Electrodeposition for Low-Cost, Water-Based Electrode Manufacturing

Vehicles Technology Office
2017 Annual Merit Review

Project ID: ES263

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Overview

Timeline

- Project start date: January 1st, 2016
- Project end date: December 31st, 2018
- 33% complete

Barriers

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

Budget

- Total project funding:$3,999,034
  - DOE share: $1,399,275
  - FFRDC: $1,600,000
  - Contractor share: $999,759
- Project is fully funded.

Partners

- Argonne National Lab
- Oak Ridge National Lab
- Navitas System

Relevance

Objectives

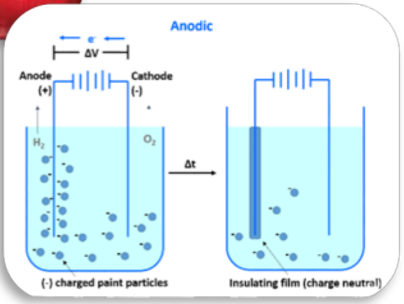
- Reduce the processing cost of electrode manufacturing using electrocoat processing.
- Improve the environmental friendliness of battery processing using water-based electrocoat.

Objectives this Period

- Design and synthesize flexible binders that electrodeposit.
- Explore high energy density cathode materials for these binders
- Formulate water-based cathode coating systems.
- Demonstrate electrochemical storage for coated cathodes

Impact

- Successful electrocoated cathodes reduce cell cost by at least 20%. Adoption of electrocoat into automotive battery supply chain will facility automotive OEM and consumer acceptance of electric vehicles and allow for the creation of the next generation of US advanced battery manufacturing.



Milestones

Budget Period	Task	Description	Target	Status
1. Materials Development	1	Electrochemical Performance Demonstrated	3/31/16	Complete
	1	Active Material Identified	9/30/16	Complete
	1	Candidate Resin Identified	12/30/16	Complete
	2	Development Process Established	6/30/16	Complete
2. Process Development	3	Parameters optimized	6/30/17	BP2
	3	Cell Testing Complete	9/29/17	BP2
	4	Mini-Coater Built	12/29/17	BP2
	5	Cost Estimate Updated	3/31/17	BP2
3. Scale-up and Demo.	6	Electrodes Produced	3/30/18	BP3
	7	Build 1 Complete	6/29/18	BP3
	7	Build 2 Complete	9/28/18	BP3
	7	Failure Mechanisms Identified	12/31/18	BP3

1. Approach: Manufacturing Process Design

Eliminate Toxic Solvent Exposure Costs

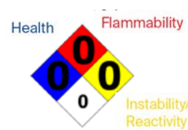
NMP Solvent – GHS Hazards Label

Signal Word: WARNING!
Hazard Statements:
- H227 - Combustible liquid and vapor.
- H316 - Causes mild skin irritation.
- H320 - Causes eye irritation.
- H335 - May cause respiratory irritation.
- H360 - May damage fertility or the unborn child.

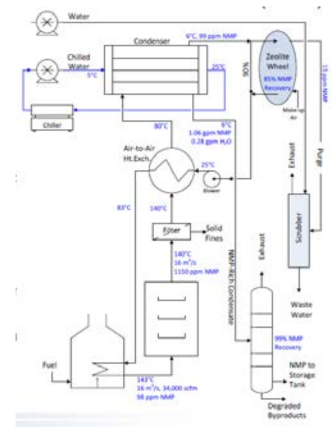
Water – GHS Hazards Label

No GHS Warnings

Health	0
Flammability	0
Physical Hazard	0
Personal Protection	X

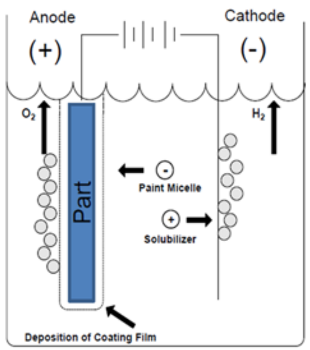


Eliminate Toxic Solvent Recovery Costs



NMP Recovery Process
Shabbir Ahmed, ANL
2015 VTO AMR ES228

Reduce Drying Costs



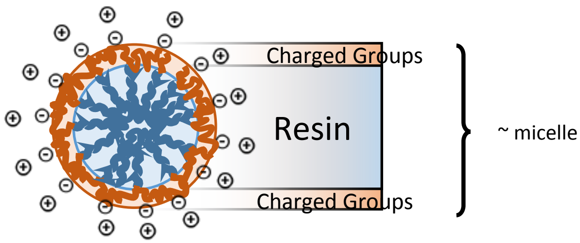
Wet Film going into oven –
- High solids
- Low solvent
- Low VOC
- No LEL limitation

Eliminate 2-Step Coating Process

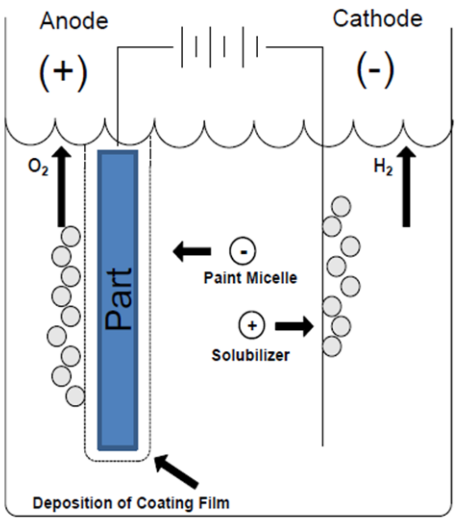
Electrocoat both sides simultaneously -
- One pass through oven
- Resistance controls uniformity
- Particle assembly controls porosity

2. Approach: Electrocoat System Design

- **Design, synthesize and screen electrocoat resin chemistries for the following properties:**
  - Stable bath with suitable binder compositions for cathode films (< 5 weight %).
  - Coalesce into a porous and conductive film at the surface of the current collector.
  - Robust cathode coating for out-of-cell manipulation
  - Outstanding performance when operated as a cathode.



- **Develop electrocoat process parameters to perform rapid, double-sided coating of uniform films with high areal capacities using water-based processing.**



- **Design and manufacture a pilot scale roll-to-roll electrocoat system.**

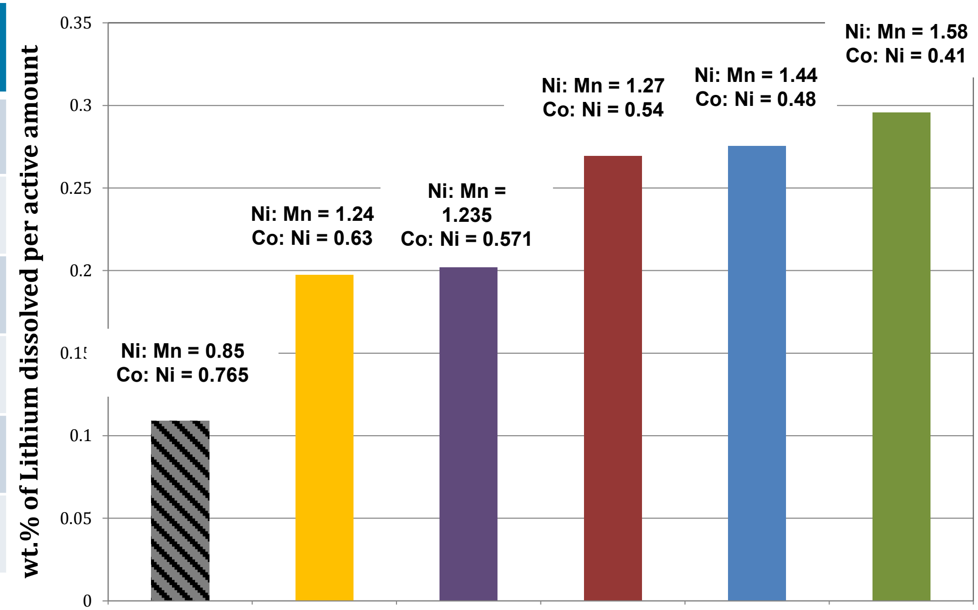
- **Demonstrate a battery supply chain model that mirrors traditional automotive Original Equipment Manufacturer (OEM) supply chain models to reduce the risk of electric vehicles (EVs) and increase adoption.**

1. Technical Accomplishments: Cathode Active Material Synthesis

- Lithium leaching from NMC in water varies with source
- Some commercial materials show consistent performance before and after water soaking

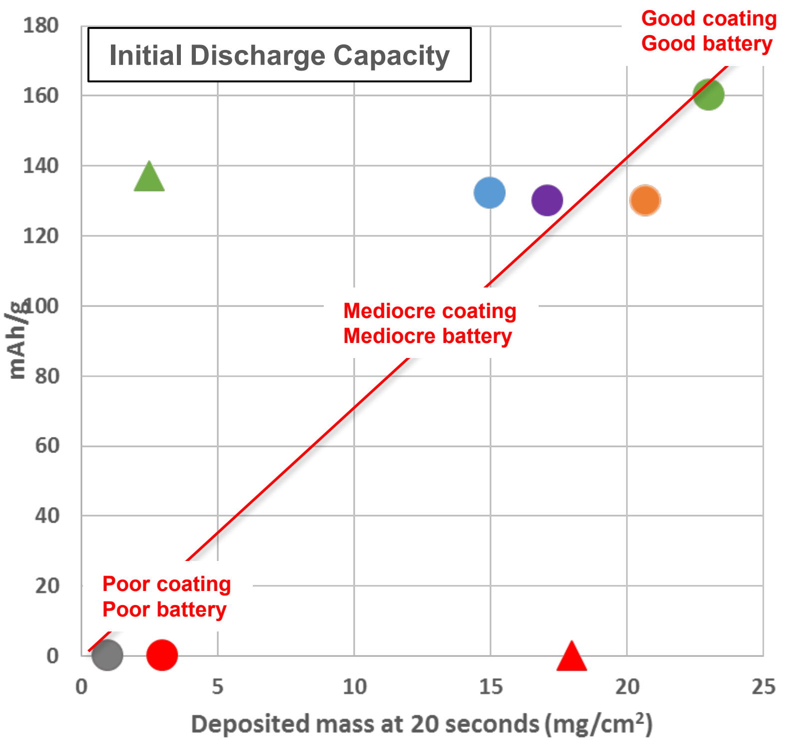
Type	Source	Particle Size	Soaking	Specific Charge Capacity	Specific Discharge Capacity	Coulombic Efficiency
NMC-532	Commercial	6 µm	Before	188	168	89%
NMC-532	Commercial	6 µm	After	189	170	91%
NMC-532	TVR	5 µm	Before	183	175	91%
NMC-532	TVR	5 µm	After	170	147	89%
NMC-532	TVR	6 µm	Before	180	166	92%
NMC-532	TVR	6 µm	After	170	131	77%

Lithium leaching increases with nickel content

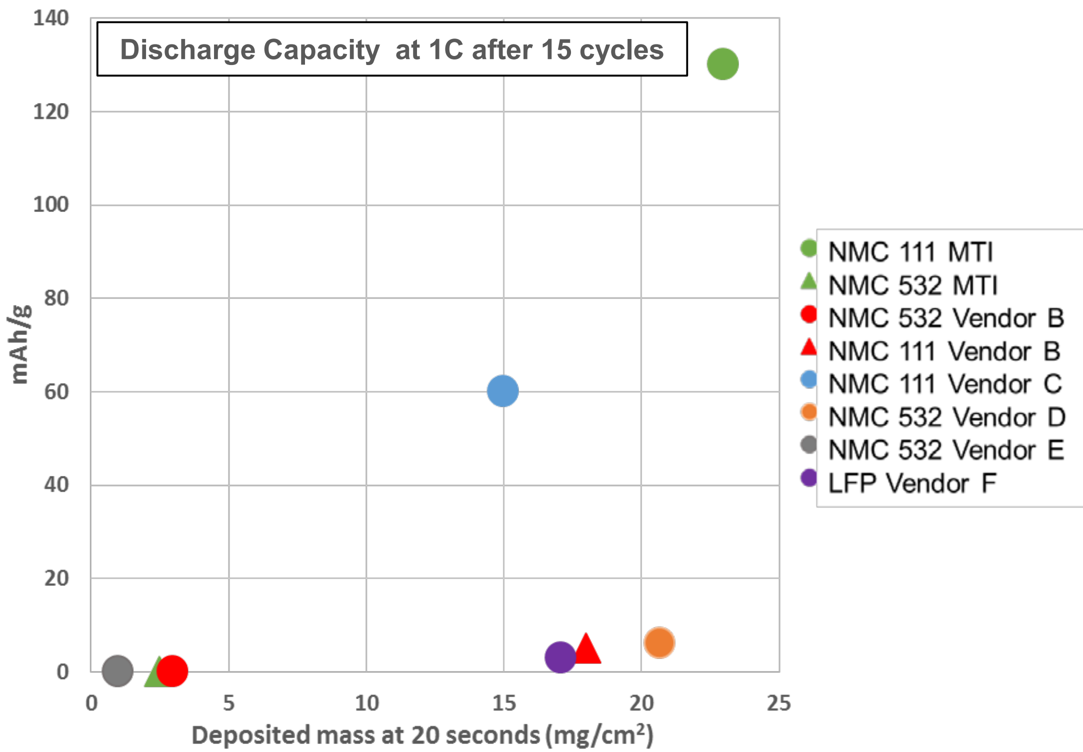


2. Technical Accomplishments: Commercial Material Screening

Initial storage capacity and electrocoat deposition rate depend on NMC composition and commercial supplier



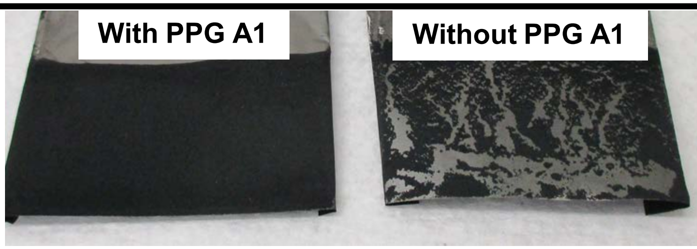
Storage stability after only 15 cycles reveals one commercial NMC is viable

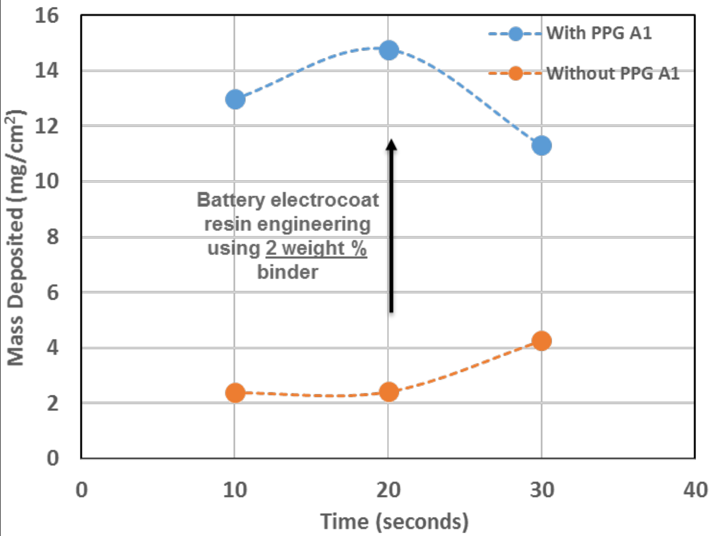


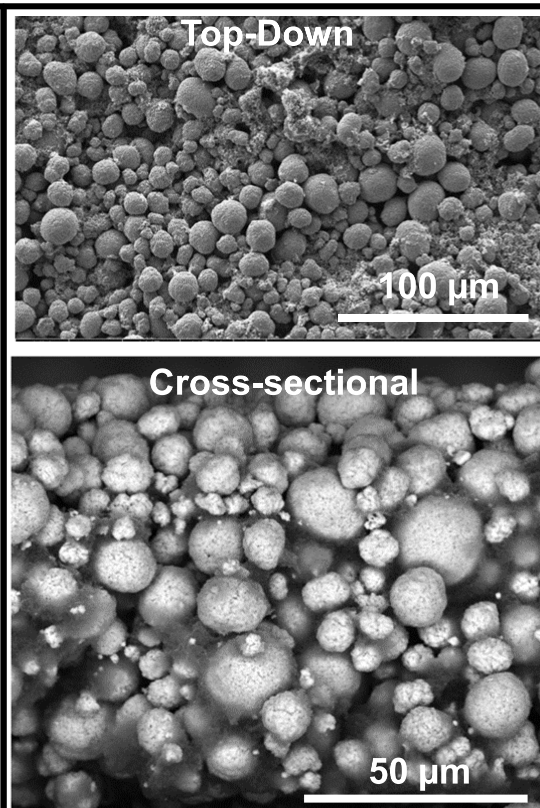
3. Technical Accomplishments: Coating System Development

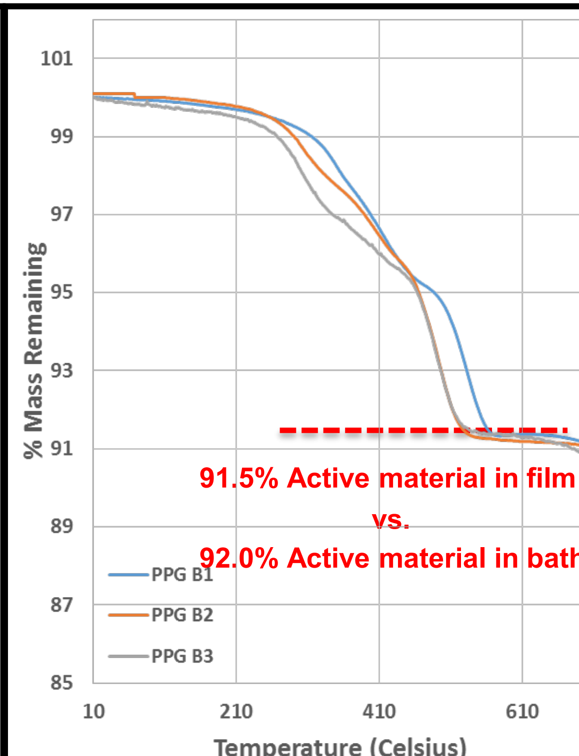
High deposition rates were achieved at battery-relevant binder compositions

TGA and SEM analysis indicate that the film composition is directly controlled by the bath composition









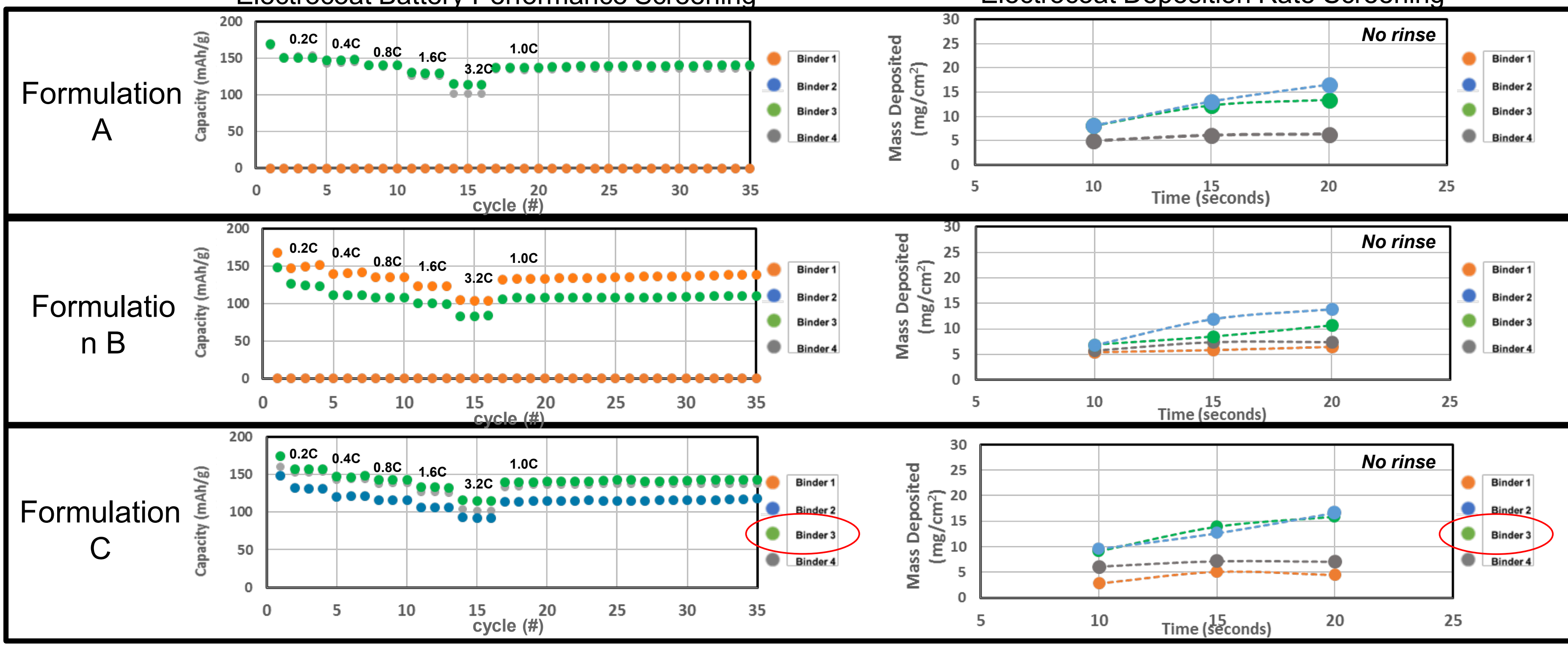
4. Technical Accomplishments: Coating System Development

Electrocoat resins were screened for the ability to both electrocoat and perform as a cathode

Binder 3 in Formulation C was down-selected from a pool of 12 unique binder and formulation combinations

Electrocoat Battery Performance Screening

Electrocoat Deposition Rate Screening

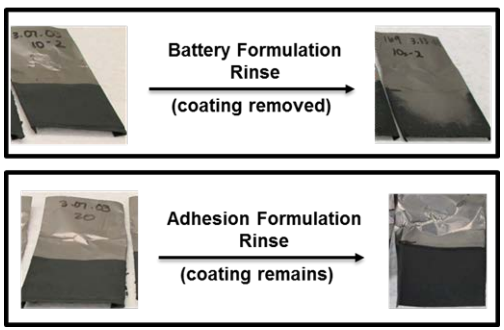


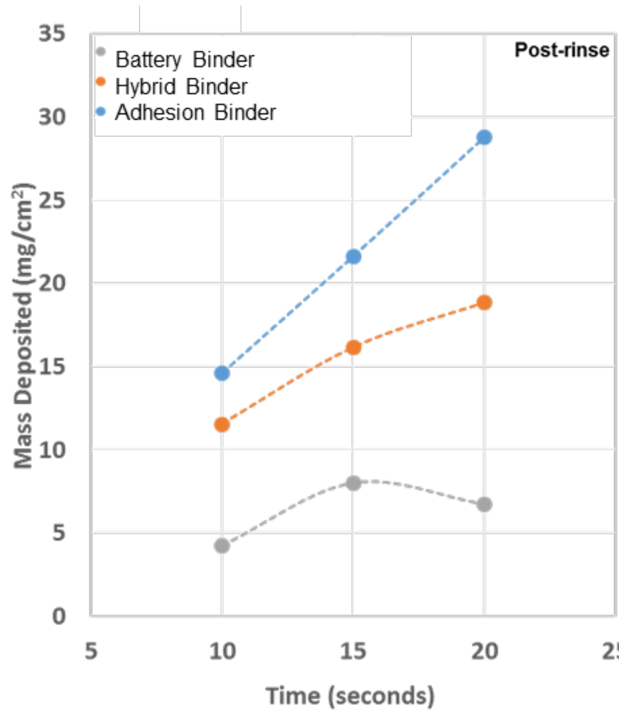
5. Technical Accomplishments: Coating Process Development

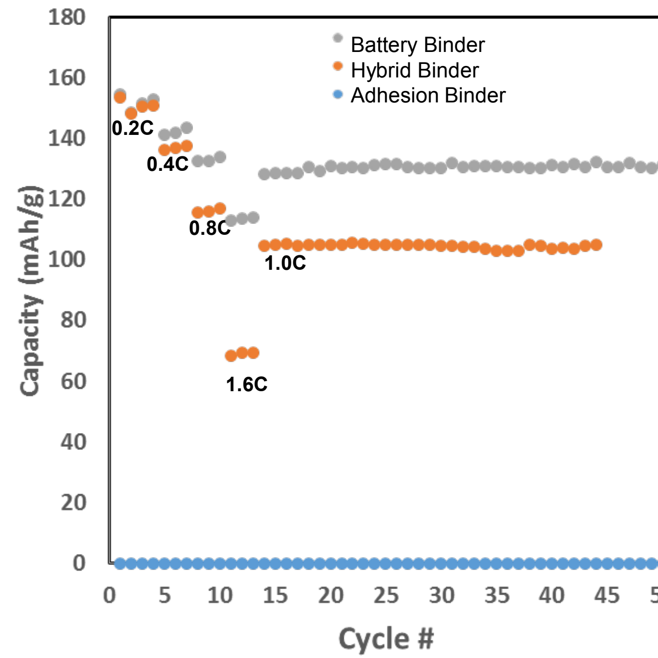
Electrocoat requires sufficient wet-adhesion to endure a rinsing step

Binders were synthesized that both deposit and adhere during the rinse step

Currently focused on mitigating capacity loss from these adhesion formulations



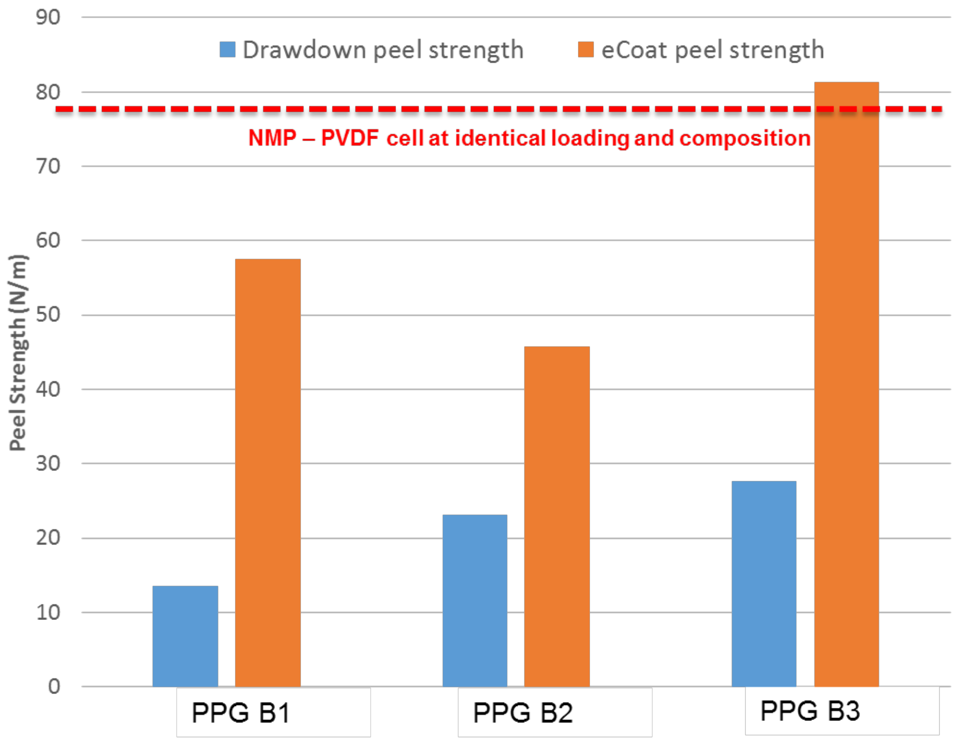


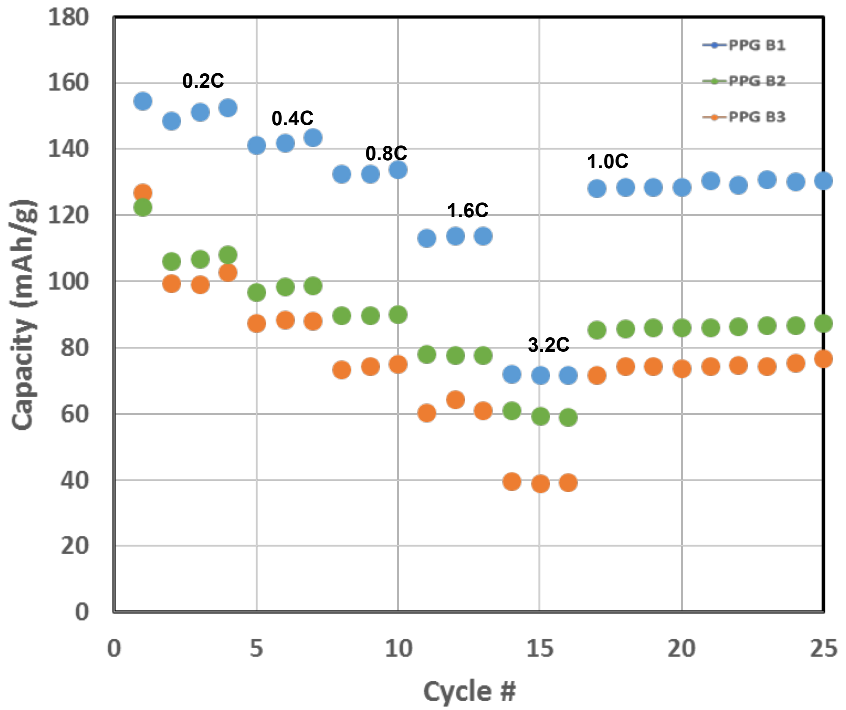


6. Technical Accomplishments: Coating System Development

Electrocoat increases adhesion of cathode compared to identically formulated films applied by drawdown

- Many binder chemistries which possess exceptional adhesion suffer a drawback in battery performance
- Resin was identified which demonstrated both adhesion and battery performance

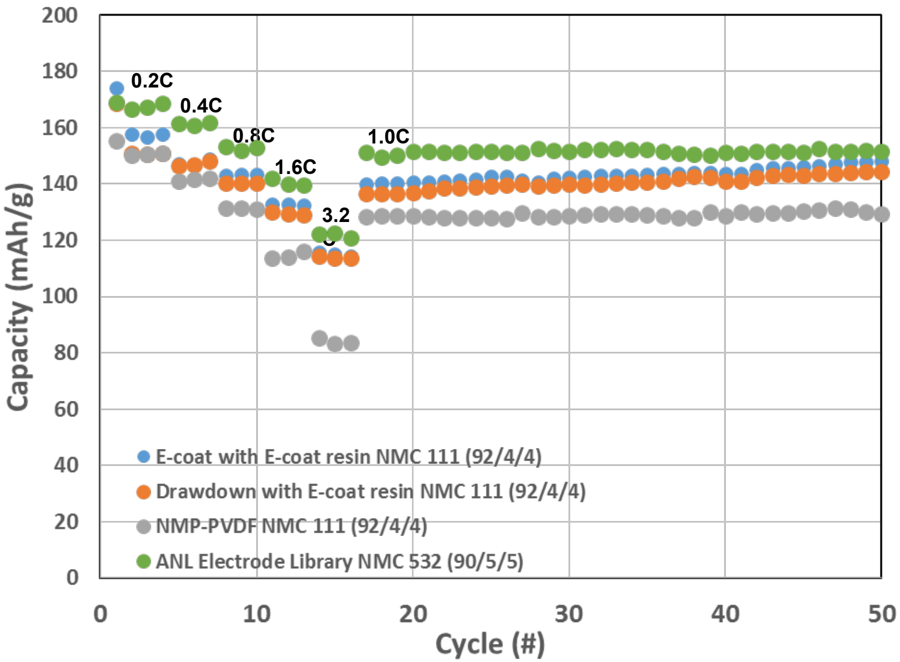


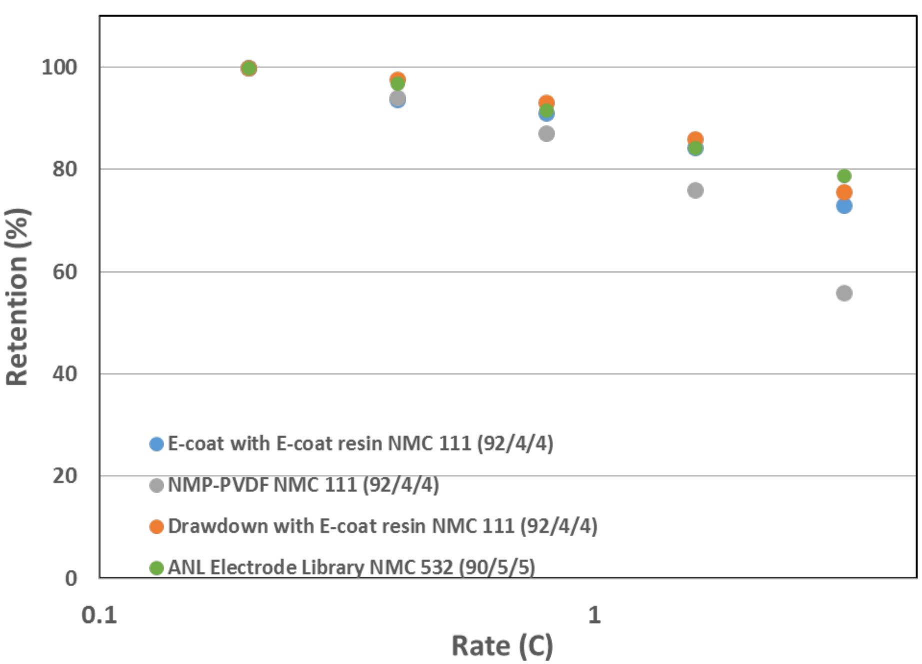


7. Technical Accomplishments: Battery Performance of Electrocoat Cathodes

- Battery performance of cathode coatings applied by electrocoat indicate no loss in capacity or cycleability due to the process of electrocoat
- Similar battery performance is achieved between electrocoat and conventional electrode manufacturing processes

- Improved rate performance compared to NMP-PVDF cells of identical composition and formulation is observed
- Comparable rate performance to NMC 532 materials obtained from the ANL electrode library is also observed





Response to Reviewer Comments

Comments from 2016 AMR	Response to Comments
"The reviewer expressed concern about the deposition of multiple materials to create working electrodes ever working well."	This is a very important to point to highlight and is critical to the ability to controllably deposit battery coatings of known composition. In this budget period, we have carried out extensive work to verify that the composition of the bath is identical to the composition of the deposited film. TGA analysis has conclusively verified that the amount of active material may be reliably engineered through tuning of the bath composition (TA 3) . Future efforts in this regard will be made to distinguish between binder materials and carbon materials which decompose at similar temperatures.
"The reviewer said that it is important to assess the chemical/electrochemical stability of the resins (binders) in the battery electrolytes; the electrochemical stability of the substrate (the reviewer wondered if this is Al) at these high deposition potentials (and in aqueous medium); the effects of gas evolution during deposition; the effects of residual water, if any; and demonstrate the feasibility of coating both sides simultaneously. The reviewer also commented that a preliminary cost estimate needs to be made to make the argument that this is indeed economical and will lead to a noticeable reduction in battery cost, which is one of DOE's goals."	The reviewer has addressed a number of excellent points in this comment that we will address individually - - The chemical/electrochemical stability of the resins was identical to the stability of traditional NMP-PVDF systems in half cell configurations. Long term stability will be investigated this budget period using large format full-cell, pouch-cells. - The electrochemical stability of carbon-coated aluminum substrate is adequate in half-cell, coin-cells. - The effects of residual water content can play a critical role in battery degradation and will be a main focus of the investigations to be performed by ORNL over this upcoming budget period. - Dual-sided coating is commonly done with these systems and we have produced a number of double-sided coatings on the lab scale. The data presented in this update is for single-sided coatings to enable the production of coin cells from the foils, however, the upcoming budget period will transition to double-layer pouch cells in which double-sided coatings will be used. - Preliminary cost models for the implementation of water as the solvent predict significant cost reductions approaching 20%. Future cost modeling will rely on characterizations of the electrocoat process on the large scale which is a focus of this upcoming budget period.
"The PI should put more attention to the feasibility evaluation, e.g., can the uniform coating be achieved at a practically adequate rate."	- The coating deposition rate is directly tunable through process parameters such as the applied voltage, bath conductivity, and bath temperature. In budget period 1, thick films exceeding the current state of the art were deposited with acceptable adhesion at these thicknesses. In the upcoming budget period, the ability to produce these coatings using a continuous roll-to-roll process will be a primary focus.

Collaboration with Others

Team Member	Role	Significance
PPG	Coating system and manufacturing process development	E-coat commercialization expertise coupled with automotive manufacturing relationships will drive adoption by battery manufacturers
Argonne	Active materials development	Custom active materials enable the development of the coating system as well as optimize the performance of resulting electrodes
ORNL	Aqueous coatings development expertise	Challenges unique to aqueous formulations will be identified and addressed
Navitas	Cell build and testing, manufacturing and commercial insight	Experience in implementing novel technologies to meet specific customer requirements will align technology with battery needs and overcome implementation barriers

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Remaining Challenges and Barriers

Active material

- Compatibility differences of commercially available NMC materials is not understood.
- Methods to improve the compatibility of higher nickel content NMC are needed.

Film uniformity

- Process development for uniform, double-sided coating.

Scale-up production to large format cells

- Lab-scale to pilot-scale coating requires new design.

Supply chain

- Supplying a fully formulated solution is disruptive to the current battery supply chain.

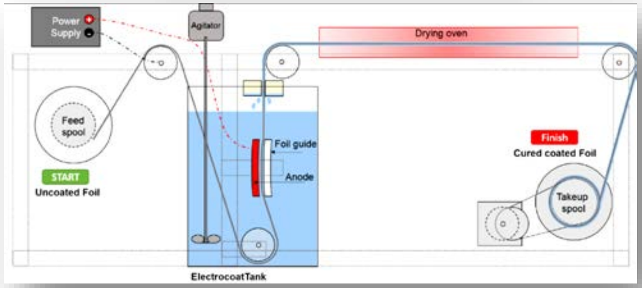
Future Work

Remainder of FY17

- **Coating system formulation and application refinements:**
  - Transition from lab-scale, batch coating processes to a continuous roll-to-roll design.
  - Coat large format substrates and achieve film uniformity enabling transition from coin cell to pouch cell level testing
- **Cathode film and related battery performance:**
  - Thorough investigation related to the mechanism of variable battery performance between commercially available active materials.
  - Investigate the drying processes and the effect of residual moisture on cell performance.

Into FY18

- **Coating system formulation and application refinements:**
  - Refine the coating process parameters on the roll-to-roll system to coat thick, uniform films over large area.
  - Characterize large format cells (> 1Ah) produced using the continuous roll-to-roll process at loading densities ranging between 1-3 mAh/cm^2.
- **Cathode film and related battery performance:**
  - Broaden the available active materials to include high energy density materials.
  - Achieve sufficient film adhesion with high areal loadings (>1 Ah) to compete with traditional NMP-PVDF processes.



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

Summary

Electrocoat is a viable method for assembling cathode films comprised of Ni-rich active materials

- The process rapidly coats films with high areal loading using as little as 2 weight % binder
- Composition can be controlled through resin and bath parameters
- Electrocoat with LFP or Ni-rich active
- Exceptional adhesion for coatings with a thickness as high as 30 mg/cm^2

Commercially available active materials are chemically stable in the electrocoat bath and retain excellent energy storage capabilities after coating

- Electrocoated cathodes exhibit comparable performance to cells fabricated using traditional NMP-PVDF processing methods