Thicker Cathode Coatings for Lithium-Ion Electric Vehicle Batteries

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Vehicles Technology Office
2018 Annual Merit Review

Project ID: BAT357

Overview

Timeline

- **Project start date:** June 2017
- **Project end date:** June 2019
- **Percent complete:** 50%

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:**	$ 571,168
• **Funding for 2018:**	$ 666,141

Partners

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

1. Relevance - Objectives

Overall Objective

- **Increase cathode energy density** to 40mg/cm^2 while maintaining good 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

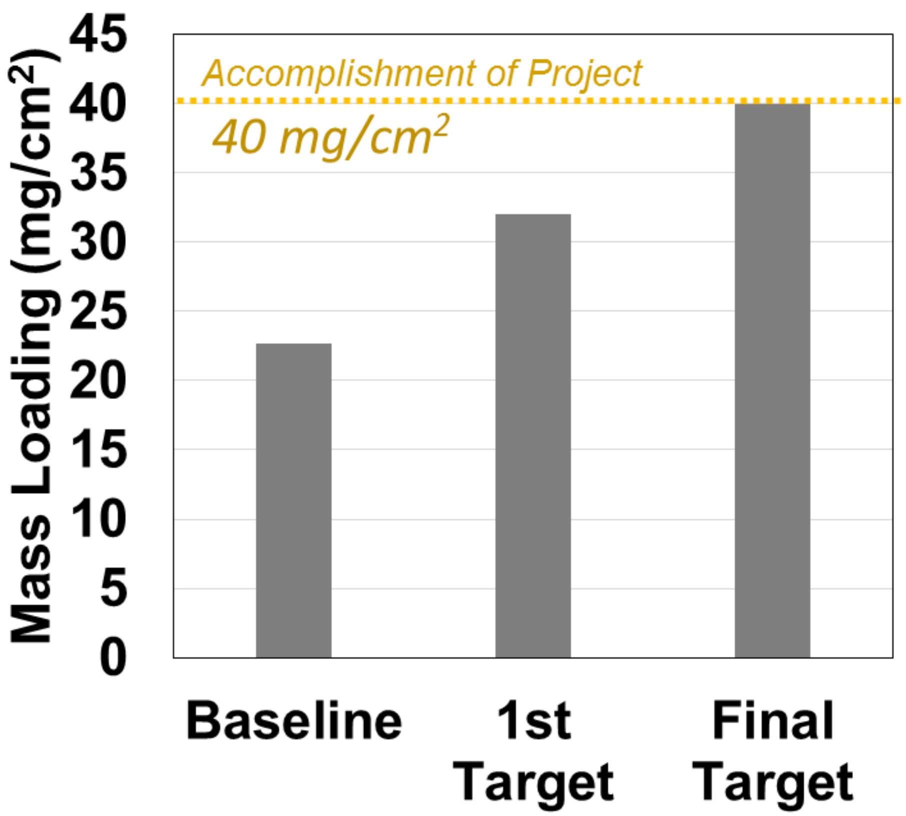
- **Design and improve NMP-free binder formulation to produce cathodes:**
  - Thicker
  - Flexible
  - Good adhesion
- **Evaluate pouch cell performance of cathodes made with these NMP-free binders**
- **Evaluate formulation made with high Ni-based cathode active pigments**

2. Relevance and Targets

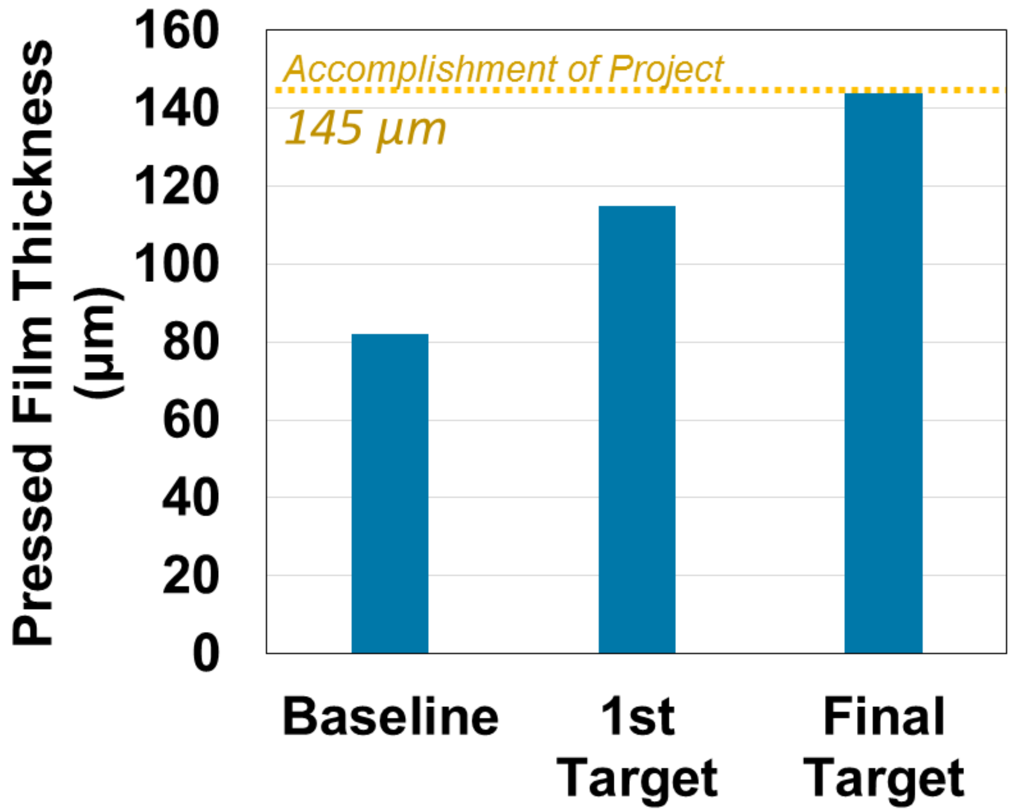
Impact

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

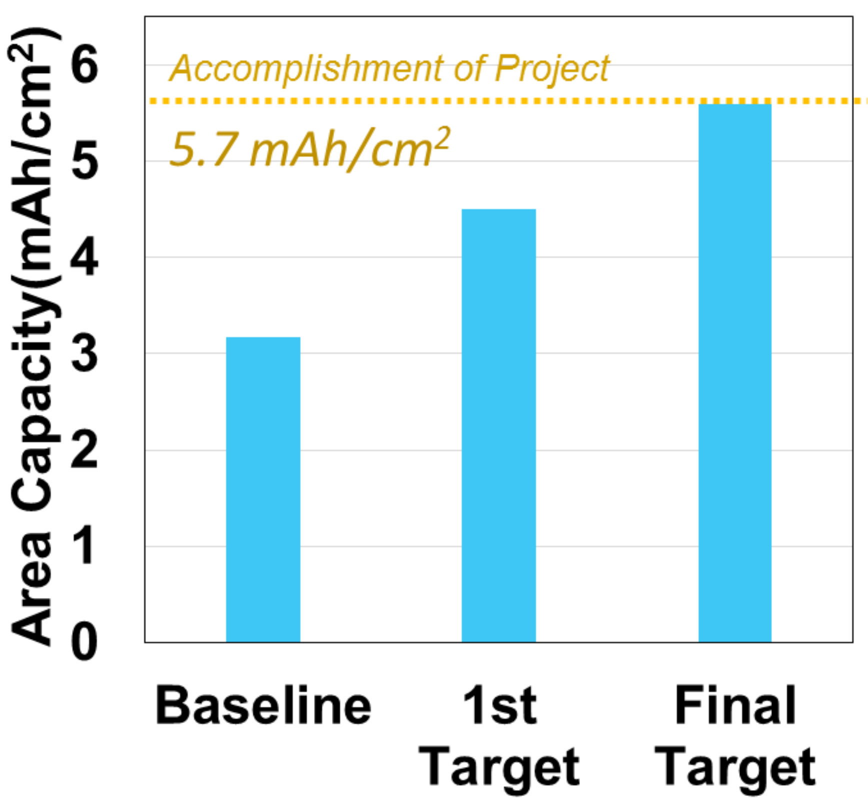
Mass Loading



Pressed Film Thickness



Energy Density



- NMC111: Carbon: Binder ratio 93:3:4
- Nominal Active Energy Density: 158 mAh/g

- Peel strength: 66 N/m (100 gf/15 mm)
- Flexibility: Pass 1/2” mandrel bend

Milestones

Date	Milestone	Status
---	Baseline solventborne data obtained	On going
2Q2018	110µ solvent-borne coatings meeting target coating performance	Finished
3Q2018	Electrochemical performance data obtained on 110µ coated electrodes obtained	On track
3Q2018	110µ electrodes meet minimum electrochemical performance targets	On track
3Q2018	150µ coatings meeting target coating performance	Finished
4Q2018	Electrochemical performance data on 150µ coated electrodes obtained	On track
2Q2019	Waterborne baseline data obtained	On track
3Q2019	110µ waterborne coatings meeting target coating performance	On track
4Q2019	Electrochemical performance data on 110µ waterborne coated electrodes obtained	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

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 cathode slurry with high solid content.
  - Easy to process rheology property for coating application.
  - 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:

- Baseline Cathode Coating

Coating

- Cathode made with PPG proprietary binder
- NMC-111 active
- Active:Carbon:Binder 93:3:4
- Slurry solids 72%
- Double-side coated tape via slot-die coater
- Coated over 100 linear feet
- Oven bake 74°C to 93°C with 4 zones

Coating Properties

- Energy density 3.17 mAh/cm^2
- Approx. thickness 79 microns per side
- Coating weight 23 mg/cm^2 per side
- Pressed porosity 32%
- Peel strength 377 N/m (576 gf/15 mm)
- Flexibility Pass 1/8' mandrel bend

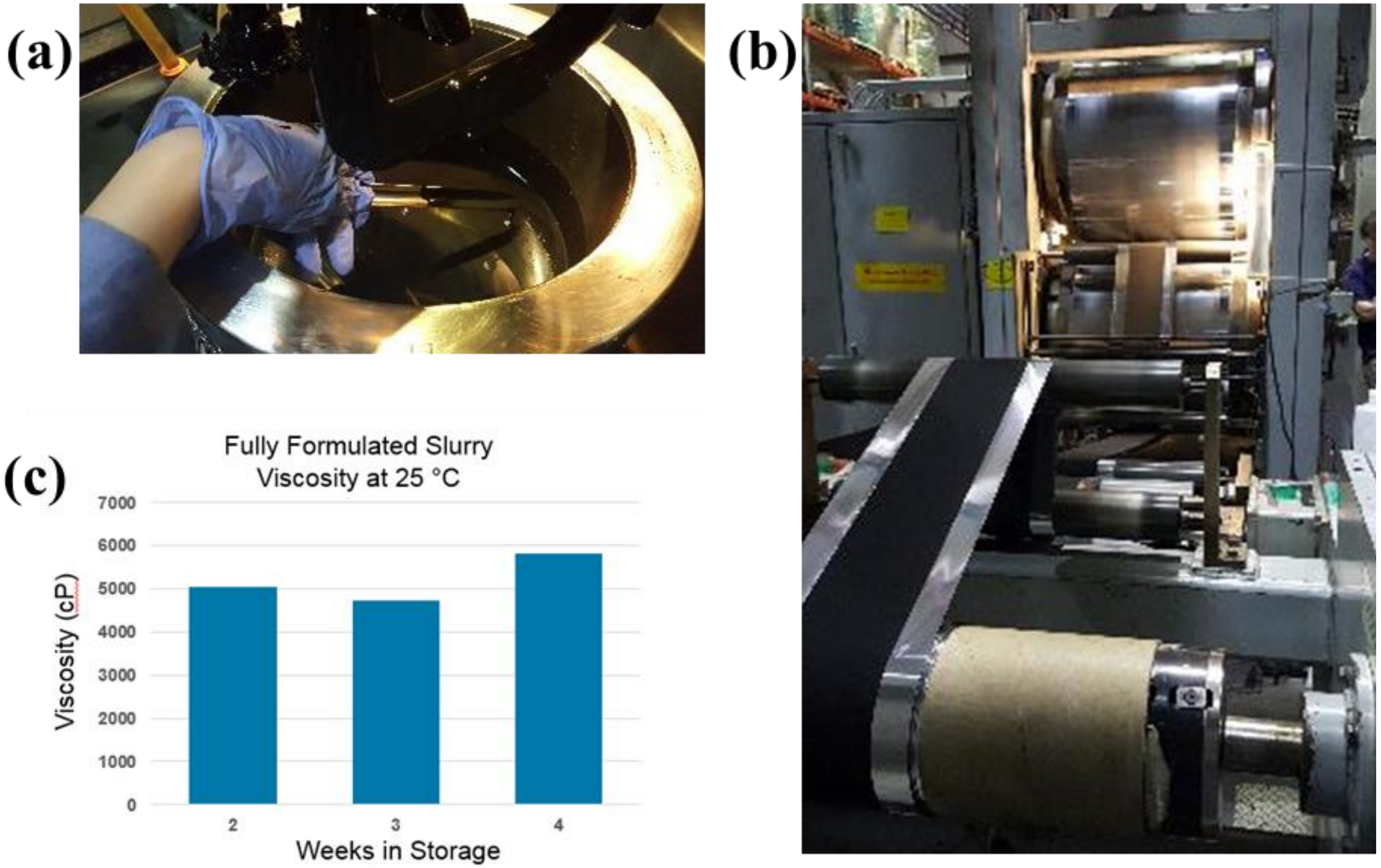


Figure: (a) PPG binder scale up. (b) Calendaring. (c) Shelf life stability test results. Mixing and coating done at Coulometrics.

2. Technical Accomplishment:

- Baseline Cathode Evaluation

LG Chem pouch cell testing

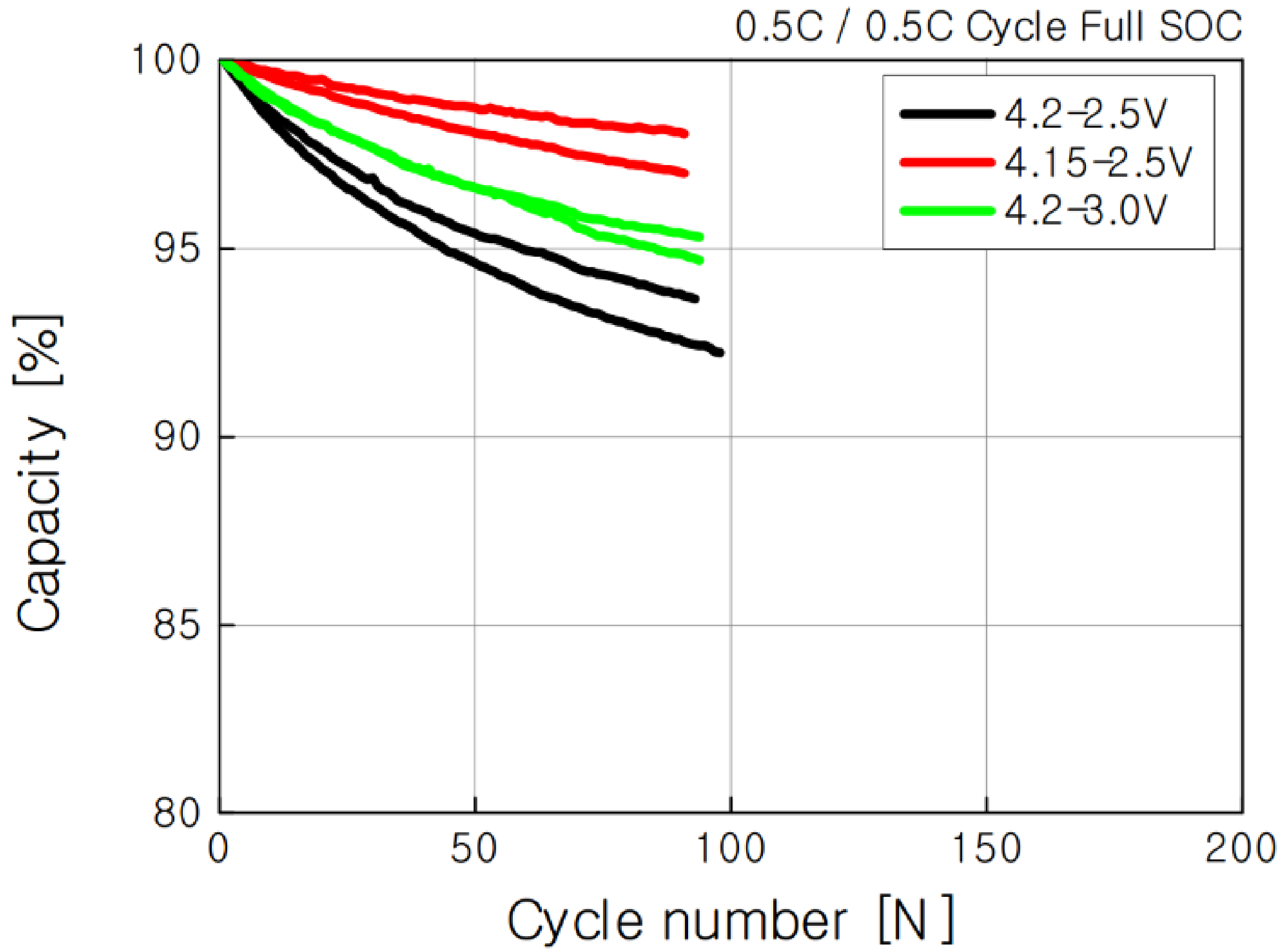


Figure: LG Chem pouch cell testing of cathode made using PPG Binder under different voltage window at C/2.

Possible Root Causes

- Binder side reaction
- Cathode-anode mismatch
- Aged active material
- Electrolyte additives side reaction
- Bad coatings made
- International shipping
- Moisture exposure

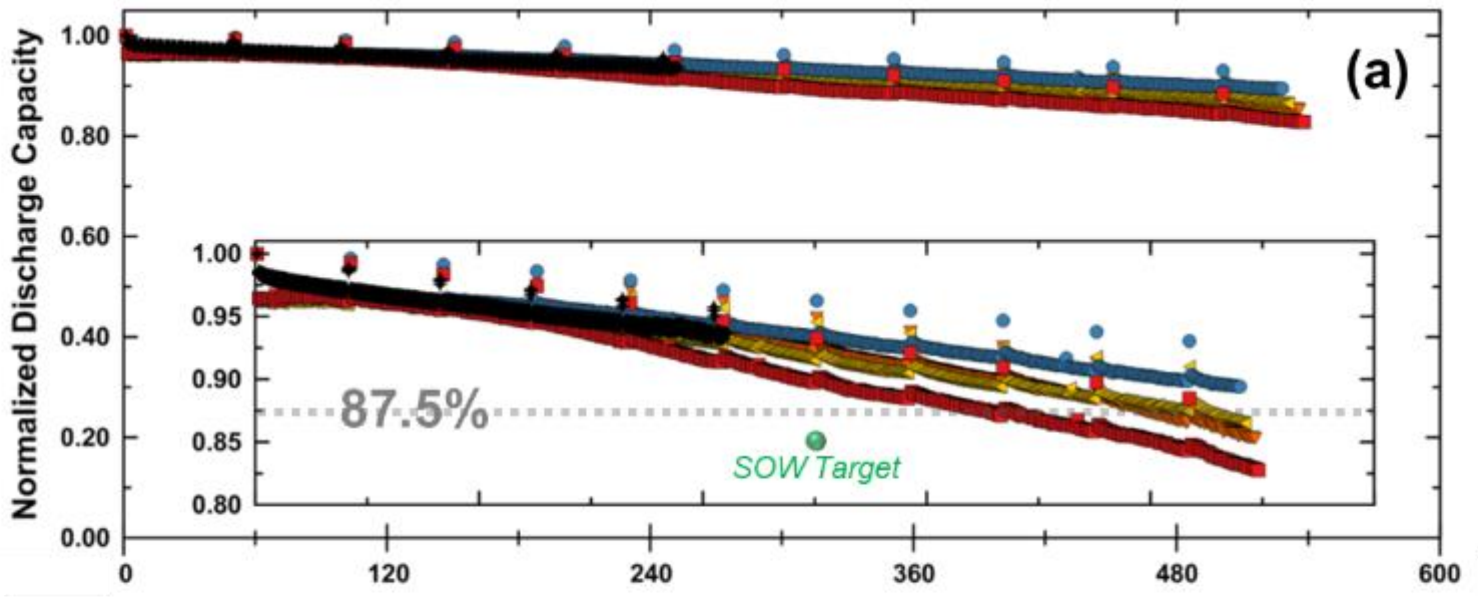
3. Technical Accomplishment:

- Baseline Cathode Repeat

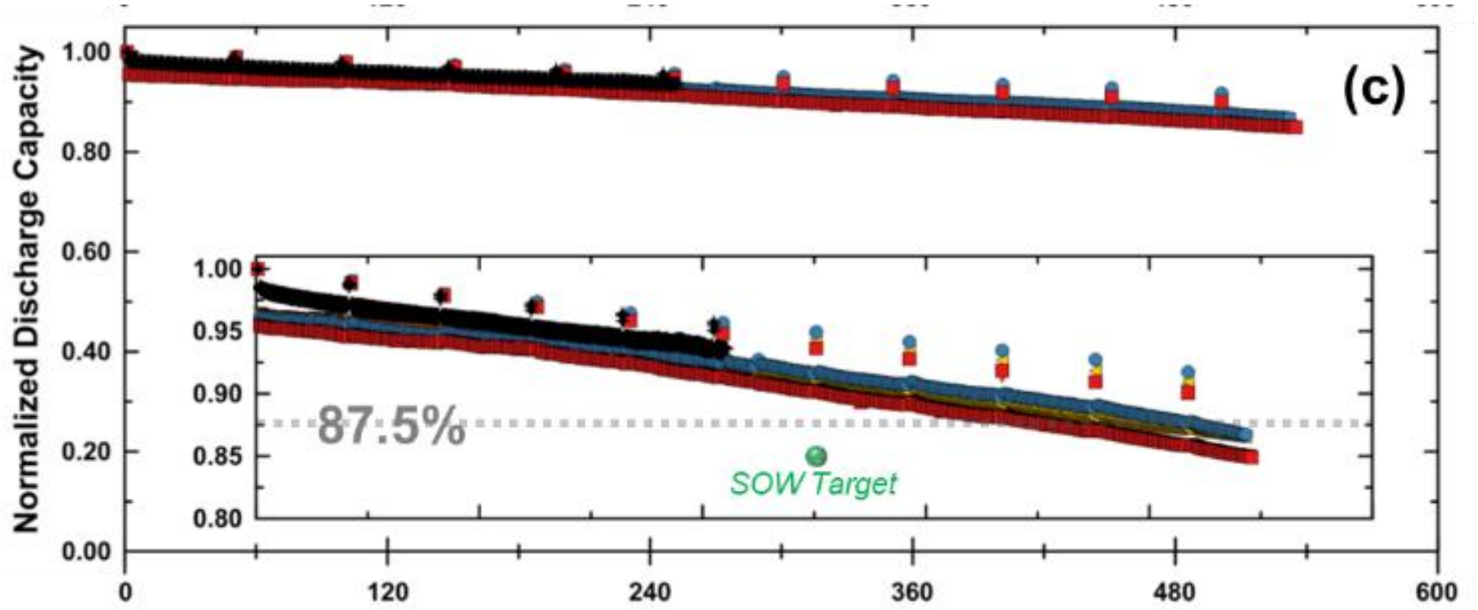
Investigating Root Causes with 18650 Cell Evaluation

- Repeat baseline at same areal density
- Anode was matched to cathode
    - 1.05*(reversible + irreversible capacity)
- NMC 111 fresh shipment from supplier
- Cathode coated, battery built and tested onsite instead of shipping coated foil internationally.
- Electrolyte contained additives common to Ni-based battery

Baseline with PPG Binder



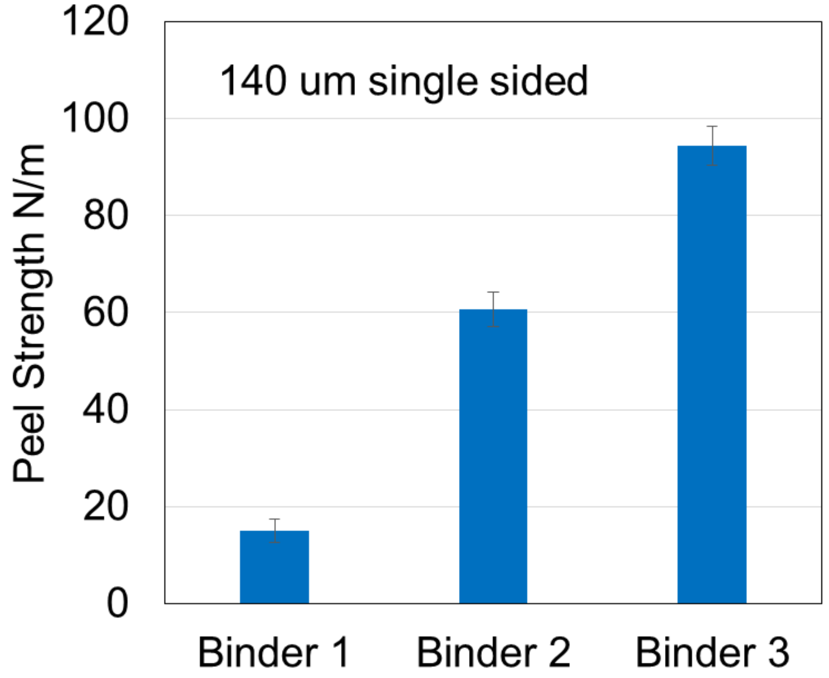
Baseline with PVDF NMP Control



5. Technical Accomplishment:

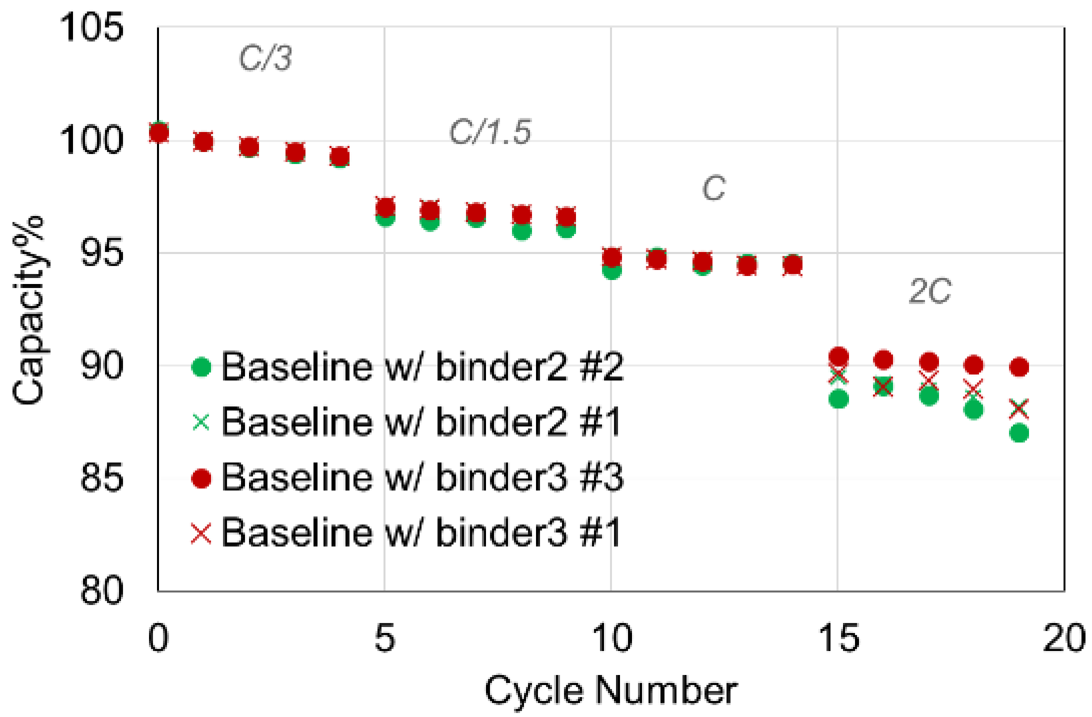
- Binder Optimization

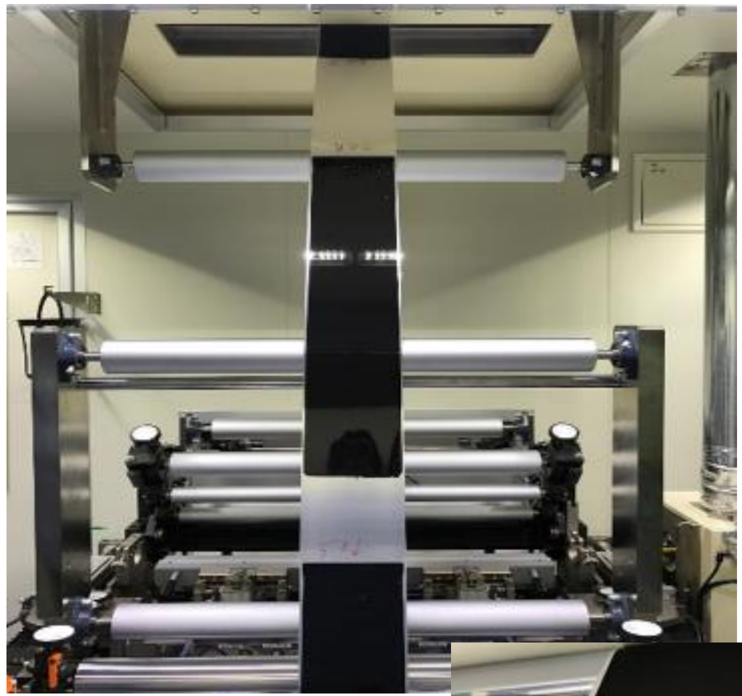
Effect of Binder on Adhesion

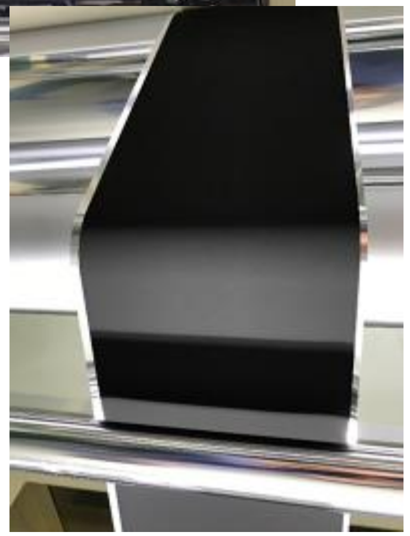


Rate Capability with 2.9 Ah Pouch Cells - Baseline (3.03 mAh/cm²)

Pouch cells are made at University of Michigan Battery Lab.







PPG can change binder to improve cathode coating adhesion and rate capability.

4. Technical Accomplishment:

- Cathodes to Reach Thick Coating Targets

PPG Binder	Areal Capacity	Peel Strength	Flexibility
1st TARGET	4.5 mAh/cm^2	66 N/m	1 inch bend
Current Status	5.3 mAh/cm^2	70±4 N/m	3/8 inch bend

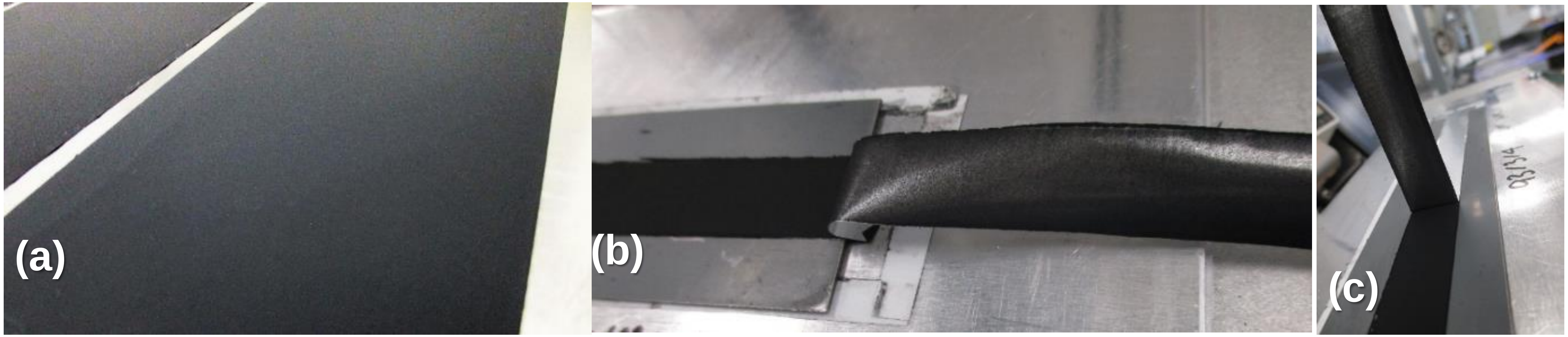
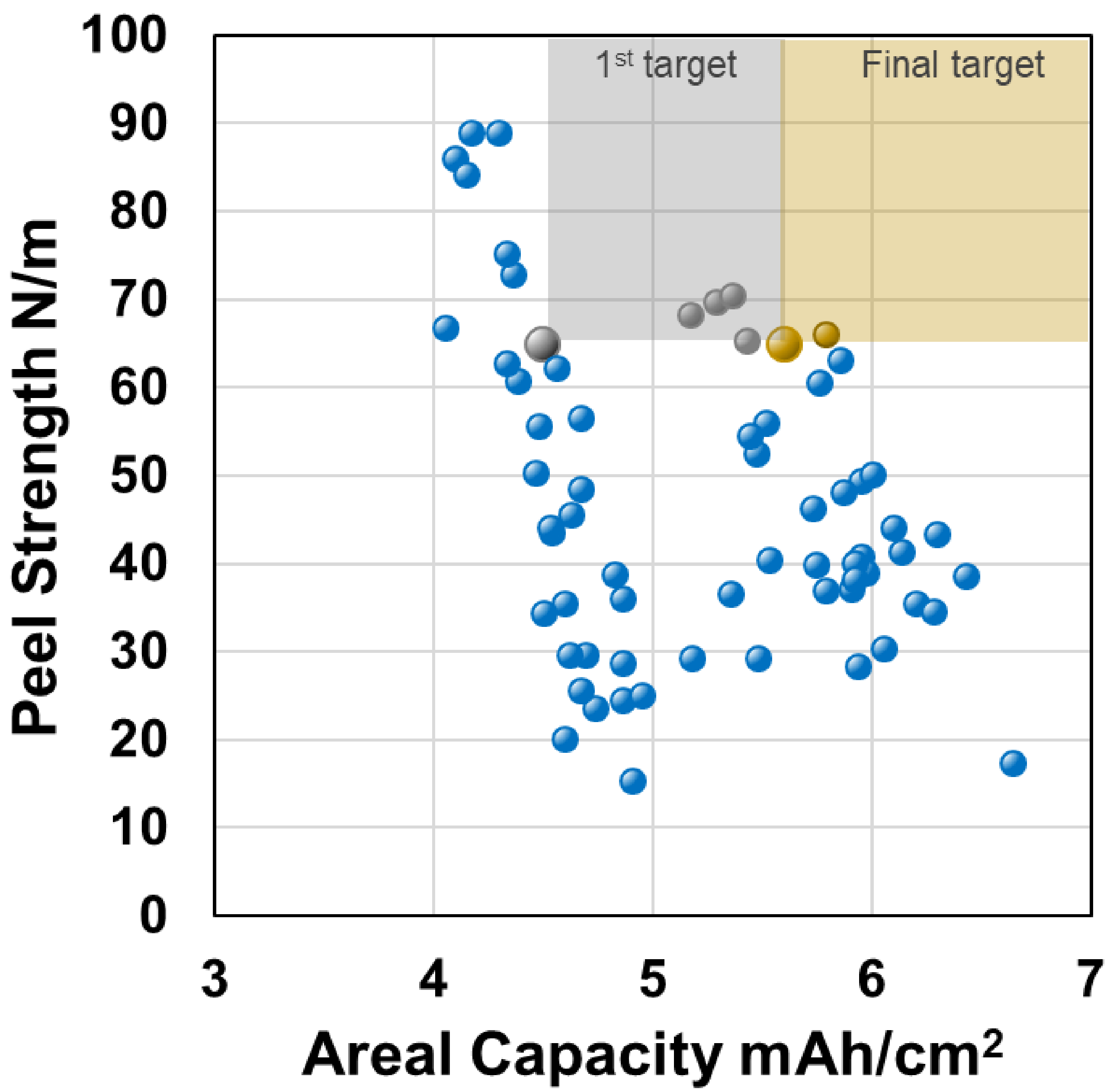


Figure: (a) Coating appearance, (b) Peel testing, (c) After peel.

6.Technical Accomplishment:

- Increasing areal density to 40mg/cm² cathode

Final Target Out of Cell Property Achieved by Q4 2017



PPG Binder	Areal Capacity	Peel Strength	Flexibility
Final Target	5.6 mAh/cm²	66 N/m	1 inch bend
Current Status	5.8 mAh/cm²	68 N/m	3/8 inch bend

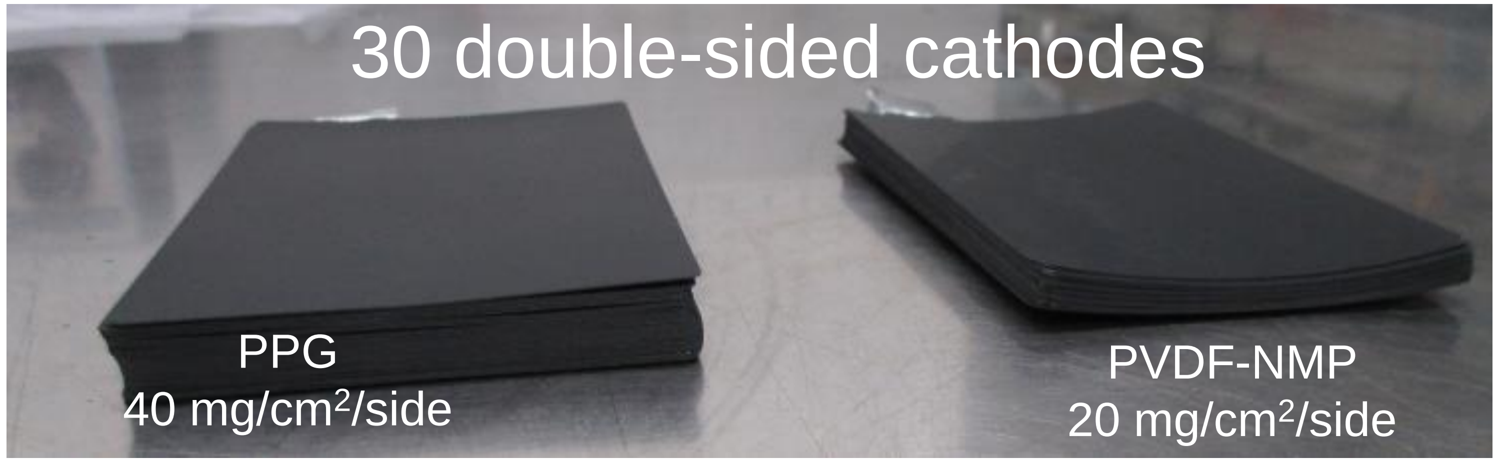
Actions to achieve target

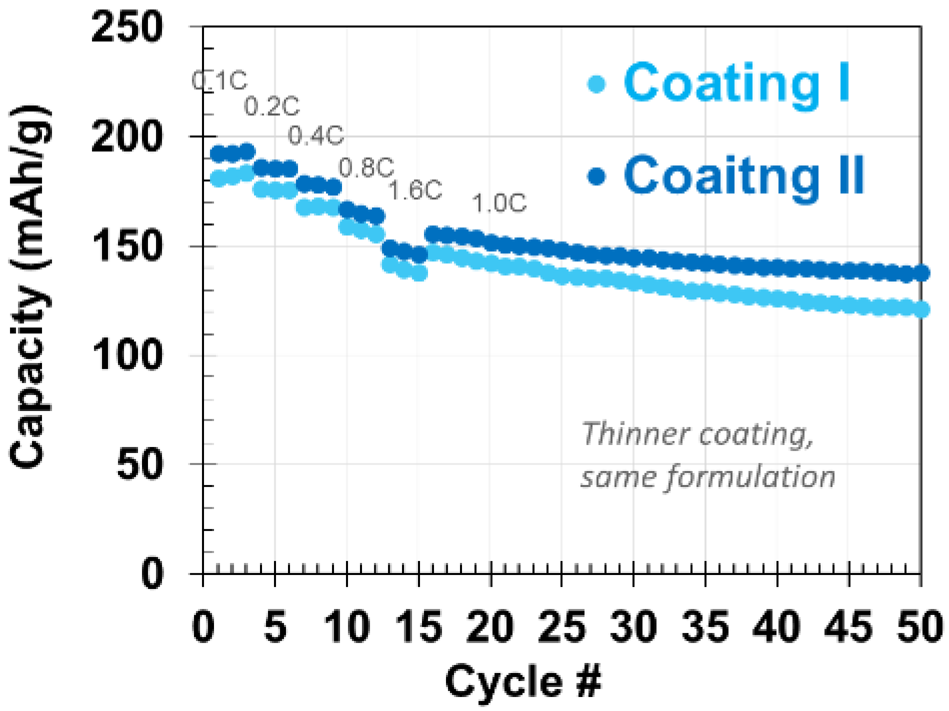
- Custom resin synthesis
- Binder formulation optimization
- Process controls

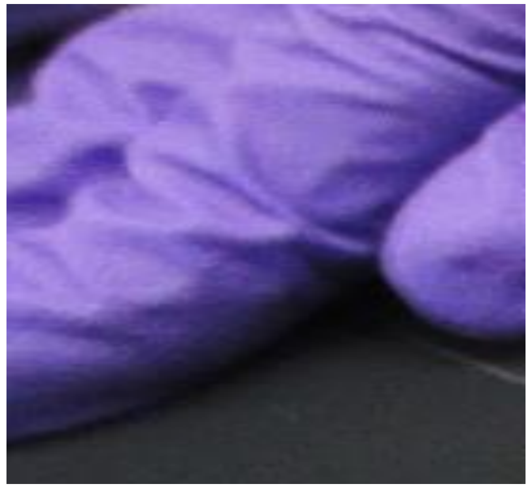
7. Technical Accomplishment:

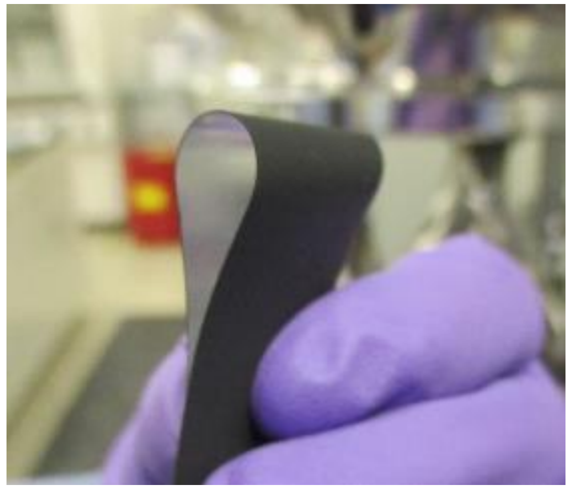
- High nickel cathode with areal density of 40mg/cm^2

Parameters	Coating I	Coating II
Active: Carbon: Binder	93:3:4	93:3:4
Carbon Type	Carbon I	Carbon II
Mass Loading (mg/cm^2)	37.4	39.3
Areal Capacity (mAh/cm^2)	6.95	7.31
Thickness (um)	130	136
Porosity	32-35%	32-35%
Peel strength (N/m)	67	98
Flexibility (Mandrel test)	¼ R	¼ R









Response to Previous Years Comments

This project is a new start.

Collaboration

LG Chem	Assembled baseline 1Ah pouch cells, provided feed back on cell performance. Conducted full pouch cell and single layer pouch cell performance characterization under different voltage windows.
Idaho National Laboratory	Provided feed back on project during quarterly meeting. Will be testing milestone for pouch cell deliverables with baseline and thicker coatings.
PennState	Join project in May 2018. Will be providing anode formulation for high energy density thicker cathode formulation and assemble pouch cells to screen cathode for deliverables to LG.
Michigan Engineering, University of Michigan	Provided facility for slurry mixing, coating, calendar press, and pouch cell assembly.
Coulometrics	Mixed and coated initial PPG baseline cathode coating. Evaluated PPG baseline cathode coating in 18650 cells.

Remaining Challenges and Barriers

Electrochemical Stability

- A discrepancy arose between battery performance with18650 cells and pouch cells made with baseline cathodes using PPG binders but assembled and tested at different locations.
- PPG is investigating the root causes for this discrepancy through extensive pouch cell testing. Evaluations include *in-situ* cathode stability, anode development, electrolyte effects.

Matching Anode for Thicker Cathode

- PPG has developed thick cathode up to 9mAh/cm^2 that pass ½ inch bend. However, thicker anode has not yet been developed for full cell evaluation.
- PPG has identified Penn State as a partner for thicker anode development.

Future Research

Pouch Cell Testing with Anodes Designed for Thicker Cathode Coatings

- Collaborate with Penn State to develop anode for high capacity thicker cathode.
- Screen cathode formulations at Penn State
- Optimized electrode pairs based on the feed back from Penn State testing.
- Validate performance of cathode and anode pair at LG Chem
- Final deliverable is pouch cell testing conducted at INL with cells from LG Chem

Thicker Coating Development with High Nickel Active Materials

- Apply and optimize NMP-free binders for high Ni active materials.
- Anode will be developed and electrolyte will be identified through collaboration with Penn State.

Waterborne Thicker Coating Development

- Resin design to optimize current waterborne cathode binder system.
- Identify water stable active materials for good battery performance.

Summary

PPG NMP-free binders enable manufacturing of high areal density cathode coatings

- ☑ Up to 145 um per side double sided coating with pilot-scale roll-to-roll coater
- ☑ Up to 40 mg/cm^2 mass loading
- ☑ Pressable to 32-35 % porosity

Good flexibility and peel strength for cathode coatings to meet final target specification

- ☑ Peel strength (90°) exceeding 66N/m.
- ☑ Flexibility for thicker double-sided coating passes ½ inches Mandrel bend test.

Evaluation of pouch cell performance ongoing

- ☑ Lab scale coin cell testing
- ☑ Pouch cell fabrication at University of Michigan accomplished
- ☐ Pouch cell testing in progress
- ☐ Anode development at Penn State as future work.

PPG