Thicker Cathode Coatings for Lithium-Ion Electric Vehicle Batteries

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Vehicles Technology Office
2020 Annual Merit Review

Project ID: BAT357

Overview

Timeline

- **Project start date:** February 2017
- **Project end date:** June 2020
- **Percent complete:** 90%

Budget

- **Total project Funding:** $1,393,397
  - **DOE Share:** $ 618,654
  - **Contractor Share:** $ 774,743
- **Funding for 2017:** $ 290,738
- **Funding for 2018:** $ 736,828
- **Funding for 2019 :** $ 322,255
- **Funding for 2020:** $ 43,576

Barriers

- **Low areal density electrodes limit overall energy density**
- **Difficult to manufacture high areal density cathodes and matching anodes**
- **Cell manufacturers use toxic solvents**

Partners

- **Idaho National Lab**
- **LG Chem Power**

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 collectors
- **Improve the safety during manufacturing** by eliminating NMP in cathode slurry

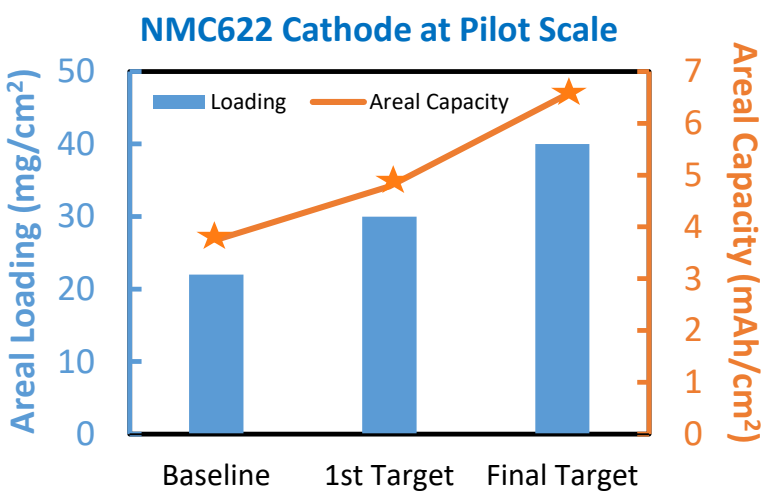
Objective this Period

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

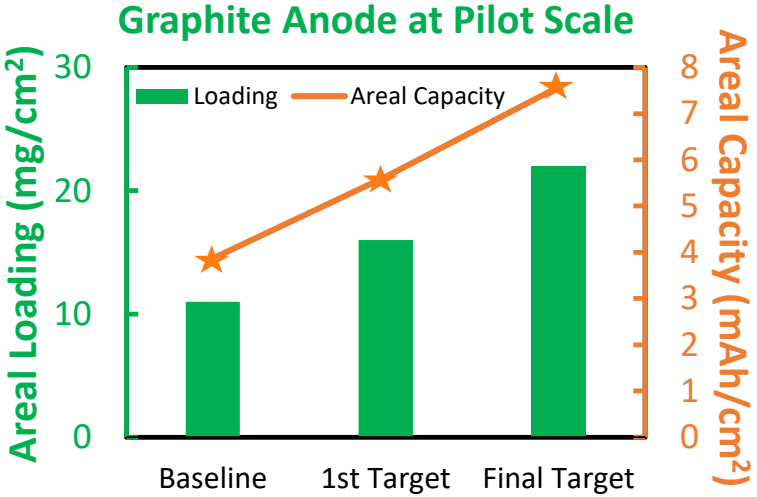
2. Relevance and Targets

Impact

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



Cathodes



Anodes

- Maintaining the peel strength and flexibility
- Further improvement in anode could be made by utilizing the Si based active material

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	110 µm solventborne cathode coatings meeting target coating performance	Finished 4Q2018
1.3 Milestone	Electrochemical performance data obtained on 110 µm coated electrodes obtained	Finished 2Q2019
3.2 Milestone	Demonstrate 1st target cell performance of 85% capacity retention at 300 cycles tested at C/3 at 30°C.	Finished 4Q2019
2.1 Milestone	150 µm solventborne cathode coatings meeting target coating performance	Finished 2Q 2019
2.2 Milestone	Electrochemical performance data on 150 µm coated electrodes obtained	Finished 4Q 2019
*2.3 Milestone	Develop thick graphite anode to match the final target (150 µm) cathode	Finished 1Q 2020
3.3 Milestone	Demonstrate final target cell performance of 85% capacity retention at 300 cycles tested at C/3 at 30°C.	On-going
2.2 Deliverable	20 1Ah cells based on 150 µm solventborne coating delivered to USABC for evaluation	On track

* New milestone added for 2019-2020

1. Approach

EHS Benefits

NMP Solvent → PPG Solvent

Improved Manufacturing Cycle Time

NMP Binder **11 hours**

PPG Binder **5 hours** ← 50% Reduction in mix time

Lower Baking Temperature

NMP Binder	PPG Binder
120°C	**110 °C**

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

Higher Flexibility

NMP Coating

PPG Coating

**For 100 micron coating thickness.*

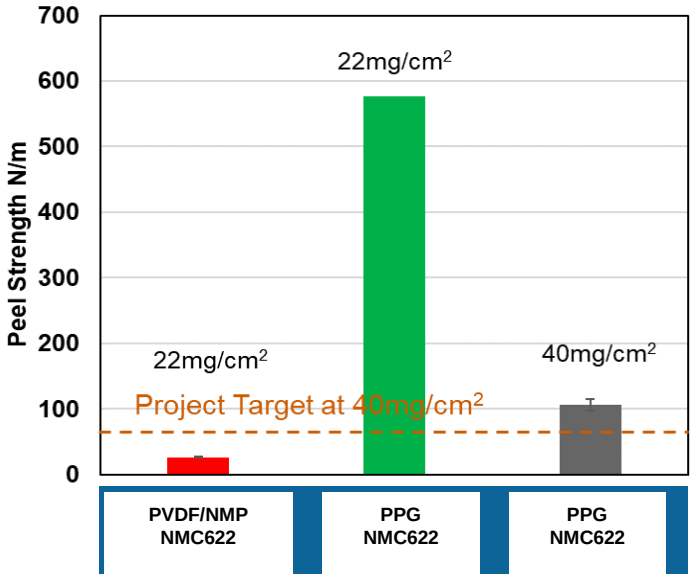
2. Approach: NMP-free Cathode Design

- **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:

- High Nickel Cathodes with High Peel Strength

NMC 622 at baseline and final target loadings

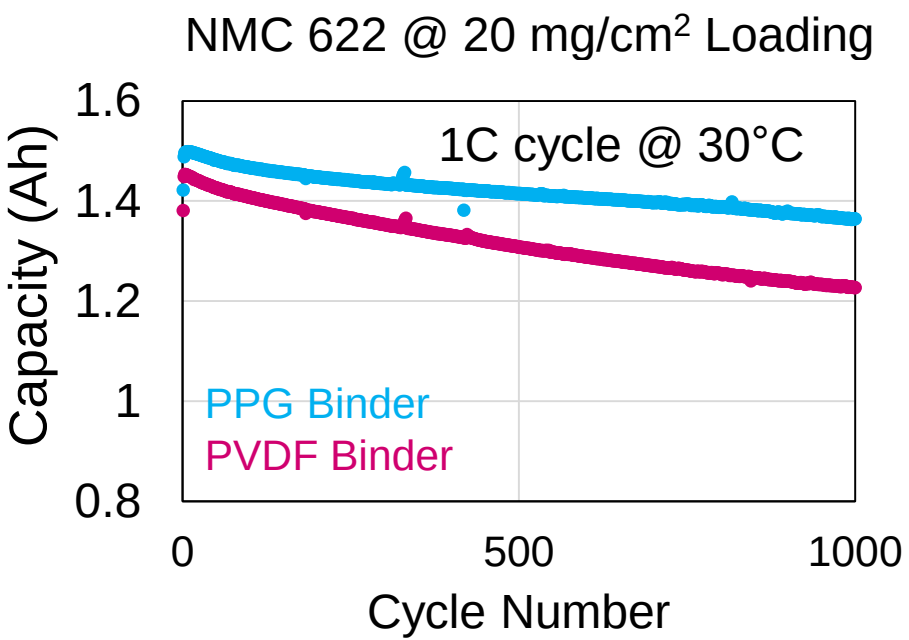


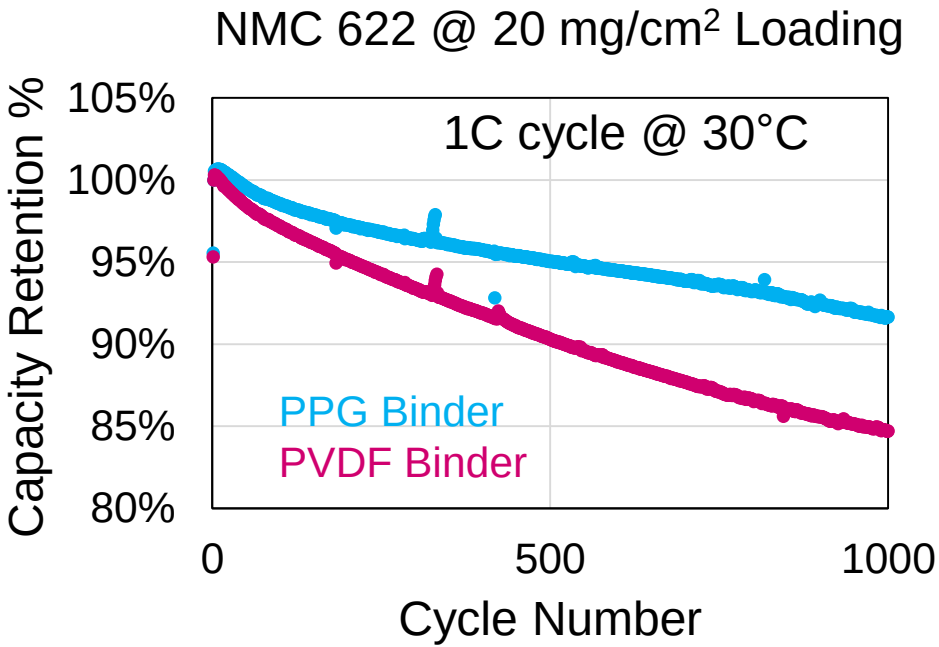
Pilot-scale roll to roll coatings

- Coatings with PPG binders have outstanding peel strength compared with PVDF-NMP system
- 50% higher peel strength than target with 40 mg/cm^2 thick coating

2. Technical Accomplishment:

- Baseline Cathode (22 mg/cm^2) Performance





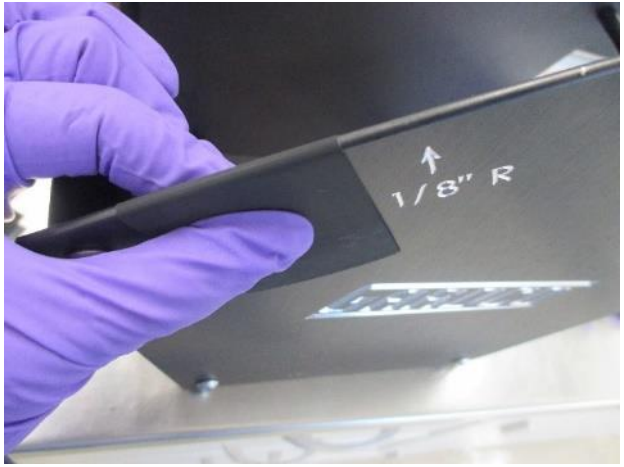
Cells with PPG binder significantly outperformed cells with PVDF-NMP binder

- 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.

3. Technical Accomplishment:

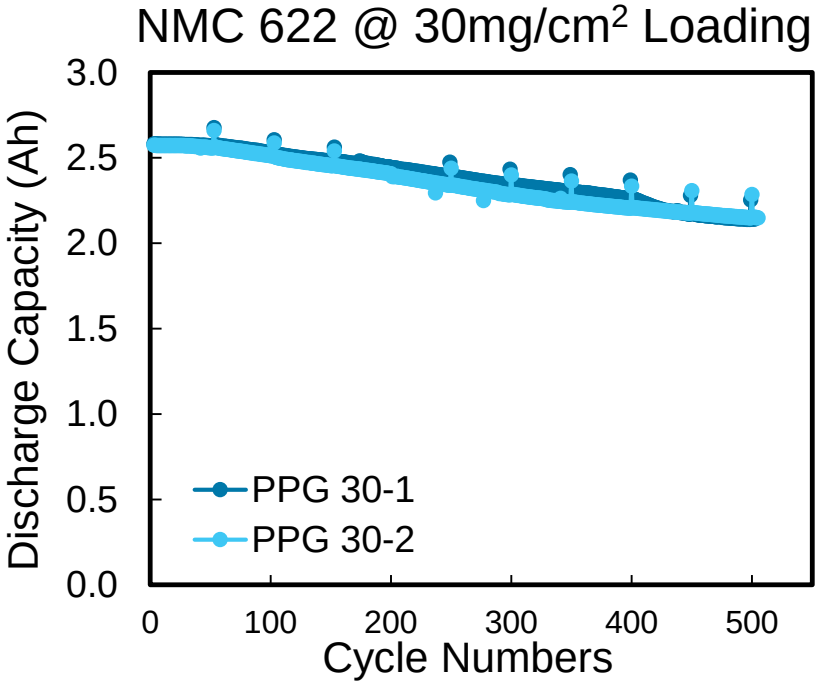
- Cells with 1st Target Thickness Cathode

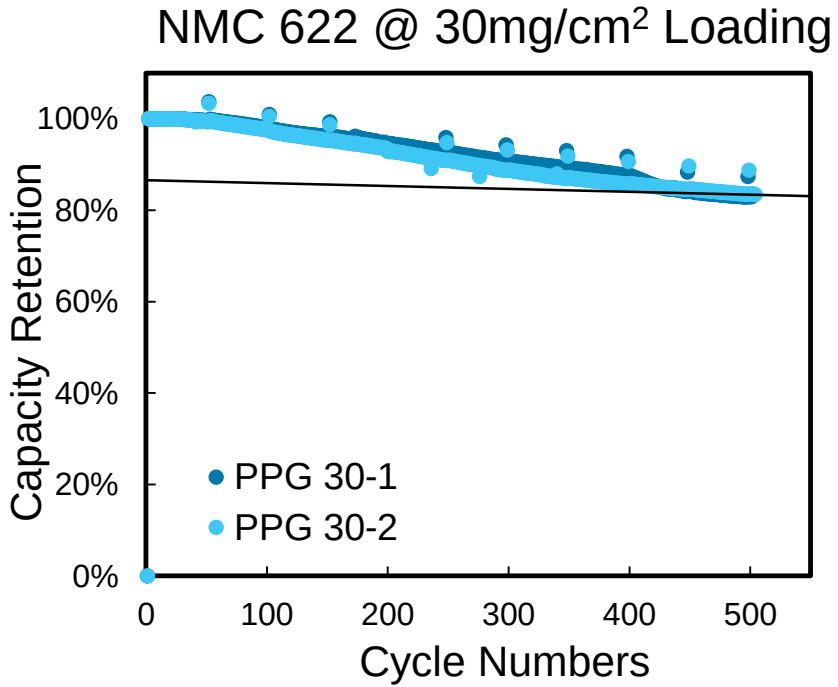
- Successfully coated NMC 622 cathode with mass loading of 30 mg/cm^2 on roll to roll pilot line
- Peel strength > 66N/m, flexibility pass 1/8” R
- An anode with mass loading of 15.8 mg/cm^2 was fabricated to match the cathode, using CMC/SBR formulation
- First- Batch Pouch cells fabricated and being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



4. Technical Accomplishment:

- 1st Target Cathode (30 mg/cm²) Performance





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

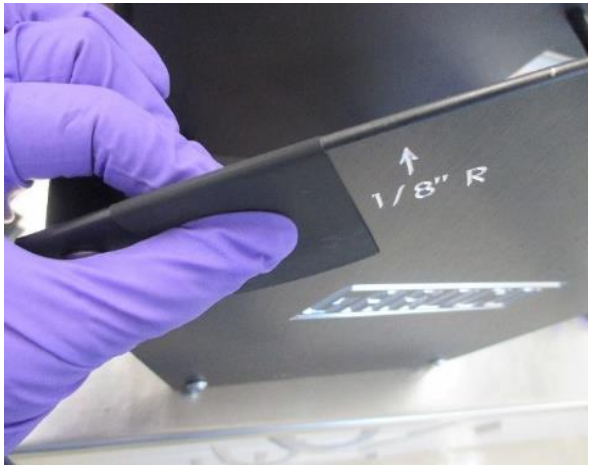
- 93/3/4 coatings and cells fabricated at UMEI. Finished testing at BIC under C/3 at 30°C
- At 500 cycles cells with PPG coating and NMC 622 was at 85% capacity retention
- PVDF/NMP control cathodes of 30 mg/cm² cells were not able to complete the 100 cycles with 85% capacity retention

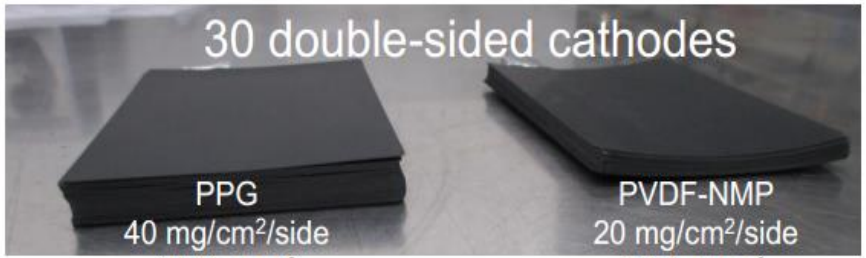
5. Technical Accomplishment:

PPG Thick Coatings with NMC622 – 40mg/cm^2

Job Description	Mass Loading	Areal Capacity (mAh/cm^2)
Hi-Ni 40mg/cm^2	40mg/cm^2	6.02
Hi-Ni 40mg/cm^2 with additives	40mg/cm^2	6.02

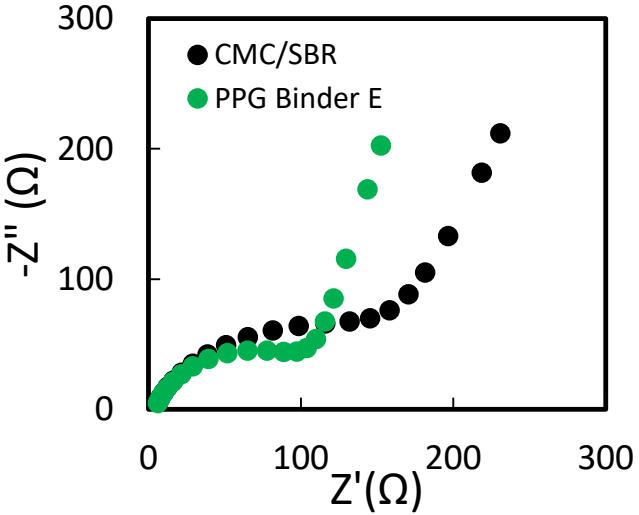
- Pilot scale roll-to-roll double-sided coated up to 40 mg/cm^2 per side
- Pouch cells fabricated
- Lab scale draw down up to 49 mg/cm^2
- Good flexibility - ½ inch
- Outstanding peel strength

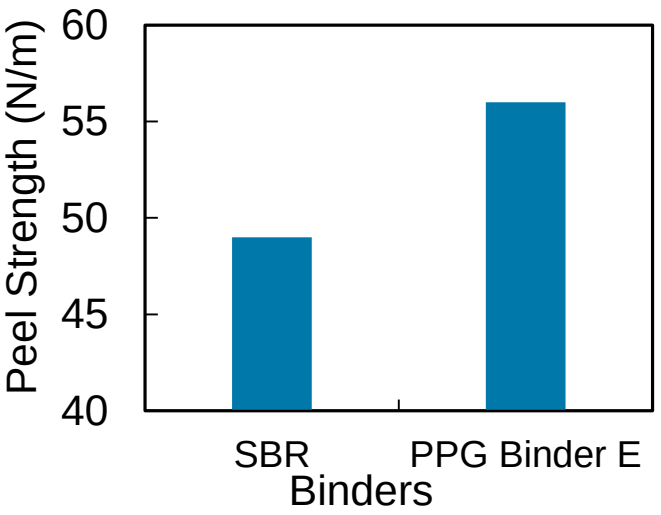


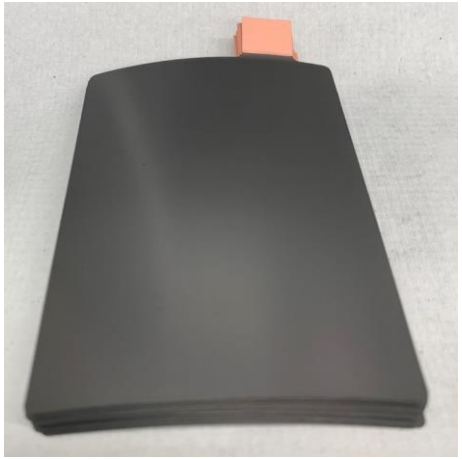


6. Technical Accomplishment:

- High Energy Density (7.5 mAh/cm^2) Anode Development



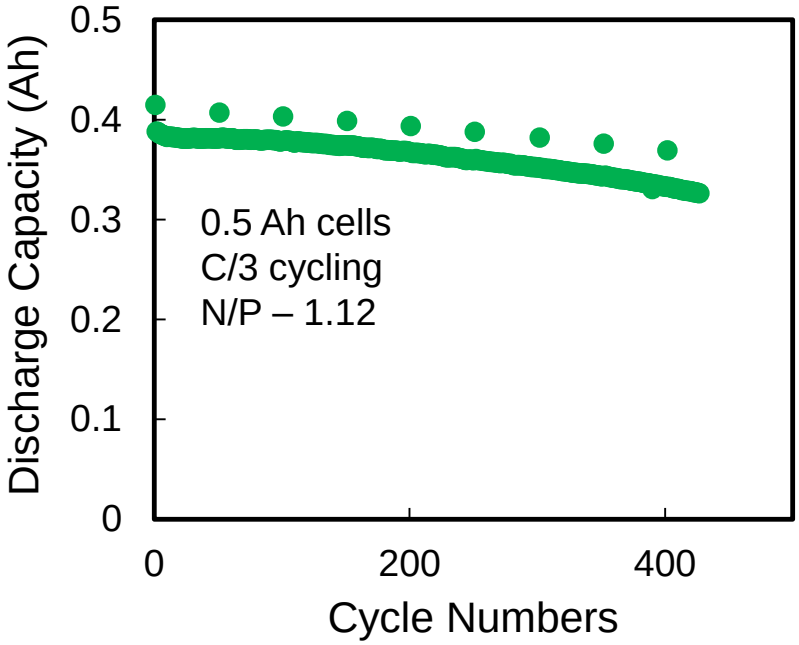


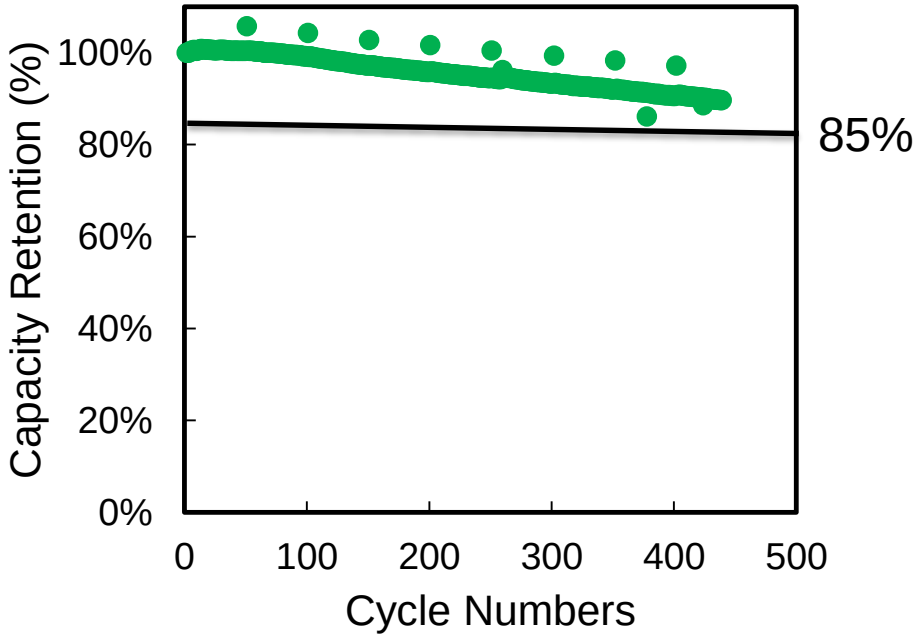


- PPG novel waterborne binder (PPG Binder E) outperform than CMC/SBR anode
- PPG successfully coated 22 mg/cm^2 (7.5 mAh/cm^2) anode at pilot scale

7. Technical Accomplishment:

- Final Target Cathode (40 mg/cm^2) Single Layer Pouch Cells (SLP)





- SLP of final target cathode and anode showed acceptable performance
- These cells completed 450 C/3 cycles with 90% capacity retention

8. Technical Accomplishment:

- Status for Final Target

- Final target cathode and anode successfully coated at pilot scale
- SLP of final target cathode and anode showed acceptable performance
- 2 Ah Multi-layer pouch cells were fabricated
- PPG will deliver these final target cells after initial cycling

Response to Previous Years Comments

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 2: The reviewer observed excellent progress on the process ability 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. PPG developed anode formulation at pilot scale UMEI facility.

Reviewer 3: The reviewer asked the difference between the baseline cell performance at PPG and deliverables.

PPG performed the root cause analysis and find out that the deliverable cells made in November trial at UMEI. In November trial, there was an issue of inappropriate mixing (smaller planetary mixer and lower mixing time) of cathode slurry and over-pressing of anode to lower porosity.

Collaboration

LG Chem	Cell performance target and 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.
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.

Remaining Challenges and Barriers

Final Target Cell Performance Evaluation

- PPG is optimizing the cell performance of final target cathode (40 mg/cm^2) and matching anode.

Future Research

Pouch Cell Testing with Anodes Designed for Thicker Cathode Coatings

- PPG assembled the final target cells but these cells didn't go for prolonged C/3 cycling due to lab shut-down from COVID-19.
- PPG will deliver final target pouch cell to INL for testing after COVID-19 situation is over and UMEI lab will come into normal operation.

*Any proposed future work is subject to change based on funding levels and COVID-19 situation

Summary

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

- ☑ Up to 150 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- >91% capacity retention after 1000 C/3 cycles
- ☑ Baseline pouch cell delivered to INL
- ☑ Cell evaluation by INL

Evaluation of 1st target pouch cell performance completed

- ☑ 1st target NMC622 cathode (30 mg/cm^2) with PPG binder meet the project goals
- ☑ Anode formulation of 1st target developed by PPG
- ☑ Pouch cells fabricated at UMEI met the qualification criteria

Final-target cells in progress

- ☑ Final target NMC 622 cathode at 40 mg/cm^2 cathode meeting project goals
- ☑ Anode formulation of final target (22 mg/cm^2) successfully coated at pilot scale
- ☑ Final-target pouch cell assembled at UMEI
- ❑ Final target cells will be delivered to INL for evaluation