

# **Vehicles Technology Office 2016 Annual Merit Review**

## **Electrodeposition for Low-Cost, Water-Based Electrode Manufacturing**

**Stuart Hellring (PI)**  
**June 9, 2016**

### **Contributors:**

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**ORNL - David Wood III**

**Navitas – Mike Wixom, Pu Zhang**

**Project ID: ES263**

# Overview

## Timeline

- January 1, 2016
- December 31, 2018
- Eight Percent Complete

## Budget

- Total Funding: \$3,999,034
- DOE Share: \$1,399,275
- FFRDC: \$1,600,000
- Cost Share: \$999,759
- 2015 DOE Funding: \$0
- 2016 DOE Funding: \$437,430

## Barriers

- High material processing costs
- High manufacturing costs

## Partners

- Argonne National Lab
- Oak Ridge National Lab
- Navitas Systems

# Relevance

## Advances in Electrode and Cell Fabrication Mfg.

**Current:** NMP-based slot-die coating system

**Proposed:** Efficient water-based electrocoat system

### Project Objectives

- Utilize Advanced Electrodeposition Coating Materials and Application Process to Produce Battery Electrodes

### Materials

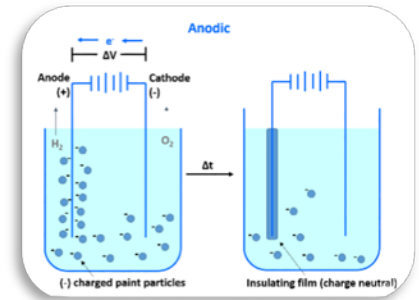
- Novel binders and cathode active materials

### Electrocoat Application

- Improve battery performance
- Reduce cell costs by at least 20%

### Supply Chain Model

- Mirror traditional automotive OEM supply chain
- Reduce risk and increases adoption



# Relevance

## Leveraging the Advantages of Electrocoat

### Formulation

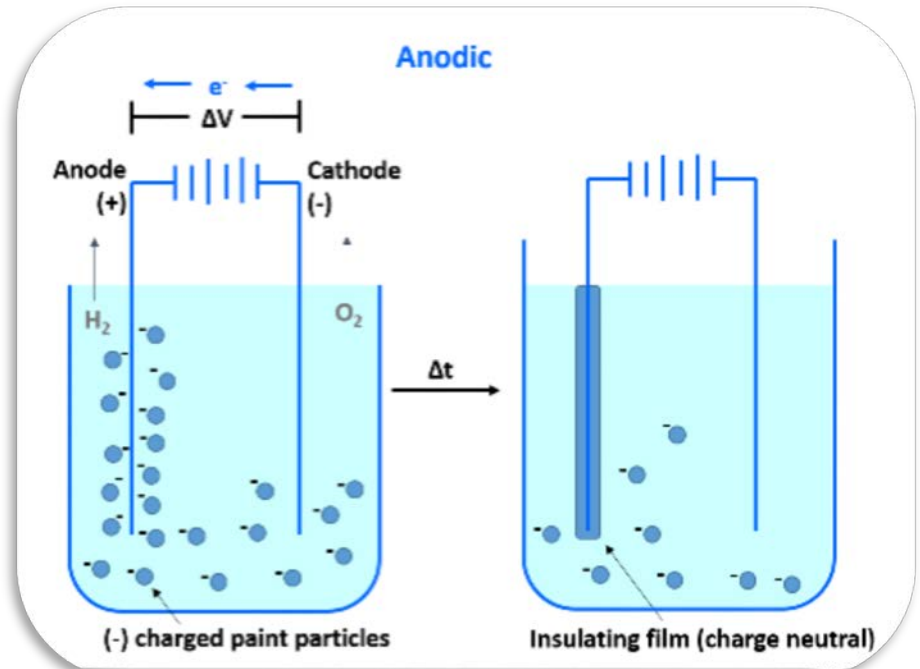
- Low solids, low viscosity bath
- Waterborne, NMP-free

### Coating

- Uniform film build
- High density film
- Additives stay in bath

### Process

- Coat both sides simultaneously
- High transfer efficiency
- Highly automated
- Scalable, High throughput
- Low cost, low emissions

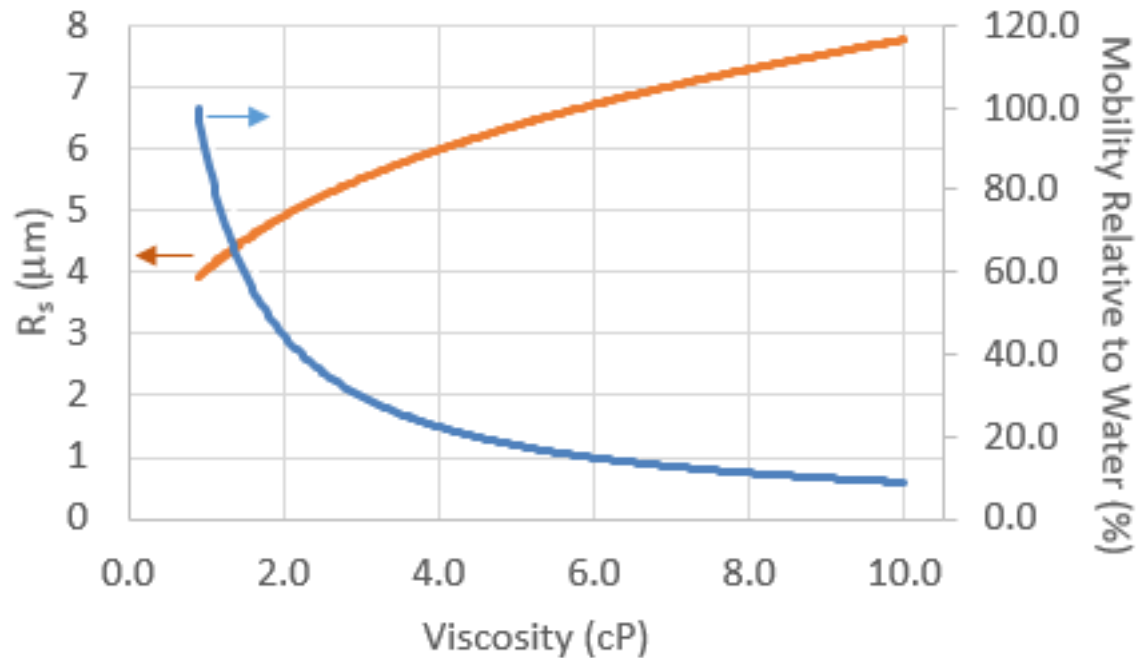


# Project Milestones

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

# Approach/Strategy

## Active material particle size will matter



A theoretical plot of particle electrophoretic mobility relative to water (blue) and the radius of sedimentation (orange) for idealized monodisperse particles that do not aggregate or agglomerate shows that a stable suspension can be obtained with a low-viscosity bath for particles  $< 5\mu\text{m}$ .

# Approach/Strategy

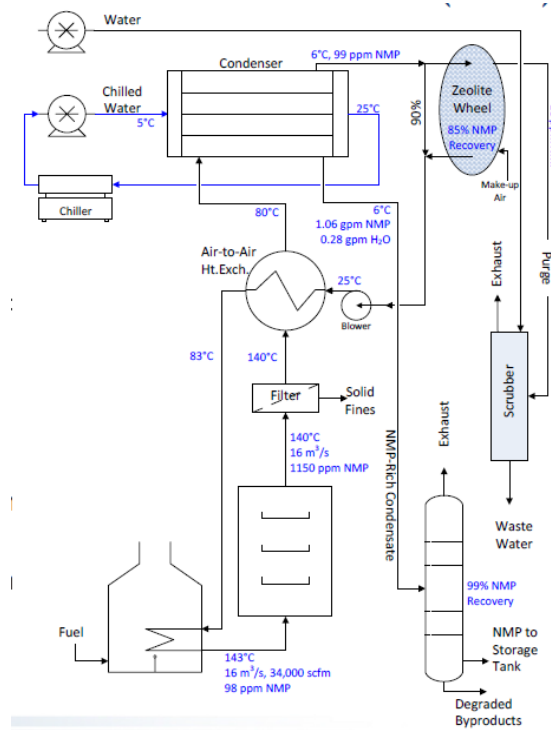
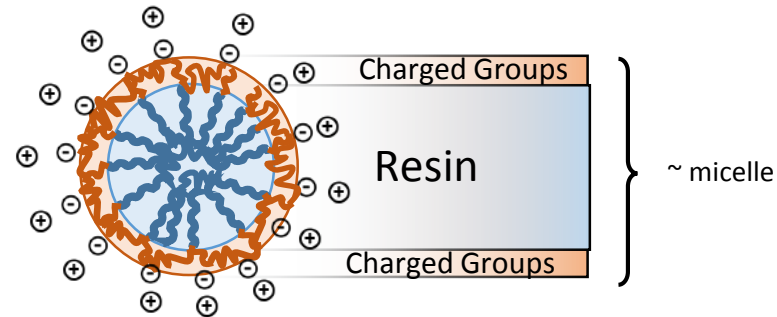
## Formulation will matter

### Binder

- Holds it all together
- Effects mobility
- Deposition rate
- Hygroscopic nature of dry film

### Water

- Lithium dissolution
- Transition metal dissolution
- Permanent damage

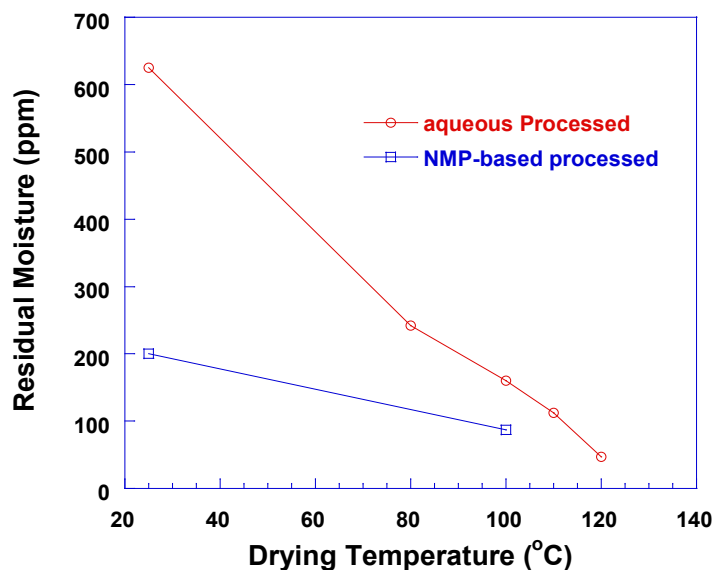


NMP Recovery Process  
Shabbir Ahmed, ANL  
2015 VTO AMR ES228

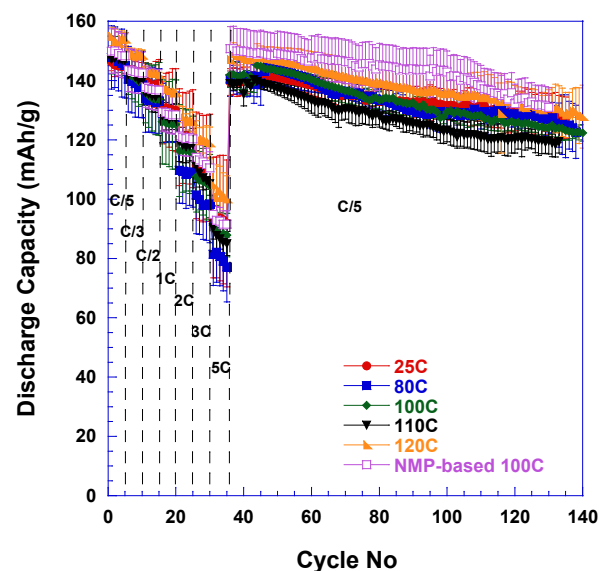
# Approach/Strategy

## Post-deposition processing will matter

Karl Fischer Titration Results



Half Coin Cell Results



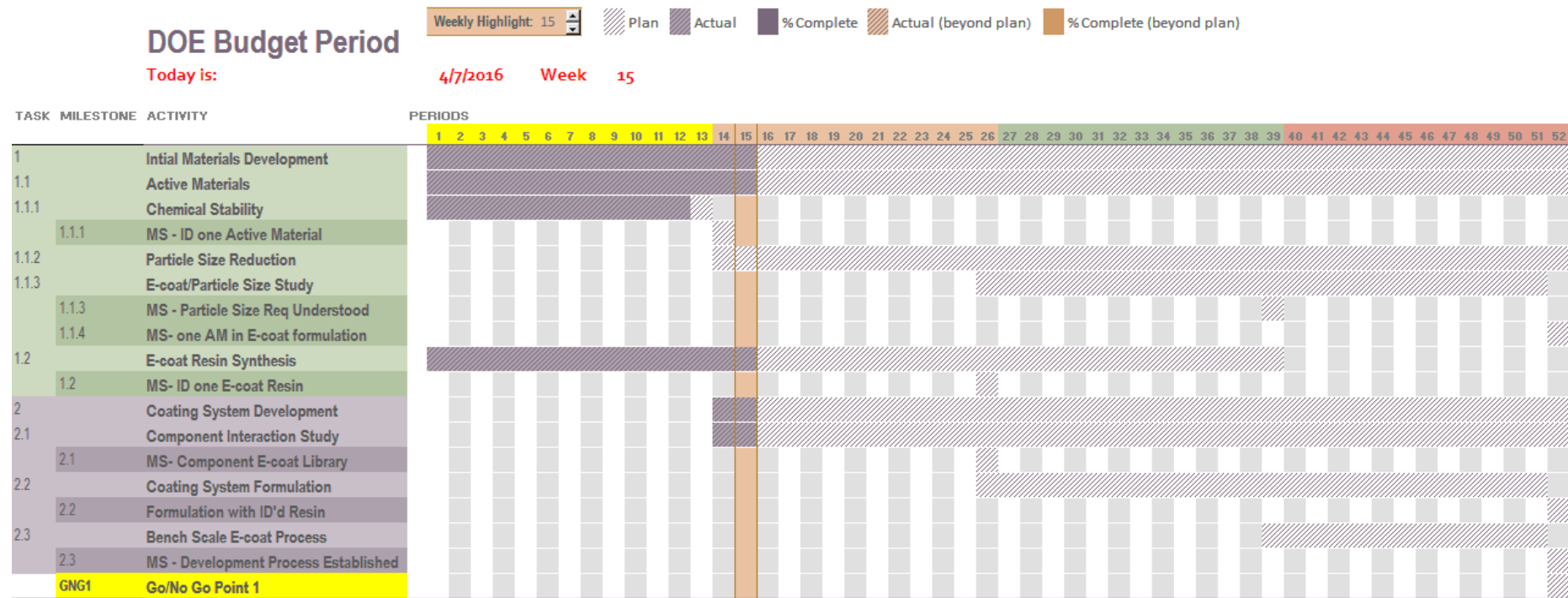
Secondary drying to reduce adsorbed water content

J. Li, C. Daniel, S.J. An, D. Wood, *MRS Advances*, DOI: 10.1557/adv.2016.6 (2016).



## Technical Accomplishments

## Budget Period 1 Plan



# Technical Accomplishments

## Small Cathode Particle Synthesis

		TVR-5 $\mu\text{m}$	Commercial-6 $\mu\text{m}$	Commercial-11 $\mu\text{m}$
Composition by ICP	P*	Li <sub>1.0199</sub> Ni <sub>0.48</sub> Mn <sub>0.31</sub> Co <sub>0.21</sub> O <sub>x</sub>	Li <sub>1.0668</sub> Ni <sub>0.47</sub> Mn <sub>0.34</sub> Co <sub>0.19</sub> O <sub>x</sub>	Li <sub>1.0139</sub> Ni <sub>0.51</sub> Mn <sub>0.29</sub> Co <sub>0.20</sub> O <sub>x</sub>
	W-T**	Li <sub>1.0076</sub> Ni <sub>0.48</sub> Mn <sub>0.31</sub> Co <sub>0.21</sub> O <sub>x</sub>	Li <sub>1.0562</sub> Ni <sub>0.47</sub> Mn <sub>0.34</sub> Co <sub>0.19</sub> O <sub>x</sub>	Li <sub>1.0005</sub> Ni <sub>0.51</sub> Mn <sub>0.29</sub> Co <sub>0.20</sub> O <sub>x</sub>
Tap density, g/cc	P*	2.083	1.852	2.592
	W-T**	2.370	2.007	2.641
D <sub>10</sub> / D <sub>50</sub> / D <sub>90</sub> , $\mu\text{m}$	P*	3.12 / 5.09 / 8.58 (Mean dia.: 5.53 $\mu\text{m}$ )	3.26 / 6.08 / 11.34 (Mean dia.: 6.80 $\mu\text{m}$ )	6.33 / 10.85 / 18.76 (Mean dia.: 11.85 $\mu\text{m}$ )
	W-T**	3.11 / 5.05 / 8.51 (Mean dia.: 5.49 $\mu\text{m}$ )	3.19 / 5.78 / 10.44 (Mean dia.: 6.37 $\mu\text{m}$ )	6.41 / 10.99 / 18.97 (Mean dia.: 12.00 $\mu\text{m}$ )
BET, m <sup>2</sup> /g	P*	0.559	0.814	0.217
	W-T**	2.949	1.701	1.018

\*: P; pristine - untreated cathode sample

\*\* : W-T; water treated cathode sample

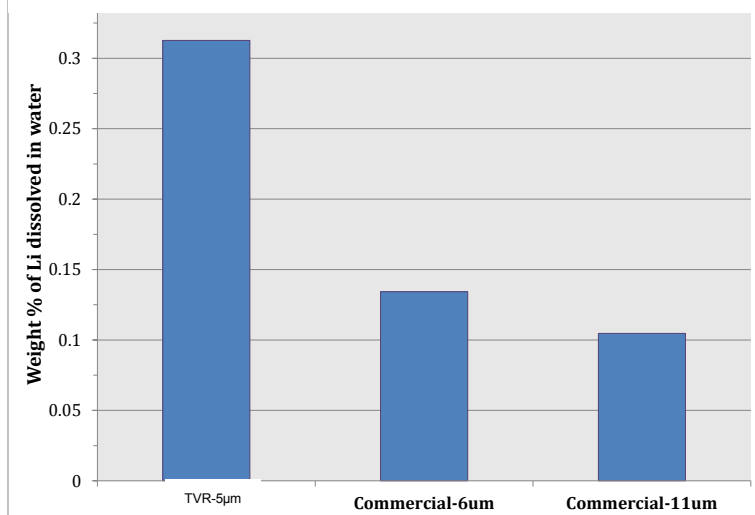


# Technical Accomplishments

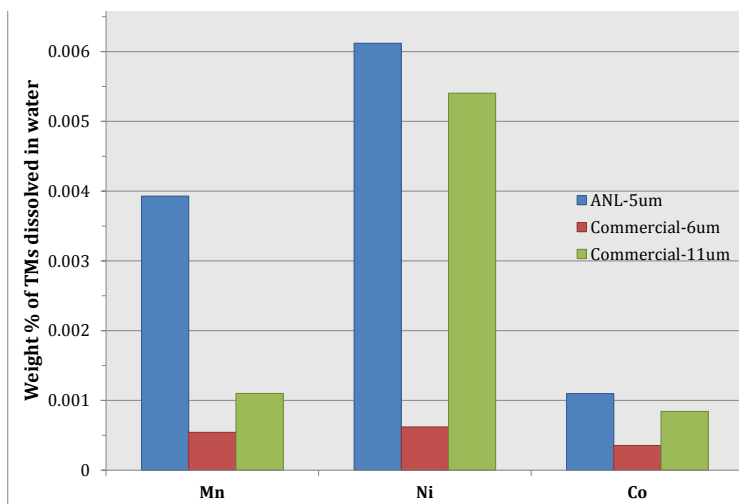
## Active Particle Soaked in Water

### Dissolution behavior and pH trend

Lithium ion dissolution



Transition metals dissolution

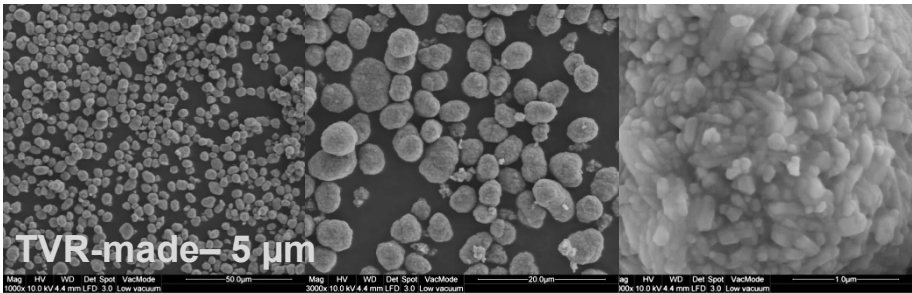


	pH water-only soak			pH water-soak with binder
Soak Time (h)	TVR 5μm	Commercial 6μm	Commercial 6μm	Commercial 6μm
2 min	10.72	10.30	10.30	
1	10.91	11.06	11.06	
2	11.25	11.19	11.19	
24	11.59	11.40	11.40	
48	11.85	11.62	11.62	10.25

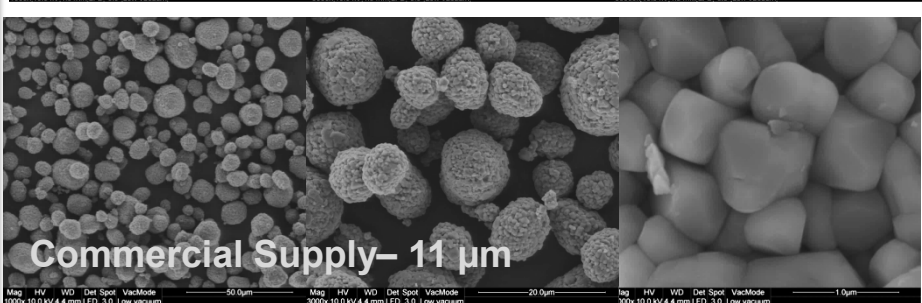
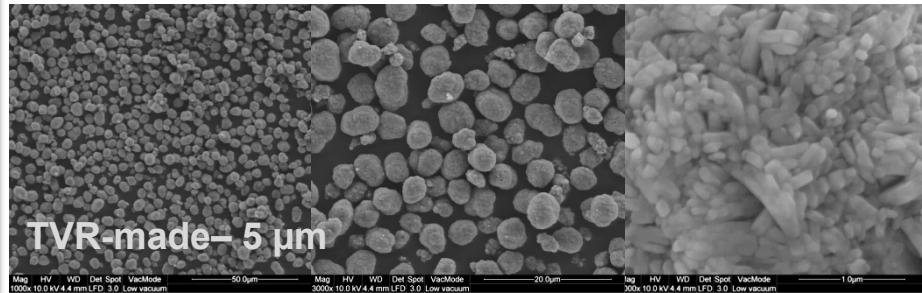
# Technical Accomplishments

Material's morphology – no significant changes after water soak

Before Water Soak



After Water Soak



# Technical Accomplishments

## Resin Synthesis

Relative Properties					
Binder ID	T <sub>g</sub>	Function	MW	Particle Size	Zeta Potential
PPG-072	0.8	0.0	1.4	0.9	-63
PPG-059	0.8	0.0	1.2	1.2	-65
PPG-060	0.8	0.0	1.3	1.1	-68
PPG-042	1.0	1.0	1.0	1.0	-63
PPG-036	0.8	1.0	2.5	0.5	
PPG-063	0.8	0.5	1.5	1.1	-71
PPG-025	0.8	0.3	1.4	1.2	-70



# **Response to Previous Year Reviewers' Comments**

**This is the first AMR presentation for this project**

# 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





# Remaining Challenges and Barriers

## Materials

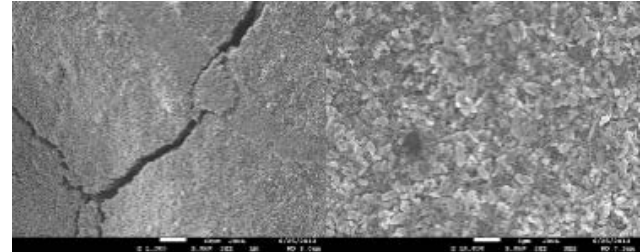
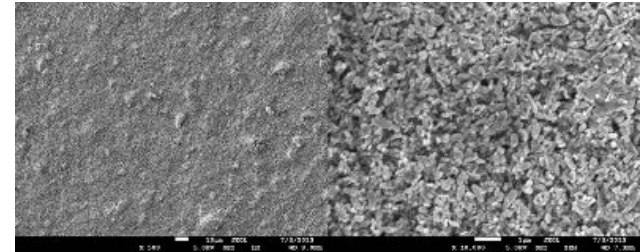
- Active synthesis
- Small Particle Performance

## Formulation

- Bath Stability
- Bath vs Coating Composition
- Deposition Rates

## Cathode Performance

- Film Quality
- Energy Density
- Battery Performance





# Future Work

## Budget Periods 2 & 3

### Process & Equipment Development

- Refinement & scale-up of active material
- Coatings system formulation optimization
- Impact assessment: Electrode coatings drying process

### Design & Build Lab-scale Coater

- Continuous two-sided coating
- In-process drying

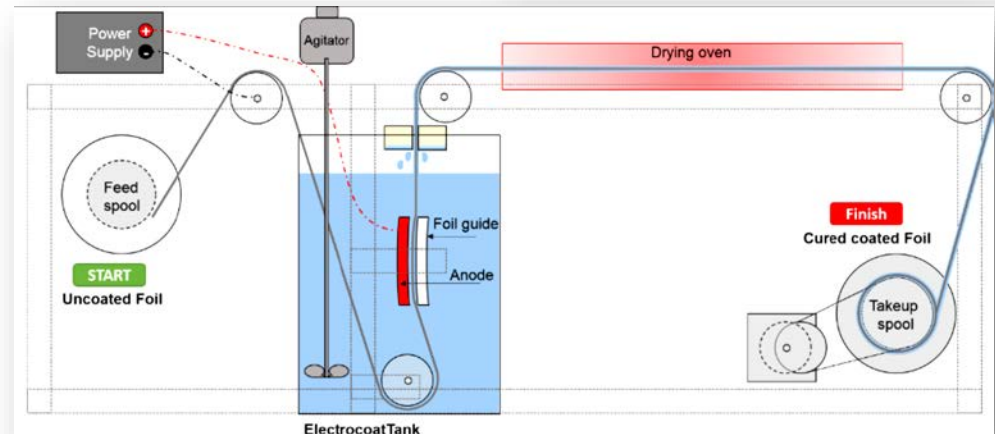


## Other Possible Work

Electrocoat Silicon Anode

Electrocoat Polymer Separator

Binders for Thick Films



# Summary Slide

## Relevance

- Enable lower-cost production of longer-lasting, reproducible battery electrodes with reduced environmental footprint

## Approach

- Utilize Advanced Electrodeposition Coating Materials and Application Process to Produce Battery Electrodes

## Technical Accomplishments

- 5 micron NCM-523 synthesized
- Lithium dissolution characterized
- 8 binders synthesized

## Partners

- Argonne National Lab, Oak Ridge National Lab, Navitas Systems





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