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 #
ES266

Overview

Timeline

- Project start date:
  December 17, 2015
- Project end date:
  Aug 31, 2019
- Percent complete:
  30%

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 2016 Funding (DOE): $360,232
- FY 2017 Funding (DOE): $451,231

Partners

Project Lead

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Project Partners

Oak Ridge National Laboratory

Ford

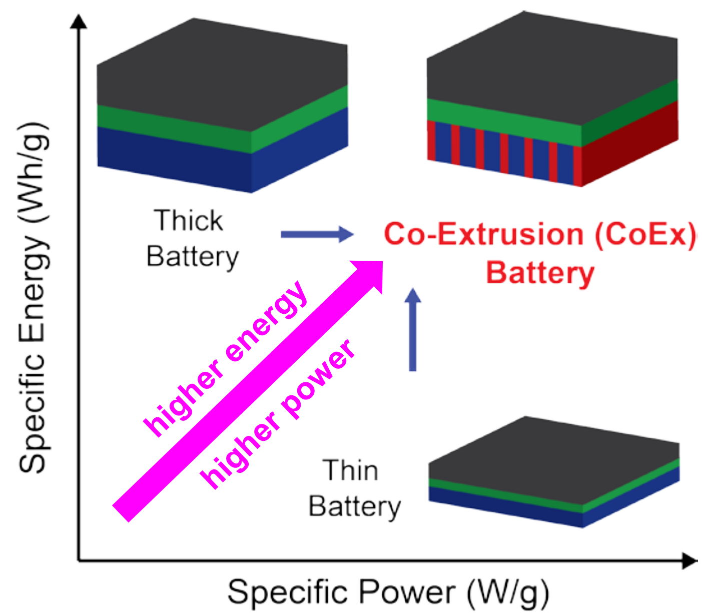
Collaborations

Navitas Systems

Argonne National Laboratory

Relevance and Project Objectives

- Overall Project Objectives:
  - Demonstrate pilot scale, electric vehicle (EV)–relevant 14 Ampere hours (Ah) 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
- FY2016/2017 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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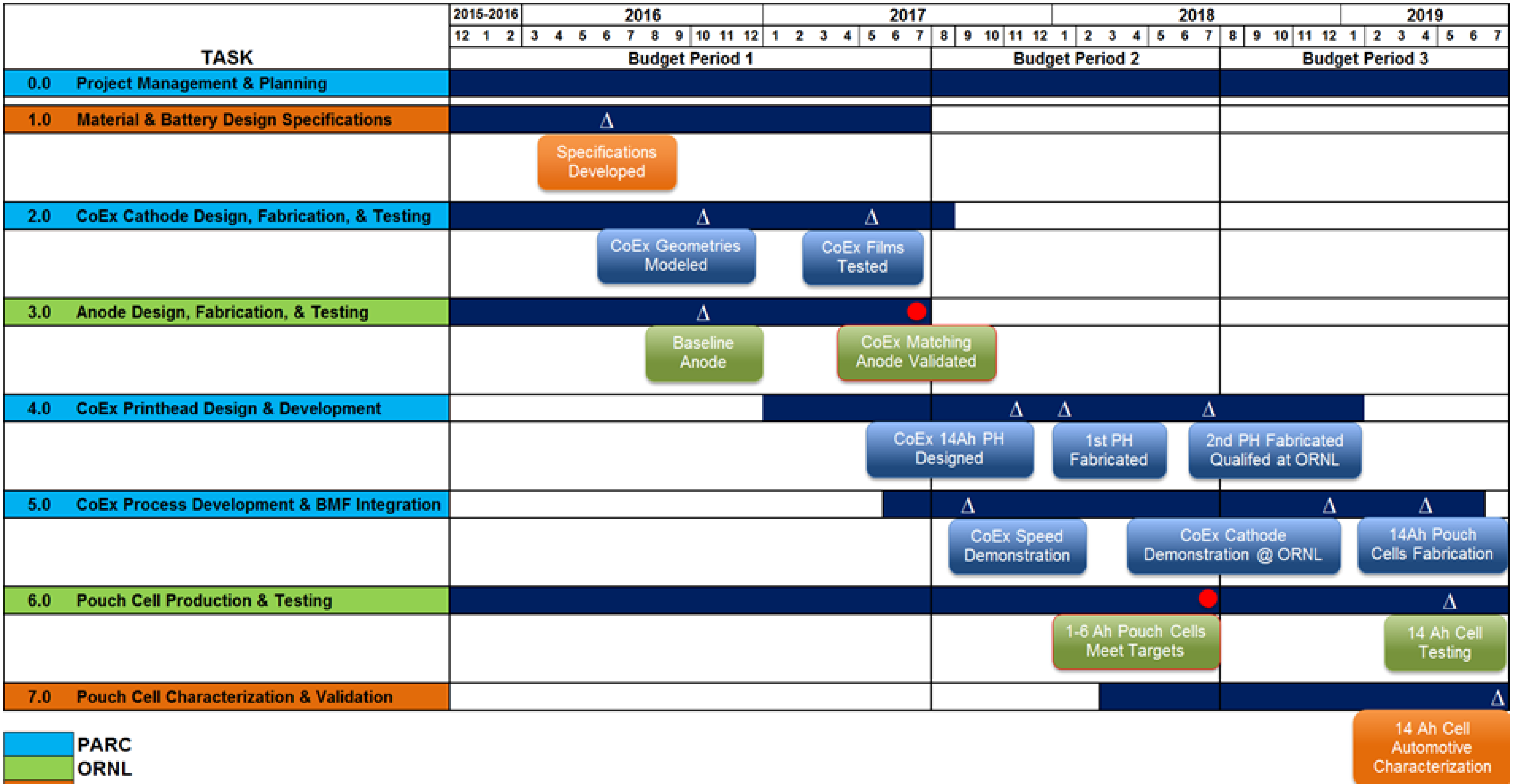
Milestones: FY 2016/2017

Milestone	Type	Description	Due Date	Status
Specifications developed	Technical	Recommended cell targets for a Nickel Manganese Cobalt (NMC)-graphite materials set are identified.	5/20/2016	Completed
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.	10/14/2016	Completed
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.	5/12/2017	Completed
Baseline Validated	Technical	Baseline anode meets specifications.	10/18/2016	Completed
Capability Demonstrated	Go/No Go	A homogenous ≥120μm anode film demonstrates the capacity required to balance the CoEx cathode.	8/16/2017	On Track

Approach and Strategy: Timeline



PARC
ORNL
Ford

PH = Printhead
BMF = Battery Manufacturing Facility

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Approach and Strategy: Co-extrusion (CoEx)

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.

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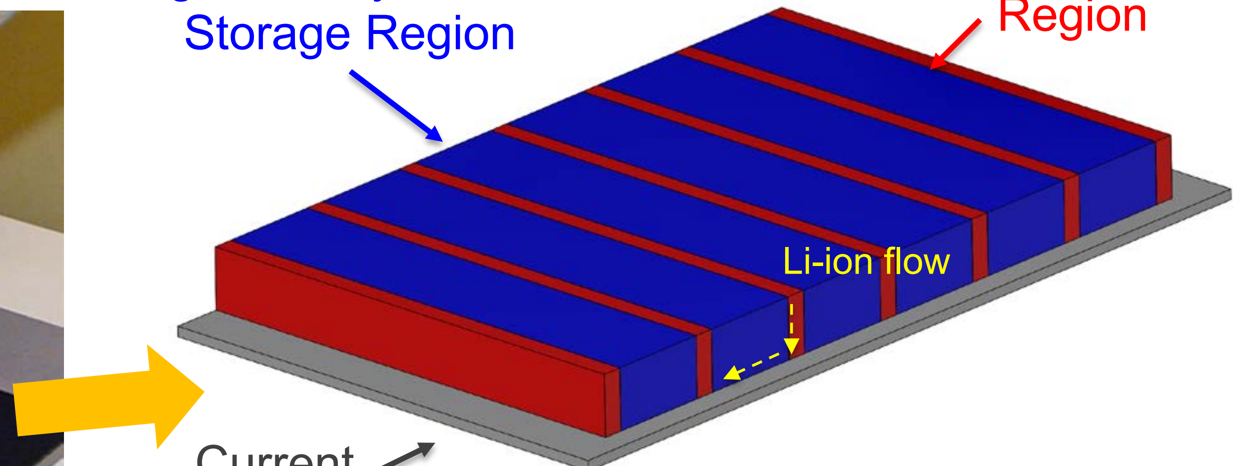

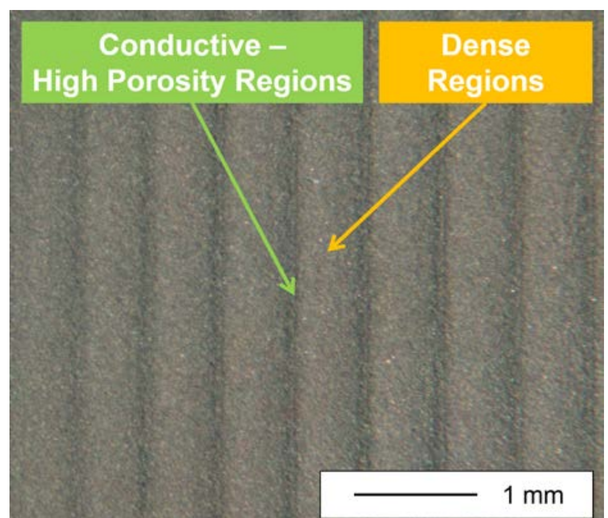

Top View
Dried CoEx Cathode Sample

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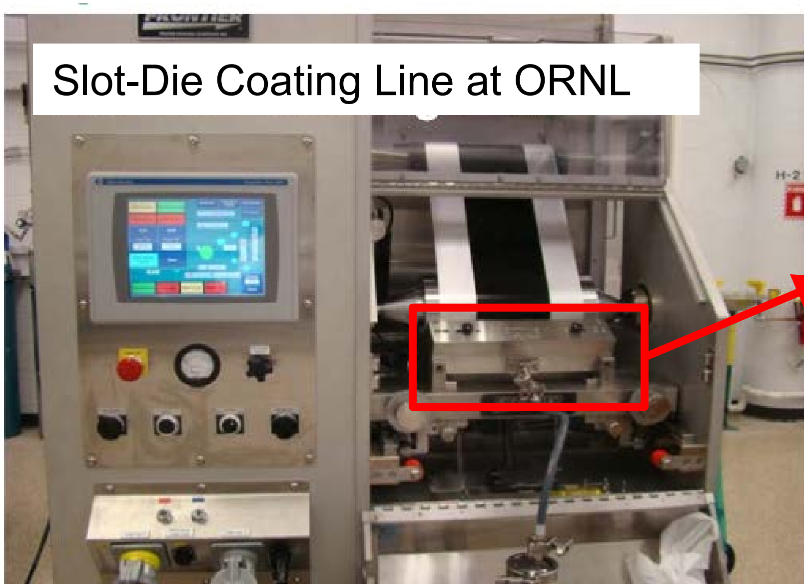
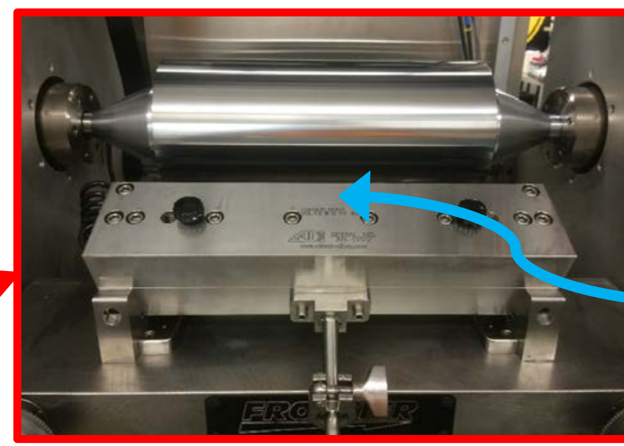
Approach and Strategy: High Capacity Anode

- 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

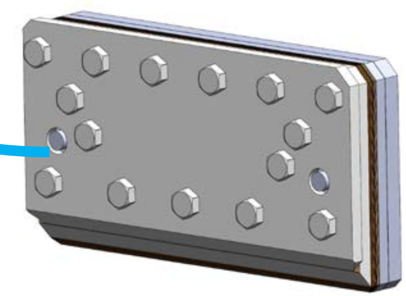
- End of Project Goals:
  - Integration of pouch cell scale CoEx printhead equipment at ORNL Battery Manufacturing Facility (BMF)
  - Production and characterization of 14 Ah pouch cells
  - Develop a plan for commercialization of the CoEx technology with potential end-users and suppliers





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

PARC CoEx printhead



Electrochemical Modeling Summary

Design rules established for ≥ 10% improvement in pouch cell capacity

***Assuming a 90/5/5 NMC532/CB/PVDF composition**

CoEx 1: Corrugated Design

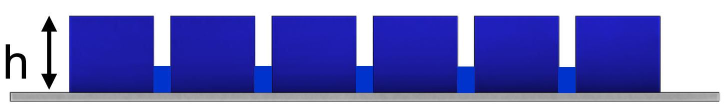
h ≥ 2X over monolithic reference

CoEx 2: Two-material Design

h ≥ 2X over monolithic reference

CoEx 3: Corrugated Design

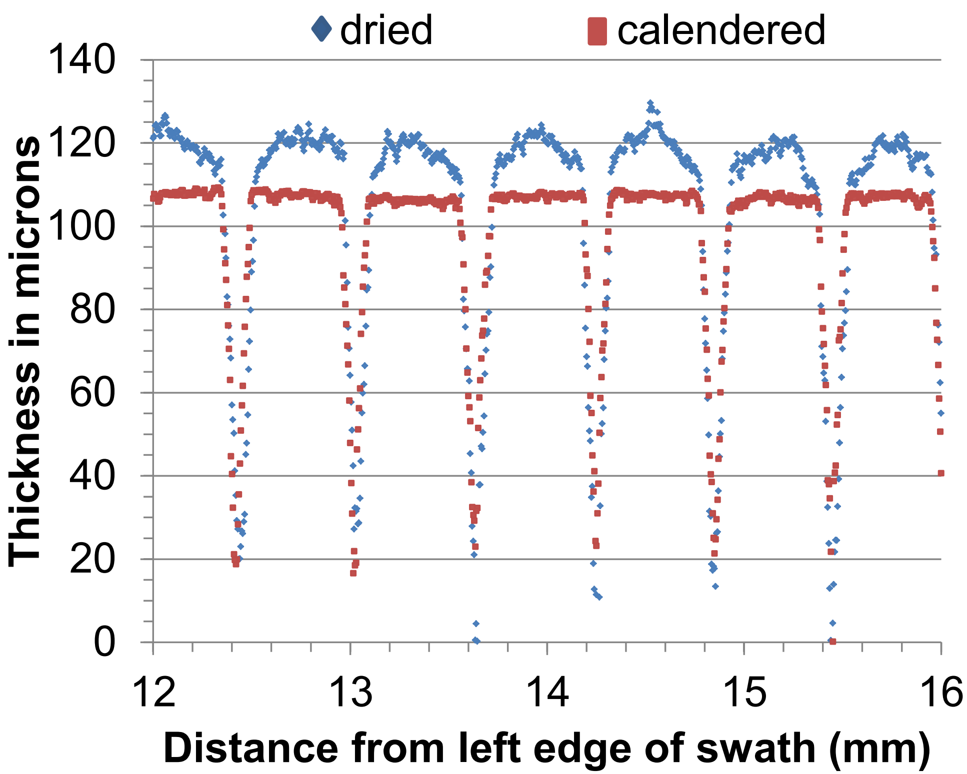
(with pad of cathode material in Region 2)



h ≥ 2X over monolithic reference

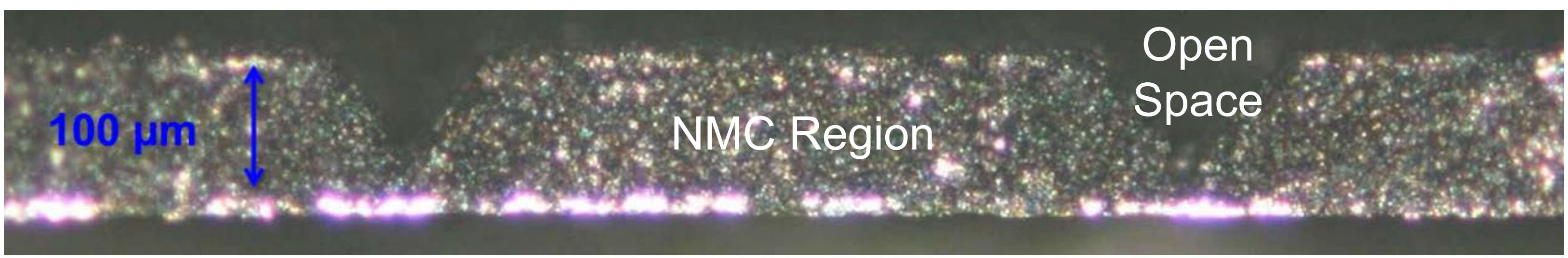
Description	Stripe Width Ratio	*Region 1 Porosity (%)	Region 2 Porosity (%)
Corrugated design with open channels for electrolyte	≥ 4	34	100
Two-material design with a porosity differential between the stripes	≥ 2	34	≥ 55
Corrugated design with open channels for electrolyte & a small pad of cathode material to compensate for volumetric losses	≥ 4	34	100 (not including pad of cathode material)

Printing of CoEx Structured Cathodes



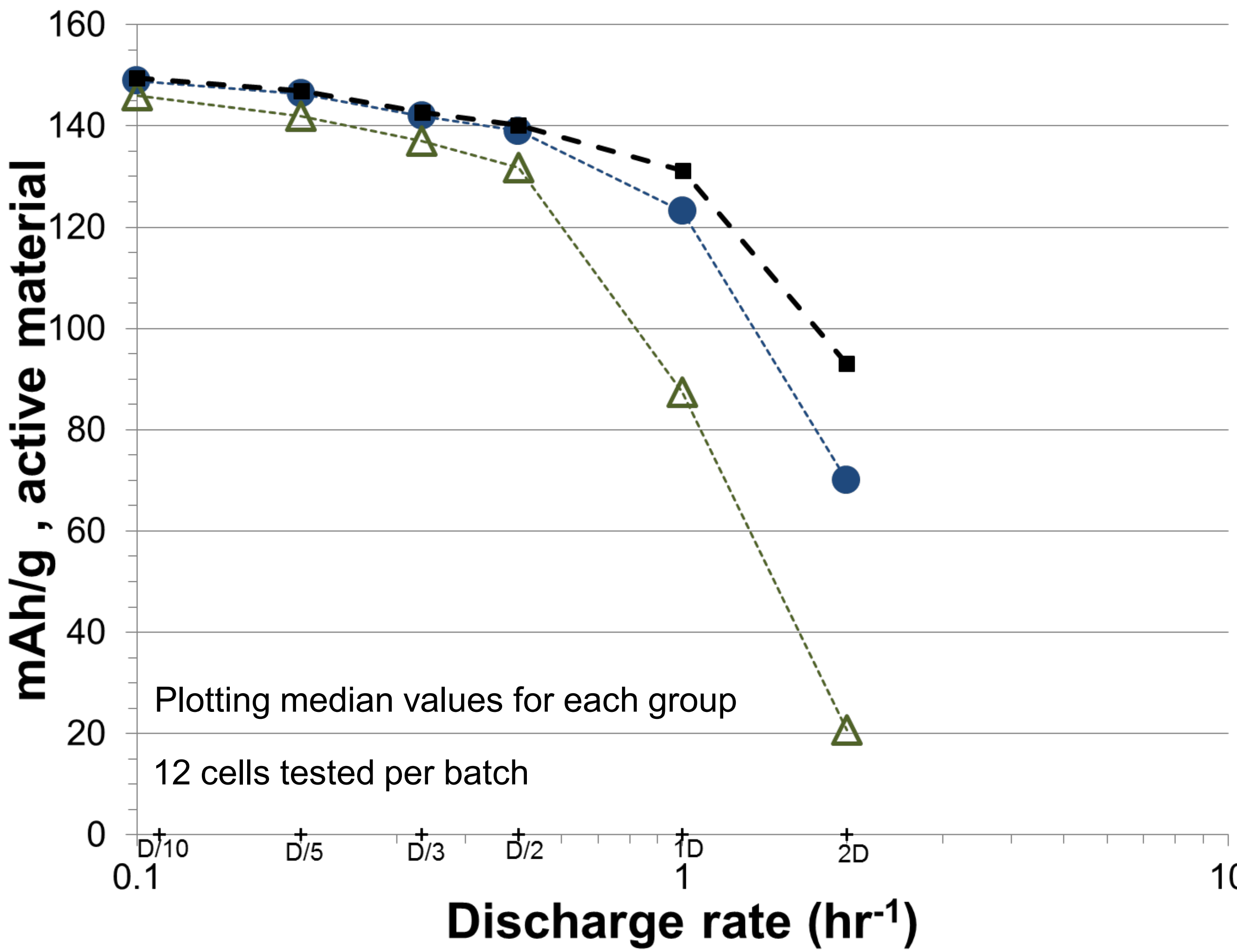
The first geometries attempted were CoEx 1 structures. This latest print (shown) is for a hybrid structure with loading in between that of the ideal CoEx 1 and CoEx 3 structures

Chromatic confocal profilometry (left) and cross-sectional micrograph (below) shows that CoEx "open space" structure is maintained after calendering



Current Collector

Specific Capacity of CoEx Cathodes



Half-cells:

Cathode	Thick µm	Active material mg/cm^2
PARC CoEx Cathode	108	23.2
Thick Conventional Cathode	102	21.6
PARC Baseline Cathode	69	15.8

CoEx electrodes show significant improvement over unstructured electrodes with similar loading

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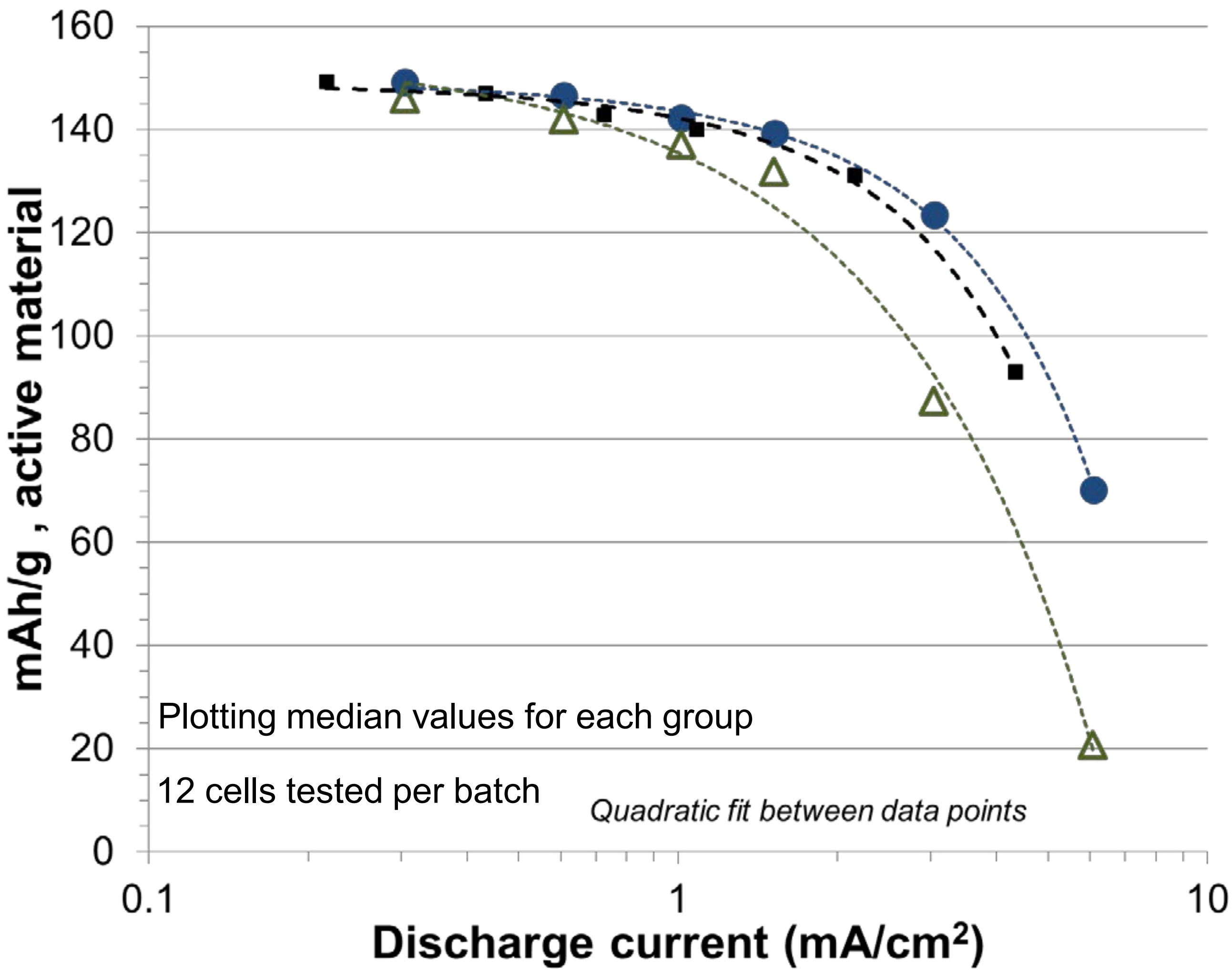
Technical Accomplishments

Areal Current Capability of CoEx Cathodes



Half-cells:

Cathode	Thick µm	Active material mg/cm²
PARC CoEx Cathode	108	23.2
Thick Conventional Cathode	102	21.6
PARC Baseline Cathode	69	15.8

CoEx electrodes show improvement in discharge current capability compared to unstructured cathodes

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

CoEx Cathode Vs Baseline Conventional Unstructured Cathode

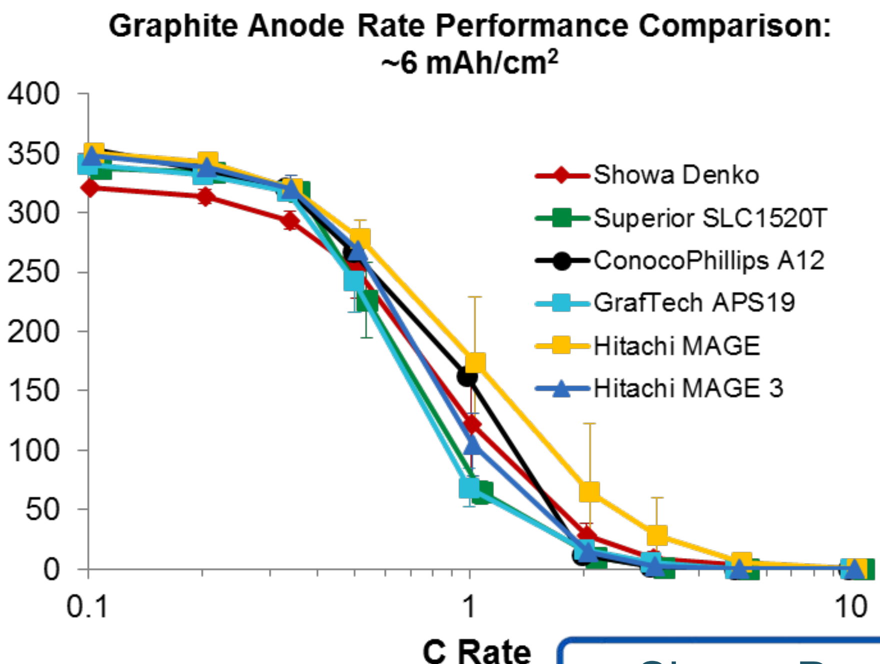
D-rate	mAh/g	Wh/ kg	Wh/ L	W/kg
D/10	7%	7%	6%	7%
D/5	6%	6%	5%	6%
D/3	7%	6%	5%	6%
D/2	7%	5%	4%	5%
1D	1%	**-3%**	**-4%**	**-3%**
2D	**-19%**	**-23%**	**-24%**	**-23%**

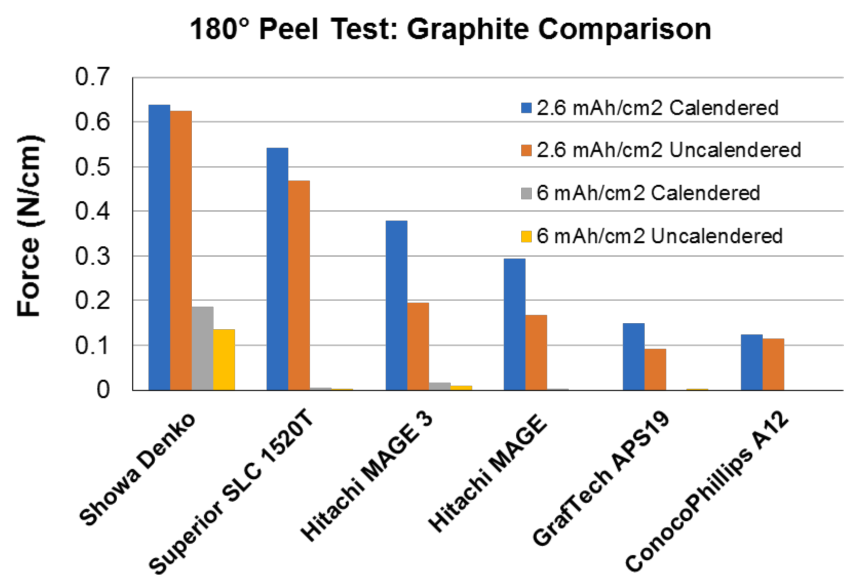
CoEx Cathode Vs Thick Unstructured Cathode

D-rate	mAh/g	Wh/ kg	Wh/ L	W/kg
D/10	**-1%**	**-1%**	**-4%**	**-1%**
D/5	**0%**	**-1%**	**-3%**	**-1%**
D/3	1%	**0%**	**-3%**	**0%**
D/2	2%	3%	**0%**	3%
1D	33%	37%	33%	37%
2D	227%	221%	213%	221%

Initial Anode Selection and Testing

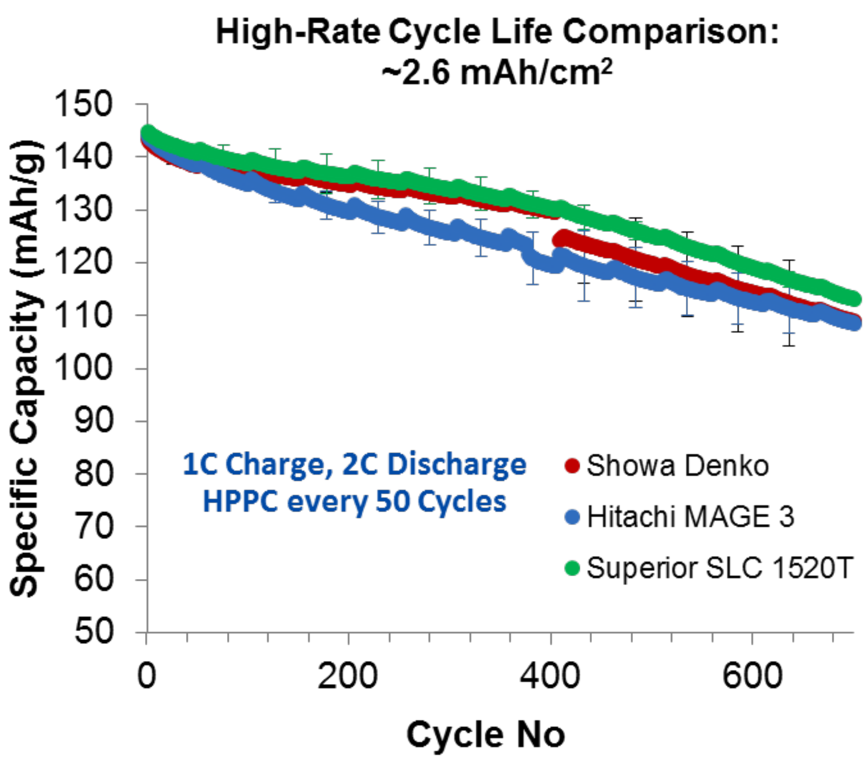
- 11 different graphite anode materials were tested and evaluated for their electrochemical performance
- A downselect process was used to identify the top 3 materials based on electrochemical performance and then adhesion in thick calendered films

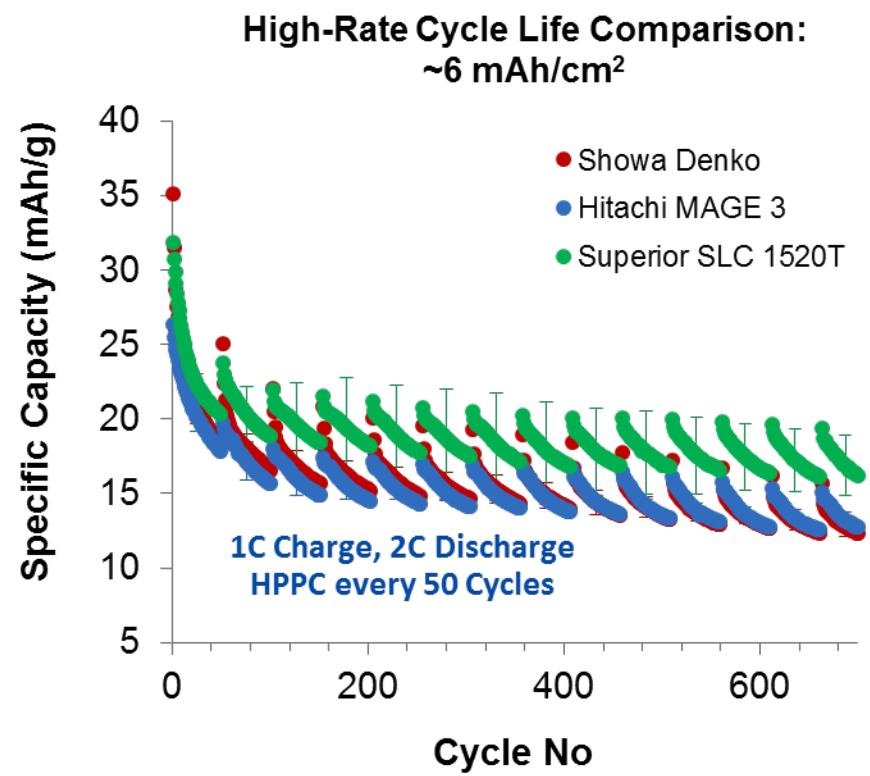




Showa Denko, Superior SLC 1520T, and Hitachi MAGE 3 are top anode candidates

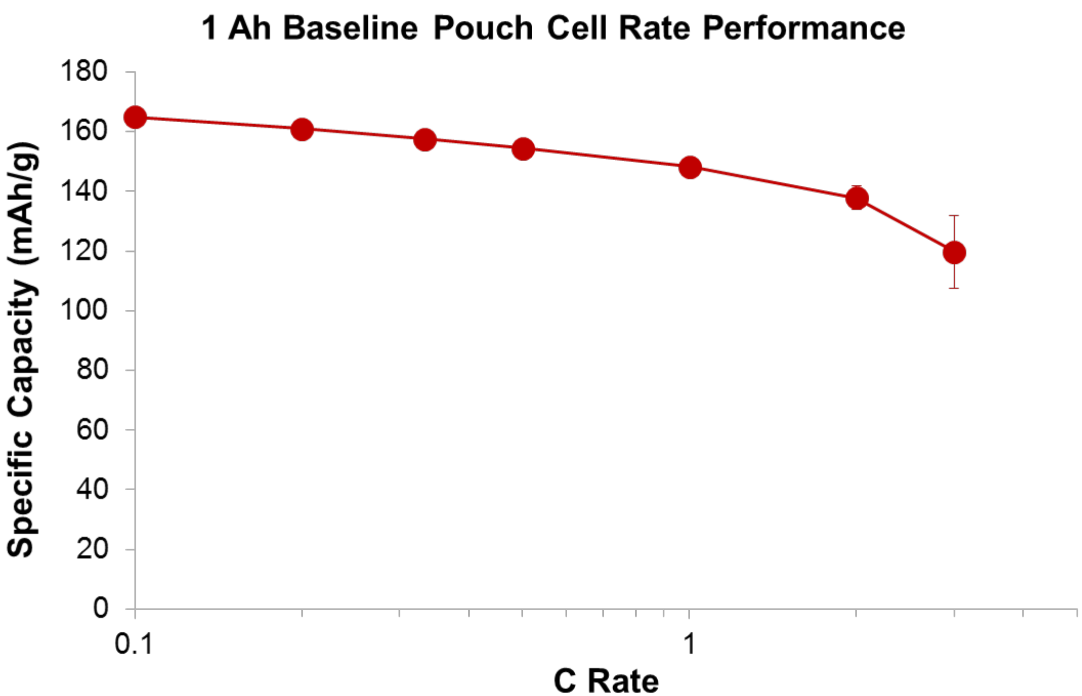
Anode Cycle Life Comparison





Based on cycle life data (single layer pouch cells) at low and high sample loadings, Superior SLC 1520T was chosen as the graphite for pouch cell investigations moving forward

Anode Pouch Cell Performance



<u>Cathode Parameters</u>

- NMC 532
- Loading: 14.7 mg/cm^2
- 35% porosity (calendered)
- Thickness: 55 µm

<u>Anode Parameters</u>

- Superior SLC 1520T
- Loading: 7.45 mg/cm^2
- 45% porosity (calendered)
- Thickness: 63 µm

Graphite anode performance was verified in multilayer pouch cells by matching with an appropriate NMC 532 cathode and testing in 1 Ah full cells

Responses to Reviewer Comments

Comment From 2016 Merit Review	Response
Multiple reviewers expressed concern that the structuring within the cathodes would not be able to survive the calendering process	The cathode samples shown in this presentation do not have any observable cracks via optical inspection. For the CoEx samples with alternating "ridges" and "valleys", we have not observed cracking problems either after drying or calendering. The ridges maintain their integrity even though calendering broadens the ridge regions. This broadening of the ridges partially compensates for thickness reduction but still results in overall film compression (densification). At the coin cell level, we have not noticed any issues with mechanical integrity.
One reviewer did not see how a 30% decrease in cost is possible on a kWh basis	Our estimates of 30% decrease in cost were derived from BatPAC calculations (v2.2.2), assuming that CoEx (150 μm thick) would be able to achieve comparable performance to the baseline conditions, and accounting for the reduced costs of NMP drying and recovery due to the relatively higher solids loading of CoEx slurries.
Multiple reviewers would like to see a more comprehensive cost model	The cost and performance estimates given in our 2016 Merit Review presentation reflected calculations from BatPAC as well as preliminary modeling based on LCO cathodes. We plan to refine the cost model based on the electrochemical modeling done in the current budget period, the electrochemical data from proposed pouch cell work, and assistance from our industrial partners

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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

Upcoming Work & Challenges

Key Challenges

- CoEx Cathode (NMC 532)
  - Enabling further increases in electrode thickness and current density without sacrificing performance
  - Scaling existing CoEx printhead and support hardware from 65 mm wide prints to 150-200 mm wide for 14Ah pouch cell production
- Anode (Graphite)
  - Developing the matching high capacity anode for the CoEx cathode and maintaining good electrode integrity
  - Designing anode architecture for desirable power performance

Future Work

- Ongoing (FY 2017)
  - Optimization of CoEx open space regions to enable better high rate performance
  - Speed demonstration of CoEx printing at 400 mm/s
  - Printing of CoEx cathodes for 1 Ah pouch cells
  - Development of high energy anodes for matching thicker CoEx Cathodes
  - 1 & 6 Ah pouch cell testing of baseline anodes
- Proposed (FY 2018)
  - Hardware development for full-width printhead
  - Fabrication of 1-6 Ah pouch cells using CoEx cathodes and high energy anodes (Go/No-Go)

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

Summary

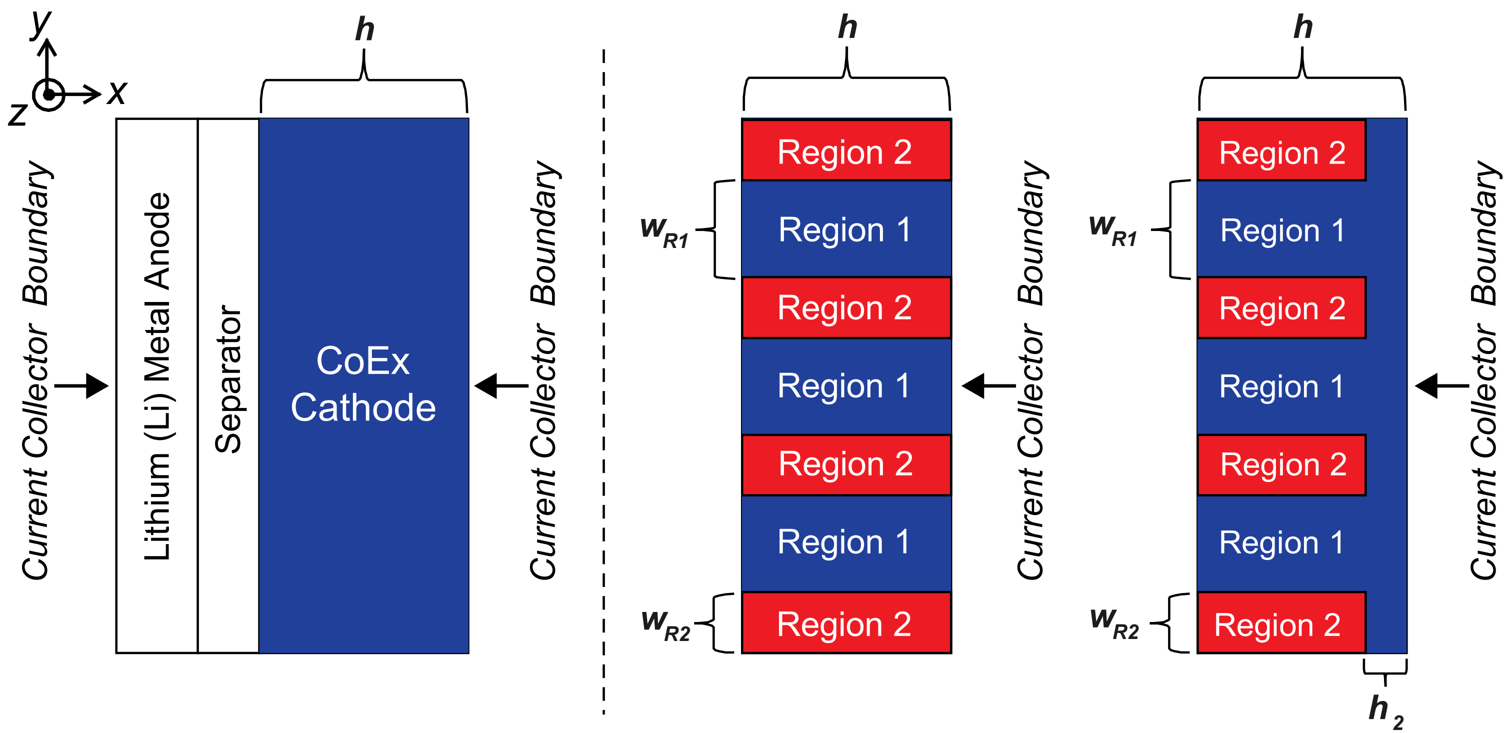
- **Relevance**
  - Demonstrate pilot scale, electric vehicle (EV)–relevant, 14 Ampere hours (Ah) pouch cells via Co-extrusion (CoEx), with goals of 30% reduction in cost and gravimetric energy density improvement of ≥ 20%
- **Approach**
  - Develop thick structured electrodes with CoEx to mitigate power 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**
  - Electrochemical modeling has identified target geometries needed to achieve a minimum of 10% improvement in pack level energy density
  - Initial CoEx structures show substantial improvement in performance compared to unstructured electrodes of comparable loading and moderate improvement in calculated pack level energy density compared to conventional cells
  - A series of graphite materials have been evaluated and downselected to identify anode materials with the best performance at high loading and c-rate
- **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

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%

Half Cell Model Setup in COMSOL



(a) Half Cell Geometry

(b) Cathode Structure for CoEx A

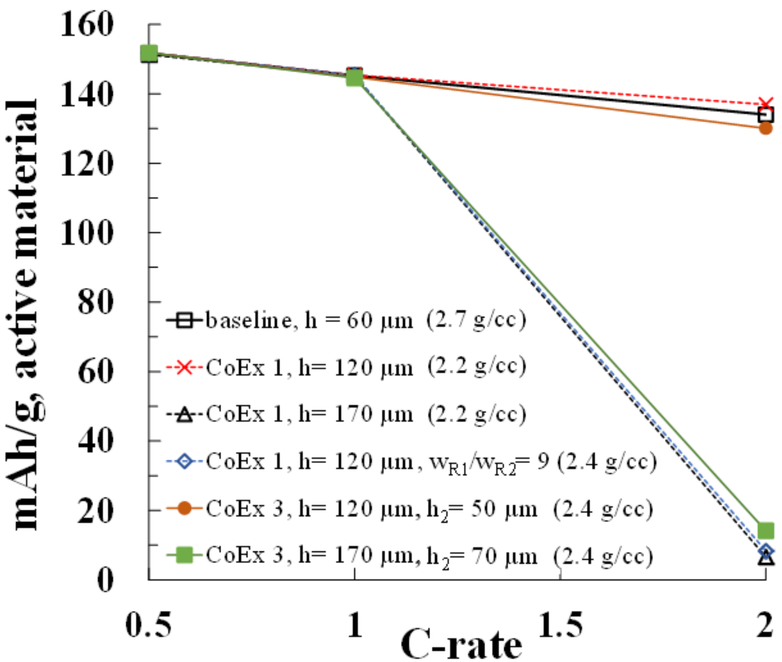
e.g. CoEx 1

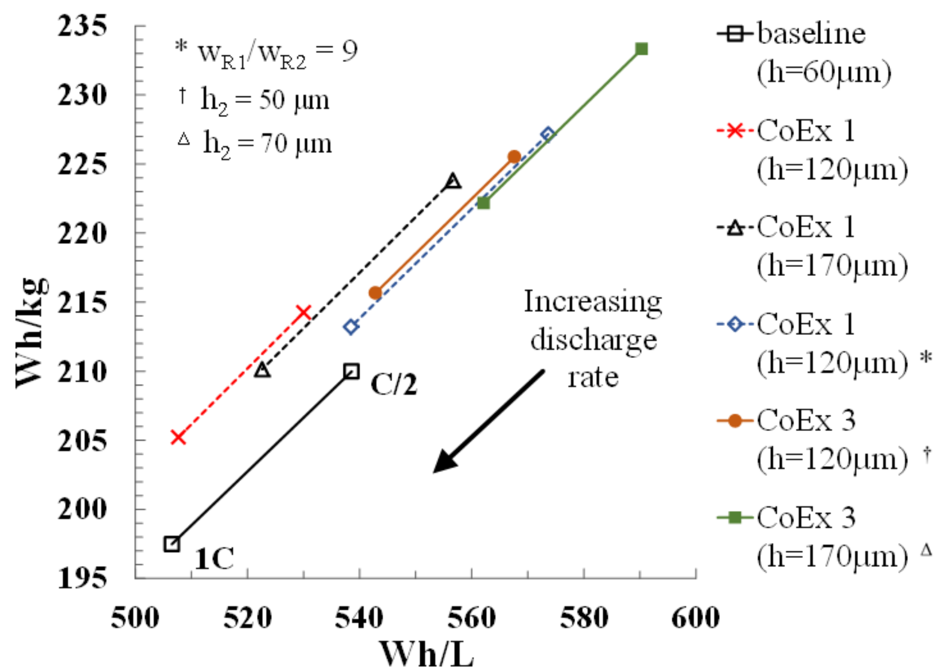
(c) Cathode Structure for CoEx B

e.g. CoEx 3

Modeling Results Summary

w_{R1}/w_{R2} = 4 unless otherwise noted, and the width of the electrolyte region (w_{R2}) is 25 µm





Modeling results show that CoEx 1 and CoEx 3 structures that are 2x the thickness of the conventional baseline have comparable performance, resulting in improvements in energy density at the pack level. CoEx 3 structures are the most promising from a energy and power density perspective.