Thicker Cathode Coatings for Lithium-Ion Electric Vehicle Batteries

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June 20, 2019

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Vehicles Technology Office
2019 Annual Merit Review

Project ID: BAT357

Overview

Timeline

- **Project start date:** February 2017
- **Project end date:** December 2019
- **Percent complete:** 74%

Barriers

- **Low areal density electrodes limit overall energy density**
- **Difficult to manufacture high areal density cathodes**
- **Cell manufacturing uses toxic solvents**

Budget

- **Total project Funding:** $1,237,309
  - **DOE Share:** $ 618,654
  - **Contractor Share:** $ 618,654
- **Funding for 2017:** $ 290,706
- **Funding for 2018:** $ 740,181

Partners

- **Idaho National Lab**
- **LG Chem Power**
- **Pennsylvania State University**

1. Relevance - Objectives

Overall Objective

- **Increase cathode energy density** to 40mg/cm^2 while maintaining good flexibility and adhesion
- **Match thick anodes** to 22 mg/cm^2 while maintaining flexibility and adhesion
- **Reduce the battery size, volume, cost** by decreasing the usage of separator and current collector
- **Improve the safety during manufacturing** by eliminating NMP in cathode slurry

Objective this Period

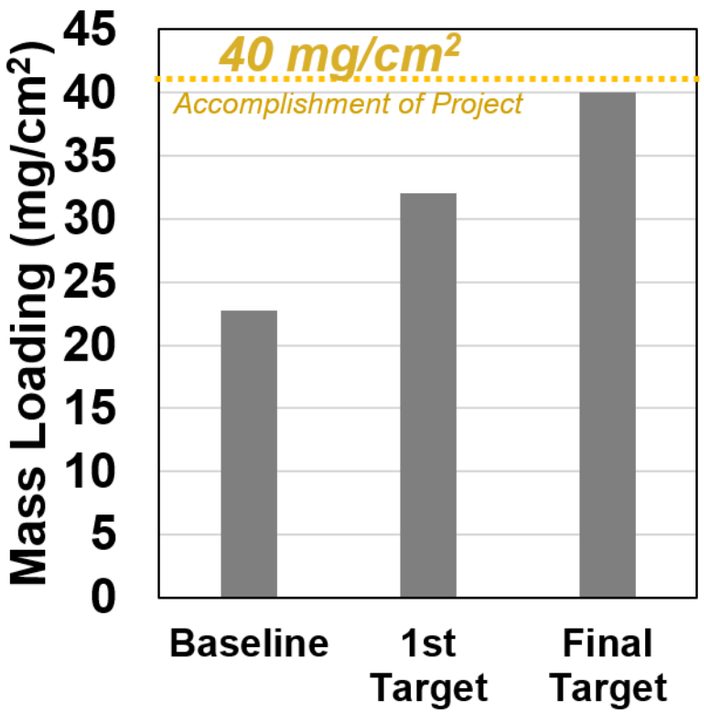
- **Design and improve NMP-free binder formulation for Ni-rich cathodes NMC622**
  - Flexible
  - Good adhesion
- **Evaluate pouch cell performance of cathodes at baseline loading and deliver**
- **Develop thick anodes to assemble full cells with 30 – 40 mg/cm^2 cathodes**

2. Relevance and Targets

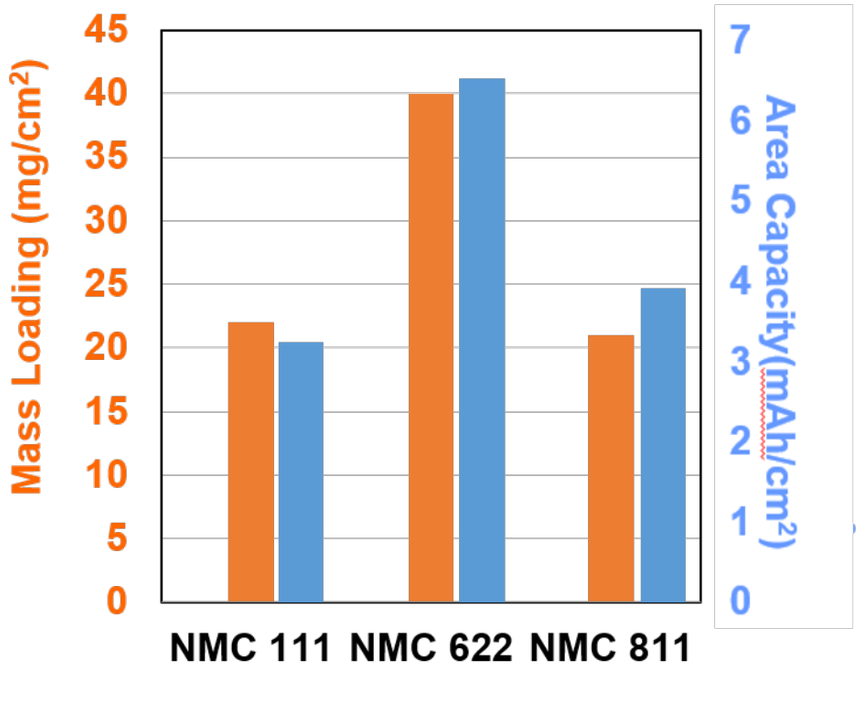
Impact

NMP free thicker coating with high energy density can reduce the overall battery cost, reduce battery size/weight, improve safety during production process.

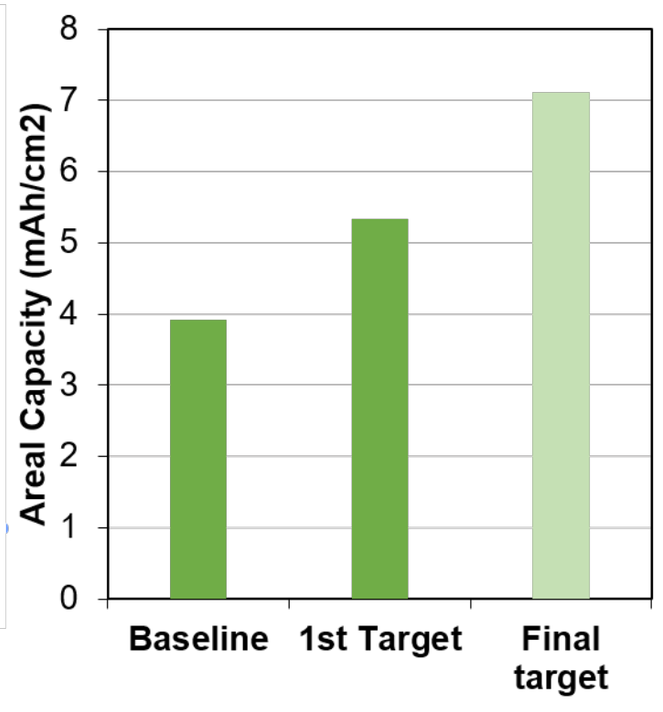
Mass Loading



Cathode Status in Pilot Scale



Anode Status in Pilot Scale



- NMC622: Carbon: Binder ratio 93:3:4
- Nominal Active Energy Density: 175 mAh/g
- Peel strength: 106 N/m
- Flexibility: Pass 1/2” mandrel bend

Milestones and Deliverables

Target	Description	Status
1.1 Milestone	Baseline solventborne data obtained	Finished 2Q2018
3.1 Milestone	Demonstrate baseline cell performance of 85% capacity retention at 300 cycles tested at C/3 at 30°C.	Finished 3Q2018
1.1 Deliverable	14 cells to be provided to INL for baseline evaluation	Finished 4Q2018
1.2 Milestone	110µ solventborne cathode coatings meeting target coating performance	Finished 4Q2018
1.3 Milestone	Electrochemical performance data obtained on 110µ coated electrodes obtained	On-going
3.2 Milestone	Demonstrate 1st target cell performance of 85% capacity retention at 300 cycles tested at C/3 at 30°C.	On-going
2.1 Milestone	150µ solventborne cathode coatings meeting target coating performance	Finished 2Q2018
2.2 Milestone	Electrochemical performance data on 150µ coated electrodes obtained	On-going
3.3 Milestone	Demonstrate final target cell performance of 85% capacity retention at 300 cycles tested at C/3 at 30°C.	On track
2.2 Deliverable	20 1Ah cells based on 150µ solventborne coating delivered to USABC for evaluation	On track

1. Approach

EHS Benefits

NMP Solvent → PPG Solvent

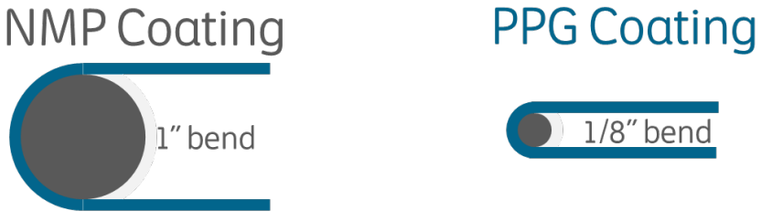
Improved Manufacturing Cycle Time

Lower Baking Temperature

NMP Binder	PPG Binder
120°C	**90°C**

**For solvent evaporation rate of 50 g/m²min.*

Higher Flexibility



**For 100 micron coating thickness.*

2. Approach: NMP-free Cathode Design

- **Design, synthesize, and screen binder resin chemistry for the following properties:**
  - Stable
  - High solids
  - Easy to process rheology properties
  - High quality carbon dispersion.
  - Improved electrochemical stability.

- **Further improve the film property by optimizing processing conditions such as baking and calendar press.**

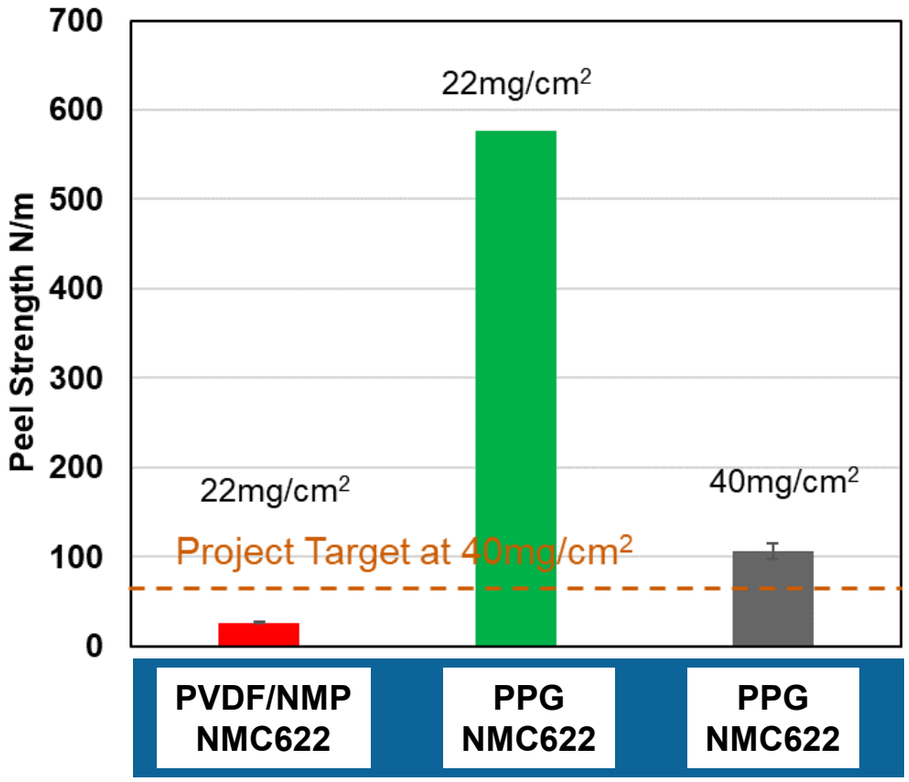
- **Reduce battery size/weight/cost by using thicker coating technology.**

- **Increase EHS compatibility by switching to NMP-free solvent.**

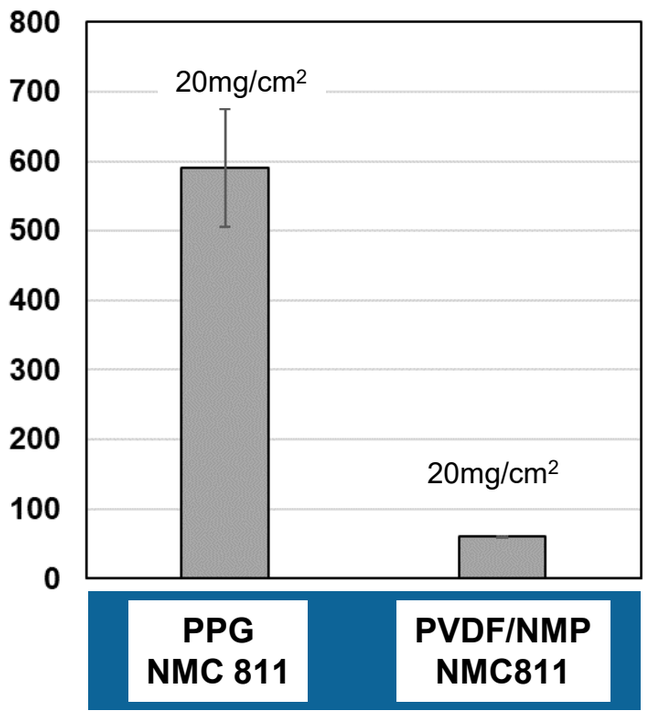
1. Technical Accomplishment:

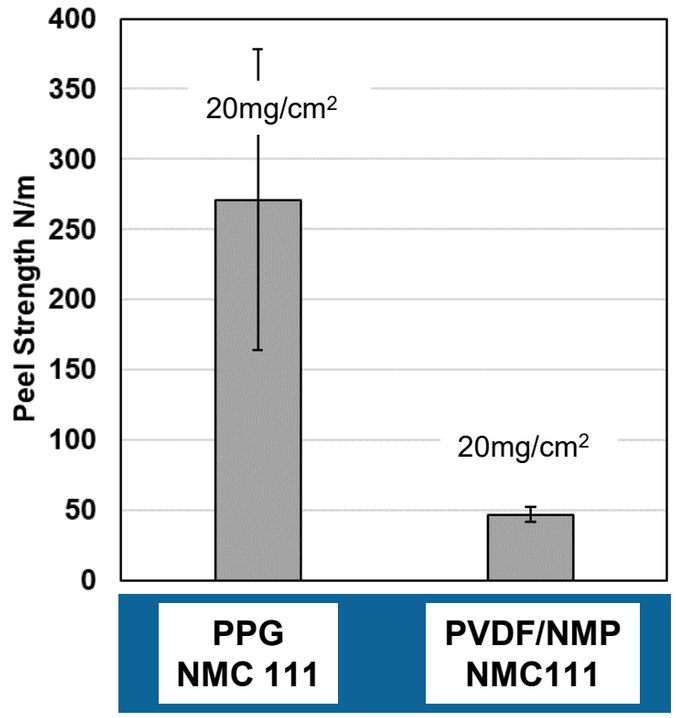
- Pilot-Scale High Nickel Cathodes with High Peel Strength

NMC 622 at baseline and final target loadings



Coatings at ~ 20 mg/cm² loading fabricated at UMEI

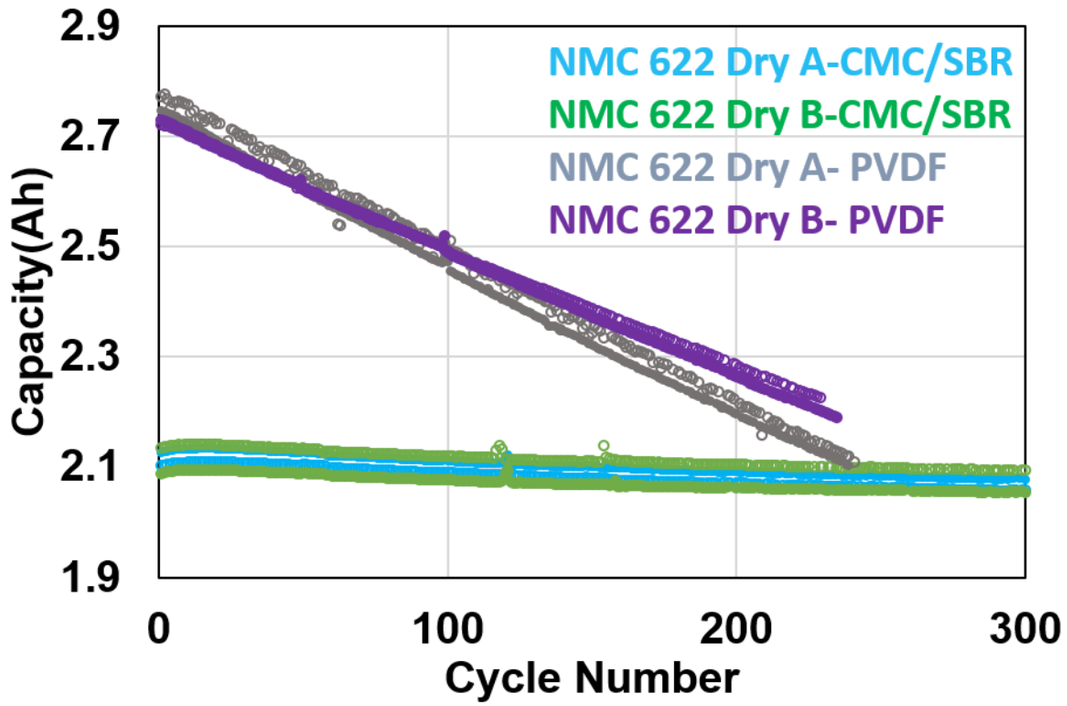


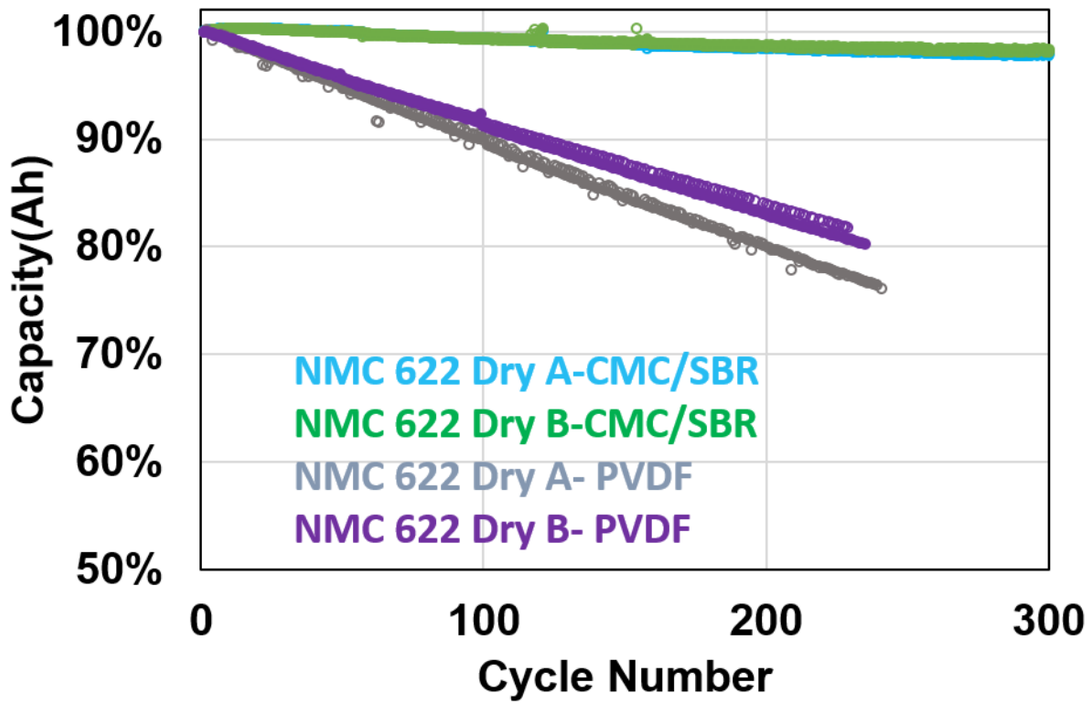


- Coatings with PPG binders have outstanding peel strength compared with PVDF-NMP system
- 50% higher peel strength than target with 40 mg/cm² thick coating

2. Technical Accomplishment:

- Acquisition of Anode Formulation Tools

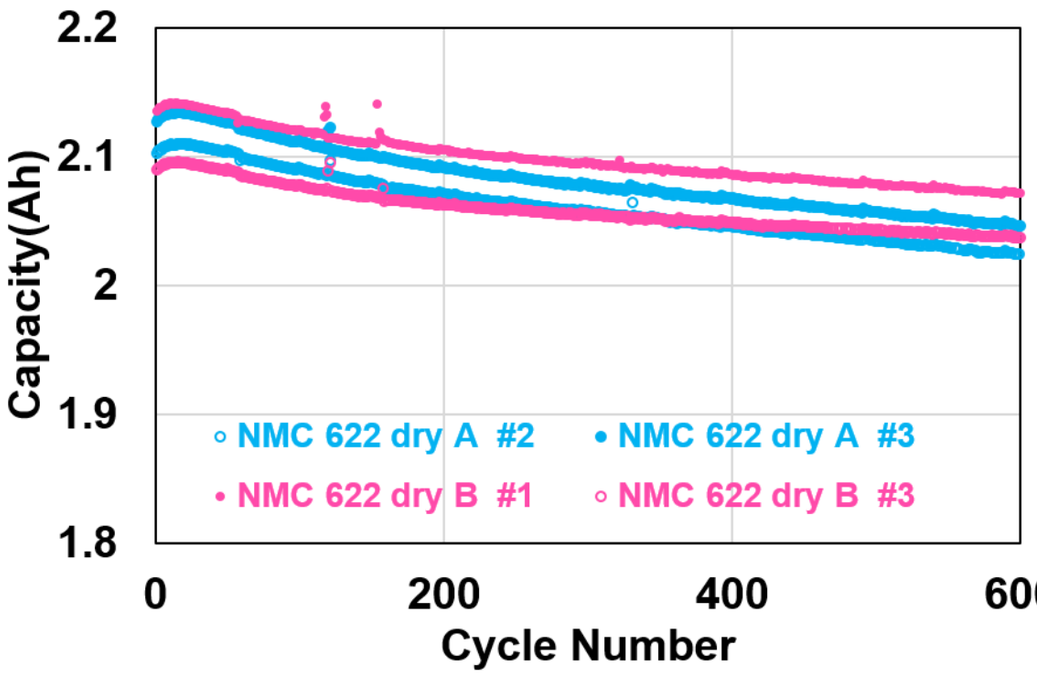


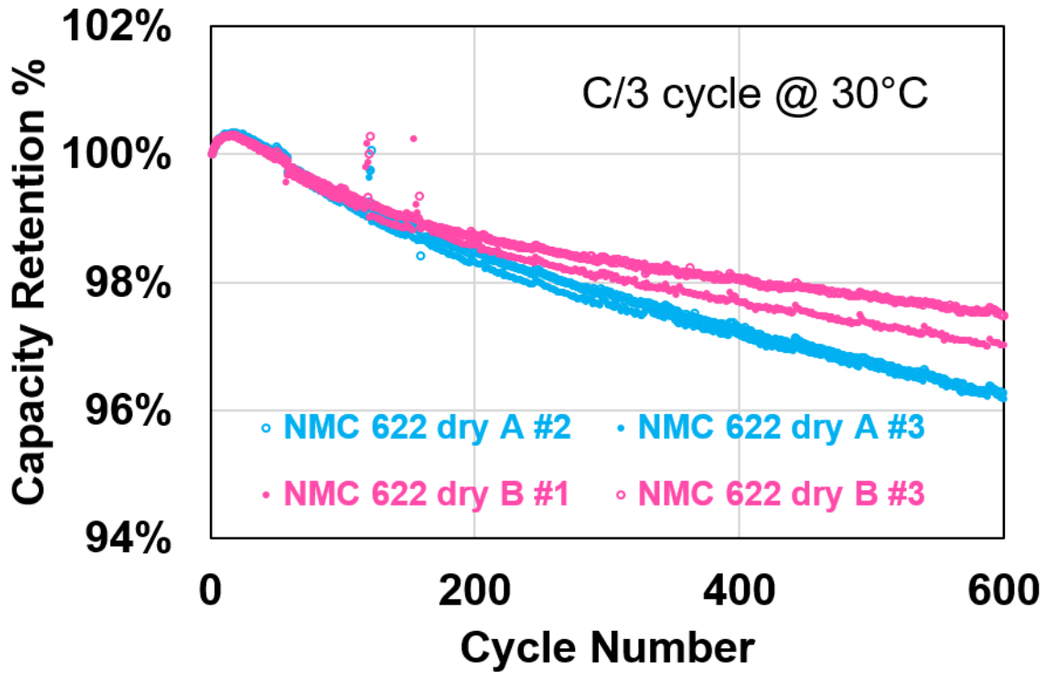


- By changing to waterborne anode, UMEI cells with NMC 622 now show good battery performance
- Cathodes with PPG binders are matched with anodes with both PVDF-NMP binder and CMC/SBR binder
- Cells with PVDF-NMP binder show 76-80% capacity retention after 240 cycles. Cells with CMC/SBR anodes showed significantly higher capacity retention, 97-98% after 330 cycles.

3. Technical Accomplishment:

- Cathode Binder Optimization





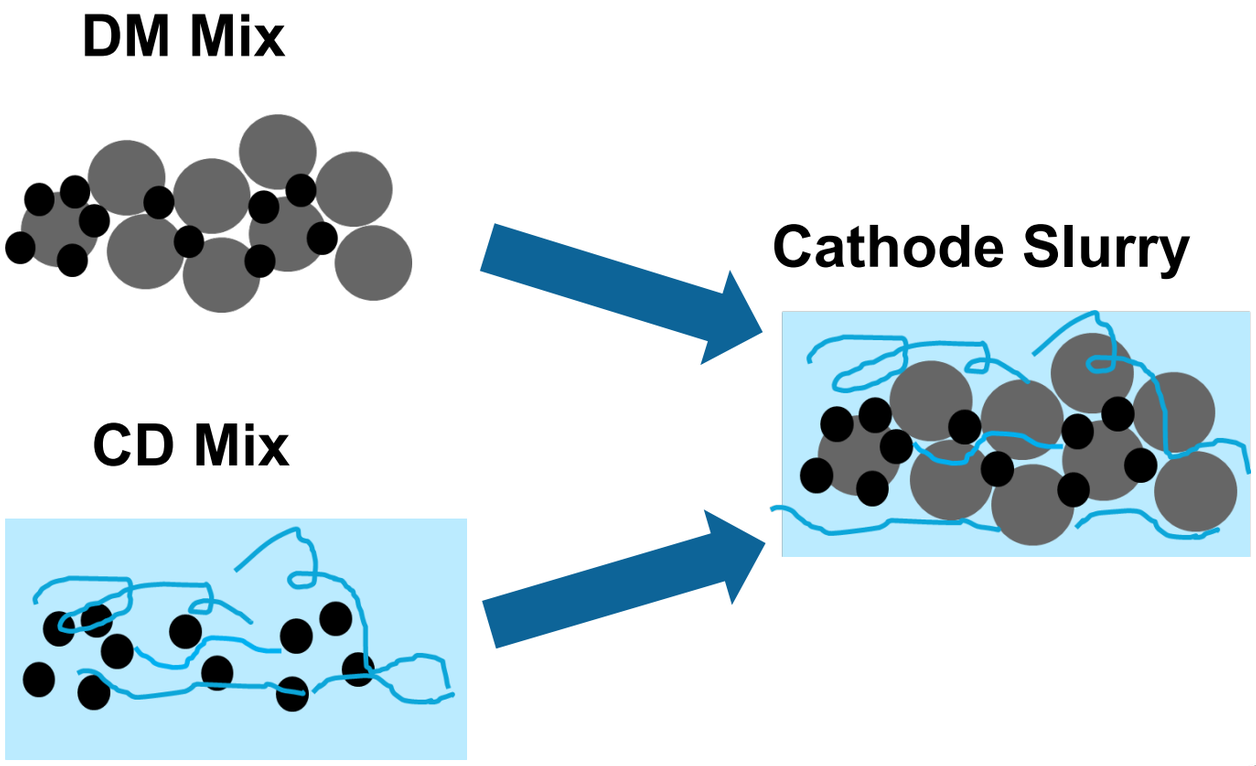
* Coatings and cells fabricated at UMEI. Currently being testing at BIC under C/3 at 30°C

PPG can change binder to improve cathode coating cycle performance

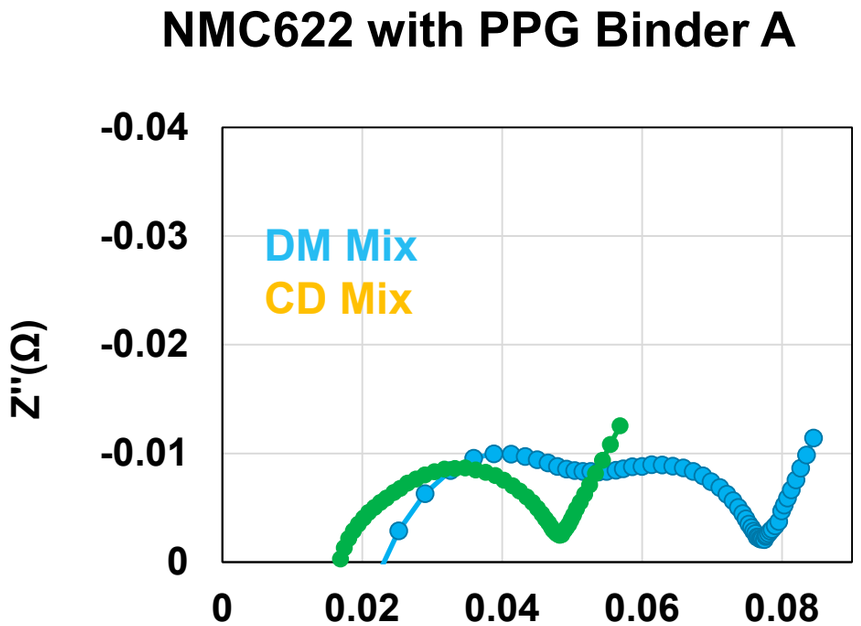
- **NMC 622 cathode with binder B has >97 % retention after 600 cycles at C/3**
- **Binder B starts to show advantages over PPG coatings with binder A after 150 cycles**

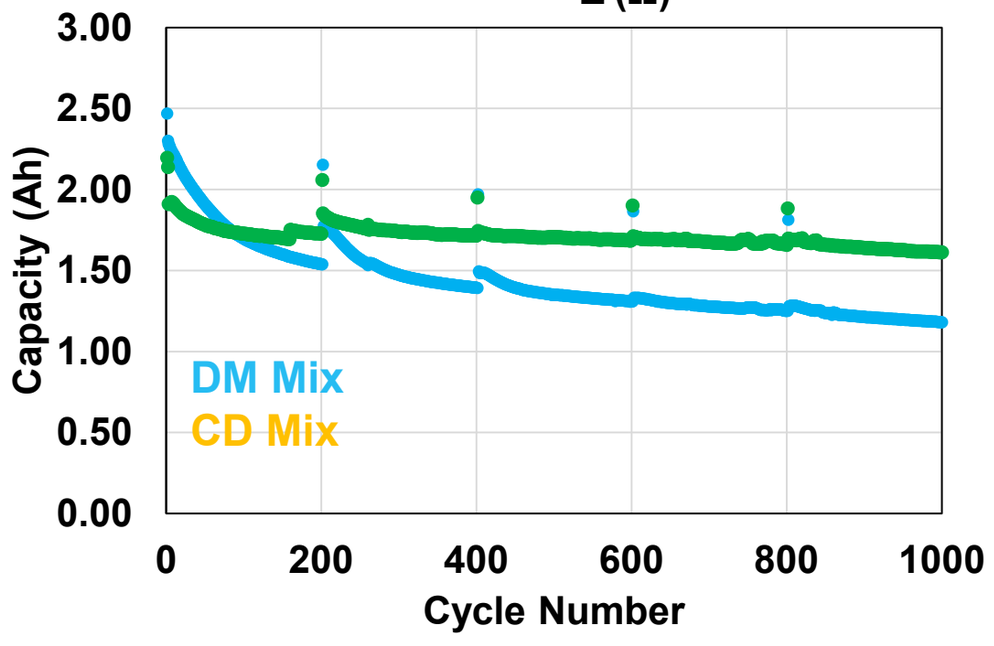
4. Technical Accomplishment:

- Cathode Coating Process Optimization



PPG can tune mixing procedure to improve cathode performance

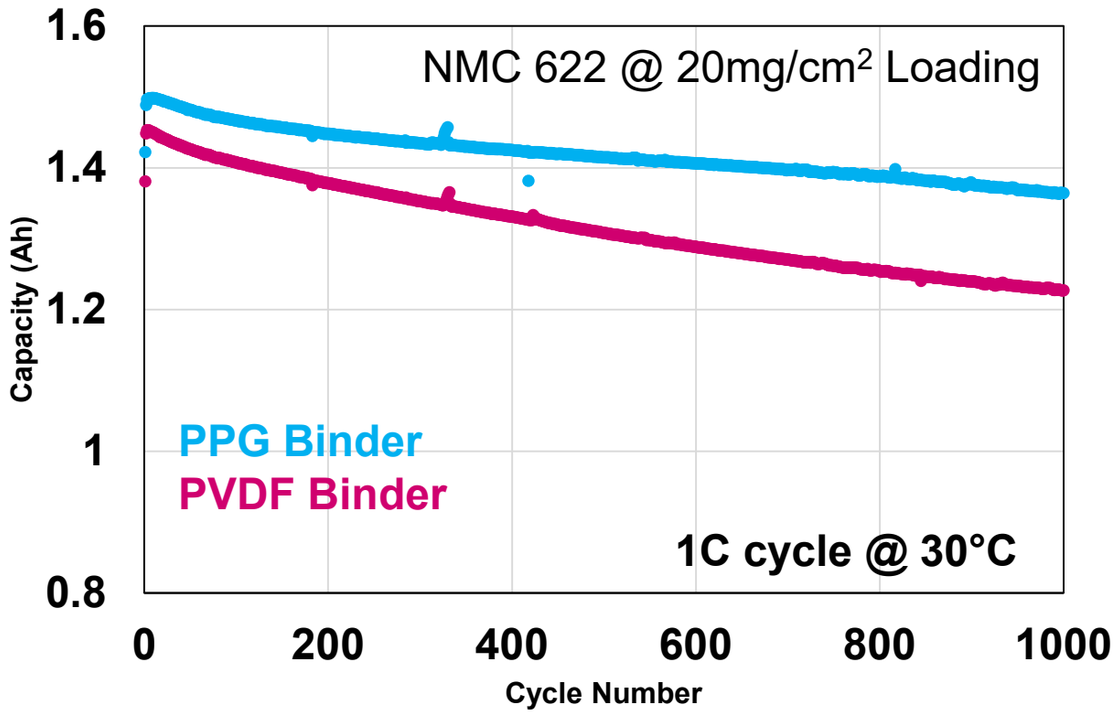


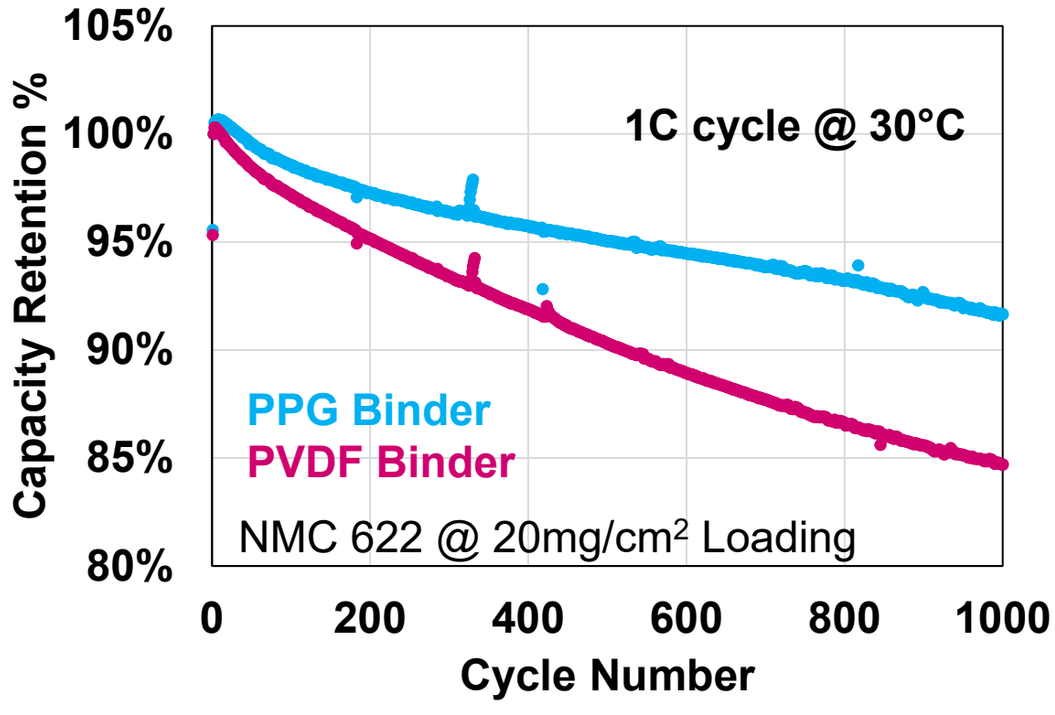


Cathode fabricated at UMEI; anode and cells made and tested at PSU

5. Technical Accomplishment:

- Baseline Cathode Performance





Cells with PPG binder significantly outperformed cells with PVDF-NMP binder

- 95/3/2 coatings and cells fabricated at UMEI. Finished testing at BIC under 1C at 30°C
- At 1000 cycles cells with PPG coating and NMC 622 was at 91.5% capacity retention, and the control cells was at 84.7% capacity retention.

6. Technical Accomplishment:

- Baseline Cell Delivery

14 deliverable cells were fabricated and delivered to INL in Q4 2018

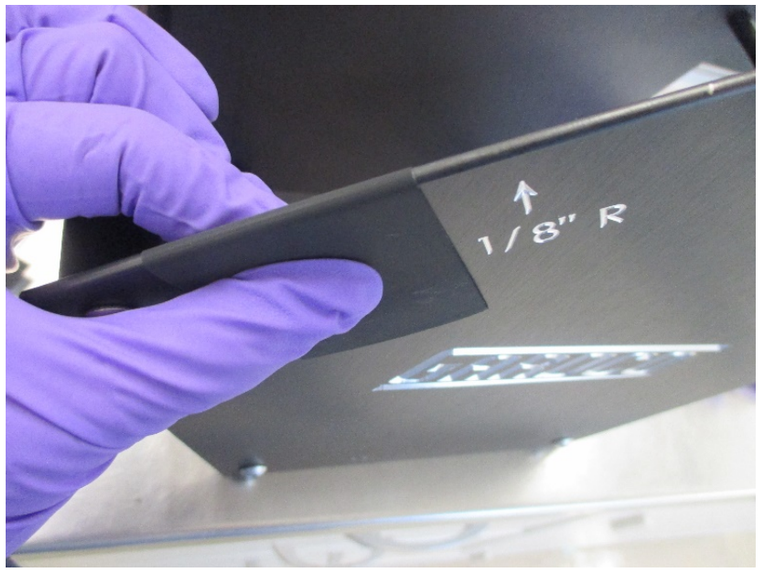
- Cathode with Binder B using a carbon dispersion mixing
- CMC/SBR anode to match the cathode. N/P ratio ~1.1
- 1.9Ah pouch cells with 4 layers of cathode and 5 layers of anode
- Testing plan, fixture information finalized

Electrodes	Coating Facility	Active/Carbon/Binder	Binder	Active Material	Carbon	Mix	Mass Loading mg/cm^2
NMC 622 B	UMEI	93/3/4	PPG Binder B	NMC622 supplier 1	LITX 200	Carbon dispersion	22.2
Anode I	UMEI	97/1.5/1.5	CMC/SBR	graphite	N/A	N/A	11.7

7. Technical Accomplishment:

- Cells with 1st Target Thickness Cathode

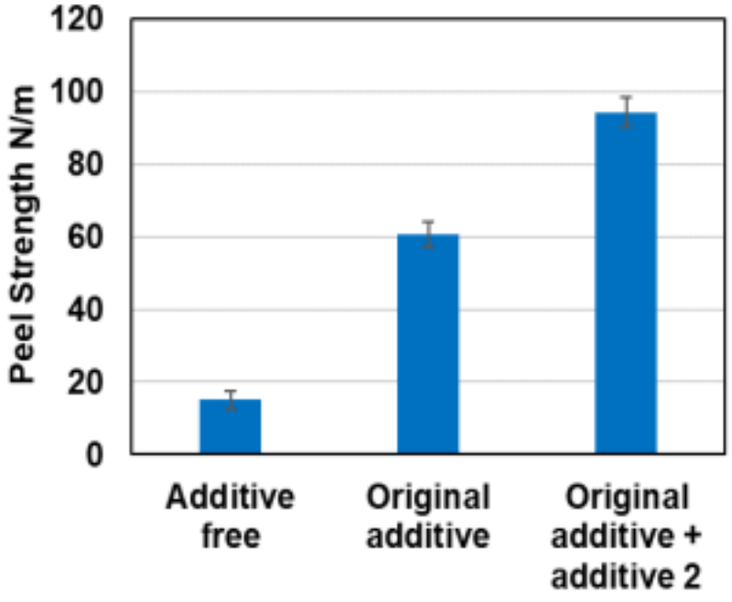
- Successfully coated NMC 622cathode with mass loading of 30 mg/cm^2 on roll to roll pilot line
- Peel strength > 66N/m, flexibility pass 1/8'' R
- An anode with mass loading of 15.8 mg/cm^2 was fabricated to match the cathode, using baseline formulation
- First- Batch Pouch cells fabricated and being tested at UMEI

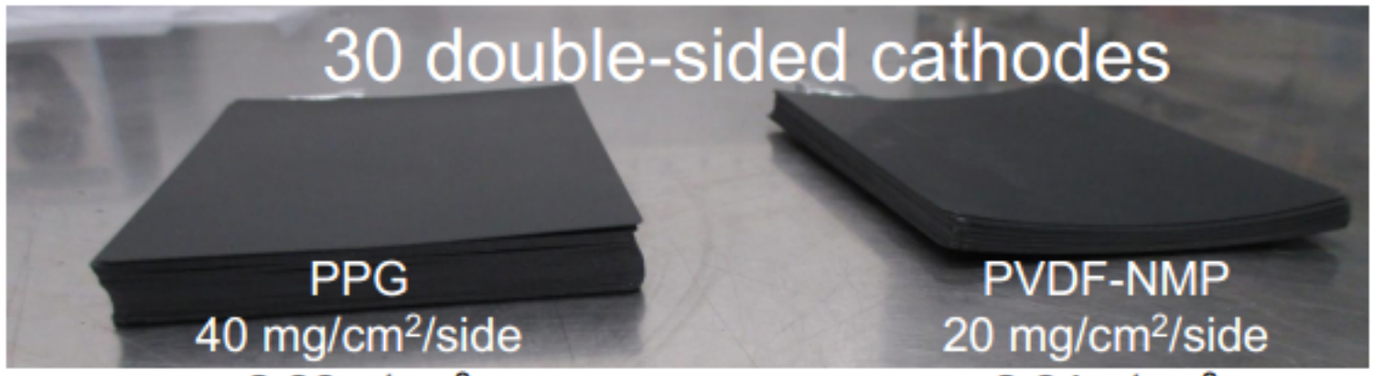


PPG Thick Coatings with NMC622 – 40mg/cm²

Job Description	Mass Loading	Areal Capacity (mAh/cm²)
Hi-Ni 40mg/cm²	40mg/cm²	6.02
Hi-Ni 40mg/cm² with additives	40mg/cm²	6.02

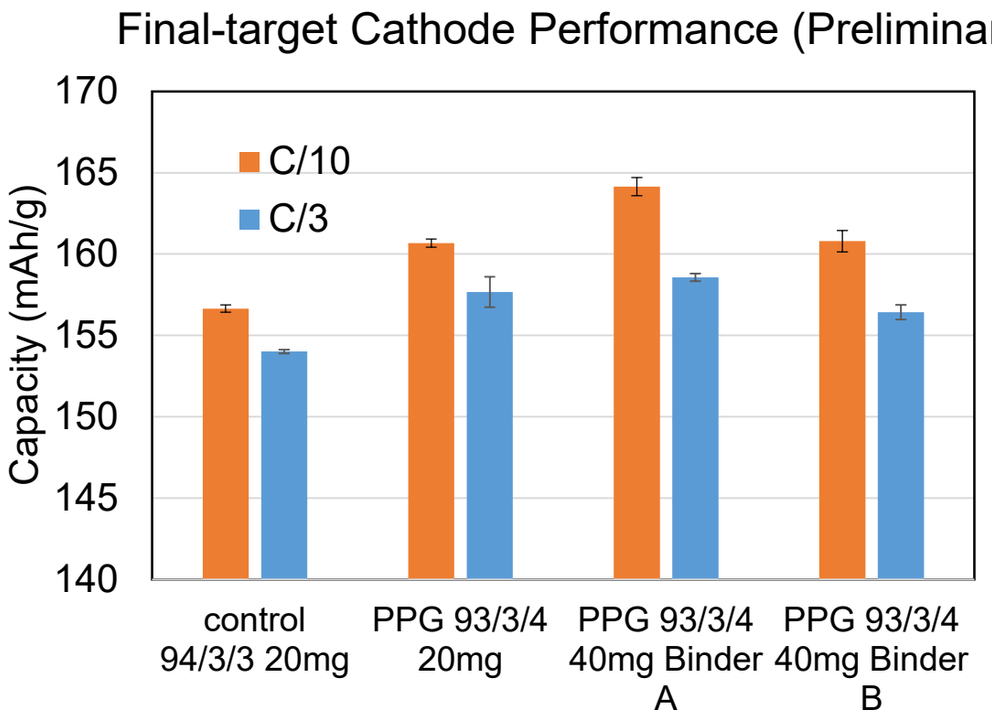
- Pilot scale roll-to-roll double-sided coated up to 40 mg/cm² per side
- Pouch cells fabricated
- Lab scale draw down up to 49 mg/cm²
- Good flexibility - ½ inch
- Outstanding peel strength



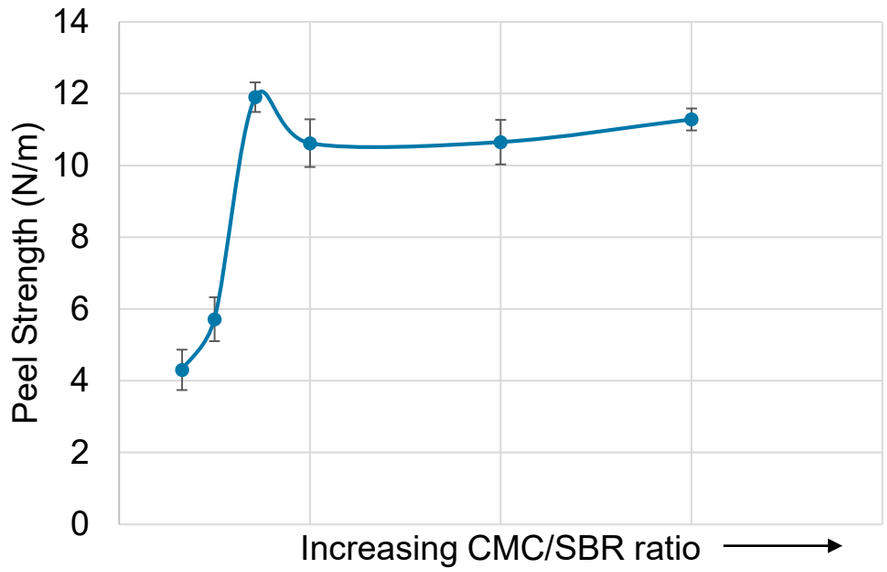


8. Technical Accomplishment:

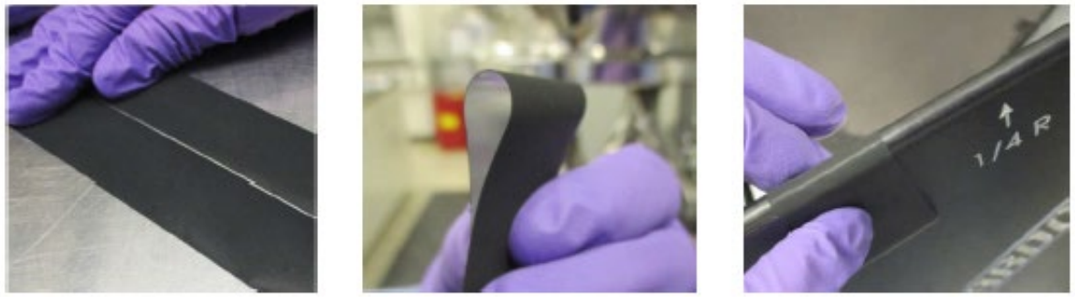
- Status for Final Target



Cathode: PPG Binder- NMC 622
Anodes: PVDF-NMP-graphite (not optimized)



Anode Formula: 97% graphite, 3% binder
Loading: 22 mg/cm^2 (final target)



- **PPG final-target cathodes delivered reasonable capacity when paired with un-optimized anode**
- **On-going formulation work to optimize thick anodes**

Response to Previous Years Comments

Reviewer 1: There is no plan in this work on how to address the high-power limitations of these coatings (i.e., 5-9 mAh/cm2).

Agree but power is not a focus of this project, project focuses on C/3 which is a practical rate in electric vehicles.

Reviewer 1: No details were provided on how to switch from the solvent system (which is of limited benefit over conventional NMP) to a pure water system.

Impending regulation will limit manufacturing with NMP. PPG Binder/Solvent has several benefits over NMP. The statement of work was modified to remove waterborne cathode work in favor of anode development.

Reviewer 2: The PPG binder has some advantages over the NMP binder. However, the reviewer was unclear why the PPG binder is better than the NMP binder. It is unclear why the PPG binder-based cathode can be thick without sacrificing the power density and cyclic performance.

PPG binder produces higher solids, thicker, flexible coatings, low toxicity, lower production costs than PVDF in NMP.

Reviewer 3: The reviewer said that the percentage of NMC in the electrode needs to be increased to 96% or above to further increase the areal capacity. The reviewer asked if the minimum amount of PPG binder is 4%, or it can be further lowered.

95/3/2 A:C:B cathodes with A=NMC622 were demonstrated in this budget period (Slide 12).

Reviewer 2: The reviewer observed excellent progress on the processability of the thick cathodes, but much work on the anode and rate performance metrics still needs to be done. It seems as though the team is having trouble with continuity of full cell builds as well.

The USABC agreed to focus work on high areal energy density anode development in this budget period rather than waterborne cathode development. Full cell development work is being performed at UMEI and we engage PSU for anode guidance.

Reviewer 3: the reviewer thought the use of “NMP-free binder” is confusing. In fact, it is not a binder-free cathode. Instead, the PPG binder is used to replace the NMP binder.

Reviewer is correct that this is not binder-free. “NMP-free” means no NMP solvent is used. Instead NMP (toxic, flammable solvent that is indicted as a reproductive hazard) is replaced with a less toxic, non-flammable solvent that also enjoys several processing advantages.

Collaboration

LG Chem	Cell performance validation by commercial battery producer
INL Idaho National Laboratory	Provided feed back on project during quarterly meeting. On-going tests for baseline pouch cells. Will be testing milestone for final-target thick coatings.
PennState	Join project in May 2018. Assemble baseline pouch cells with existing anodes at PSU to screen cathode for deliverables to INL.
MICHIGAN ENGINEERING UNIVERSITY OF MICHIGAN	Provided facility for slurry mixing, coating, calendar press, and pouch cell assembly.
OBIC	Provided test channels for electrochemical performance evaluation.

PPG

Remaining Challenges and Barriers

Developing Thick Anode

- PPG has developed thick anode for 1st target. However, thicker final-target anode is still under development
- PPG has decided to develop anode formulations in its own lab

Optimizing Final-Target Cathode

- PPG may need to optimize cathode formulation and cell design further based on electrochemical performance of thick anode-thick cathode pairs

Future Research

Thick Anode Development

- Optimize thick anode formulation by tuning CMC/SBR ratio, dosage and type
- Optimize fabrication process of thick anode- mixing, coating and heating procedure

Pouch Cell Testing with Anodes Designed for Thicker Cathode Coatings

- Optimize electrode pairs based on lab formulation practice
- Final deliverable is pouch cell testing conducted at INL with cells fabricated by PPG
- Validate performance of cathode and anode pairs at LG Chem

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

Summary

PPG NMP-free binders enable manufacturing of high areal density, high- Ni cathodes

- ☑ Up to 145 um per side double sided NMC 622 coating with pilot-scale roll-to-roll coater
- ☑ Up to 40 mg/cm^2 mass loading
- ☑ Peel strength (90°) up to 100 N/m
- ☑ Flexibility for thicker double-sided coating passes ½ inch Mandrel bend test

Evaluation of baseline pouch cell performance completed

- ☑ Anode formulation developed at PPG and PSU
- ☑ Pouch cell fabrication at University of Michigan
- ☑ Baseline pouch cell testing completed- >97% capacity retention after 600 C/3 cycles
- ☑ Baseline pouch cell delivered to INL
- ☐ Cell evaluation by INL

Evaluation of 1st target pouch cell performance in progress

- ☑ 1st target pouch cells fabricated at UMEI with 30 mg/cm^2 cathode loading
- ☐ 1st target pouch cell testing in progress at UMEI

Final-target cells in progress

- ☑ Final target NMC 622 cathode at 40 mg/cm^2 cathode meeting project goals
- ☐ Anode formulation in progress at PPG
- ☐ Final-target pouch cell delivered to INL
- ☐ Cell evaluation by INL and LG Chem