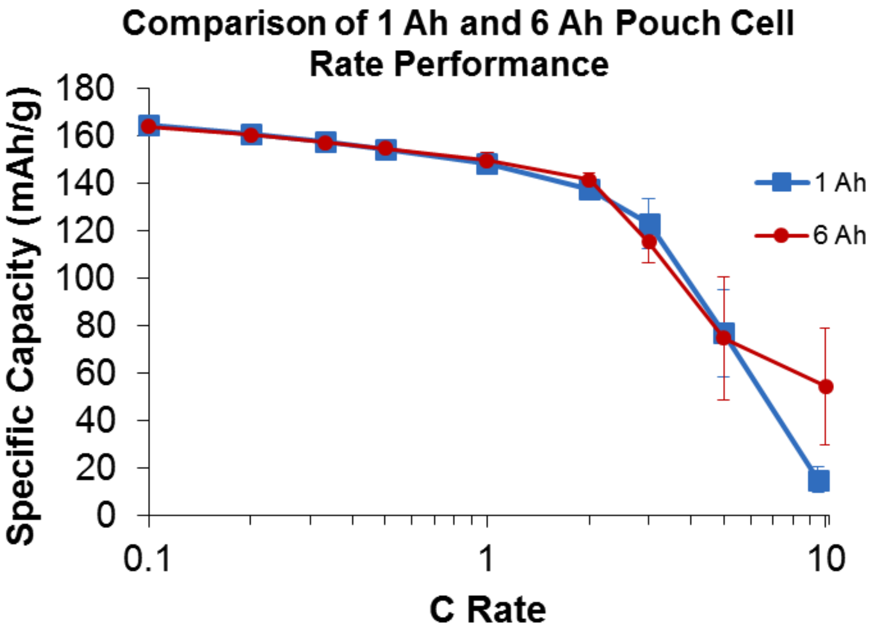


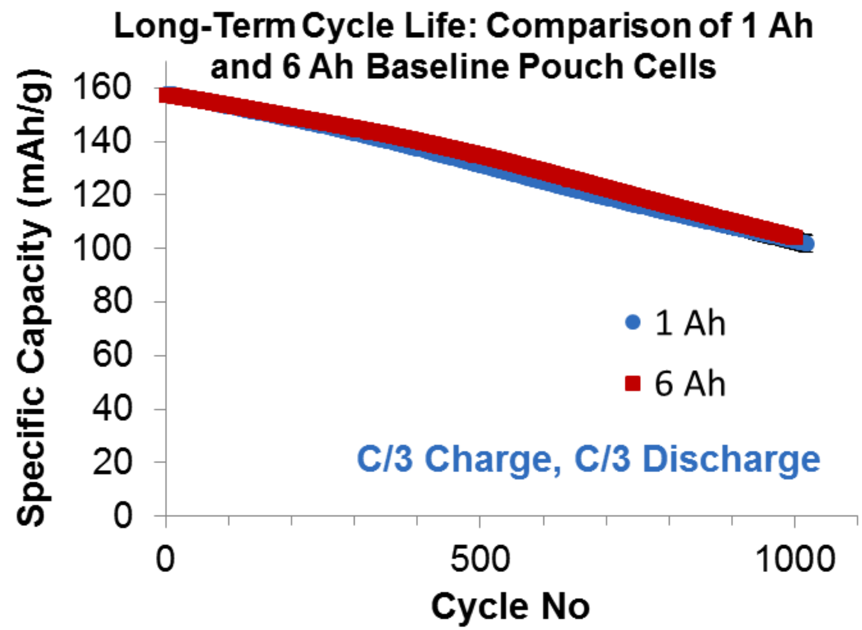
To enable cathode fabrication for pouch cells (84.4 x 56 mm), a new printhead was designed and fabricated

Baseline Pouch Cell Development

Baseline materials were used to create 1 and 6 Ah pouch cells

	Active Material	Active Material Loading (mg/cm^2)	Calendered Porosity	Electrode Thickness (µm)
Cathode	NMC 532	14.70	35%	55
Anode	Superior SLC 1520T Graphite	7.45	45%	63





Both 1 and 6 Ah cells show similar C-rate and cycle life performance

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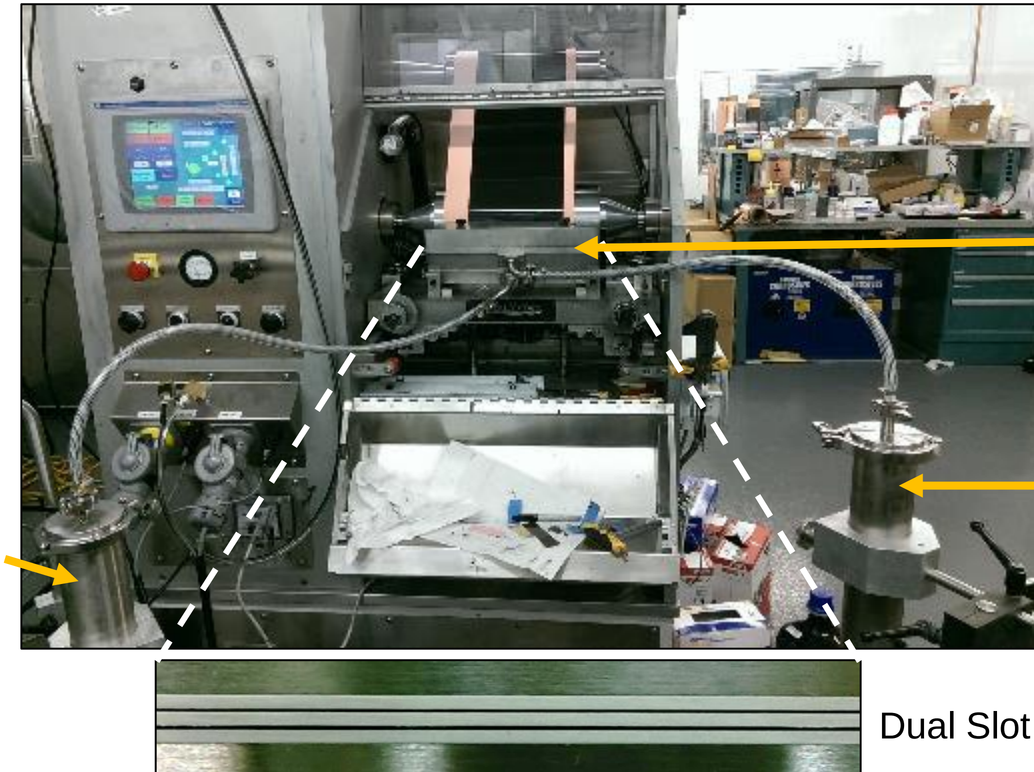
Two-Layer Thick Anode Coating Design

Dual Slot Die Setup



Dual Slot Die

Slurry for Top Coating

Slurry for Bottom Coating

Dual Slot Die Edge

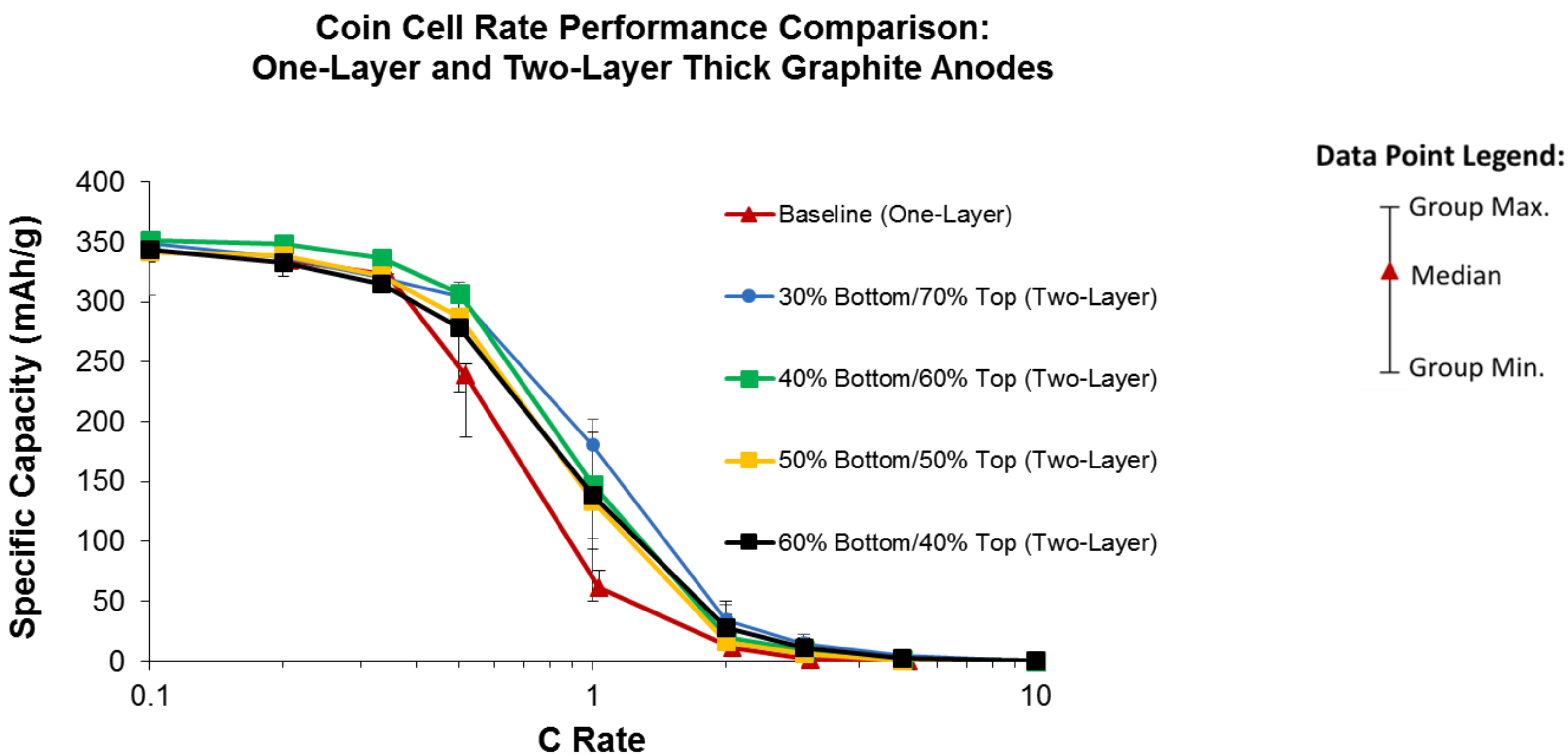
Thick Graphite Anode to match CoEx cathode (~6 mAh/cm^2)

4 wt% Binder
8 wt% Binder
Cu

(Baseline binder composition is 6 wt%)

- Coated 2-layer thick anodes with more binder in the bottom layer (8 wt%) to improve rate performance and adhesion to the current collector without sacrificing actives loading
- Dual slot die setup allows two different compositions to be coated on top of each other simultaneously, and for different ratios of top and bottom composition

Two-Layer Anode Rate Performance



- Two-layer anodes with more binder in the bottom layer show better rate performance at high C rates compared to the one-layer baseline
- Performance of all two-layer anodes is similar, but the ~40% Bottom/60% Top Coating is slightly better at C/5 and C/3, and the ~30% Bottom/70% Top Coating is slightly better at 1C

Responses to Reviewer Comments

Comment From 2017 Merit Review	Response
One reviewer warned that regions of high porosity mixed with regions of low porosity in the CoEx cathode will result in uneven current density and possibly lead to Li plating on the anode	We understand the risk that current inhomogeneities may eventually lead to plating on the anode, especially at the high current densities seen with the thicker cathode. In order to help assess the effect that differential porosity will have on lithium transport and plating, we have applied for and received time on beamlines at the SLAC National Accelerator Laboratory. We intend to use the SSRL beam line to interrogate CoEx structures through tomography, with the goal of eventually performing in operando measurements to track the motion of lithium ions.
Multiple reviewers would like to see a more comprehensive cost model developed	The cost and performance estimates given in our relevance statements reflected calculations from BatPAC as well as preliminary modeling based on LCO cathodes. We feel that a more meaningful TEA will be possible after small pouch cell tests are complete, as this will be the first instance where CoEx cathodes will be paired with thick, matching anodes and tested in full cells.
Multiple reviewers are concerned that as thickness and electrode area are increased, the quality of the fabricated cathodes will be reduced	We recognize that as our goal is to scale up the production of thick, structured cathode electrodes from coin cell to pouch cell (over 30x increase in electrode area), having uniformly patterned electrodes is critical. This is reflected in our BP2 and BP3 milestones which include demonstrations of print quality and thickness uniformity

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

SLAC National Accelerator Laboratory (Collaboration)
Using synchrotron radiation source to perform microstructural analysis of CoEx electrodes

Upcoming Work & Challenges

Key Challenges

- Small Pouch Cell Electrode Fabrication
  - Moving from coin cell electrode fabrication to pouch cell electrode fabrication requires maintaining same degree of electrode uniformity over 30x greater area
  - Developing the matching high capacity anode for the CoEx cathode and maintaining good electrode integrity and adhesion
- Large Pouch Cell Electrode Fabrication / Roll Coater Integration
  - We will need to adapt our current batch electrode process to the continuous web process at the BMF

Future Work

- Ongoing (FY 2018)
  - Print process development with new CoEx printhead
  - CoEx printing of cathode electrodes for pouch cells
  - Continued development of high energy anodes for matching thicker CoEx Cathodes
  - 1 & 6 Ah pouch cell testing of baseline anodes (Go/No-Go)
- Proposed (FY 2019)
  - Hardware development for full-width (180 mm) printhead
  - Fabrication of 14 Ah pouch cells using CoEx cathodes and high energy anodes

Any proposed future work is subject to change based on funding levels

Summary

- **Relevance**
  - Demonstrate pilot scale, electric vehicle (EV)–relevant, 14 Ampere hours (Ah) pouch cells via Co-extrusion (CoEx), with goals of 30% reduction in cost and gravimetric energy density improvement of ≥ 20%
- **Approach**
  - Develop thick structured electrodes with CoEx to mitigate power and energy trade-offs in cathodes
  - Use advanced slot coating capabilities to develop a matching, thick, high capacity anode
  - Leverage ORNL's BMF to scale up CoEx production to produce 14 Ah pouch cells
- **Technical Accomplishments**
  - CoEx structures show substantial improvement in performance (at the coin cell level) compared to unstructured electrodes of comparable loading and moderate improvement in calculated pack level energy density compared to conventional cells
  - A new CoEx printhead has been designed and fabricated, scaling from coin cell to pouch cell electrode fabrication
  - We have
- **Future Work**
  - Further optimize CoEx cathode and anode for overall capacity improvement at high loading
  - Validate our half-cell performance results in full pouch cells

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Technical Back-Up Slides

Recommended Targets for Reference Cells

- Recommended Baseline Electrode Targets:
  - Cathode Loading:
    - Thickness: 64.5 μm +/- 30%
    - Capacity: 2.1 mAh/cm^2 +/- 10%
    - Mass: 0.0157 g/cm^2 +/- 10%
  - Anode Loading:
    - Thickness: 66.0 μm +/- 30%
    - Capacity: 2.4 mAh/cm^2 +/- 10%
    - Mass: 0.0081 g/cm^2 +/- 10%

Absolute Energy Density Calculations

These numbers reflect the volumetric and gravimetric energy density for a hypothetical 1 Ah pouch cell, based on the half cell, coin cell electrochemical data for each cathode

	Best CoEx Cathode			Baseline Cathode			Thick Homogenous Cathode		
D-rate	Wh/ kg	Wh/ L	W/kg	Wh/ kg	Wh/ L	W/kg	Wh/ kg	Wh/ L	W/kg
D/10	234	596	23	214	549	21	231	604	23
D/5	228	581	46	209	537	42	224	586	45
D/3	221	561	74	202	518	67	214	559	71
D/2	209	533	105	198	508	99	202	527	101
1D	176	447	176	182	467	182	129	338	129
2D	71	181	142	126	325	252	30	79	60