Electrodeposition for Low-Cost, Water-Based Electrode Manufacturing

Vehicles Technology Office
2020 Annual Merit Review

Project ID: BAT263

Stuart Hellring (PI)

June 2, 2020

Contributors:

PPG- Stuart Hellring (PI), Adam Crowe, Haley Orler, Landon Oakes

ANL- Andrew Jansen, Greg Krumdick, Ozge Kahvecioglu Feridun, Shabbir Ahmed

ORNL- David Wood III, Mengya Li

Navitas- Mike Wixom, Pu Zhang, Robert Sosik

Overview

Timeline

- Project start date: January 1st, 2016
- Project end date: December 31st, 2020
- 100% complete

Barriers

- High material processing cost
- High manufacturing cost
- Toxic material exposure

Budget

- Total project funding:$4,096,903
  - DOE share: $1,399,275
  - FFRDC: $1,600,000
  - Contractor share: $1,097,628
- Project is fully funded.
- Funding for FY 2016: $342,669
- Funding for FY 2017: $1,050,523
- Funding for FY 2018: $537,463
- Funding for FY 2019: $566,248

Collaborating Partners

- Metokote (now PPG)
  - **Role**: Roll-to-roll eCoat design and installation
  - **Project lead:** Dennis Siefer
- Navitas System
  - **Role:** Full-cell build and testing
  - **Project lead:** Mike Wixom
- Oak Ridge National Lab
  - **Role:** Electrode processing and anode support
  - **Project lead:** David Wood III
- Argonne National Lab
  - **Role:** CAD synthesis
  - **Project lead:** Greg Krumdick

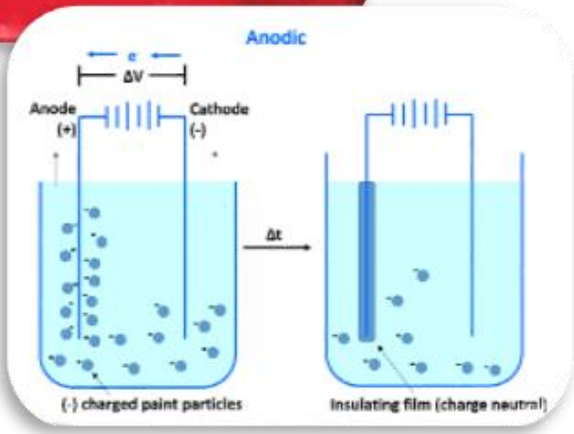
Relevance

Overall Objectives

- Reduce electrode manufacturing cost using electrocoat processing.
- Improve the environmental friendliness with water-based battery processing.

Objectives this Period

- Build and evaluate large format cells employing electrocoated cathode electrodes, compare to baseline slot-die PVDF/NMP controls.
- Identify root cause failure mechanisms for next-generation electrocoat system.



Impact

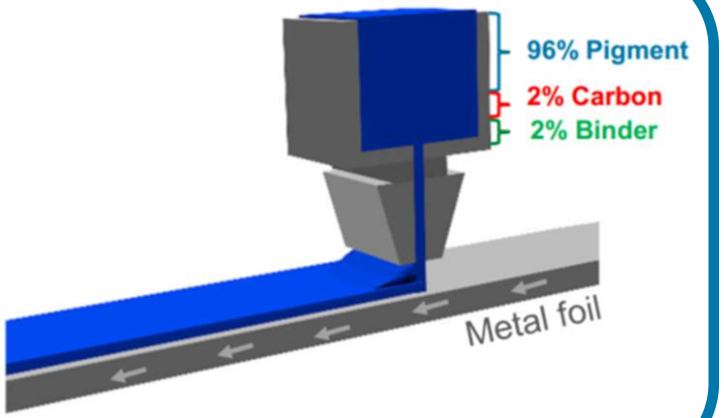
- Successful production of electrocoated cathodes to:
  - Reduce cell manufacturing cost.
  - Enable waterborne manufacturing.
  - Eliminate the need for using toxic solvents.
  - Facilitate automotive OEM and consumer acceptance of electric vehicles.
  - Allow for the creation of the next generation of US-based advanced battery manufacturing.

Milestones

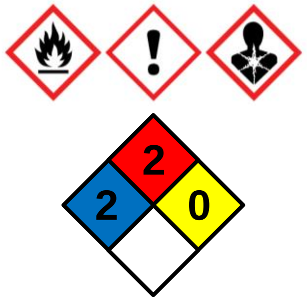
Date	Milestones and Go/No-go	Status
June 2017	**Milestone:** Formulation / application parameters are optimized sufficient to produce an electrode with an energy density of 2.5-3.0 mAh/cm^2	Complete
December 2017	**Milestone:** Pouch cells > 0.2 Ah are tested	Complete
July 2018	**Milestone:** Mini-coater is designed, built, and prepared for operation.	Complete
December 2017	**Milestone:** BatPac model updated and adjusted cost estimate obtained	Complete
December 2017	**Go/No-go:** Demonstrate ability to produce kg quantities of the active material.	Complete
December 2017	**Go/No-go:** Electrodes will either have reached a loading density of 2.0 mAh/cm^2 or a clear path to achieve metric that will be identified.	Complete
December 2018	**Milestone:** Electrodes are produced on the mini-coater that can be used for cell deliverables.	Complete
January 2018	**Milestone:** 12 baseline and 12 electrocoated cathodes will be evaluated in double layer pouch cells	Complete
August 2019	**Milestone:** 35 electrocoat and 12 baseline prismatic cells >1 Ah will be assembled and tested.18 optimized cells will be delivered to DOE for evaluation	Complete
March 2020	**Milestone:** Root cause failure mechanisms identified	Complete

Approach: Electrodeposition to Overcome Current Process Barriers

Current: Slot-Die Coating

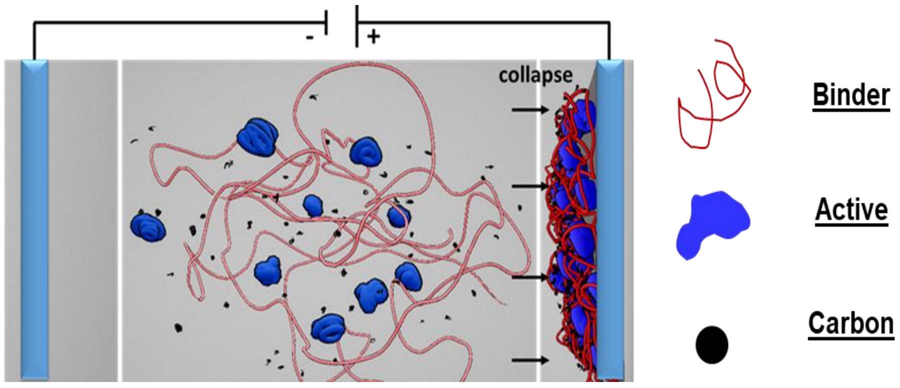


NMP Solvent
GHS Hazards Label



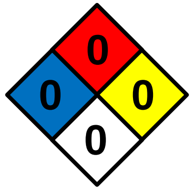
- **2-coat/2-cure process**
- **Costly solvent recovery**

Future: Electrocoat



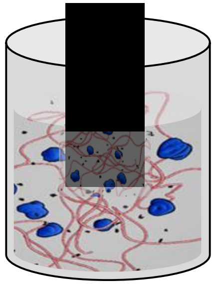
Water
GHS Hazards Label

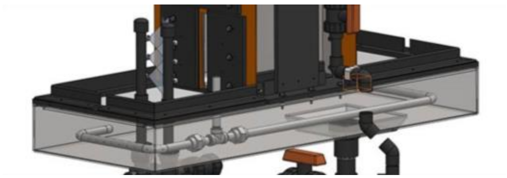
No GHS Warnings



- **Simultaneously coat both sides**
- **Lower drying costs with no LEL limitation**

Approach: *Timeline*





Formulation and application development → **Pilot system construction** → **Cell production / testing**

Year 1	***Year 2***	***Year 3***

Formulation Development

- **Design, synthesize and screen electrocoat chemistries to accomplish:**
  - Stable electrocoat bath with suitable cathode compositions.
  - Composition coalesces into a cathode coating on the current collector.
  - Bench-top, batch scale electrocoat process optimization
  - Robust cathode coating for out-of-cell manipulation.
  - Outstanding performance when operated as a cathode.

Pilot System Design and Production

- **Design, build and install a pilot scale roll-to-roll electrocoat system.**
- **Develop water-based continuous roll-to-roll electrocoat process to demonstrate production of rapid, double-sided coating of uniform cathodes with high areal capacities.**
- **Cost analysis of electrocoat manufacturing based on pilot operation data.**

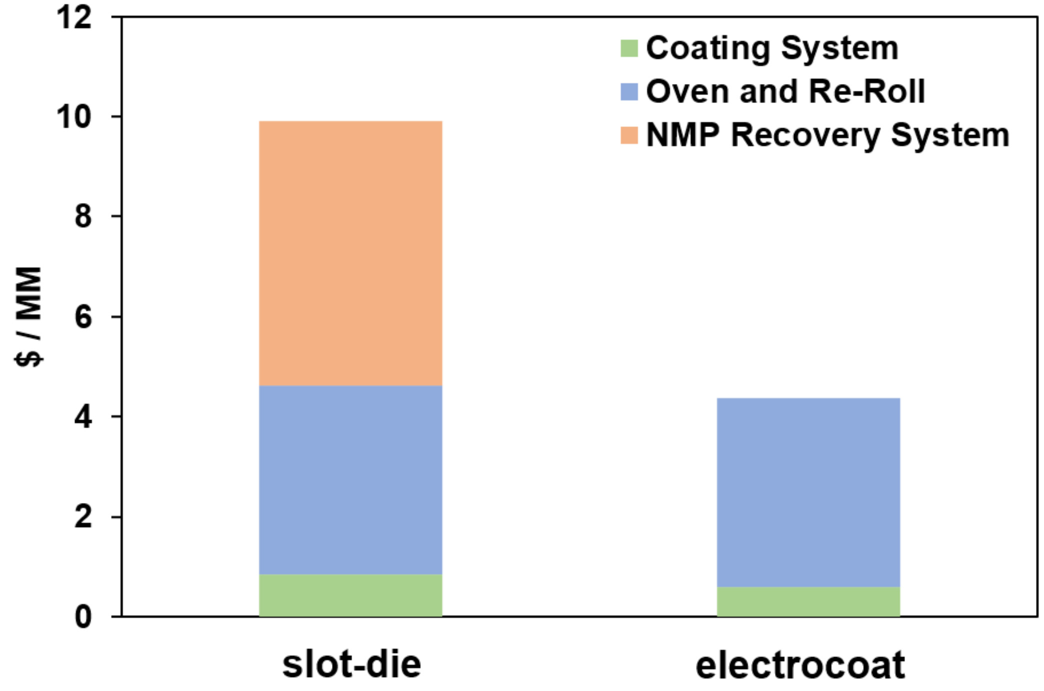
Cell Production / Testing

- **Manufacture large format pouch cells using electrodes coated on the pilot scale coater**

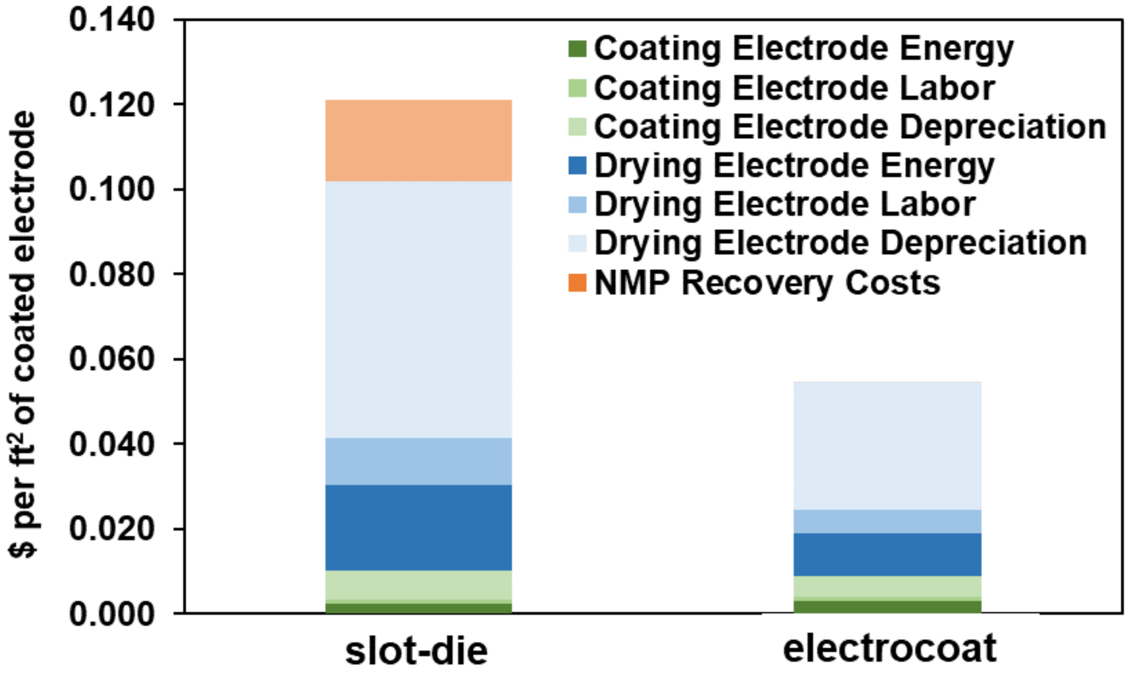
PPG

Accomplishment: Cost Model Suggests Electrodeposition Offers Decreased Capital Cost and Operating Costs

Capital Costs



Operating Costs



60% decrease in capital costs

- Removal of NMP recovery system ($5MM)*
- 30% decrease in coating system

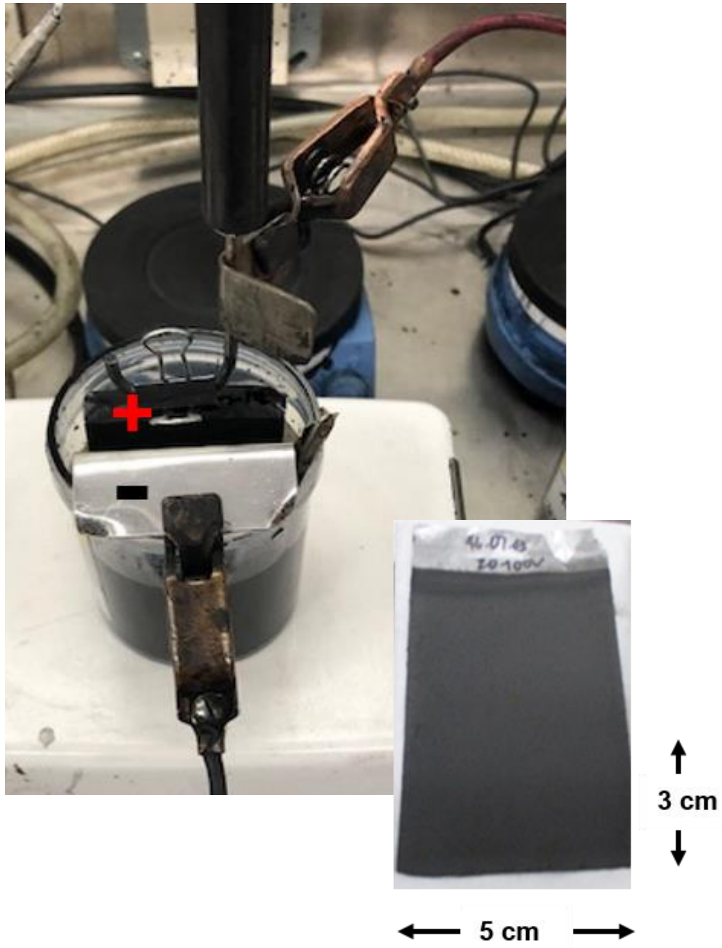
55% decrease in operating costs

- Operating costs savings of $2.36MM/GWh

Wood, D. L. *Drying Technology*, **2018**, *36*(2), 234–244.

Accomplishment: Advancing Electrodeposition from the Benchtop to a Continuous Roll-to-Roll Pilot Scale Coating System

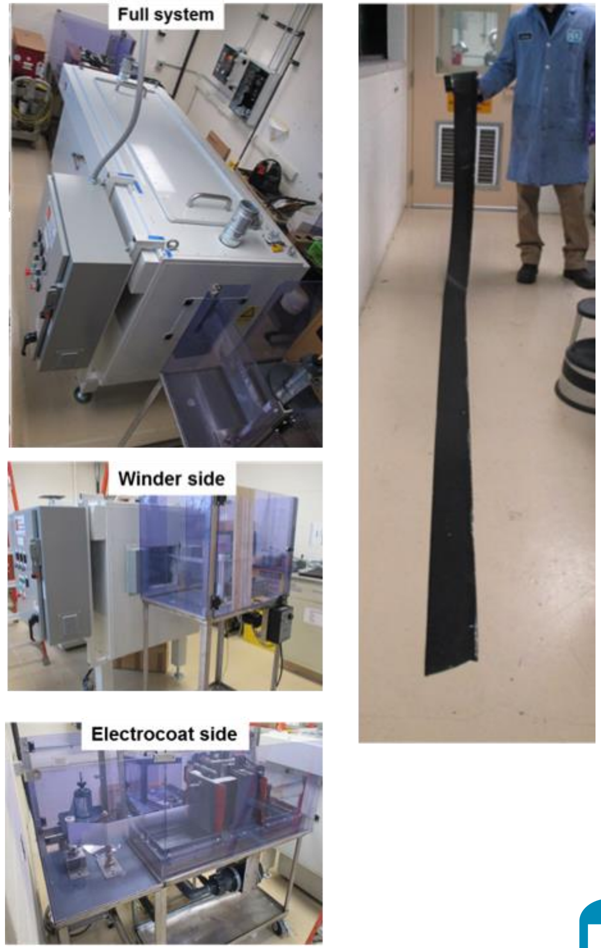
Benchtop



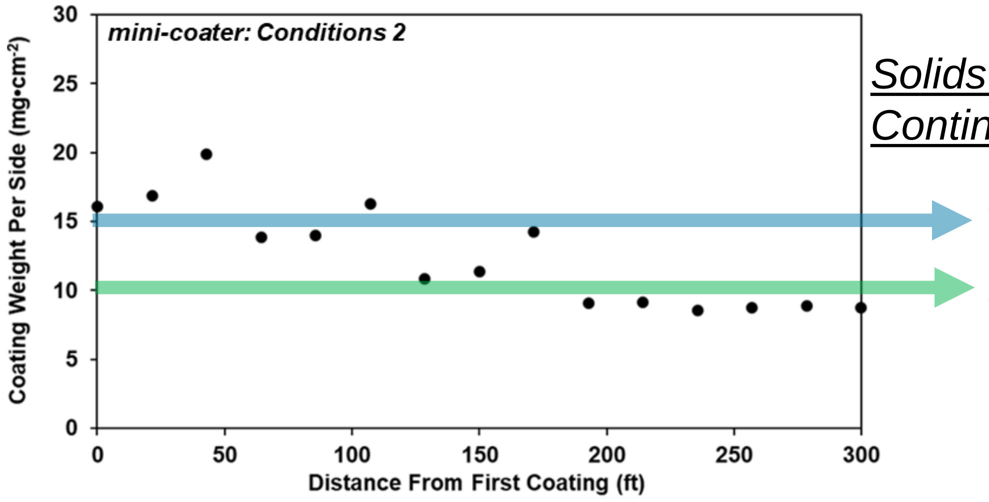
Challenges

- Pilot Coater Design
- Pilot Coater Reformulation
- Electrode Coating Quality
- Cell Performance

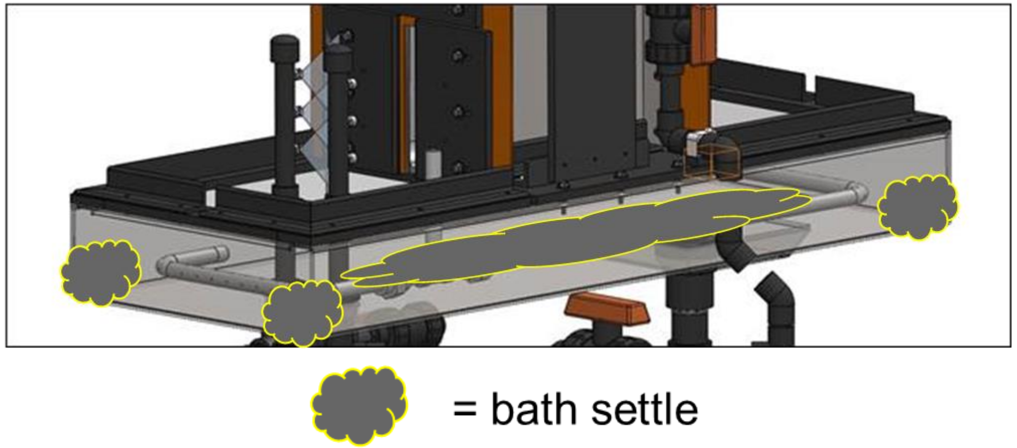
Pilot System



Accomplishment: Root-Cause Analysis of Coating Weight Reduction During Semi-Continuous Operation (300ft)



Solids Depletion Assuming Continuous Operation

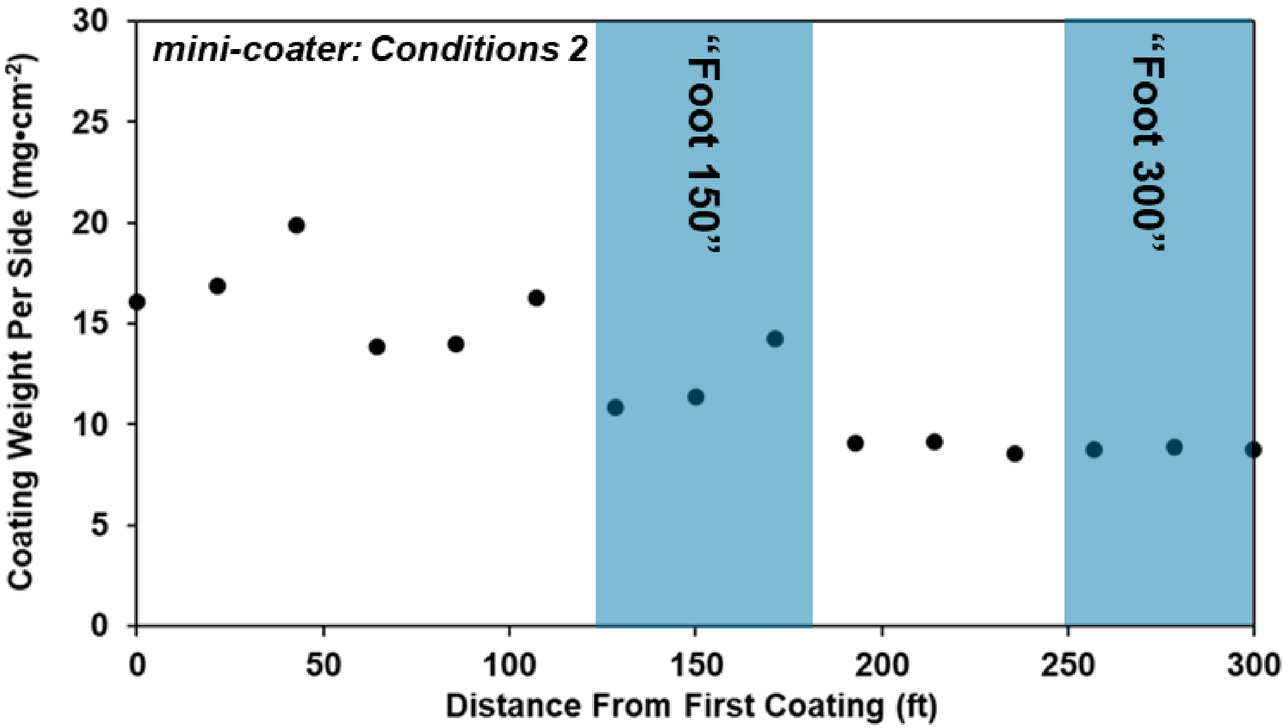


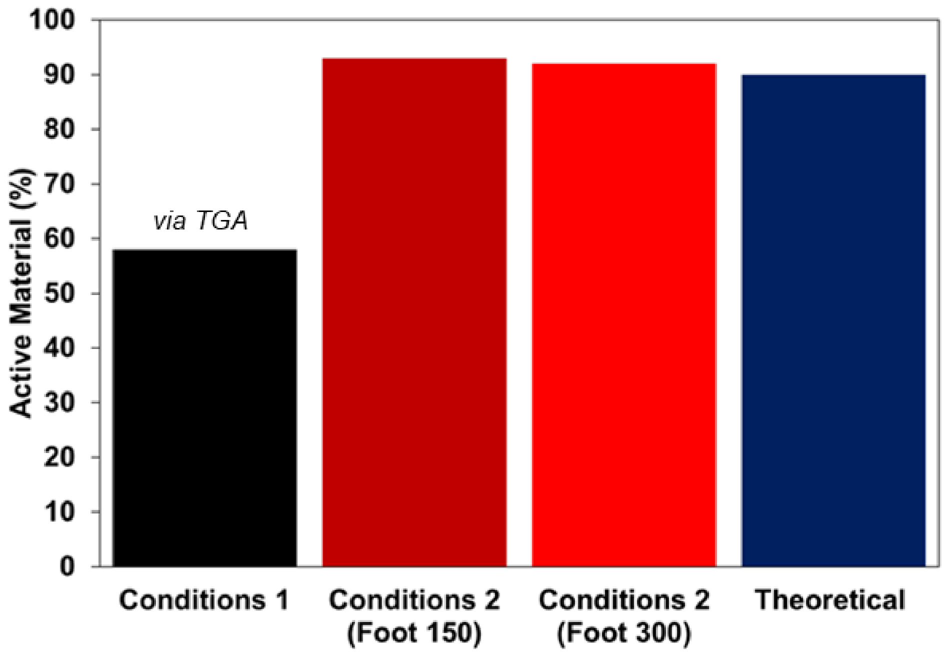
Continuous operation between 10–15 mg•cm^{-2} would deplete 55-82% of solids

- **Stabilization expected at scale with employment of bath replenishment system (commercial electrocoat lines).**

To mitigate settling, any settled material was agitated during the curing of each 8 foot section during semi-continuous operation (oven limitation).

Accomplishment: Cathode Electrode Region Analysis During Extended Operation

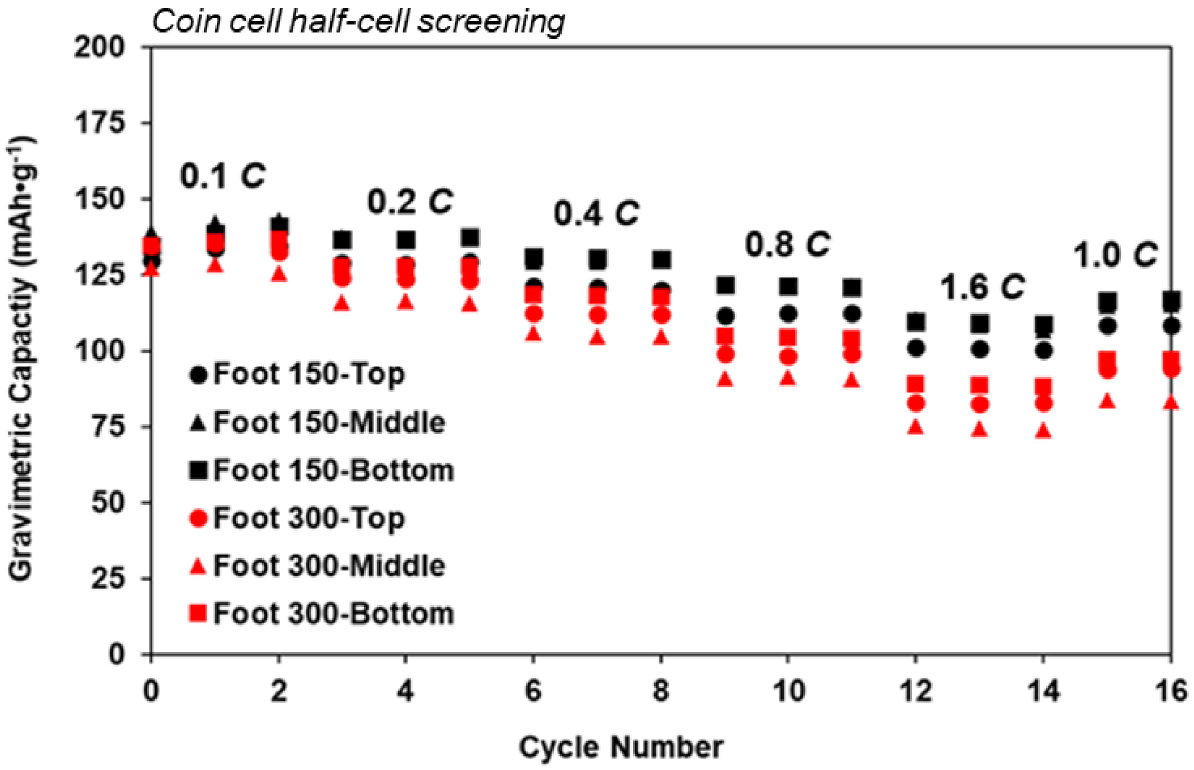


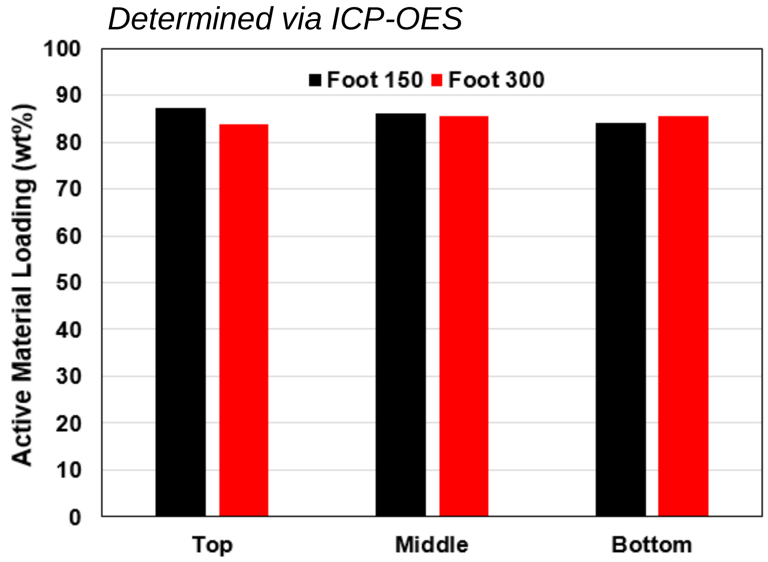


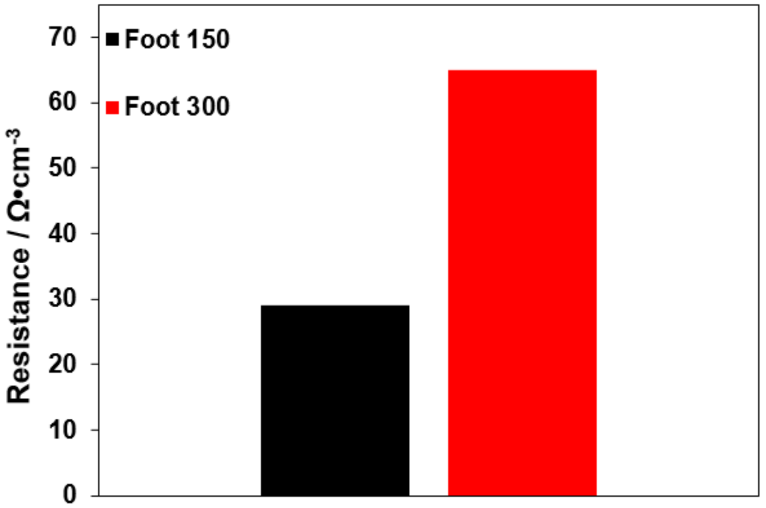
While the film build rate within the mini-coater decreased over time, ICP-OES analysis of the digested coating reveals similar active coating/bath ratio across all electrodes.

Increasing the active coating/bath ratio with Conditions 2 results in improved cell performance.

Accomplishment: Electrochemical Performance of Extended Coating Regions and Top-Middle-Bottom



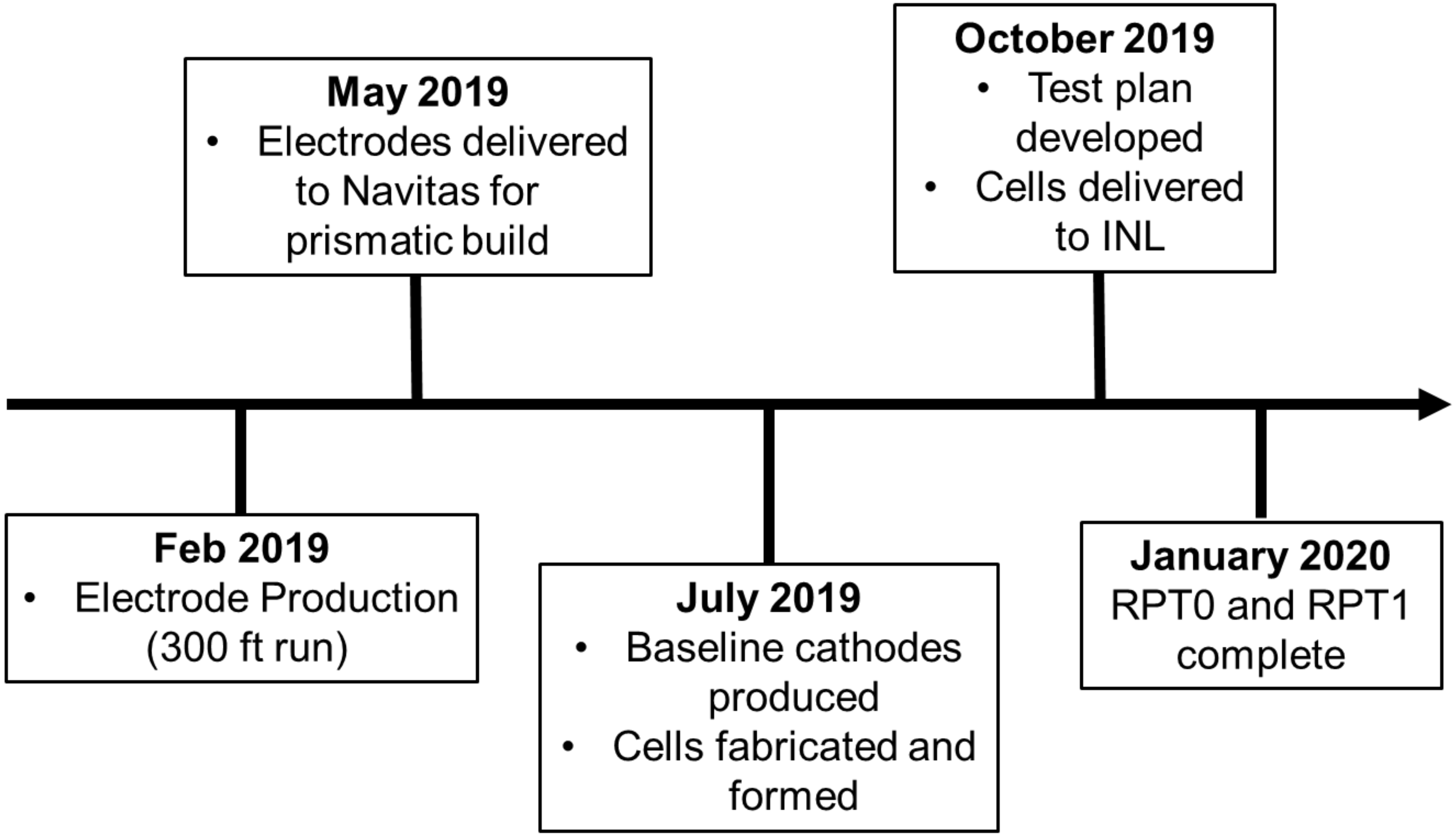




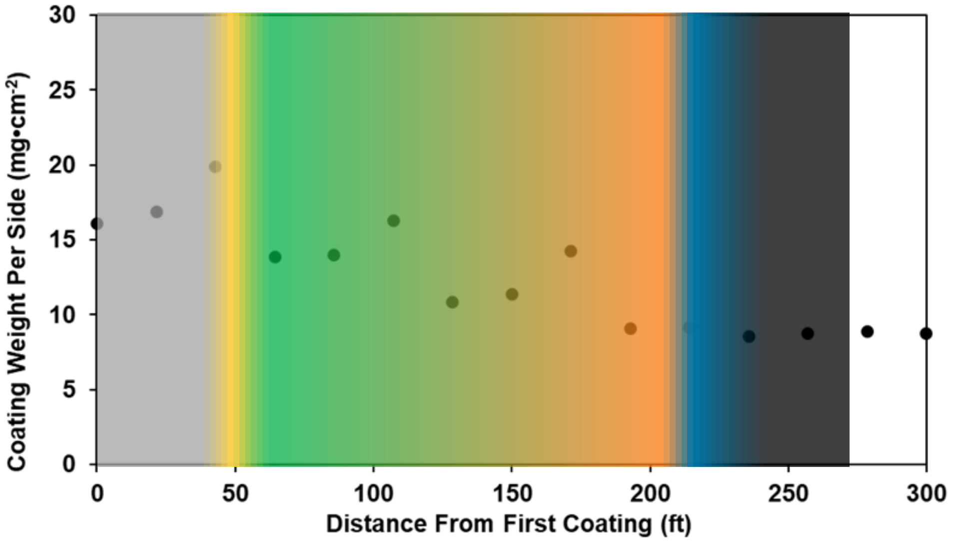
Electrodes prepared near 150ft during a semi-continuous coating session show increased performance over the last group of electrodes.

With similar active material loading (via ICP-OES) a lower through-plane resistance measurement for coatings at 150 feet compared to 300 feet suggest different carbon/binder ratios.

Accomplishment: Timeline for Large-Format Cell Fabrication and Testing



Accomplishment: Electrocoat-based and Baseline Cell Build



Last Coatings (low C) →

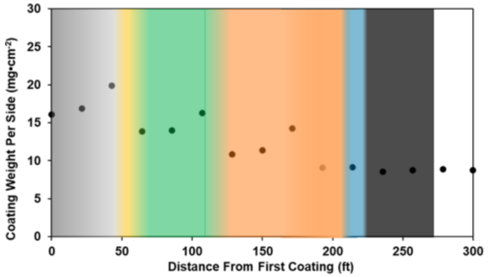
Single cell- poor build (multiple electrode groups) →

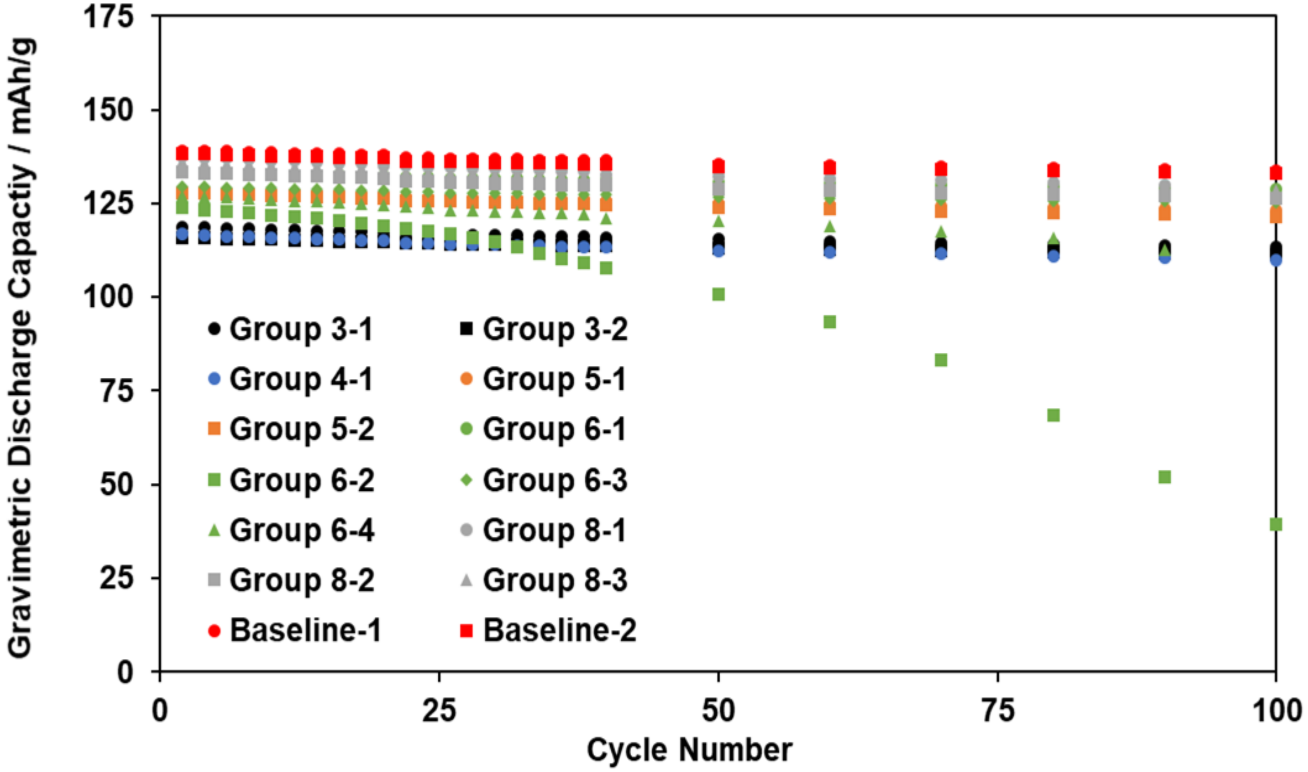
First Coatings (high coating weight) →

	mAh/cm^2	mg/cm^2	cathodes per cell	RPT0 Ah
Group 3	1.55	9.81	7	0.76
Group 4	1.71	10.82	7	0.84
Group 5	1.91	12.09	6	0.87
Group 6	2.03	12.85	6	0.93
Group 7	2.31	14.62	5	-
Group 8	2.99	18.92	4	0.97
Baseline	2.21	13.99	6	1.10

Due to bath depletion and solids settling, electrodes with a range of coating weight were produced, resulting in a complex cell build.

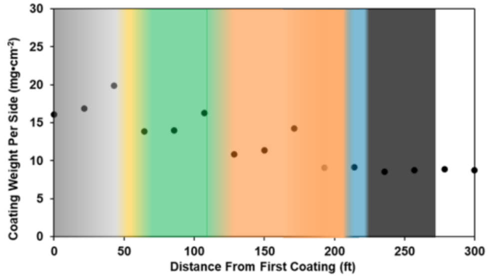
Accomplishment: Large Format Cell Performance: Gravimetric Discharge Capacity

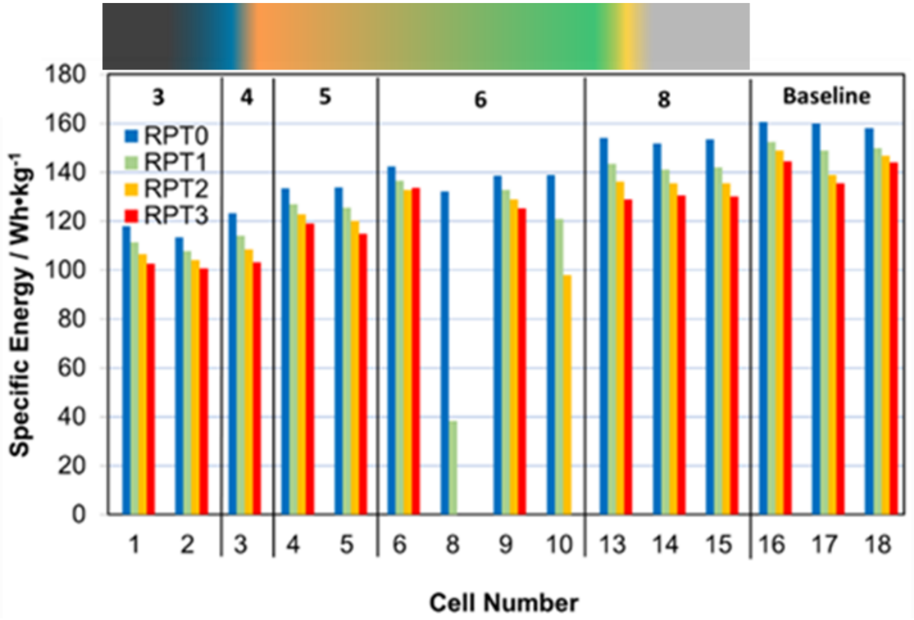


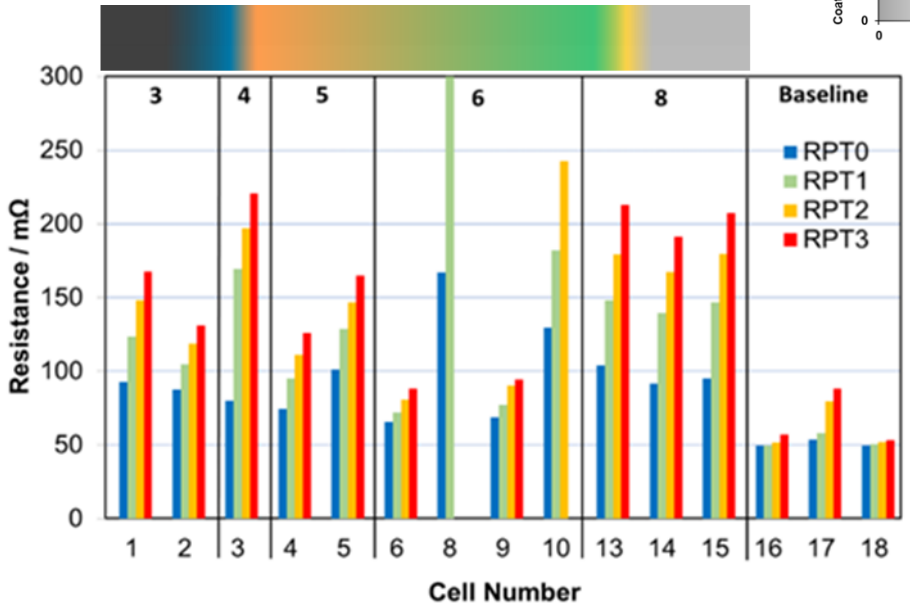


On an average gravimetric basis, electrocoating $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ from a waterborne slurry has the ability to produce high quality electrodes that deliver a C/3 specific capacity within 2.2% of slot-die produced cathode electrodes.

Accomplishment: Specific Energy and Resistance by Cell and Cathode Coating Group







>5 months of ambient storage of NMC 622-based electrodes prior to cell fabrication likely contribute to higher amounts of surface carbonates, relative to the baseline electrodes.

Aqueous processed electrodes (Group 3–8) likely show an oxygen-depleted and more resistive surface layer developed during the coating and/or drying process. In hindsight, the baseline NMC622 (prepared via solventborne slot-die) should have been washed with water prior to mixing.

Shao-Horn, Y. and Gasteiger, H. A. *J. Electrochem. Soc.* **2018**, 165 (*2*), A132-A141
Gasteiger, H *Journal of The Electrochemical Society* **2019**, 166 (*16*), A4056-A4066

Collaboration

Argonne national labs has extensive expertise in active material synthesis and characterization. This expertise was used to prepare initial cathode active materials suitable for electrocoat. Both particle size and particle-coating technologies were explored.

Oak Ridge National Lab has extensive expertise in waterborne cathode coating technology. This expertise was used to address challenges specific to cathodes produced by waterborne electrocoat systems. Unique drying conditions for waterborne cathodes were identified.

Navitas brings commercial insight with extensive experience implementing novel battery technologies and expertise in cell design. This expertise was used to produce and test cells from cathodes made by electrocoat, and make substantive comparisons against state-of-the-art cathode technologies.

Idaho National Lab has expertise in cell validation for electric and hybrid vehicle applications. This expertise was used to benchmark electrocoat produced cathodes against a baseline PVDF/NMP cathode electrode control within large format pouch cells.

2019 AMR Reviewer Comments

Question 1- Reviewer 1 indicated that this project needs more support on modeling to show the economics of it. Response: An internal cost analysis was performed, suggesting a 60% reduction in capital costs and a 55% reduction in operating costs for electrocoat technology relative to a conventional slot-die production of cathode electrodes.

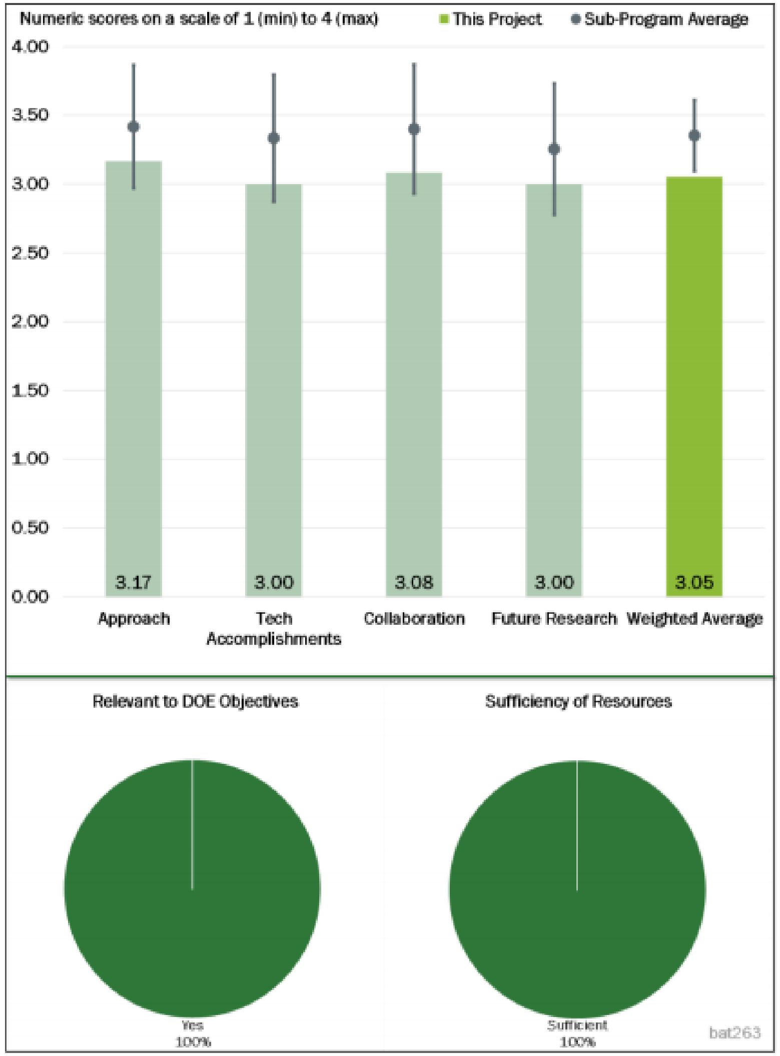
Question 2- Reviewer 5 noted the poor electrochemical performance of electrocoat produced cathodes relative to baseline electrodes. Response: Optimization of bath formulation and coating conditions (Coating Conditions 2) results in a significant increase in electrode performance.

Question 3- All reviewers offered positive comments about collaboration and coordination across project teams.

Question 4- Reviewer 2 stated that effort is needed to evaluate electrode quality comparing it to baseline electrodes. Response: A detailed investigation within 1 Ah pouch cells was employed comparing electrocoat produced cathode electrodes to slot-die produced cathode electrodes from a solventborne slurry (baseline).

Question 5- All reviewers offered positive comments about project relevance to overall DOE objectives.

Question 6- All reviewers commented that the resources are sufficient for project completion.



Remaining Challenges and Barriers

- Electrocoat system design optimization to minimize the solids settling during extended operation.
- Develop bath replenishment system to mitigate coating weight reduction due to coating-based bath depletion.

Proposed Future Research

- Continue to monitor the performance of large format cells that are benchmarking electrocoat produced cathode electrodes against a slot-die produced PVDF/NMP baseline.

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

Summary

- The viability of electrocoat to fabricate battery electrodes at commercially relevant formulations has been demonstrated using both bench-scale equipment and a small pilot-scale roll-to-roll system (mini-coater).

- Electrodeposited lithium-ion battery electrode coating system and manufacturing process is capable of reducing cell costs (>60% decrease in capital costs and >50% reduction in operating costs).

- Electrocoat technology has the ability to produce high quality electrodes that deliver a C/3 specific capacity within 2.2% of slot-die produced cathode electrodes from a solventborne slurry.

Technical Back-up Slides