# NMP-Free Lithium-Ion Electrodes for Battery Manufacturing – Phase II

## Final Report

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# Acronyms and Abbreviations

| Term | Definition |
|---|---|
| Ah | Amp-hour |
| C | Capacity |
| CA | Conductive Additive |
| CMC | Carboxymethyl Cellulose |
| Co | Cobalt |
| CTMA | Commercial Technologies for Maintenance Activities |
| DOD | Department of Defense |
| EPMA | Electron Probe Micro-Analyzer |
| Fe | Iron |
| GVSC | Ground Vehicle Systems Center |
| Mn | Manganese |
| NCMS | National Center for Manufacturing Sciences |
| Ni | Nickel |
| NMC | Lithium Nickel Manganese Cobalt Oxide |
| NMP | N-methyl pyrrolidinone |
| ODASD-MR | Office of the Deputy Assistant Secretary of Defense, Materiel Readiness |
| PVDF | Polyvinylidene Difluoride |
| SBR | Styrene-Butadiene Rubber |
| SEI | Solid Electrolyte Interface |
| TEM | Transmission Electron Microscopy |
| UMBL | University of Michigan Battery Lab |
| U.S. | United States |
| Wt.% | Weight Percent, mass of compound/mass of composite |

# 1. Executive Summary

In Q4 of 2018, PPG initiated Phase I of a project to formulate N-methyl pyrrolidinone (NMP)-free cathodes for lithium-ion batteries capable of delivering sufficient power for automotive starting, lighting, and ignition as well as adequate charge capacity for powering auxiliary electronics. Batteries that meet both demands are of interest to the United States (U.S.) Army Ground Vehicle Systems Center (GVSC), specifically those with physical format and performance requirements documented in Department of Defense's (DOD's) "6T" specification (MIL-PRF-32565). In Phase I, NMP-free energy cathodes were formulated using developmental binders and NMP-free power cathodes were formulated via iterative refinement of carbon selection, carbon/binder ratio, and slurry mix procedure. In this project, learnings from the energy and power cathode development were conceptually combined in the formulation of capacity enhanced power cathodes.

In Q2 of 2019 PPG initiated Phase II of the project described above to continue formulation of NMP-free cathode binders that improve battery performance while reducing manufacturing costs. Phase II of this project was also initiated to develop a novel coating architecture that maximizes the performance of those formulated cathode coatings, while maintaining reduced manufacturing costs. A dual layer coating architecture was evaluated composed of an energy layer (96/2/2 NMC622/ Carbon 1/PPG Binder) and a power layer (85/9/6 NMC622/Carbon 1/PPG Binder). These stratified coatings should enable lithium-ion cells to display high power performance without sacrificing energy density. In addition to studying a dual layer system, the coating application method (wet-on-wet versus wet-on-dry) and the order of layers was also evaluated for cell performance and physical properties. The dual layer application method was optimized internally on the small scale and a pilot scale evaluation was completed at the University of Michigan Battery Lab (UMBL). In addition, PPG addressed the need for anode binders with improved performance and cost reduction. These PPG cathodes and PPG anodes were evaluated electrochemically via power capability and rate capability testing in battery coin cells, 20 mAh single layer pouch cells, and 1.1 Ah multi-layer pouch cells. The goal of Phase II was to provide superior materials to currently available coatings systems for use in both 6T format lithium-ion batteries and for advanced applications such as the Next Generation Combat Vehicle.

Funding was secured for the collaborative initiative through the National Center for Manufacturing Sciences (NCMS) Commercial Technologies for Maintenance Activities (CTMA) Program and the Office of the Deputy Assistant Secretary of Defense, Materiel Readiness (ODASD-MR).

## 1.1 Results

In Task 1.1, PPG assessed the ability of formulated coatings developed in Phase I to be employed to create a novel coating architecture composed of NMP-free lithium-ion cathodes on the laboratory scale. To complete this portion of the task, PPG developed a dual layer coating system composed of a power and energy cathode that evaluated two different coating architecture methods: wet-on-dry and wet-on-wet. On the laboratory scale, the coating application process parameters including applicating method, drying requirements, and coating quality were optimized. Analytical techniques verified a stratified coating architecture with increasing nickel (Ni) content near the electrode surface. PPG identified the wet-on-dry method to demonstrate the best cell performance from half-cell coin cells, full-cell coin cells, and single layer pouch cells.

In Task 1.2 PPG evaluated internal single layer pouch cell fabrication and testing capabilities. In these experiments, 20 mAh single layer pouch cells were compared to 1.1 Ah multi-layer pouch cells (produced at the UMBL in terms of formation efficiency and discharge capacity retention at discharge rates ranging from -0.3 to -9 Capacity (C). There was no difference in performance detected between internal or external cells, highlighting the quality of PPG's single layer pouch cell fabrication capabilities. Further, a low temperature electrolyte containing propylene carbonate (E4) was identified through a series of coin and single layer pouch cell screenings. This electrolyte was used to evaluate PPG cathode and anode formulations at discharge rates of -4C and -12C and at temperatures of -18°C and -40°C. The improved power performance of PPG cathode and anode formulations was supported by longer discharge durations under these conditions prior to reaching the lower cut off voltage when compared to control binder systems. While internal testing of these cathode binder systems yields promising results, attempts to scale the stratified coating architecture developed in Task 1.1 at the UMBL were unsuccessful due to limitations of their multi-layer coating capabilities.

In Task 1.3, PPG worked with the UMBL to produce a series of 1.1 Ah multi-layer pouch cells, including 9 formulated with standard electrolyte, and 9 formulated with low temperature electrolyte E-4. As uncovered in Task 1.2, UMBL was unable to produce pilot scale qualities of dual layer cathode foils, so a single layer cathode containing PPG binder was supplied instead. PPG developed anode binders were scaled with no issues and were also included in the delivered cells.

In Task 2.1, PPG tested its capabilities for screening new anode binders for high power performance beginning with the identification of proper graphite, conductive carbon additives, and control binders for power performance. Using these materials, a novel PPG anode binder was developed which displays equivalent cycle life and power performance to a CMC/SBR control. Additional insights were used to prepare an additional PPG anode binder which improves carbon dispersion, as supported by visual images, TEM microscopy, and oscillatory rheology was identified. This PPG binder was shown to improve fast discharge performance without impacting cycle life. In Task 3, PPG continued to employ and update models developed in Phase I of this work to validate the cost reductions achievable using novel cathode and anode coatings. In this phase of the project, PPG evaluated the cost analysis and modeling using the dual layer power and energy layer cathodes which contain the novel NMP-free binders. Once again, PPG discovered an NMP-free cathode system will require about 35% less solvent and results in about 58% energy reduction compared to an NMP system.

## 1.2 Benefits

Energy storage continues to evolve as advanced materials and battery architectures are developed to address the broad need for portable, on-demand energy for a wide variety of end use applications. Goals for improvements in these devices include life extension, energy density, safety, cost reduction, and charging speed. These goals are motivated from the fact that advanced energy storage technologies have become ubiquitous in our data-driven, electronic device-powered society. From transportation to medical devices, the ability to portably power electronics with the highest reliability and longevity at the lowest cost will continue to expand and support electrification efforts without sacrificing the performance of any components. The work in this project will advance materials designed for both electric vehicles and other electrical devices which require both high power and high energy applications, maximizing the batteries' utility and function. Electric vehicles in particular, can reduce U.S. dependence on petroleum feedstocks, reduce harmful air emissions, and have a lower cost to operate. Additionally, when rechargeable batteries replace a single-use battery, or secondary batteries relying on lead-based chemistries, less waste is generated, consuming fewer resources.

One major driver for electrification is decarbonization of systems, however environmental concerns within lithium-ion battery component manufacturing need to also be addressed. This project reduces the use of hazardous solvents in manufacturing, thus minimizing the carbon footprint of these processes. Importantly, by designing a new solvent and binder system for these components, efficiency and performance improvements of the cell can be realized, in addition to reducing the environmental impact. These performance improvements will increase the adoption of lithium-ion technology into high consumption and high-cost equipment such as vehicles, as well as smaller personal electronics. A key performance metric is the lifetime of battery systems, as longer lasting batteries require less frequent replacement, reducing the number of components placed in landfills. Enhanced energy storage capability supports the use of energy resources with lower environmental impact, increasing the overall ecological benefits already obtained through less hazardous manufacturing processes and judicious selection of precursor materials.

Performance capabilities of lithium-ion batteries include charge rate, discharge rate, and cycle life continue to improve; however, they are far from optimized. In addition, they remain costly relative to incumbent technologies, particularly in devices that require exceptional performance in one aera. Consumers will benefit from batteries that can take them farther on a single charge, power more devices with ease, and recharge quickly. Improvements in battery performance and cost will speed acceptance of electric vehicles and implementation of battery technologies into new applications, bringing the environmental and economic benefits to the nation.

Specifically, in the mobility sector, more efficient batteries with higher energy and power densities reduce overall vehicle weight, reduce hazards associated with vehicle damage, and potentially lower vehicle maintenance requirements. Novel materials that enable more tailored battery performance can expand the utility of the batteries, improving consumer confidence and enabling greater global competitiveness for manufacturers. These materials may also expand the possible operating conditions that lithium-ion battery electric vehicles may operate under: including temperature extremes and high current load operations. In particular low temperature, cold-crank performance improvements are expected to expand lithium-ion electric vehicle applications to include a wider variety of climate conditions.

Through this collaboration, industry will have access to new materials and electrode coating architectures that support battery manufacturing in the U.S. Industry will be able to produce batteries that deliver a balance of power and energy without impacting device lifetime. To support advanced energy storage technology for electric vehicles and other electrical device needs, this work specifically aimed to address challenges in preparing high power cells while minimizing energy losses. Typically, a cell would require specific changes to its cathode and anode formulation to target either power or energy, which ends up sacrificing one to strengthen the other. This work sought to address this challenge by studying novel strategies for mixing and coating battery components with new binder systems. Importantly, these modifications will be designed to be drop-in replacements with as low of a barrier for adoption as possible. This project continues to further sustainable cell manufacturing by demonstrating high power performance of cells with reduced hazardous solvent use as well as reduced energy consumption and cost during manufacturing.

In addition to high power/energy considerations, electrode formulations will be designed and tested with lifetime requirements in mind. Our solutions will be developed by first understanding the key role of each component in device lifetime, such that any innovations to improve sustainability and power/energy performance do not negatively impact device lifetime or require significant modifications to existing coatings processes.

Electrification is a core component of DOD modernization initiatives. In addition, the ability to power diverse components such as weapons systems, communications, and mobile, on-board silent power generation are critical. Reducing the military's dependence on petroleum-based fuel technologies supports the ability to conduct operational missions. Vehicle power and energy needs will only increase, and electrical energy storage solutions will need to increase with them. Lithium-ion batteries are a natural solution to these requirements. With their lower maintenance costs, lighter weight, and reduced hazards compared to petroleum-based fuels, their initial costs can be justified assuming they can be reduced below current costs. As the DOD looks for efficient and safe ways to address budget challenges, renewable energy sources that can improve the outcome of operations and warfighter readiness make sense.

NCMS is interested in developing advanced battery materials that can be used in lithium ion-based replacements for 6T lead-acid batteries. In addition, these materials can be employed in high-capacity batteries designed to support Army modernization efforts such as the Next Generation Combat Vehicle. In this project, PPG continued the evaluation of novel materials for lithium-ion cathodes, including cathode design and manufacturing. PPG also expanded the evaluation to advanced materials for lithium-ion anodes, with a goal of maximizing the performance benefits while reducing manufacturing costs.

### Key Accomplishments

- Novel electrode architecture design to maximize utility and function. Specifically, to provide a high-power cathode platform with a more balanced energy and power performance than existing designs.
- Cost reduction through formulation improvements and minimizing hazardous solvent use
- Provide superior materials to currently available coatings systems for use in both the 6T format lithium-ion batteries and for advanced applications such as the Next Generation Combat Vehicle.

## 1.3 Recommendations

Research into novel battery materials and architectures has improved performance and reduced overall manufacturing costs, these advantages must be proven feasible for full-scale manufacturing. This was done by gradually increasing the scale of prototype development. Phase I focused on the need for cell-level improvements in cathode battery materials. In Phase II, PPG continued evaluation of novel materials for lithium-ion cathodes which included design and manufacturing. PPG also expanded the evaluation of advanced materials for lithium-ion anodes, with the goal of maximizing the performance benefits while reducing manufacturing costs.

This work established a series of improvements to both cathode and anode formulations and coating architectures which have demonstrated improved power performance with a specific focus on improving the cold-crank performance of lithium-ion cells. In particular, the anode binder systems are easily scaled and deployed into graphite anode formulations. Further research is necessary to understand the interplay between these novel anode binder chemistries and silicon-containing anode formulations, as there are strong signals that commercial cells will continue to implement silicon into anode at increasing levels. Further, recommended areas of additional research could include investigation of fast charging for lithium-ion batteries. Fast charge provides an advantage in the field to power various military-based applications such as devices that soldiers carry with them, batteries in military vehicles for silent watch and/or silent mobility, etc. Throughout previous funding phases provided by the GVSC, PPG has a proven track record of reducing cell manufacturing costs without sacrifice to 6T performance, However, there is still critical work to be completed to improve fast charging of lithium-ion battery cells without significant degradation to the cell's long-term performance. We propose combining the benefits uncovered in this work of graphite anode binder systems with improved cold-crank performance with next generation silicon-anode formulations to fully pair the state-of-the-art energy density materials with novel binder platforms to support both high power discharge and charge.

## 1.4 Invention Disclosure

☒ Yes Inventions ☐ No Inventions

DD882 Invention Report sent to NCMS ☒

## 1.5 Project Partners

- U.S. Army Ground Vehicle Systems Center (GVSC)
- PPG Industries
- National Center for Manufacturing Sciences (NCMS)

# 2. Introduction

## 2.1 Background

Vehicle electrification is a high priority for the DOD as an approach to mitigate fossil fuel costs, improve logistical efficiency, and reduce noise during silent watch and silent mobility missions. In Q4 2018, PPG initiated Phase I to develop cathode coatings that deliver both high energy density and high power using PPG’s NMP-free binder technology. The purpose of Phase I was to utilize PPG’s NMP-free binders, in combination with its expertise in coatings technology, to develop cathode coating formulations that reduce cost and improve the performance of lithium-ion cells utilized in the 6T battery module for military vehicles.

In Q2 of 2019, PPG initiated Phase II of the project described above to continue formulation of novel coating architectures NMP-free cathode binders that improve battery performance while reducing manufacturing costs. The novel coating architecture was developed through stratified coatings which enable lithium-ion cells to display high power performance without sacrificing energy density. In addition, in Phase II, PPG addressed the need for anode binders with improved performance and cost reduction. Improved anode binder performance was realized through improved dispersion quality of conductive carbon. As a result, power performance at both room and reduced temperatures (cold-crank) was shown to be improved compared to standard binder systems.

## 2.2 Purpose

For Phase II, the project team continued evaluation of novel materials for lithium-ion cathodes, including cathode design and manufacturing. PPG will work on these advanced architectures while maintaining the use of NMP-free cathode binder platforms. As described in Phase I, NMP-free binders are expected to reduce manufacturing cost versus PVDF-NMP binders extensively utilized in lithium-ion batteries. Phase II also expanded the evaluation of novel materials to advanced lithium-ion anodes, with a goal of maximizing the performance benefits while reducing manufacturing cost. Studies of novel cathode and anode coatings and materials will provide an understanding of the ability to fine-tune battery cell performance characteristics. Based on performance data generated in Phase I, PPG will continue development of a novel coating architecture for cathodes, eventually expanding from laboratory scale to pilot scale production. In addition, advanced materials for anodes will be evaluated for their ability to facilitate improved charge and discharge rate, a critical need for many battery applications. Notably, end-use applications may include lithium-ion based replacements for 6T lead-acid batteries and high-capacity cells designed to support vehicle electrification.

## 2.3 Scope/Approach

This project was divided into three tasks.

### Task 1 – Cathode Coating Architecture

1.1 – PPG assessed the ability of formulated coatings developed in Phase I to be employed to create a novel coating architecture in NMP-free lithium-ion cathodes on a laboratory scale. Coating application process parameters, including application methodology, drying requirements, and coating quality were optimized. PPG characterized the electrochemical performance of these cathodes using GVSC specified testing protocols, working with partner participants as needed. Testing was accomplished in coin and small-format pouch cells.

1.2 – After suitable novel coating architecture was achieved, PPG assessed the feasibility of scaling the coating architecture from laboratory to continuous pilot scale. This was anticipated to be achievable using existing slot-die coating equipment with minimal modifications. Again, coating application process parameters were optimized to ensure that coating quality and battery performance properties were retained on scale-up. Partner participants were employed for scale-up capabilities and process optimization. Screening testing were accomplished using coin and small-format pouch cells, with larger pouch cells for process/electrode validation.

1.3 – PPG prepared sufficient cathodes using the NMP-free cathode formulation(s) and novel coating architecture to partner participants, with the goal of preparing 6 NMP-free deliverable cells meeting GVSC specifications. These were compared to 6 baseline cells prepared with standard NMP containing cathodes.

### Task 2 – Anode Binder Development

2.1 – PPG evaluated the use of novel formulated lithium-ion anode binders for improved cold-crank performance and ability to facilitate more rapid battery charging. Anodes developed in this task were graphite-based. Screening was accomplished using coin cell half-cells, with promising candidates evaluated using small-format pouch cells. Analysis was conducted to support the enhanced coating performance (flexibility, adhesion, temperature stability, etc.) and electrochemical performance (reduced impedance, charge rate, cycle/calendar life, etc.)

### Task 3 – Cost Analysis and Modeling

3.1 – PPG continued to employ and update models developed and employed in Phase I of this work to validate the cost reductions achievable using novel cathode and anode coatings.

# 3. Project Narrative

In Phase II, PPG led the formulation and optimization of NMP-free cathodes and anodes tailored for high power and high energy applications. Cathode and anode formulations were prepared, coated onto current collectors, calendared, and then used in the assembly of half-cell coin cells, full-cell coin cells, and single layer pouch cells which were subject to electrochemical analysis via rate capability and cycle life testing internally. Candidate cathodes and anodes exhibiting promising performance were selected for pilot scale evaluation of multi-layer pouch cell testing under conditions relevant to 6T specifications at the UMBL.

## 3.1 Task 1.1 – Cathode Coating Architecture

In Task 1.1, PPG committed to assessing the ability of formulated coatings developed in Phase I to be deployed to create a novel coating architecture in NMP-free lithium-ion cathodes on a laboratory scale. Coating application process parameters, including application methodology, drying requirements, and coating quality were optimized. PPG characterized the electrochemical performance of these cathodes using GVSC specified testing protocols and working with partner participants as needed. Testing was accomplished in coin and small-format pouch cells.

### 3.1.1 Application Method

Phase II focuses on the idea of a dual layer cathode coating. The dual layer coating is made up of a power layer and an energy layer. The power layer is a formulation containing 85 NMC622/9 Carbon 1/6 Binder B13H. The energy layer is a formulation of 96 NMC622/2 Carbon 1/2 Binder B13H. This dual layer cathode coating can be applied in one of two ways. The first application method is a wet-on-dry method which applies a layer (power or energy) and then dries the film. Then, the second layer is applied directly on top of the first layer, and the film is dried again. The second application method is a wet-on-wet method. In this method, a layer is applied (power or energy), and the second/other layer is applied immediately onto the first layer. Then the dual layer system is dried one time.

Both application methods utilize the same energy on power on current collector application with an energy: power ratio of 2. This ratio represents the areal capacity loading of both electrodes, the total capacity of these electrodes is 2 ~ 2.4 mAh/cm$^2$. To further understand the dual layer coating system. The schematic shown below further outlines a dual layer system and the corresponding formulation properties including total solids, coat weights, and capacities (Figure 1).

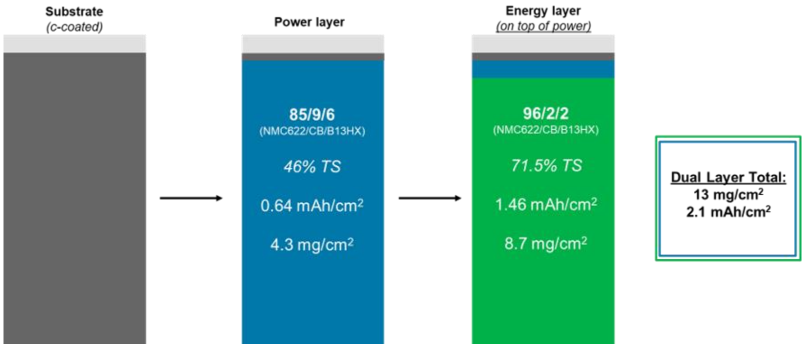


***Figure 1. Representative Dual Layer Cathode Coating on Carbon-Coated Substrate***
*The target loadings and coat weights used in the project are also shown*

### 3.1.2 Wet-on-Dry Application Method

First, PPG evaluated the wet-on-dry dual layer application method. In addition, the effect of layering scheme on cell performance was also evaluated. The layering scheme is denoted as “Layer 1|Layer 2”, where Layer 1 is the electrode formulation coated on the current collector, and Layer 2 is the electrode formulation coated on Layer 1. Samples of the dual layer cathodes with 85/9/6 NMC 622/Carbon 1/B13H as power (P) and 96/2/2 NMC 622/Carbon 1/B13H as energy (E) cathode formulations were pressed to approximately 20% porosity. Half-cell coin cells were assembled and subject to rate capability testing. For this cathode system (raw materials, and wet-on-dry application), the cells with the P|E layering scheme exhibited slightly superior rate capability when compared to the E|P layer scheme. This data is represented in Figure 2.

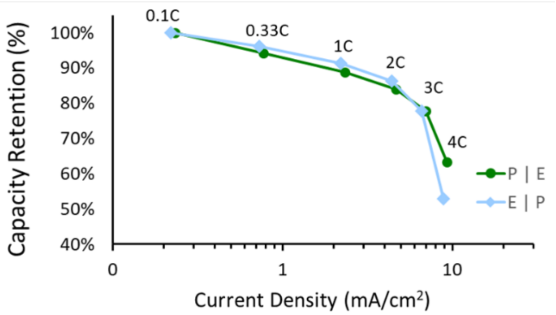


***Figure 2. Capacity Retention Plot Evaluating P|E and E|P Layering Schemes in Half-Cell Coin Cells***
*Higher rate capacity was observed at high discharge rates with the P|E (power on current collector) layering scheme*

To further evaluate the layering scheme, peak power testing was performed to determine whether this effect could be observed in testing comparable to 6T cold crank. Half-cell coin cells were assembled using dual layer cathodes with P|E and E|P layering schemes. Formation occurred over five cycles of 0.1C/0.1C charge to 4.3V/discharge to 3.0V. Peak power testing was performed in accordance with the United States Advanced Battery Consortium (USABC) test manual. Cells were charged again at 0.33C to 4.3V. After a 3-hour rest, cells were continuously discharged in ten consecutive increments comprising of a 30-second 4C discharge and 40-minute 0.1C discharge. Power delivered for each 4C pulse was calculated and normalized to the coating loadings. An advantage in power capability was observed in cells with the P|E layering scheme versus the E|P layering scheme, which is consistent with the rate capability data (Figure 3).

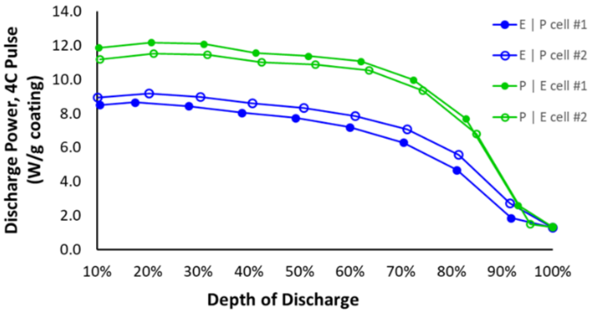


***Figure 3. Peak Power Testing Evaluation of P|E and E|P Layering Schemes in Half-Cell Coin Cells***
*Higher rate capacity was observed at higher discharge rates with the P|E layering scheme*

### 3.1.3 Wet-on-Wet Application Method

After the wet-on-dry method was evaluated, PPG evaluated the second application method, wet-on-wet. Once again, the wet-on-wet application method first applies the power layer. Then the energy layer is applied immediately onto the power layer. After the energy layer is applied, the dual layer system is dried.

For the wet-on-wet application, the dual layer cathodes were prepared using the modified power cathode formulation, 85/9/6 NMC 622/Carbon 1/B13H, and the energy cathode 96/2/2 NMC 622/Carbon 1/B13H. Samples were prepared using E|P and P|E layering schemes with areal capacities of 1.99 and 1.75 mAh/cm$^2$. The films were pressed to 20% porosity. Half-cell coin cells were assembled and subjected to rate and peak power testing.

For the rate capability testing, formation was performed using 5x 0.1C/0.1C charge/discharge cycles. In addition, an asymmetric rate ladder with 5x 1C charge and 5x 0.33C, 1C, 2C, 3C and 4C discharge cycles, followed by a cycle life 50x 1C/1C charge/discharge cycle was deployed. The specific discharge capacities were in-line with NMC 622, ~175 mAh/g. Higher capacity retention was observed with dual layer cathodes layered on the current collector as opposed to energy on current collector (Figure 4). In addition, the cycle life of half cells with dual layer cathodes prepared with P|E layering scheme exhibited 80% capacity retention after 80 cycles, whereas the cell with dual layer cathode prepared with the E|P layering scheme exhibited about 70% capacity retention after 80 cycles.

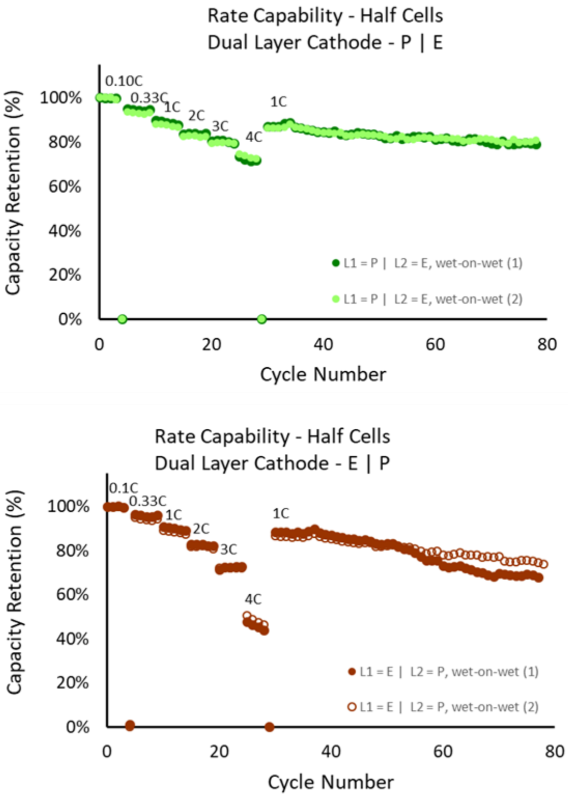


***Figure 4. Capacity Retention Plot Evaluating P|E and E|P Layering Schemes in Half-Cell Coin Cells for Wet-on-Wet Application Method***
*Higher rate capability was observed with the P|E layering scheme*

Peak power testing of half-cell coin cells assembled with dual layer cathodes was performed in accordance with the USABC test manual, as previously described. After formation, cells were charged to 4.3V at 0.33C, and then subject to a continuous discharge in 10 increments comprising alternating high-rate discharge pulse at 4C, and base current discharge at 0.1C. The discharge power was calculated for each pulse. Again, the superior power capability was observed with P|E layering scheme (Figure 5). Collectively, rate and power capability consistently show that coating a power layer on the current collector yields superior performance.

Both coating application methods (wet-on-wet and wet-on-dry) yield superior rate capabilities when the power cathode was coated on the current collector. This data is summarized in Figure 6.

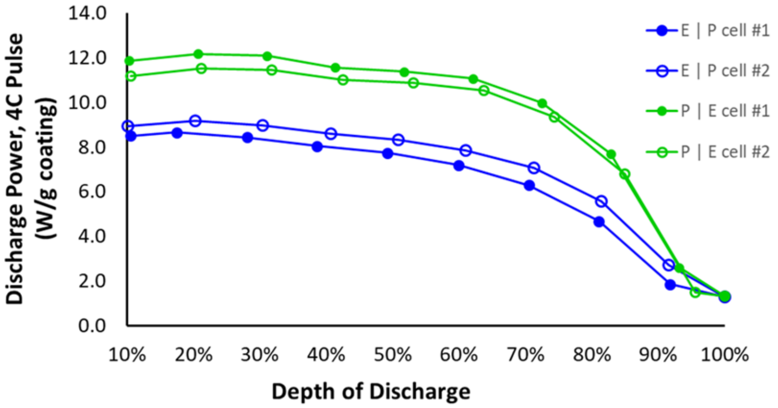


***Figure 5. Peak Power Testing Plot Evaluating P|E and E|P Layering Schemes in Half-Cell Coin Cells***

*The P|E layering scheme continues to outperform the E|P scheme*

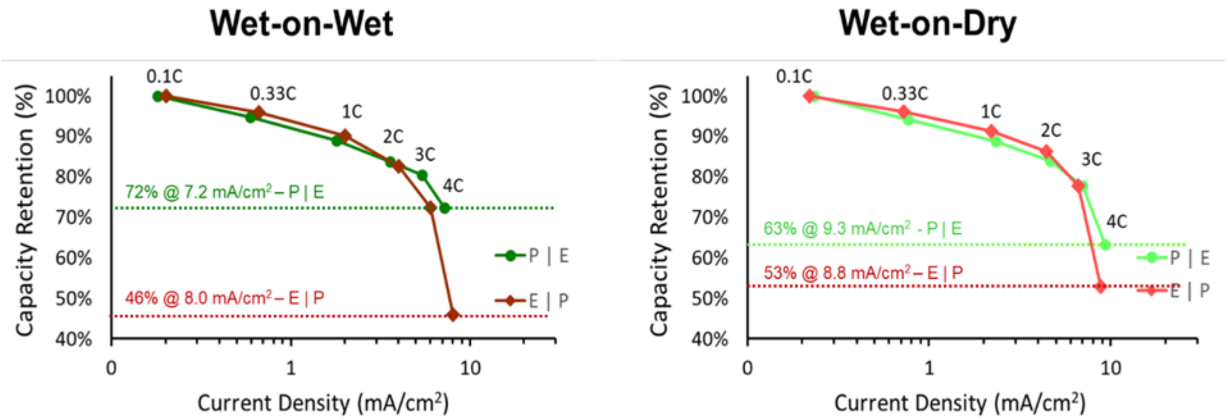


***Figure 6. Capacity Retention Plots Evaluating P|E and E|P Layering Schemes in Half-Cell Coin Cells for Each Application Method***

*Higher rate capability was observed with the P|E layering scheme for both application method*

### 3.1.4 Coating Architecture Evaluation

Rate capability and peak power testing both indicated that the Al|P|E (power cathode on current collector) layering scheme outperformed the Al|E|P layering scheme (energy cathode on current collector). This finding was consistent with test data from half cells assembled with dual layer cathodes prepared using both a wet-on-dry and a wet-on-wet application method.

PPG then began comparative testing of dual layer cathodes against single layer controls. New batches of dual layer cathodes (power cathode = 85/9/6; energy cathode = 96/2/2 NMC 622/Carbon 1/B13H) were prepared, along with a single layer control (92/4.5/3.5 NMC 622/Carbon 1/B13H). This single layer formulation has been utilized to prepare single layer control cathodes because it is a very close approximation to the experimental compositions of dual layer systems previously made with target E/P ratio ≈ 2. Films were coated to an areal capacity of 2 ~ 2.4 mAh/cm$^2$ (first charge basis) and pressed to approximately 20 ~ 25% porosity. Half-cell coin cells were assembled to evaluate and compare the cathode samples. Cells with dual layer cathodes having the P|E layering scheme exhibited improved rate capability (~55% capacity retention @ 6C) versus those with the E|P layering scheme and the single layer control (~30% capacity retention @ 6C) (Figure 7). This observation is consistent with previous testing where dual layer cathodes with the P|E layering scheme have consistently exhibited better performance.

To further compare the performance of dual layer cathodes versus single layer cathodes, the capacity retention data at 6C was also plotted against porosity. It was determined that the half cells assembled using dual layer cathodes exhibited superior capacity retention across the entire applicable porosity range (6C capacity retention of 76% at 57% porosity – 6C capacity retention of 62% at 20% porosity). The single layer cathodes exhibited a more dramatic decline in rate capability as a function of porosity (6C capacity retention of 75% at 60% porosity – 6C capacity retention of 45% at 27% porosity) (Figure 8).

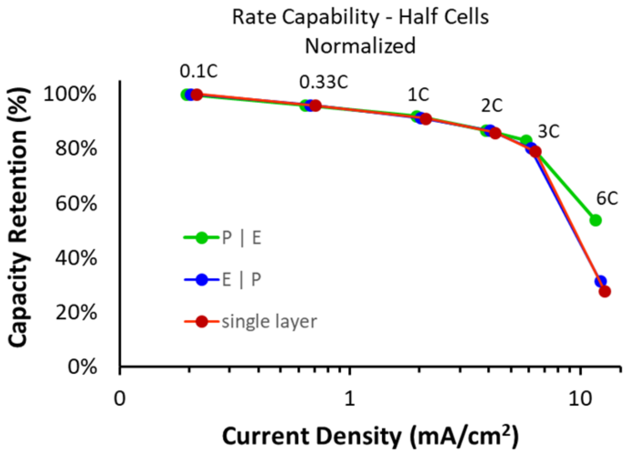
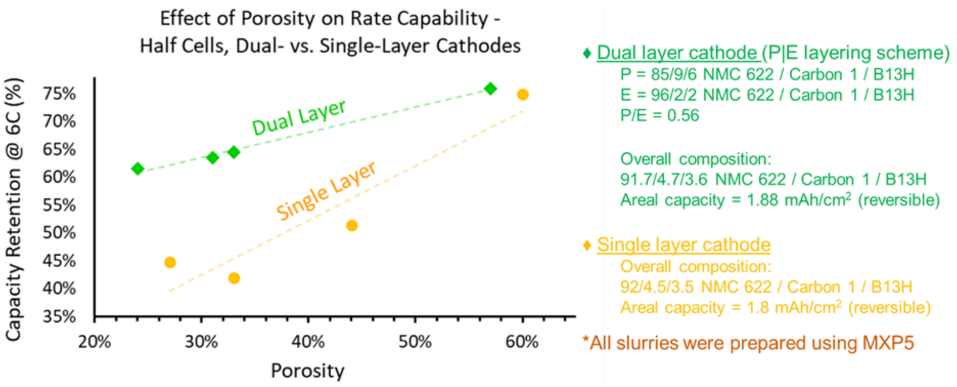


***Figure 7.   Rate Capability Results for P|E and E|P Layering Schemes and Single Layer Cathode in Half-Cell Coin Cells***



♦ Dual layer cathode (P|E layering scheme)
P = 85/9/6 NMC 622 / Carbon 1 / B13H
E = 96/2/2 NMC 622 / Carbon 1 / B13H
P/E = 0.56

Overall composition:
91.7/4.7/3.6 NMC 622 / Carbon 1 / B13H
Areal capacity = 1.88 mAh/cm$^2$ (reversible)

♦ Single layer cathode
Overall composition:
92/4.5/3.5 NMC 622 / Carbon 1 / B13H
Areal capacity = 1.8 mAh/cm$^2$ (reversible)

*All slurries were prepared using MXP5

***Figure 8.   Capacity Retention at 6C Compiled to Understand Effect of Porosity on Rate Capability in Dual Layer Cathodes Compared to Single Layer Cathodes***

The benefit of the dual layer architecture is much more apparent at high C-rates and lower cathode porosities. The data indicates that there is relatively good contact between the active material and carbon particles – this is likely due to highly effective mixing, carbon selection and/or binder facilitating good contact between the active material and the carbon.

The effect of porosity was further investigated on rate capability in full-cell coin cells assembled using PPG dual layer cathodes and a PPG graphite anode. Figure 9 demonstrates the impact of capacity retention and energy density that were plotted against porosity. The performance differentiation that was previously observed with the dual layer cathode in half-cells concerning porosity was not observed in the full cell data shown.

Full-cell coin cells comprising of dual layer cathodes and single layer control cathodes were paired with optimized PPG graphite anodes and were subject to rate capability testing (Figure 10). The dual layer full cells exhibited 40% capacity retention at 6C. The single layer cathodes only exhibited 25% capacity retention at 6C.

To further evaluate the effect of coating architecture, full-cell coin cells were assembled for peak power testing using a modification of the USABC peak power test. Cells were subject to continuous

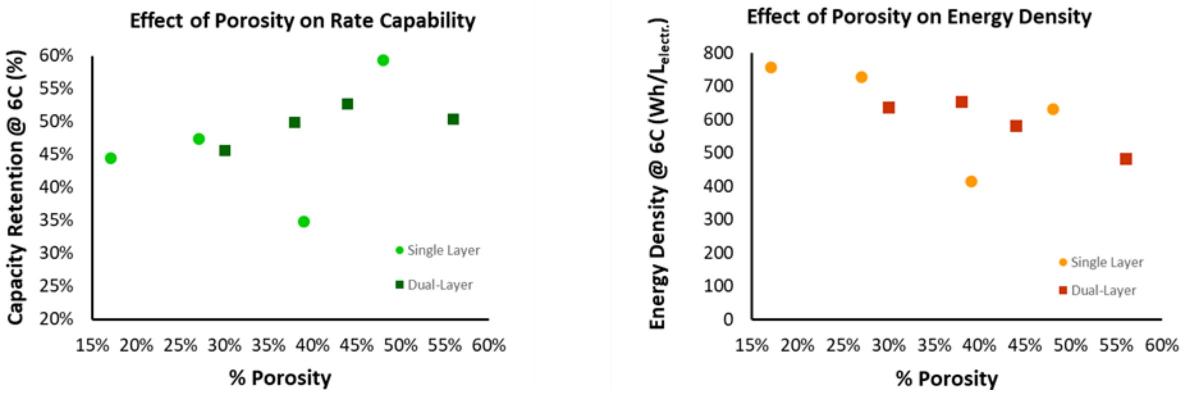


*Figure 9. Impact of % Porosity on Rate Capability and Energy Density for Dual Layer and Single Layer Cathodes Evaluated in Full-Cell Coin Cells*

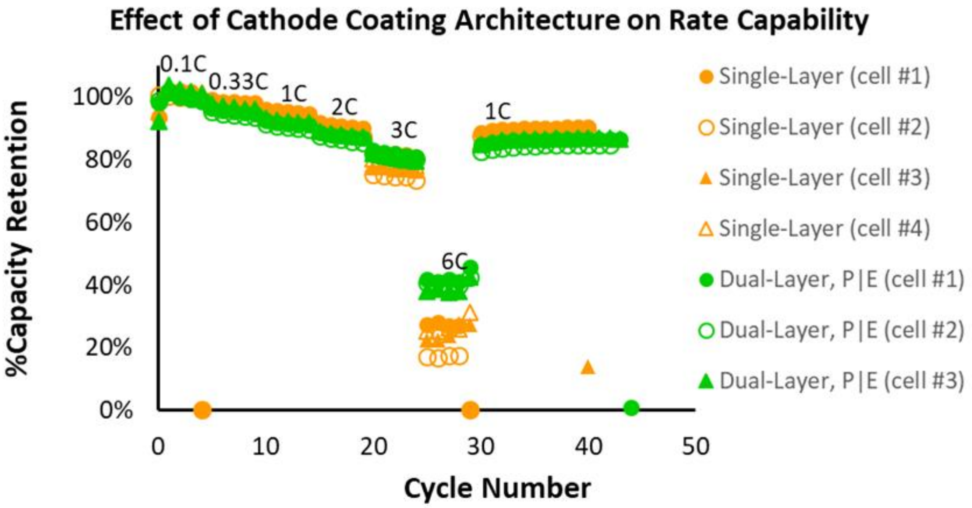


*Figure 10. Capacity Retention Plot Evaluating Full-Cell Coin Cells of Dual Layer System (P|E layer scheme) and Single Layer Cathode*

discharge divided into 10 increments comprising of a high-rate (6C) discharge of 5% of the cell's capacity and low rate (0.1C) discharge of 5% of the cell's capacity. Peak power was calculated at each pulse discharge increment as previously described. Data analysis revealed that cells assembled using the dual layer cathode exhibited approximately 30% higher peak power compared to cells assembled using single layer control (Figure 11).

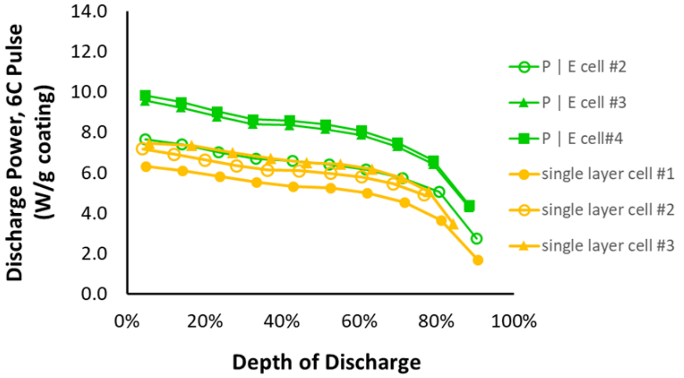


***Figure 11. Peak Power Testing Plot Evaluating P|E Dual Layer and Single Layer Cathode in Full-Cell Coin Cells***

*A higher rate capacity was observed with the dual layer cathode*

### 3.1.5 Compositional Depth Profile

The dual layer architecture was previously demonstrated by the difference in cell performance between the dual layer and single layer control cathodes. However, PPG wanted to further understand the compositional depth profile and gain insight into the extent of intermixing. Figure 12 demonstrates the different of types of intermixes that could have been occurring in the dual layer system.

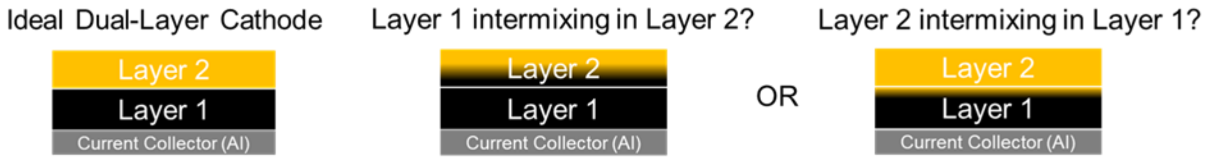


***Figure 12. Representation of Possible Intermixing That Could Occur in Dual Layer Cathode System***

PPG initiated a study using Electron Probe Microanalysis (EPMA) with the goal of determining the dual cathode composition as a function of coating height and evaluating the effect of cathode slurry viscosities on intermixing. PPG previously noted diminished differentiation between rate capability in cells assembled using dual layer cathodes compared with the single layer control in the second test-set of full cells with optimized PPG anode (Figure 14). This contrasted with the clear differentiation in rate capability of cathodes in the first test-set of full cells with optimized PPG anode (Figure 13). EPMA was used to gain insight into the role of coating composition on this occurrence. For reference, recall that for dual layer cathodes the power composition is 85/9/6 NMC 622/Carbon 1/B13H and the energy composition is 96/2/2 NMC 622/Carbon 1/B13H. The composition used for single layer controls is 92/4.5/3.5 NMC 622/Carbon 1/B13H.

Cathode samples from the first and second test-sets were provided to PPG Analytical Services in preparation for EPMA. Cathode samples were cross-sectioned and then shipped to Penn State University, where they were ion milled then analyzed via EPMA. Compositional data were reported

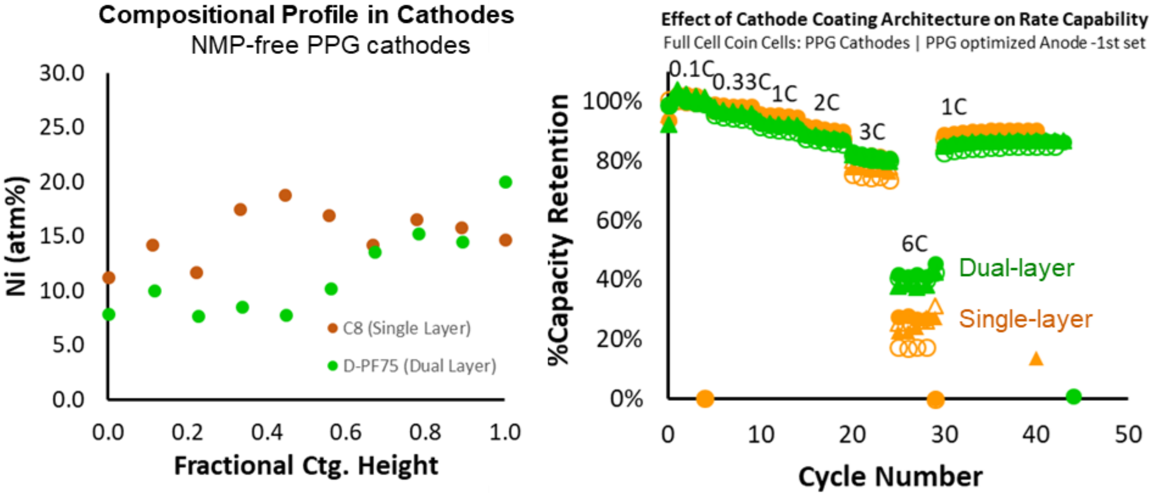


*Figure 13. First Test-Set: Compositional Profile of Single and Dual Layer Cathodes in Terms of % atm Ni and Fraction Height (left); Capacity Retention Plot of Dual Layer and Single Layer Cathodes (right)*

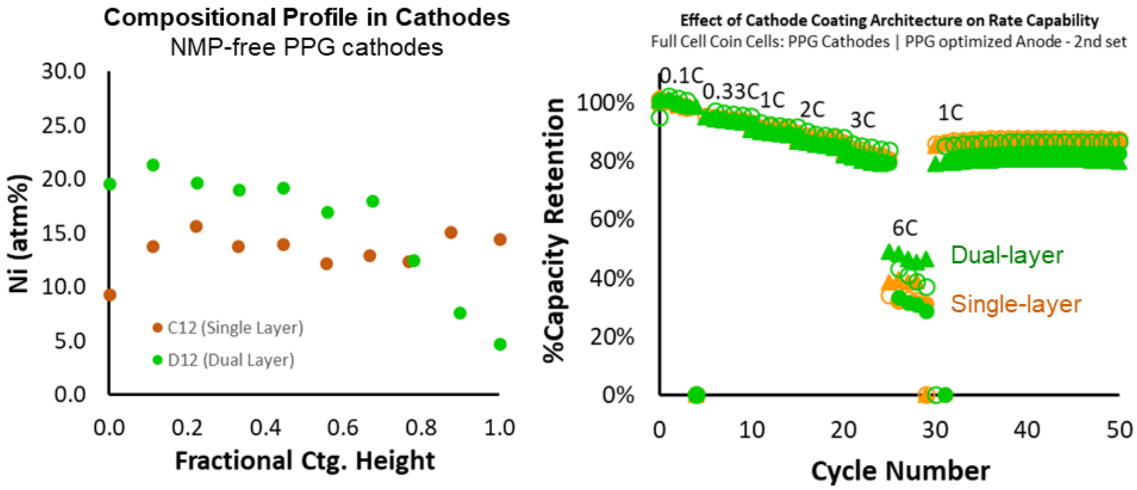


*Figure 14. Second Test-Set: Compositional Profile of Single and Dual Layer Cathodes in Terms of % atm Ni and Fraction Height (left); Capacity Retention Plot of Dual Layer and Single Layer Cathodes (right)*

in atm% nickel (Ni), cobalt (Co), manganese (Mn) and iron (Fe). Data work-up at PPG entailed generation of depth profiles by normalizing scan heights to total film thickness (fractional height) and plotting atm% of the elements of interest against fractional coating height. In these analyses a fractional coating height of 0 represents the coating touching the current collector surface, and a height of 1 represents the upper surface of the cathode coating. The first and second test set contained both a single layer and a dual layer cathode to analyze.

Cathode samples from the first and second test-sets were provided to PPG Analytical Services in preparation for EPMA. Cathode samples were cross-sectioned and then shipped to Penn State University, where they were ion milled then analyzed via EPMA. Compositional data were reported in atm% Ni, Co, Mn and Fe. Data work-up at PPG entailed generation of depth profiles by normalizing scan heights to total film thickness (fractional height) and plotting atm% of the elements of interest against fractional coating height. In these analyses a fractional coating height of 0 represents the coating touching the current collector surface, and a height of 1 represents the upper surface of the cathode coating. The first and second test-set contained both a single layer and a dual layer cathode to analyze.

In the set-comparison study, PPG compared the composition depth profiles of dual layer cathodes prepared for the first and second test-sets. In the first dual layer cathode sample, the Ni content remained nearly constant in a region of coating ranging from fractional height of 0 to 0.5, then dramatically increased up to the top of the coating (Figure 13). This observation is consistent with the P|E layering scheme (Layer 1 = power [P]|Layer 2 = energy [E]). This indicates the clear differentiation in rate capability. In addition, in the single layer system, the % Ni is approximately 15 atm%, highlighting the expected homogenous active material distribution in the single layer control.

However, this compositional profile was not observed in the second dual layer cathode, indicating that an extraneous occurrence happened during the coating process (Figure 14). This observation, coupled with the slightly lower Ni content, (indicating slightly higher carbon loading) in the single layer control could explain the diminished differentiation between dual layer and single layer cathodes in the second test-set (Figure 14). In this set, the single layer comprises just under 15 atm% Ni. In the dual layer, the % Ni is initially constant and then decreases. This is not consistent with the P|E layering but explains the diminished differentiation in the rate capability. These results further support the notion that a dual layer system outperforms a single layer cathode, but that intermixing may limit the reproducibility of dual layer preparation.

PPG also initiated a study to understand the effect of differences in the viscosity of Layer 1 (power) and Layer 2 (energy) on the compositional profile. To understand this, samples of dual layer cathodes were prepared with Layer 1 (power) at fixed viscosity with $LiFePO_4$ added as a marker, and Layer 2 (energy) coating at high, medium, and low viscosities (Figure 15). Compositional analysis by EPMA was performed as previously described. Due to scatter in the data, the effect of viscosity on compositional profile is inconclusive. However, for all three dual layer cathode samples, Fe content is observed to dramatically decline as scan height increases. This observation is consistent with the P|E layering scheme and provides direct experimental confirmation of compositional gradient in the electrode.

In addition, the effect of viscosity differences between Layer 1 (power) and Layer 2 (energy) were further probed with electrochemical testing. Half-cell coin cells were assembled using P|E dual layer cathodes with Layer 1 (power) at fixed viscosity and Layer 2 (energy) and high and low viscosities. An exceptionally high-rate capability (70% capacity retention @ 6C discharge) was observed in cells assembled using dual layer cathodes coated with a high viscosity Layer 2, whereas rate capability of the single layer control was substantially lower (40% capacity retention @ 6C discharge) (Figure 16).

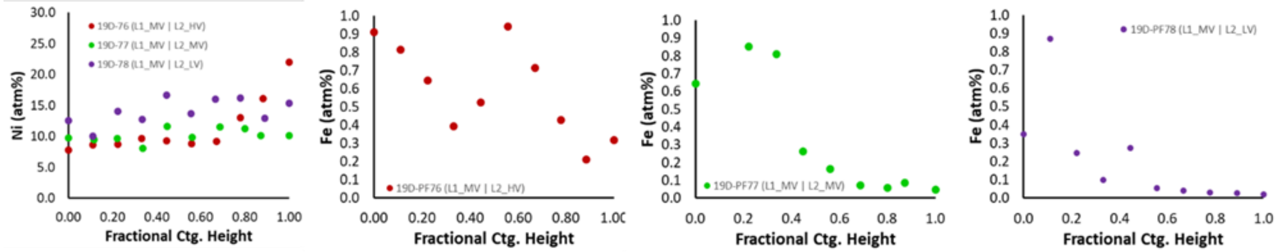


***Figure 15. Compositional Depth Profiles Generated for Ni and Fe for Dual Layer Cathodes Coated with Layer 1 (power) Having a Fixed Viscosity and Layer 2 (energy) at Low (red) Medium (green), and High (purple) Viscosities***

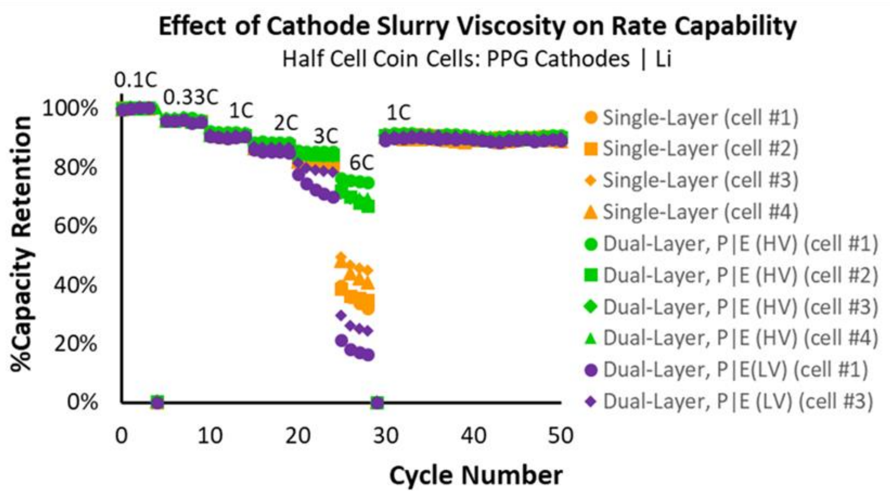


***Figure 16. Capacity Retention Plot of Half-Cells Assembled Using Dual Layer Cathodes Coated with Power Cathode Slurry at Fixed Viscosity and Energy Cathode Slurry Coated at High or Low Viscosity***

### 3.1.6 Final Internal Evaluation Prior to UMBL Pilot Trial

The wet-on-dry and wet-on-wet dual layer application methods were evaluated on the lab scale at PPG prior to the pilot trial at the UMBL. The optimized formulations, mixing procedures and dual layer application method were used. The target areal capacity for each cathode application was 2.1 mAh/cm$^2$. A single layer cathode at a formulation of 92 NMC622/3.5 Carbon 1/4.5 Binder B13H was also used for benchmarking purposes and to further understand the impact a dual layer system has on cell performance. The discharge capacity retention of the full-cell coin cells was evaluated over a series of discharge C-rates ranging from 0.3C to 12C (Figure 17). The full cell performance shows that the wet-on-wet application method demonstrates the lowest capacity retention, especially at 6C. The wet-on-dry and single layer cathodes demonstrate similar performance at each rate evaluated.

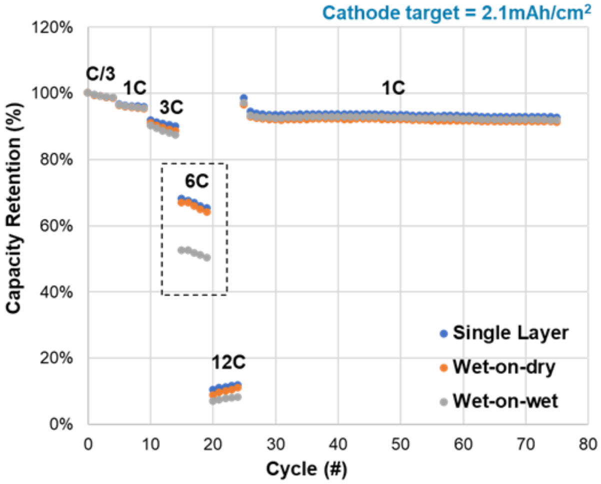


***Figure 17. Full-Cell Coin Cell Data of Dual Layer Applications Compared to Single Layer Cathode***

*The corresponding anode represent a 95/2/3 graphite/ carbon/binder (4d.4a) formulation*

The adhesion of the cathode films described above was also evaluated using a 90º peel strength method. The wet-on-wet application method demonstrates the highest peel strength; however, the error bars of the single layer and wet-on-wet method overlap with the results (Figure 18). This analysis suggests that the application method does not have a significant impact on the physical properties of the cathode electrode.

After validating the application method for the dual layer system (wet-on-dry), an internal scale-up to the kg scale on the PPG power layer cathode coating (85/9/6) was completed. At a small lab scale, the power layer was demonstrating a grainy texture on the coatings. The team wanted to understand the impact increased mixing capabilities (internal planetary mix scale-up) could have on the power layer coating quality and appearance prior to trial at the UMBL. The internal scale-up is referred to as a planetary mix and can accommodate up to 2kg of slurry. Images below demonstrate the improvement in coating appearance with high-speed dispersion on the planetary mixer (Figure 19). The grainy texture that appeared on the power layer coatings at the lab scale disappeared with the planetary mixing. The 85/9/6 formulation was successfully scaled-up with no particle aggregation or rheology issues (Figure 20).

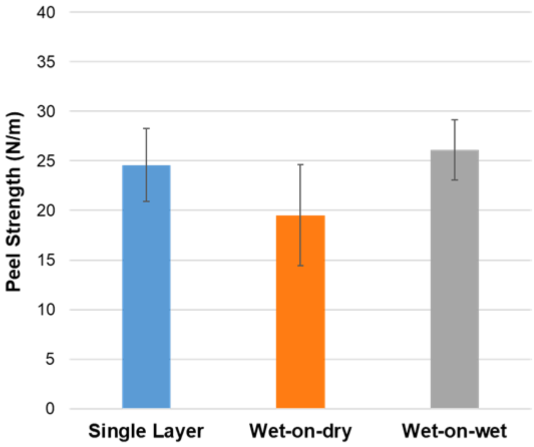


*Figure 18. Adhesion Results Via 90º Peel Strength for Each Application Method of PPG Phase II Cathode*

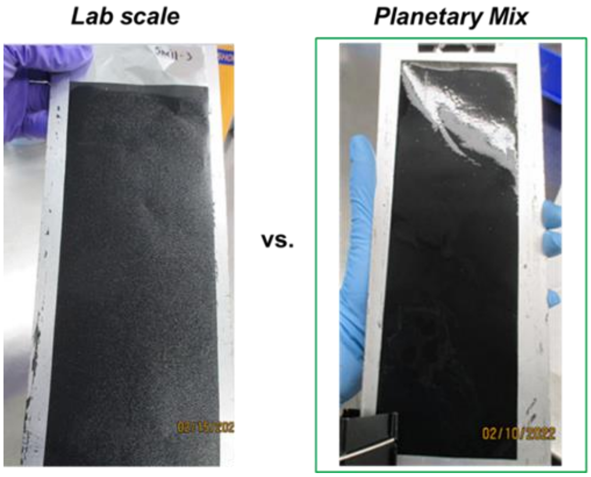


*Figure 19. Power Layer Cathode Coating Produced at Lab Scale Versus Larger Scale Planetary Mixer*

*Images are prior to drying*

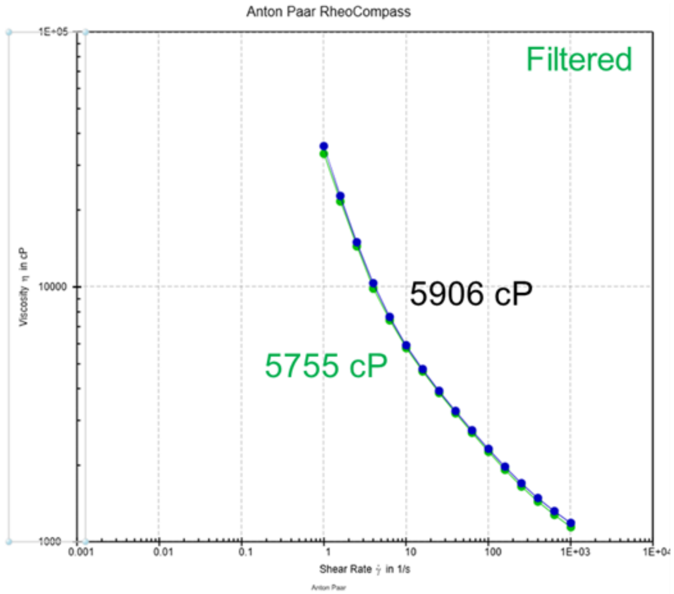


***Figure 20. Rheology Profile of Scaled-Up Power Layer (85/9/6)***
*Filtered slurry is shown in green and the unfiltered slurry results are shown in blue*

In addition, the rheology profile of the scaled-up power layer was taken (Figure 20). At a shear rate of 10s$^{-1}$, the filtered and unfiltered slurries from the planetary mix overlap almost perfectly, indicating that no significant carbon agglomeration is present, even with the higher wt.% of conductive additive present in the power layer formulation. The power layer demonstrates an acceptable and optimal rheology curve compatible with the coating capabilities of the UMBL given the % total solids of this formulation.

The corresponding cell performance data from the planetary mixer is shown in Figure 21. Qualitatively, large scale mixing significantly increases the appearance of the power layer compared to previous lab scale coatings. Full-cell coin cell testing has also verified that improved mixing capabilities provides improved cell performance, especially highlighted at 6C.

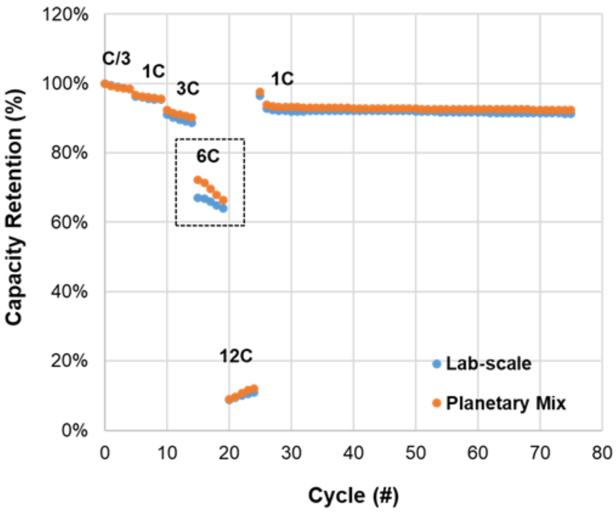


***Figure 21. Full-Cell Coin Cell Data of Power Layer Cathode (85/9/6) Compares Scale-Up Planetary Mixer to Lab Scale Results***

Additional full-cell coin cell testing verified that improved mixing capabilities provides improved cell performance of the dual layer system which is highlighted again at 6C. Compared to a single layer control (92/4.5/3.5 NCM622/Carbon/PPG Binder) and NMP/PVDF control, both at 2.1 mAh/cm$^2$, the dual layer system shows improved cell performance (Figure 22). These results highlight the improved performance of dual layer cathodes over single layer systems irrespective of the mixing conditions used to prepare these formulations.

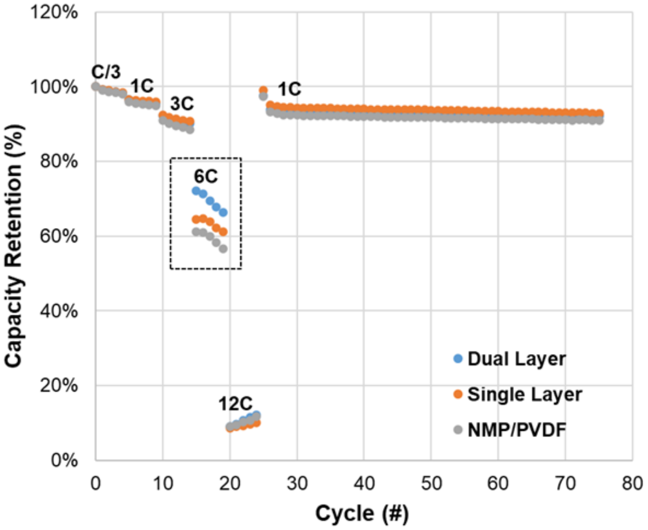


*Figure 22. Capacity Retention Plot of Full-Cell Coin Cell Data from Dual Layer, Single Layer, and NMP/PVDF Control*

## 3.2 Task 1.2 – Scalability of Cathode Coating Architectures

For Task 1.2, PPG screened single and dual layer cathode formulations using both internal single layer pouch cell capabilities and the pilot scale mixing, coating, and assembling available at the UMBL pilot facility. Cell testing was performed at both room temperature and reduced temperatures.

### 3.2.1 Single Layer Pouch Cell Testing

During this project, PPG developed internal capability for the preparation and testing of single layer pouch cells. To verify the reliability of this capability, a 92/4.5/3.5 NMC622/Super-P/B13H cathode was paired with a 95/2/3 Graphite/CA/Binder 4.4a anode and prepared at both PPG and the UMBL. Both cells were prepared using the same cathode and anode coatings at the same N/P ratio of 1.1. The capacity of the single layer pouch cells prepared at PPG were 30 mAh, while the multi-layer pouch cells from UMBL had a capacity of 1 Ah. Figure 23A highlights comparable first and last cycle efficiency during 0.1C formation cycles, and Figure 23B highlights comparable rate-capability behavior. Both cells were subjected to an asymmetric charge-discharge profile of +1C/-xC, at the discharge rate shown below. Between 0.3 and 9C, there was no change in discharge capacity retention observed, which highlights the quality of cells that can be fabricated at PPG, comparable to those prepared at UMBL.

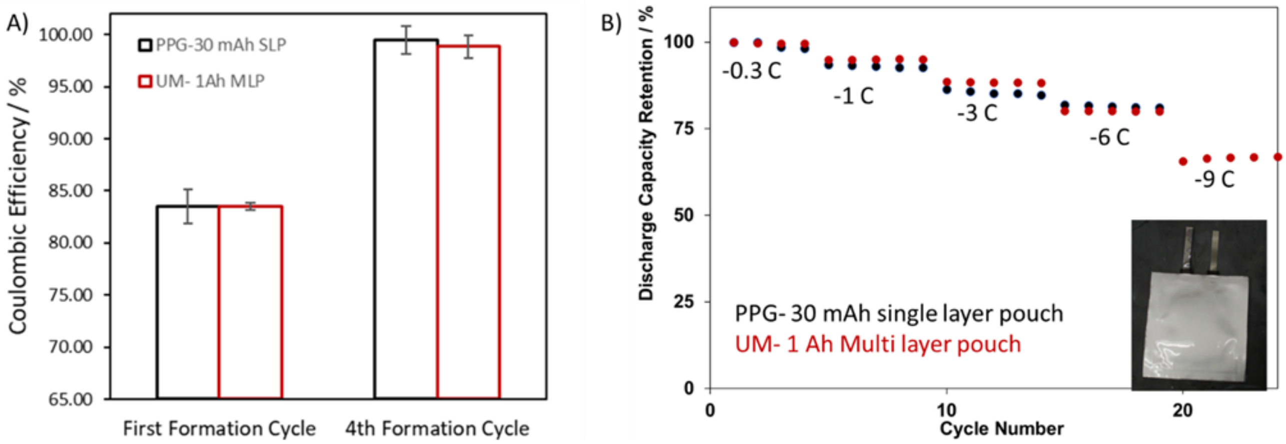


*Figure 23. Comparison of (A) Formation Efficacy and (B) Rate Capability of Pouch Cells Prepared at PPG and UMBL*

### 3.2.2 Low Temperatures Electrolyte Screening

With equivalent room temperature performance identified, PPG began to evaluate its cathode and anode binder formulations at reduced temperatures. However, the standard electrolyte used for room temperature screening is incompatible with reduced temperatures due to the high freezing point of the solvents used. A series of new electrolyte formulations E-1, E-2, E-3, and E-4 were screened in full-cell coin cells. Electrolytes E-1 and E-2 are ester-containing formulations, while E-3 and E-4 are carbonate containing. Analysis of the formation data in Figure 24 uncovered that electrolytes E-2 and E-4 have the highest first cycle efficiencies of the low temperature systems tested, while E-3 displays a noticeably reduced first cycle efficiency, and E-1 did not show any cycling behavior. This trend is reproduced in the rate capability and short-term cycle life where electrolytes E-2 and E-4 display discharge capacity retentions that most closely resemble the performance of our standard (room temperature) electrolyte. Based on the distinguished performance of electrolytes E-2 and E-4, these were down-selected for additional screening in our larger-format single layer pouch cell setup.

Single layer pouch cell testing was performed by pairing 2 mAh/cm$^2$ 92/4.5/3.5 NMC622/Carbon 1/B13H cathodes with 2.2 mAh/cm$^2$ 95/2/3 Graphite/CA/CMC + SBR anodes. Each pouch cell was

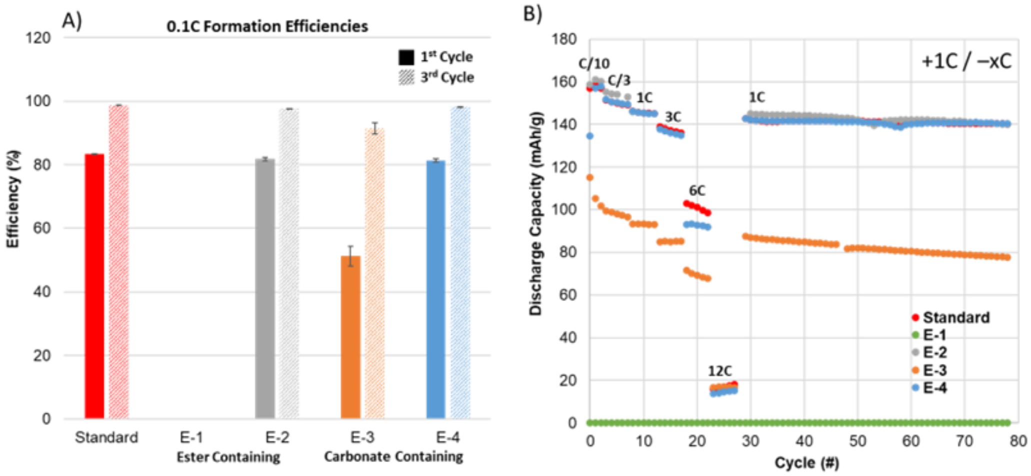


*Figure 24. Full-Cell Coin Cell (A) Formation Efficiencies and (B) Rate Ladder Discharge Capacities Data Using 4 Low Temperature Electrolytes Compared to Standard Formulation*

sealed with either standard, E-2, or E-4 electrolyte. After four symmetric 0.1C cycles with C/20 taper charges at 4.2V, the cells were subject to +1C/–1C cycle life testing. The discharge capacity retention plotted in Figure 25 demonstrates that over time, electrolyte E-4 engenders a more stable cycling profile than E-2. As such, electrolyte E-4 was selected for cold-crank testing screening of PPG cathode and anode formulation.

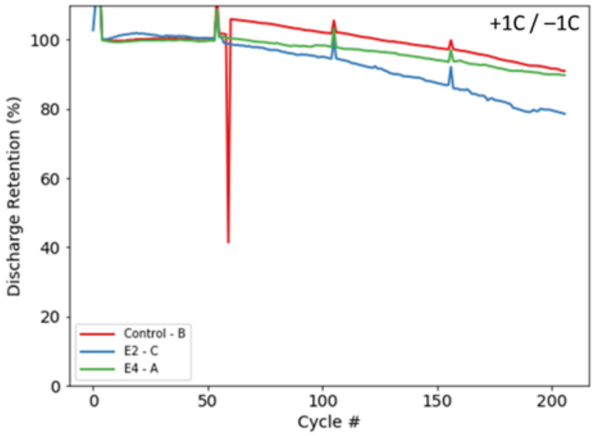


***Figure 25. Single Layer Pouch Cell Testing of Two Experimental Low Temperature Electrolyte Candidates***

### 3.2.3 Single Layer Pouch Cell Cold-Crank Testing

With equivalent room temperature performance identified, PPG began to evaluate how its cathode and anode binder formulations behaved at reduced temperatures. In the following tests, PPG cells are a pairing of a 2 mAh/cm$^2$ NMC622 NMP-free dual layer cathodes with a 2.2 mAh/cm$^2$ 95/2/3 Graphite/CA/Binder 4.4a anodes. Control cells represent a pairing of a 2 mAh/cm$^2$ 92/4.5/3.5 NMC622/Carbon 1/PVDF-NMP cathode with a 2.2 mAh/cm$^2$ 95/2/3 Graphite/CA/CMC + SBR anode. For all tests performed below room temperature, electrolyte E-4 as described in Section 3.2.2, was utilized to ensure high ionic conductivity. All electrochemical testing was performed in an ESPEC BTZ-475 thermal chamber.

PPG cells were evaluated against control cells at two different temperature/discharge profiles. In both cases, the cells were formed at room temperature and then cooled down to the specified temperature and let equilibrate for 6 hours. Once thermal equilibrium was reached, all cells were charged at 0.1C with a C/50 taper charge to 4.2V. Figure 26A displays the pulse discharge voltage profile of when PPG and control cells were subjected to a 12C discharge current while cooled to -18°C. As predicted by the improved rate capability of PPG cells compared to control formulations in Section 3.4.6, the PPG cells take around 9 seconds to reach the 2V discharge cutoff while control cells reach this cutoff within 4 seconds. This improvement likely comes from a reduced internal resistance of PPG cells due to the advanced dual layer cathode formulation and the improved conductive carbon dispersion engendered by anode Binder 4.4a. The improved internal resistance is further highlighted by the comparison of PPG and control cells at a pulse discharge rate of -4C at -40°C. The voltage profile shown in Figure 26B highlights the high internal resistance of control formulations by a rapid drop in cell voltage, which quickly crosses the 2V threshold in about 1 second, while the PPG cells provide power for approximately 7 seconds. However, it is important to note that both cells fall short of the 6T specification for performance under these conditions at 30 seconds. In this case, falling short of this target may be acceptable due to the smaller cell format of this test. It is also expected that with a larger cell format of higher capacity, cell self-heating could improve low temperature performance.

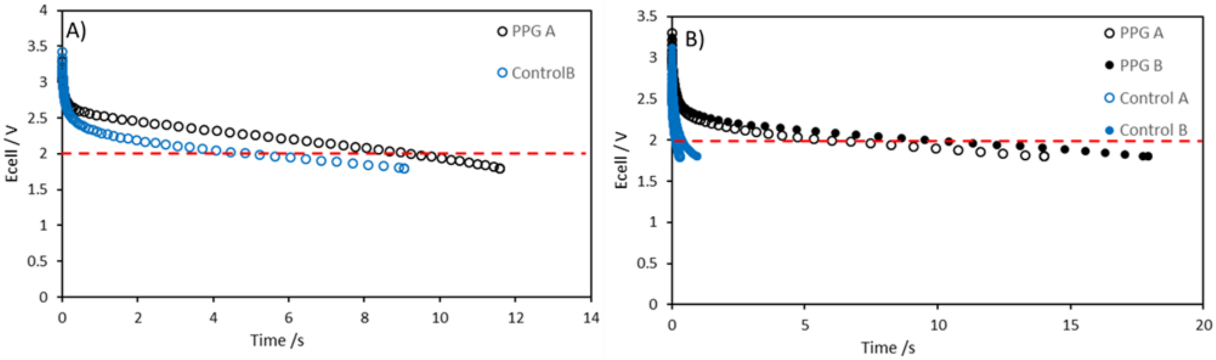


***Figure 26. Pulse Discharge Voltage Profile of PPG vs. Control 25 mAh Single Layer Pouch Cells at (A) Discharge Rate of –12C at –18°C and (B) Discharge Rate of –4C at –40°C***

### 3.2.4 PPG Dual Layer Cathode Scale-Up at UMBL

Given the promising performance of PPG dual layer cathodes when paired with PPG anode formulations, these formulations were brought to the UMBL for mixing, coating, and multi-layer pouch cell assembly on the pilot scale. As described in Section 3.1 the PPG dual layer cathode involves an either wet-on-wet or wet-on-dry coating application. Due to the coating capabilities at UMBL, a wet-on-dry coating application method was selected. Using a reverse comma coater, 4.5 mg/cm$^2$ of 85/9/6 NMC622/CA/B13Hx power layer was successfully applied to the cathode current collector, as shown in Figure 27A. The 96/2/2 NMC622/CA/B13Hx energy layer formulation was then applied to the top of the dried power layer formulation using a slot-die coater. However, due to issues with the pressure controls on the slot-die head, UMBL was unable to supply a coat weight of less than 16.6 mg/cm$^2$ which is significantly higher than the target loading of 9 mg/cm$^2$.

Due to the challenges of obtaining target loadings, PPG worked with UMBL to provide two alternative cathodes of formulation 92/4.5/3.5 NMC622/CA/Binder, where Binder = PVDF/NMP or B13H, PPG's NMP-free binder. Reverse comma coating of these formulations at 2.1 mAh/cm$^2$ proceeded with no issues to yield uniform coatings as shown in Figure 28A and B. These cathodes

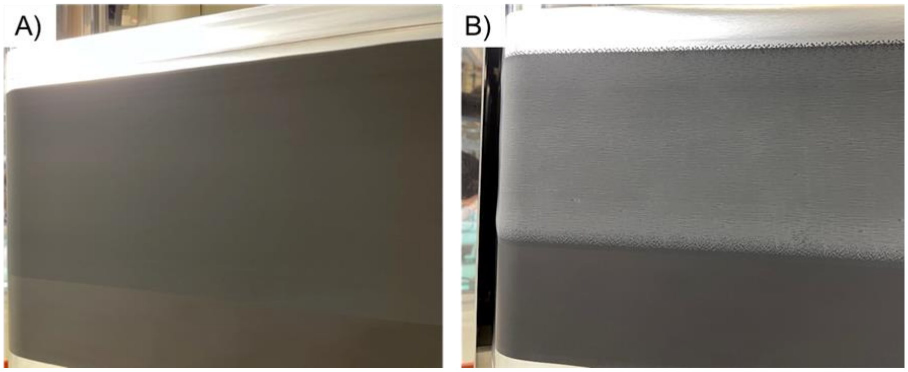


***Figure 27. Images of (A) Power Layer and (B) Dual Layer Coatings from PPG's Scale-Up at UMBL***

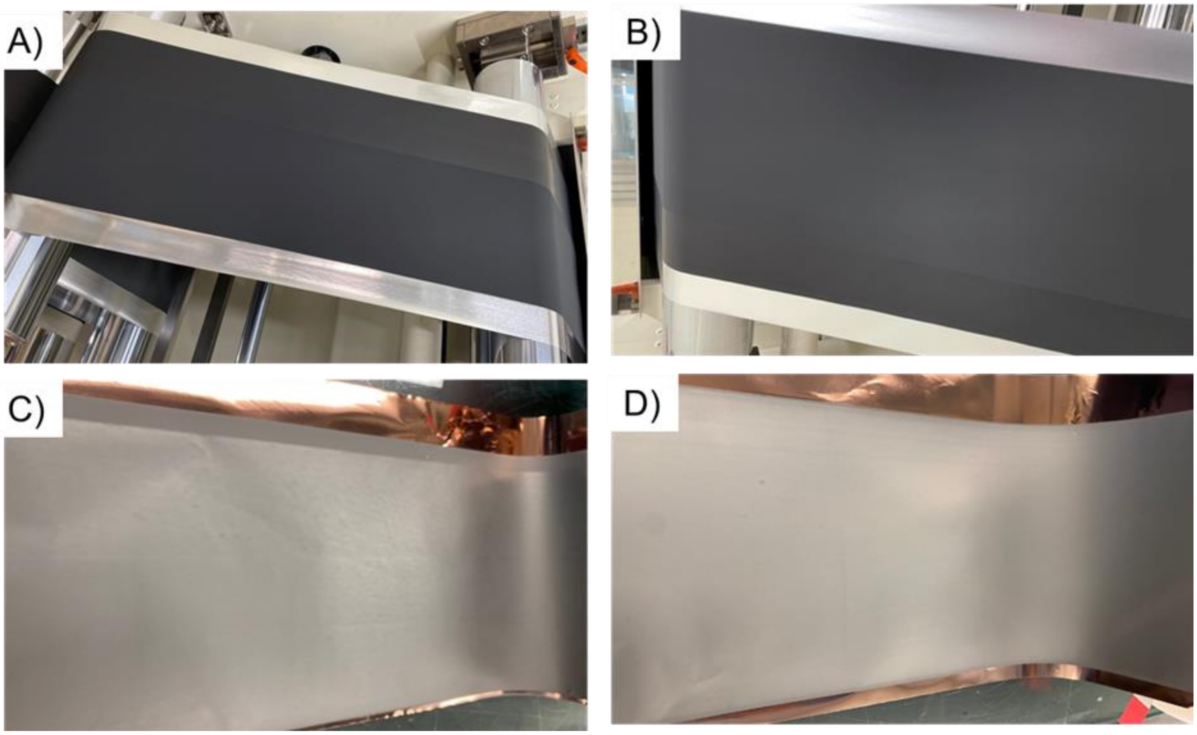


***Figure 28.*** ***Images of Dried (A) PVDF/NMP or (B) PPG NMP-Free Binder Containing Cathodes and Corresponding (C) CMC/SBR or (D) PPG Binder 4.4a Anodes***

were paired with 2.3 mAh/cm$^2$ 95/2/3 Graphite/CB/Binder anodes, where binder = CMC+SBR or PPG anode Binder 4.4a as described in Section 3.4.6. The pilot scale anode coatings proceeded with no issues to yield uniform coatings as shown in Figure 28C and D.

To verify the high quality of the coatings described above, 1.1 Ah multi-layer pouch cells were fabricated and subjected to a cycle life experiment. As shown in Figure 29, the PPG-NMP free cathodes (B13H) when paired with PPG anode Binder 4.4a display nearly identical cycle life as compared to standard cathode (PVDF/NMP) and anode (CMC + SBR) binders.

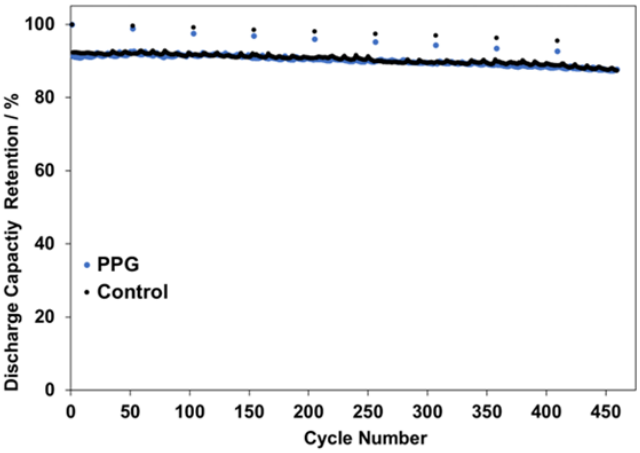


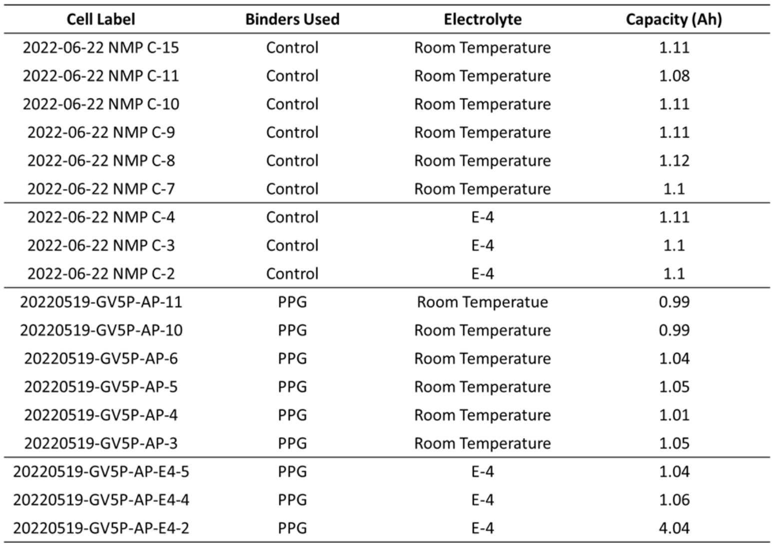
***Figure 29.*** ***Symmetric 1C Cycle Life Experiment Comparing Multi-Layer Pouch Cells Fabricated Using Control Cathode and Anode Binders (black) and PPG Cathode and Anode Binders (blue)***

## 3.3 Task 1.3 – Cell Deliverables: 9 NMP-Containing and 9 NMP-Free Multi-Layer Pouch Cells

Using the pilot scale coatings described in Section 3.2.4, PPG prepared a series of multi-layer pouch cells for evaluation by GVSC. A total of 18 cells were delivered as described in Table 1. All cells contained a pairing of 2.1 mAh/cm$^2$ 92/4.5/3.5 NMC622/CA/Binder cathodes with 2.3 mAh/cm$^2$ 95/2/3 Graphite/CA/Binder anodes. In the cells marked as “PPG”, cathode binder = B13Hx, PPG NMP-free binder and anode binder = Binder 4.4a developed in this work as described in Section 3.4. Cells marked as control contained cathode and anodes binders composed of PVDF/NMP and CMC+ SBR respectively. Cells marked for room temperature utilized standard carbonate electrolyte, while low temperature cells utilize electrode E-4 that was described in Section 3.2.2.

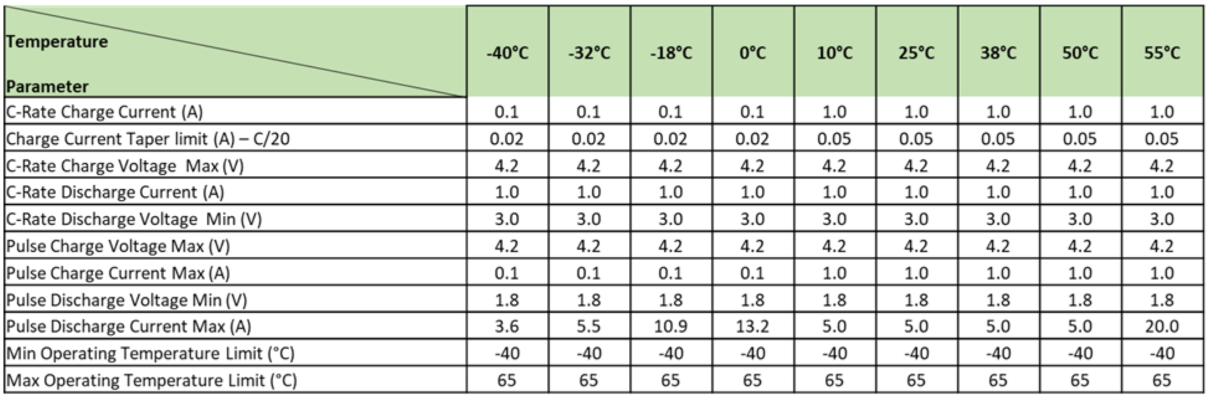
The cells listed in Table 1 were successfully delivered to GVSC at ~ 20% state of charge and were delivered on August 30, 2022. The testing plan for these cells is outlined below in Table 2. PPG and control cells will be compared at the various temperatures and discharge currents listed. Low temperature electrolyte E-4 containing cells will be used for all testing at temperatures ≤ 0°C. Testing is currently underway.

**Table 1. Multi-Layer Pouch Cell Specifications and Capacities for Project Deliverables**



| Cell Label | Binders Used | Electrolyte | Capacity (Ah) |
|---|---|---|---|
| 2022-06-22 NMP C-15 | Control | Room Temperature | 1.11 |
| 2022-06-22 NMP C-11 | Control | Room Temperature | 1.08 |
| 2022-06-22 NMP C-10 | Control | Room Temperature | 1.11 |
| 2022-06-22 NMP C-9 | Control | Room Temperature | 1.11 |
| 2022-06-22 NMP C-8 | Control | Room Temperature | 1.12 |
| 2022-06-22 NMP C-7 | Control | Room Temperature | 1.1 |
| 2022-06-22 NMP C-4 | Control | E-4 | 1.11 |
| 2022-06-22 NMP C-3 | Control | E-4 | 1.1 |
| 2022-06-22 NMP C-2 | Control | E-4 | 1.1 |
| 20220519-GV5P-AP-11 | PPG | Room Temperatue | 0.99 |
| 20220519-GV5P-AP-10 | PPG | Room Temperature | 0.99 |
| 20220519-GV5P-AP-6 | PPG | Room Temperature | 1.04 |
| 20220519-GV5P-AP-5 | PPG | Room Temperature | 1.05 |
| 20220519-GV5P-AP-4 | PPG | Room Temperature | 1.01 |
| 20220519-GV5P-AP-3 | PPG | Room Temperature | 1.05 |
| 20220519-GV5P-AP-E4-5 | PPG | E-4 | 1.04 |
| 20220519-GV5P-AP-E4-4 | PPG | E-4 | 1.06 |
| 20220519-GV5P-AP-E4-2 | PPG | E-4 | 4.04 |

**Table 2. Testing Protocol for Cell Deliverables**



| Temperature / Parameter | -40°C | -32°C | -18°C | 0°C | 10°C | 25°C | 38°C | 50°C | 55°C |
|---|---|---|---|---|---|---|---|---|---|
| C-Rate Charge Current (A) | 0.1 | 0.1 | 0.1 | 0.1 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| Charge Current Taper limit (A) – C/20 | 0.02 | 0.02 | 0.02 | 0.02 | 0.05 | 0.05 | 0.05 | 0.05 | 0.05 |
| C-Rate Charge Voltage Max (V) | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 |
| C-Rate Discharge Current (A) | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| C-Rate Discharge Voltage Min (V) | 3.0 | 3.0 | 3.0 | 3.0 | 3.0 | 3.0 | 3.0 | 3.0 | 3.0 |
| Pulse Charge Voltage Max (V) | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 | 4.2 |
| Pulse Charge Current Max (A) | 0.1 | 0.1 | 0.1 | 0.1 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| Pulse Discharge Voltage Min (V) | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 |
| Pulse Discharge Current Max (A) | 3.6 | 5.5 | 10.9 | 13.2 | 5.0 | 5.0 | 5.0 | 5.0 | 20.0 |
| Min Operating Temperature Limit (°C) | -40 | -40 | -40 | -40 | -40 | -40 | -40 | -40 | -40 |
| Max Operating Temperature Limit (°C) | 65 | 65 | 65 | 65 | 65 | 65 | 65 | 65 | 65 |

## 3.4 Task 2 – Anode Binder Development

### 3.4.1 Test Method Optimization

To successfully screen viable anode binder candidates, PPG first sought to ensure the internal ability of collecting high quality electrochemical data. Nearly every area of cell design and formulation was considered and tested. From the coin cell level testing performed, the team found that separator identity, conductive additive identity, and graphite quality to be main determining factors for performance. Full-cell coin cells were constructed pairing 1.45 mAh/cm$^2$ 92/4/4 NMC532/CA 1/PVDF/NMP cathodes with 1.72 mAh/cm$^2$ 95/2/3 Graphite/CA2/CMC+ SBR anodes. Figure 30 displays the discharge capacity from an asymmetric charge/discharge rate capability ladder with +1C/–xC current, where x = 0.1, 0.3, 1, 3, 6, and 12. Both Separators 1 and 2 showed an expected decrease in discharge capacity as the discharge current was increased, however Separator 1 (orange) showed a much higher discharge capacity retention compared to Separator 2 (blue). Based on the higher performance of Separator 1, this separator was used in all future coin cell tests.

With an appropriate separator for high-rate performance identified, PPG then evaluated various commercial conductive additives in anode formulations. Figure 31 displays the rate capability ladder from 1.45 mAh/cm$^2$ 92/4/4 NMC532/CA 1/PVDF/NMP cathodes paired with 1.72 mAh/cm$^2$ 95/2/3 Graphite/CAX/CMC+ SBR anodes. Four conductive additives were evaluated, and as expected given the key role these materials play in the conductivity of electrode formulations different additives showed better performance owing to their surface area, particle size and surface functionality. Based on its high discharge capacity retention at a discharge rate of -12C, CA3 was selected to be used in further full cell and pouch cell testing.

In addition to separator and conductive additive, graphite identity is expected to play a major determining role in the final performance of any lithium-ion cell. In agreement with this hypothesis, there was a distribution in discharge rate capability when evaluating the discharge capacity from full-cell coin cells composed of 1.45 mAh/cm$^2$ 92/4/4 NMC532/CA 1/PVDF/NMP cathodes paired with 1.72 mAh/cm$^2$ 95/2/3 Graphite/CA3/CMC+ SBR anodes. Figure 32 displays the discharge capacity retention normalized to the 0.3 discharge capacity. From this analysis, Graphite 2b displays the best performance for fast discharge and will be used in further screenings of PPG anode binder systems.

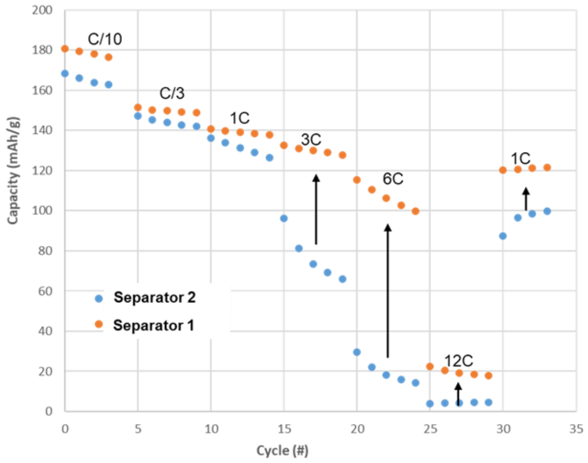


*Figure 30. Full-Cell Coin Cell Analysis Utilizing Two Different Coin Cell Separators*

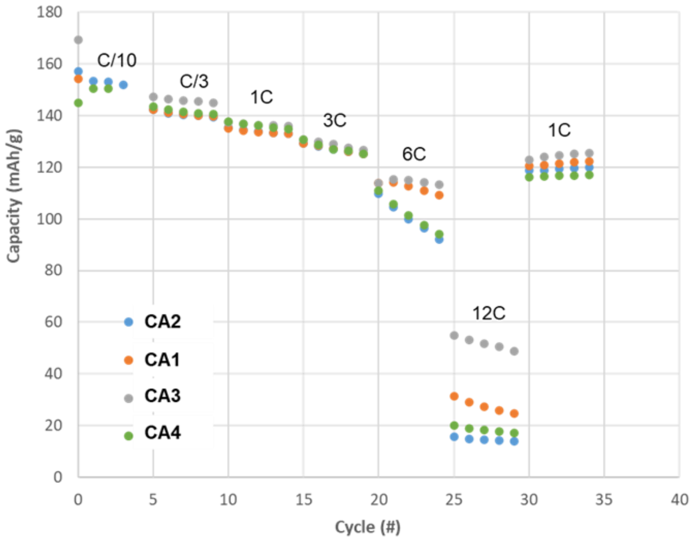


*Figure 31. Rate Capability Analysis of Full-Cell Coin Cell Data Using Various Anode Conductive Additives*

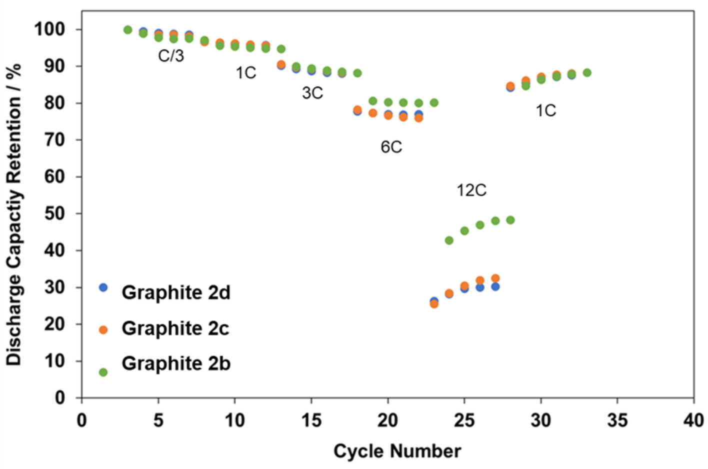


*Figure 32. Rate Capability Analysis of Full-Cell Coin Cell Data Using Various Graphite Active Materials*

### 3.4.2 PPG Anode Binder Screening

Following the identification of appropriate anode active materials, conductive additives and separators, PPG next sought to understand the key properties of anode binders which engender key anode binder properties. These properties include good dispersion of active materials and conductive additives, high adhesion to the copper current collector, good flexibility, and a polymer backbone that resists electrochemical degradation under the lithium-ion battery charging and discharging conditions. There are a variety of different anode binder chemistries and polymer properties that can be deployed as graphite anode binders; therefore, PPG began investigation into these materials by screening alternative binder systems visually after coating and drying onto a copper current collector. Binders were used to formulate 95/2/3 graphite/carbon/binder slurries which were drawn down onto copper foil at an aerial loading of 2.3 mAh/cm$^2$. The electrodes were then pressed to 35% porosity and were ready for screening. A total of 41 aqueous binder systems were prepared and evaluated in this manner and were compared to the physical and electrochemical properties of a commercial CMC+SBR control binder package. Many binder candidates showed obvious adhesion issues, as shown in Figure 33A and B, which represent PPG anode Binders F and T. These anodes are representative of binder candidates which failed in the first stage of screening – any visual indication of delamination or edge cracking disqualified candidates from further analysis. Acceptable adhesion includes no obvious flaking or delamination from the current collector either before or after calendaring as demonstrated by PPG anode Binder Z-B in Figure 33C.

Any electrodes which displayed acceptable visible appearance similar to PPG anode Binder Z-B in Figure 33C, were further screened for flexibility by bending across a series of mandrels ranging from 1-3 mm in diameter, as shown in Figure 34. This test is necessary to ensure that the PPG anode binders provide sufficient flexibility for roll-to-roll processing and compatibility with a wide variety of cell formats. Any cracking along the coating or its edge would disqualify the anode binder from further screening.

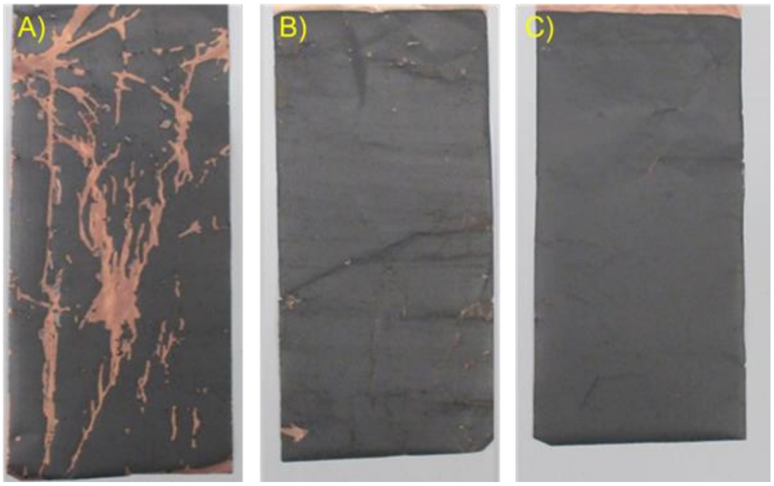


***Figure 33. Images of 95/2/3 Graphite/CA/Binder Anode Films After Drying as Representative Examples of Unacceptable (A, B) and Acceptable (C) Electrode Appearance***

*Binders shown here include PPG anode Binders F, T, and Z-B*

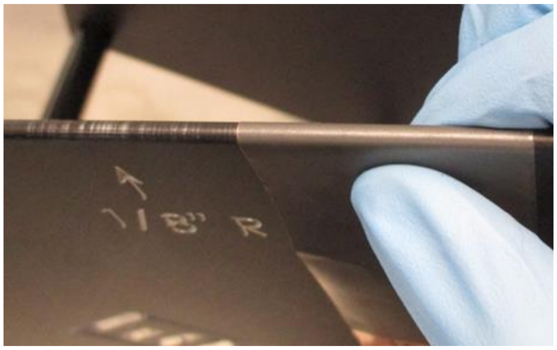


***Figure 34. Example of Good Electrode Flexibility as Determined by Mandrel Bend Test***

### 3.4.3 Half-Cell Screening of PPG Anode Binders

Binders which were determined to have good physical properties, as described in Section 3.4.2, were then used to assemble lithium-ion half-cells coin cells. All cells were assembled against a lithium metal cathode and formed with a symmetric charge/discharge rate of 0.1C with a C/20 CV taper at the top of charge. Formation efficiency and discharge capacity for tested binders was measured to determine the initial electrochemical quality of the electrode as well as look for potential incompatibilities between binders, active material, and lithium-ion battery electrolyte. Figure 35 is a representative example of the various trends observed when screening PPG anode binders. The differences in binder composition cause some anodes to have higher or lower first cycle efficiency; however, the cells all reached approximately 99% efficiency after the third formation cycle. Cells which displayed reduced first cycle efficiency, compared to the CMC+SBR control, such as PPG anode Binder C as shown in Figure 35, were disqualified from additional testing. This selection process attempted to screen out any candidates which would significantly interfere with Solid Electrolyte Interface (SEI) formation, or those that may have other electrochemical incompatibilities.

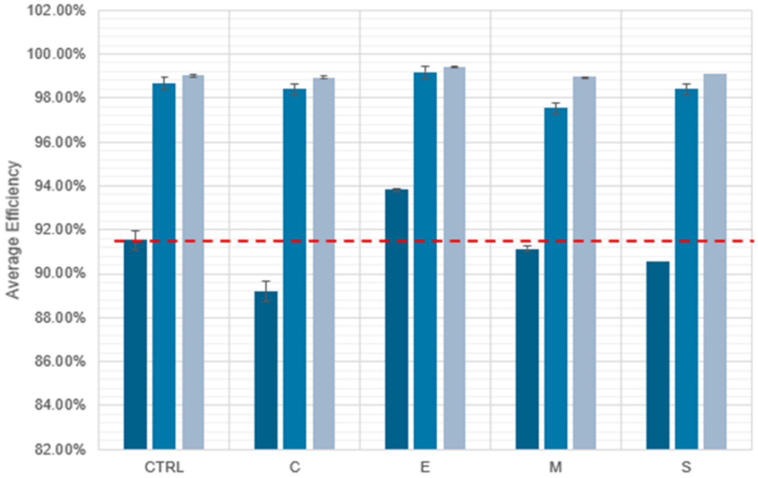


***Figure 35. Measured Formation Efficiencies of a Selection of Tested PPG Binders in Half-Cells***

*This figure shows the average efficiency and standard deviation over 3 cycles for 3 trials of each binder. The red line represents the first cycle efficiency of CMC+SBR control anode*

### 3.4.4 Single Layer Pouch Cell Cycling of PPG Anode Binders

Using the screening tools outlined in Section 3.4.2 and Section 3.4.3, the team was able to down-select PPG anode binder chemistries for cycle life in single layer pouch cells. It is critical that any anode binder does not interfere with standard cycle life performance, regardless of any benefits to processability or power. To evaluate the lifetime of these systems, pouch cells were prepared by pairing a 95/2/3 Graphite/Carbon/Binder anode at 2.3 mAh/cm$^2$ with a 92/4.5/3.5 NMC622/CA 1/B13Hx cathode at 2.1 mAh/cm$^2$. For every new set of cells prepared, a fresh CMC+SBR control anode was generated and tested at the same time as the PPG anode binder to account for any temperature fluctuations during cycling. In order for a binder to successfully pass this test, its discharge capacity retention must closely mirror that of the CMC+SBR control when subjected to a series of 1C/1C charge/discharge cycles. Several of the PPG anode binders tested display an increased loss in discharge capacity retention as compared to control formulations as shown in Figure 36A. In this case, the discharge capacity retention of anode cells formulated with PPG anode Binder V reached < 80% within 80 cycles while the control CMC+SBR anode system maintained a much flatter capacity profile. Cell teardown analysis of this electrode reveals significant edge delamination and surface morphological changes, including ripples and bumps across the anode. This finding suggests that the wet adhesion of this binder may be poor and is a possible explanation for the reduced cycle life.

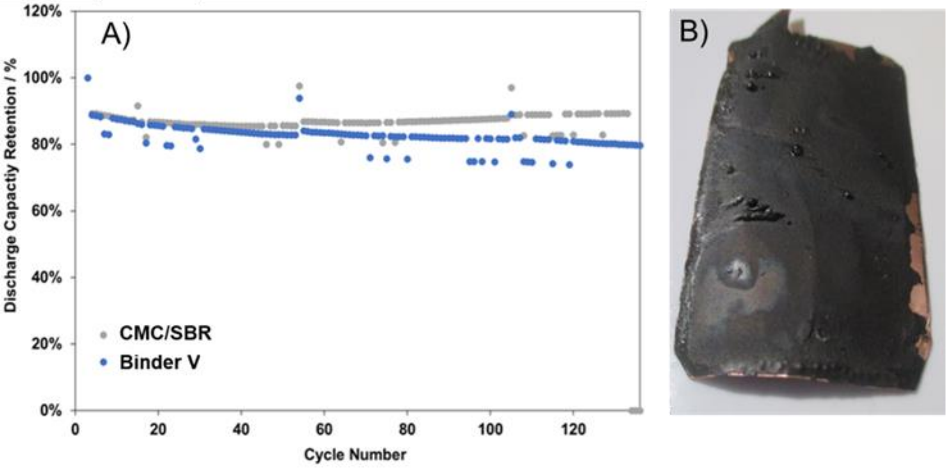


*Figure 36. Single Layer Pouch Cell Cycle Life of PPG Anode Binder V Compared to CMC+SBR Control (A) and Image of Anode After Cycling (B)*

Using insights based on the chemical functionality present on PPG anode binders, such as Binder V, a series of new binders were prepared to address the issues suggested by the postmortem anode in Figure 36B. One such anode which was prepared and passed all the previous screening methods in described in Section 3.4.2 and, was PPG anode Binder Z-B which was subsequently used to prepare a series of single layer pouch cells. Figure 37A displays the discharge capacity retention of PPG anode Binder Z-B in comparison to CMC/SBR over the course of 200 1C/1C charge/discharge cycles. As demonstrated by the nearly identical discharge capacity retention over time, PPG anode Binder Z-B has been identified as a potential candidate which does not interfere with the cycling performance of lithium-ion cells. In agreement with the hypothesis that wet adhesion resulted in cell failure in previous anode binder systems, images collected of PPG anode Binder Z-B after cycling shows no edge delamination or surface defects, as shown in Figure 37B.

With the successful identification of PPG anode Binder Z-B as a component which does not interfere with the long-term cycle life capability of lithium-ion cells, the team next sought to evaluate its baseline ability to improve the power performance of anodes. The power performance was evaluated by performing a rate capability ladder with increasing discharge rates, ranging from C/3 to 12C as

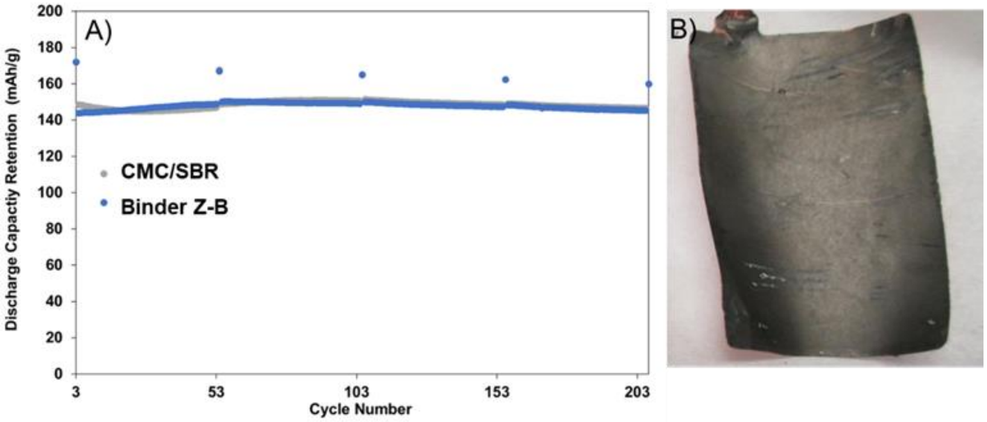


*Figure 37. Single Layer Pouch Cell Cycle Life of PPG Anode Binder Z-B Compared to CMC+SBR Control (A) and Image of Anode After Cycling (B)*

shown in Figure 38. Given the design of these anodes was to first yield a material with good physical and electrochemical properties as compared to the CMC/SBR control platform it was unsurprising that cells containing PPG anode Binder Z-B closely mirrored the discharge capacity of CMC/SBR over a wide variety of discharge rates. The value of this initial investigation was to develop internal understanding of how various polymer chemistries and properties translate to electrode properties such as wet and dry adhesion and electrochemical performance. The next sections will expand this knowledge to develop anode binders which display both excellent physical properties and improved power performance.

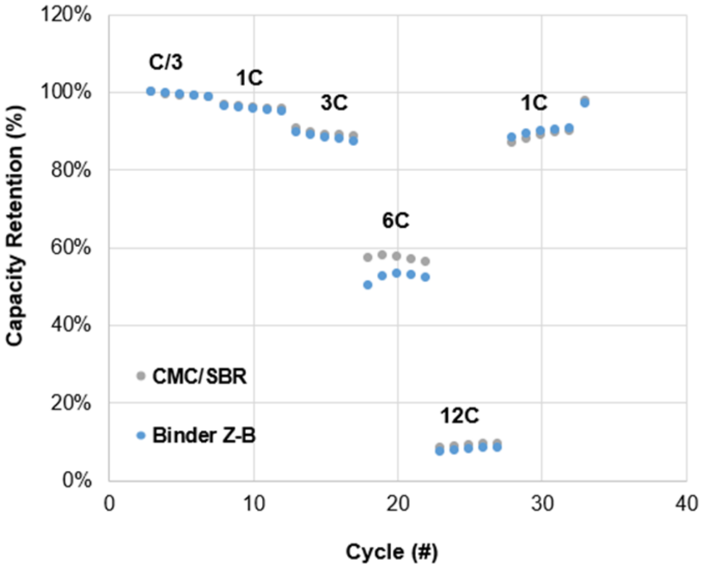


*Figure 38. Rate Capability Performance of 2.1 mAh/cm² NMC622 Cathodes Paired with 2.3 mAh/cm² Graphite Anodes Formulated with Listed Binders with Asymmetric Charge/Discharge Profile where Charging Rate was 1C and Discharge Rate as Listed*

### 3.4.5 Impact of Binder Chemistry on Physical Properties

To improve the power performance of cells, our approach was to formulate and test additional PPG anode binders which can improve the dispersion of carbon particles in the aqueous anode formulation. As such, various PPG anode binder chemistries were evaluated for their ability to improve carbon dispersion quality while maintaining physical properties similar to CMC + SBR control binders. Carbon dispersion quality was screened by centrifugal mixing a conductive additive with the various PPG binders at 2000 rpm for 30 seconds. The aggregated carbon black particle size was assessed using a Hegman fineness of grind gauge. This device determined if the carious anode binder formulations tested provided any benefit to the carbon dispersion quality. The images in Figure 39 are of these carbon slurries after 30 seconds of mixing. The appearance of a speckled surface like that in Figure 39A indicate a large quantity of carbon black agglomerates along the entire gauge, starting at a grind value around 5, corresponding to a particle size of > 100 mm. In contrast, the carbon agglomerates in PPG Binders 4.4 or 7.6 are significantly smaller, as shown in the Hegman images of Figure 39B and C.

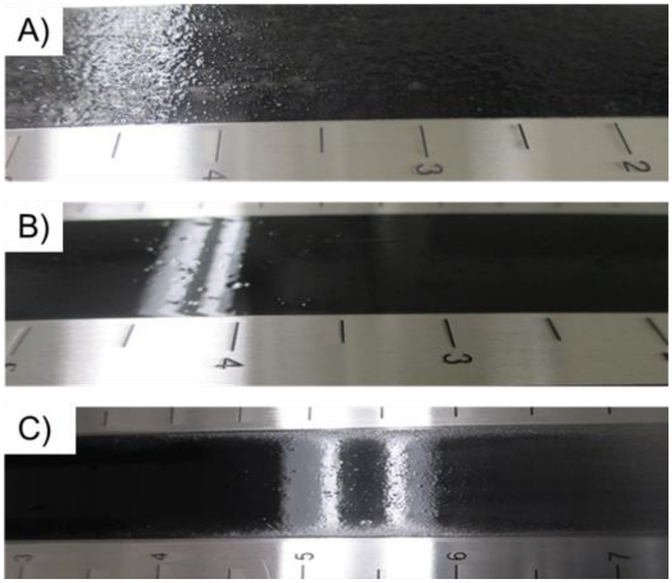


*Figure 39. Hegman Grind Gauge Images of CA3 Dispersed into (A) CMC Control, (B) PPG Binder 4.4, or (C) PPG Binder 7.6*

Various other binder chemistries and PPG formulations were tested, some of which showed similar improvements to carbon dispersion quality; however, none could match the physical properties of the CMC + SBR control binder systems as shown in Figure 40. Binders with peel strength values significantly below the CMC + SBR threshold of ~ 10 N/m are typically difficult to handle and show a high degree of edge delamination during cell preparation.

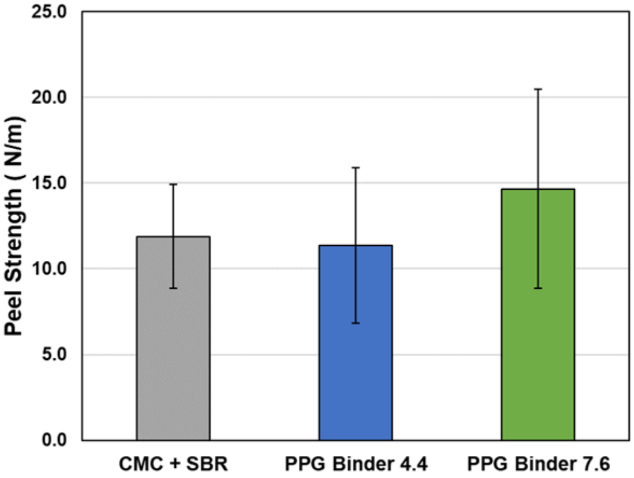


*Figure 40. 90° Peel Strength of Various 95/2/3 Graphite 2b/CA3/Binder Formulations*

## 3.4.6 Electrochemical Testing

After identifying PPG anode Binders 4.4 and 7.6 as viable candidates based on their physical properties (Figure 40) and improved ability to disperse conductive carbon (Figure 39), full-cell electrochemical testing was performed on multi-layer pouch cells. In these cells, 2.1 mAh/cm$^2$ 92/4.5/3.5 NMC622/CA/B13Hx cathodes were paired with 2.3 mAh/cm$^2$ 95/2/3 Graphite/CA3/binder anodes, where binder = CMC+ SBR, PPG Binder 7.6 or PPG Binder 4.4. The cycle life of these anode binders was evaluated using the same cell configurations that were now subjected to a symmetric 1C/1C cycling protocol. As shown in Figure 42, PPG Binder 4.4 (green) displays similar capacity retention compared to CMC + SBR control (grey) after 500 cycles indicating an electrochemically stable binder system. Unfortunately, PPG Binder 7.6 (blue) presents a much steeper capacity fade profile, with < 80% capacity retention after 400 cycles.

Given the electrochemical stability of PPG anode Binder 4.4 shown in Figure 41, the rate capability of these cells constructed using this anode binder was evaluated. In this testing procedure, each cell was charged at 1C with a C/20 tapper charge followed by discharge at increasing rates of: C/3, 1C, 3C, 6C, and 9C. Analysis of the capacity retention of these cells reveals that at the modest discharge rates of 0.3 C, 1C and 3C there is no discernable difference between CMC + SBR control binder and PPG anode Binder 4.4. At 6C, a slight increase in discharge capacity retention is observed, and at 9C this difference is magnified such that the PPG anode binder displays 9% higher capacity retention than the control binder. This improved performance is due to the improved conductive carbon network as described above and as further supported in Section 3.4.7 by microscopy and rheology.

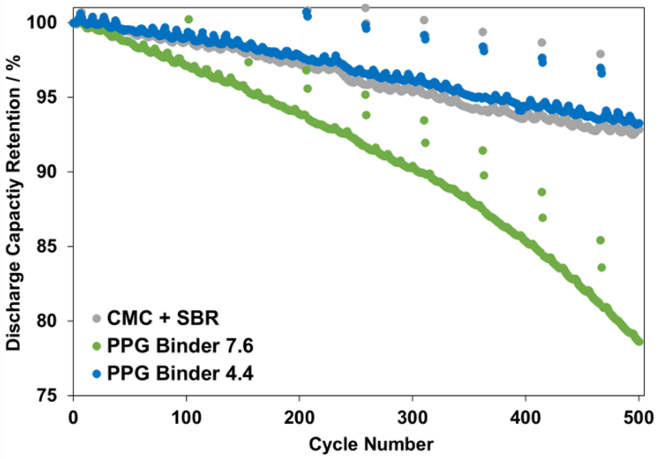


*Figure 41. 1 Ah Multi-Layer Pouch Cell Cycle Life of 2.1 mAh/cm² NMC622 Cathodes Paired with 2.3 mAh/cm² Graphite Anodes at Charge/Discharge Rate of 1C*

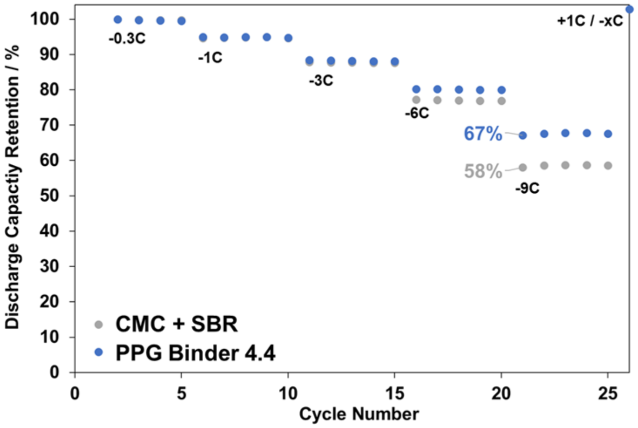


*Figure 42. 1.1 Ah Multi-Layer Pouch Cell Rate Capability Performance of NMC622 Cathodes Paired with Graphite Anodes with Asymmetric Charge/Discharge Profile where Charging Rate was 1C and Discharge Rate as Listed*

### 3.4.7 Carbon Dispersion Qualification

To fully characterize the effect of PPG Binder 4.4 on the quality of conductive carbon dispersion a series of analytical techniques including oscillatory rheology and TEM imagining were performed. In oscillatory rheology, a small sinusoidal oscillation is applied to the sample that does not affect the bulk structure of the sample. By this process, the storage modulus (G', solid circles) which indicates solid-like behavior and the loss modulus (G", open circles), which indicates viscous-like behavior can be determined for the same slurry. The crossover frequency, where G'=G", can empirically be understood as the minimum shear rate to cause the sample to flow (i.e., to exhibit more liquid-like behavior than solid-like behavior). This occurs once the solid conductive carbon network is disrupted. Figure 43 presents the oscillatory rheology of conductive carbon dispersed into either control binder or PPG anode Binder 4.4. The crossover frequency of the control is at ~ 0.5 rad/s, whereas the CMC+PPG additive is > 100 rad/s, showing the carbon particle network is much stronger for the latter sample set. Additionally, in the black data (control) there is a signal that at low frequencies a carbon network forms in the dispersion by the region where G'>G". This network is weaker and easily disrupted as the difference in moduli is no longer visible at frequencies > 0.5 rad/s. In contrast, PPG anode Binder 4.4 imparts G'>G" at a wide range of frequencies, including those much higher than the control.

To support the improved conductive carbon network suggested by the oscillatory rheology data shown in Figure 43, TEM images were collected of conductive carbon dispersed into either control or PPG anode Binder 4.4. Figure 44 displays these TEM images, which illustrate how the conductive carbon is distributed through either a control binder (A) or PPG anode Binder 4.4 (B). The dark sphere with a diameter of ~ 50 nm are the conducive carbon particles, and the light grey regions represent the binder network. In Figure 44A, there are binder rich regions (red circle) that form discontinuities in the conductive carbon network. In contrast, Figure 44B reveals a much more continued carbon network in PPG anode Binder 4.4 films. When the PPG anode binder is utilized, it is easy to draw several different direct paths from top to bottom on these images, which indicate a larger, more fractal carbon network as suggested by oscillatory rheology, as well as a more electrically conductive network as supported by the improved discharge capacity retention of these binders. By contrast, in control TEM image, the discontinuous carbon network can impact the high-power performance of the resulting anode films.

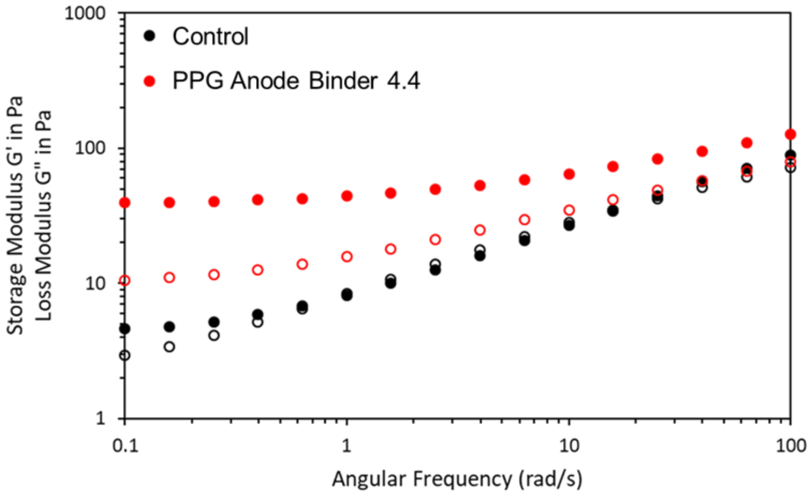


*Figure 43. Oscillatory Rheology of Conductive Carbon Dispersed in Either Control or PPG Anode Binder 4.4 Solutions*

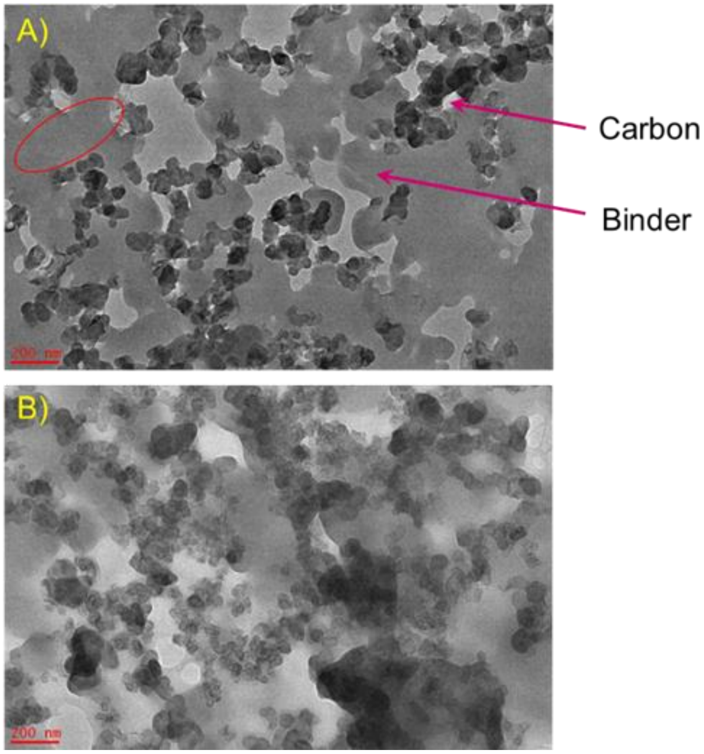


***Figure 44. TEM Images of Conductive Carbon Dispersed in Either (A) Control Binder, or (B) PPG Anode Binder 4.4***

*The dark black circles represent conductive carbon particles, and the lighter regions denote binder-rich areas*

## 3.5 Task 3 – Cost Analysis and Modeling

Task 3.1 stated that PPG continue to employ and update models developed and employed in Phase I of this work to validate the cost reductions achievable using novel cathode and anode coatings.

In Phase I, PPG was able to determine a 58% reduction in energy demand for a PPG system (single layer) compared to an NMP system. The calculation was based on an 8-zone oven with 5 meters in length per zone and a baseline speed of 25 meters/minute. It is important to note that there was a 10% increase in slurry solids for a PPG system compared to an NMP system. In addition, 39% less solvent was used in the PPG system.

In Phase II, PPG assumed the same approach to validate the cost reductions achievable using their novel cathode coating. However, in Phase II, PPG is accounting for a dual layer system, which requires some additional measurements to verify the same level of cost reduction. The dual layer system contains a power layer and an energy layer. The power layer system has a formulation of 85 NMC622/9 Carbon 1/6 Binder. The energy layer system has a formulation of 96 NMC622/2 Carbon/2 Binder. The PPG Binder system contains the NMP-free binder package, while the NMP system contains an NMP/PVDF binder package. To complete the cost modeling, cathode slurries at the given power and energy formulation ratios were formulated using the same binder package. To determine the percent solids advantage and corresponding energy reduction of the PPG system versus an NMP system, the viscosities of these cathodes were matched.

For the power layer, a target viscosity of 4000 cP at a shear rate of $10s^{-1}$ was used. From this target, PPG was able to determine how much solvent is needed in comparison to an NMP system. Figure 45 shows the flow curve for the NMP cathode and PPG power cathode slurries and their overlapping viscosity values at $10s^{-1}$.

For the PPG power layer slurry and NMP power layer slurry to overlap at the $10s^{-1}$ shear rate, the total slurry solids needed to be adjusted. A NMP power layer cathode requires 35% total slurry solids, and a PPG power layer cathode requires 45% total slurry solids to reach the same viscosity. There is a 10% total slurry solids advantage for a PPG power layer system. Table 3 demonstrates the calculations and overall solvent savings by using a PPG system. At the power layer formulation, the PPG system will use 34% less solvent.

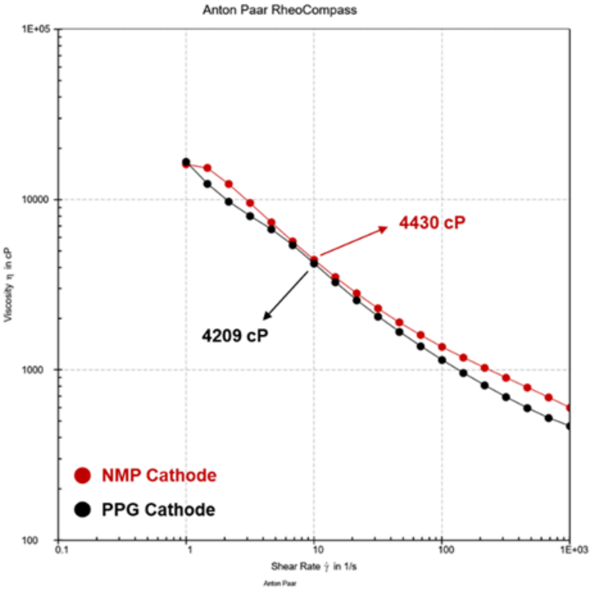
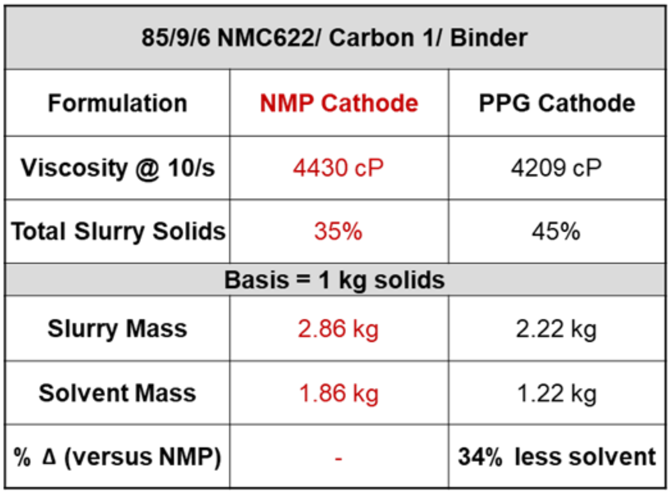


*Figure 45. Rheology Flow Curve for Power Layer Cathodes (85 NMC622/9 Carbon1/6 Binder) Slurries Formulated Using PPG and NMP Systems*

**Table 3. Comparison of NMP Cathode and PPG Cathode Systems at Power Layer Formulation Ratios**


<table>
<tr><th colspan="3">85/9/6 NMC622/ Carbon 1/ Binder</th></tr>
<tr><th>Formulation</th><th>NMP Cathode</th><th>PPG Cathode</th></tr>
<tr><td>Viscosity @ 10/s</td><td>4430 cP</td><td>4209 cP</td></tr>
<tr><td>Total Slurry Solids</td><td>35%</td><td>45%</td></tr>
<tr><td colspan="3">Basis = 1 kg solids</td></tr>
<tr><td>Slurry Mass</td><td>2.86 kg</td><td>2.22 kg</td></tr>
<tr><td>Solvent Mass</td><td>1.86 kg</td><td>1.22 kg</td></tr>
<tr><td>% Δ (versus NMP)</td><td>-</td><td>34% less solvent</td></tr>
</table>

For the energy layer, a target viscosity of 3000 cP at a shear rate of $10s^{-1}$ was used. From this target, PPG was able to determine how much solvent is needed in comparison to an NMP system. Figure 46 shows the flow curve for the NMP cathode and PPG energy cathode slurries and their overlapping viscosity values at $10s^{-1}$.

For the PPG energy layer slurry and NMP power layer slurry to overlap at the $10s^{-1}$ shear rate, the total slurry solids needed to be adjusted. A NMP power layer cathode requires 65% total slurry solids, and a PPG power layer cathode requires 75% total slurry solids to reach the same viscosity. Once again, there is a 10% total slurry solids advantage for a PPG power layer system. Table 4 demonstrates the calculations and overall solvent savings by using a PPG system. At the energy layer formulation, the PPG system will use 39% less solvent.

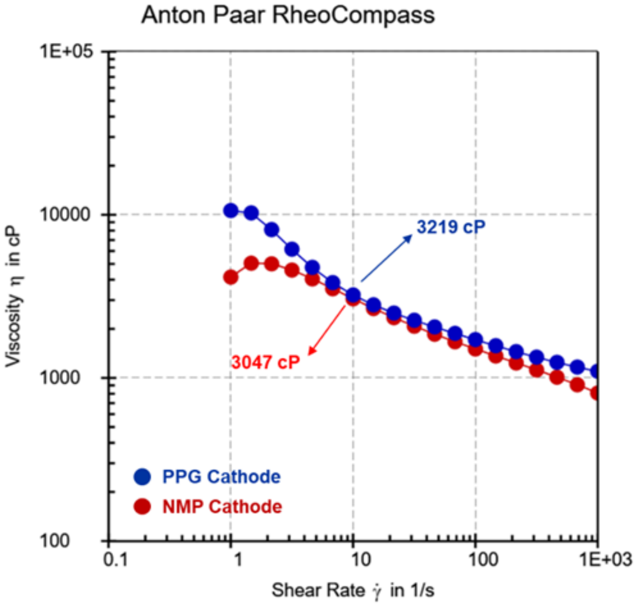


*Figure 46. Rheology Flow Curve for Energy Layer Cathodes (96 NMC622/2 Carbon1/2 Binder) Slurries Formulated Using PPG and NMP Systems*

**Table 4. Comparison of NMP Cathode and PPG Cathode Systems at Energy Layer Formulation Ratios**

| 96/2/2 NMC622/ Carbon 1/ Binder | | |
|---|---|---|
| **Formulation** | **NMP Cathode** | **PPG Cathode** |
| **Viscosity @ 10/s** | 3047 cP | 3219 cP |
| **Total Slurry Solids** | 65% | 75% |
| **Basis = 1 kg solids** | | |
| **Slurry Mass** | 1.54 kg | 1.33 kg |
| **Solvent Mass** | 0.54 kg | 0.33 kg |
| **% Δ (versus NMP)** | - | **39% less solvent** |

Since the PPG power and energy layer cathodes demonstrate almost the same percentage of less solvent when compared to an NMP system as demonstrated in Phase I, it is safe to assume that there will once again be a greater than 50% energy reduction. Deploying a PPG system with an NMP-free binder technology will increase the total solids of the cathode slurry and greatly reduce the energy consumption when compared to an NMP-containing system. The reduction in solvent demand for both power and energy cathodes formulated using the PPG binder technology will also yield additional benefits like high efficiency and productivity because of the ability to operate at higher line speeds. This could result in additional cost-savings.

# 4. Conclusions and Benefits

PPG evaluated NMP-free cathodes tailored for use in lithium-ion batteries designed for automotive starting, lighting, and ignition in military vehicles in accordance with 6T specifications. In Phase II, PPG developed a dual layer coating system composing of a power and energy cathode that evaluated two different coating architecture methods: wet-on-dry and wet-on-wet. PPG identified the wet-on-dry method to demonstrate the best cell performance from half-cell coin cells, full-cell coin cells, and single layer pouch cells. PPG also addressed the need for anode binders with improved performance and cost reduction. PPG tested its capabilities for screening new anode binders for high power performance beginning with identification of proper graphite, conductive carbon additives, and control binders for power performance. This binder was shown to improve fast discharge performance without impacting cycle life. PPG then worked with the UMBL to produce a series of 1.1 Ah multi-layer pouch cell, including 9 formulated with standard electrolyte, and 9 formulated with low temperature electrolyte. As uncovered in Task 1.2, UMBL was unable to produce pilot scale qualities of dual layer cathode foils, so single layer cathode containing PPG binder was supplied instead. PPG anode binders developed were scaled with no issues and were also included in the delivered cells. Pouch cell testing revealed that the performance that was in-line with 6T requirements. Validation of these results by GVSC is currently underway.

### Benefits

- Formulation of novel cathode coating architecture slurries using PPG’s NMP-free binder technology requires about 40% less solvent than control slurries
- Cell production cost savings of about 5.7% are expected based on reductions in mixing, coating, and drying times when cathodes are formulated using PPG’s NMP-free binder technology compared to PVDF-NMP
- Multi-layer pouch cell testing of novel PPG anode binder improves carbon dispersion and fast discharge capacity retention without impacting more most cycling up to 500 cycles
- Combining novel PPG cathode coating architecture and anode binders yields cells with improved power performance from room temperature down to -40°C