Final Project Report

Project Title: Advanced electrode manufacturing to enable low cost PEM electrolysis

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Recipient: Proton Energy Systems Inc. d/b/a Nel Hydrogen US

Working Partners: National Renewable Energy Laboratory (NREL), Oak Ridge National Laboratory (ORNL), General Motors (GM), and Kodak

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Executive Summary:

This project focused on one of the highest cost components of proton exchange membrane (PEM)-based electrolyzers, the catalyst-coated membrane (CCM). A PEM electrolyzer typically operates through splitting of water at the anode to form oxygen, with protons flowing through the membrane to form hydrogen on the other side. Catalysts contacting each side of the membrane reduce the energy required for the generation of the hydrogen and oxygen, while the membrane separates the gases. The CCM contributes substantially to both the capital cost and efficiency of the electrolyzer; the high cost is due to the high loadings of platinum group metals and the thick fluorinated membranes, while the efficiency is also highly dependent on the membrane thickness, as well as the activity of the catalysts.

Currently, the CCM manufacturing is more limited by the process capability of the methods of manufacture used, rather than the requirements of the device. For example, catalyst layer deposition methods which require the use of screens or similar templating to form the layers are limited in going to lower loadings because the layer has to be as thick as the screen. Methods which put significant stress on the membrane also result in thicker membranes being used in the manufacturing process, even if they are not needed in the cell. Moving to alternate manufacturing methods which can produce uniform layers at lower thicknesses and use more controlled handling methods can therefore reduce material usage significantly. In this project, the goal was over 75% reduction in catalyst loading, while showing a pathway to over 50% reduction in membrane thickness.

The fuel cell industry, including General Motors, has already done significant work in transitioning electrode manufacturing to roll to roll methods which provide the required process capability to meet these goals. The National Labs also have a roll to roll consortium to study ink and coating fundamentals, which includes Oak Ridge National Lab and the National Renewable Energy Lab. Nel had all three partners on the project to assist in material selection, process refinement, and quality inspection. Kodak was also a part of the team as a toll manufacturer with roll to roll capabilities that could be applied to Nel's materials and formulas.

Within the project, the team first evaluated catalyst materials and different methods of mixing, using a benchtop coater, and then transitioned to the larger equipment at the National Labs and Kodak. For the cathode, a platinum catalyst supported on high surface area carbon was selected, with the carbon support chosen for appropriate hydrophilicity and resistance to corrosion in the electrolyzer application. For the anode, an iridium oxide catalyst was chosen for its processability in the ink as well as activity at lower loadings. Various mixing equipment was evaluated to determine the impact of mixing on coating quality, and different coating machines were used to coat and dry the electrodes. Ultimately, slot die printing was downselected due to the better coating quality at Kodak with that method, and the coating substrate was selected based on coating properties on the substrate and release of the ink to during lamination.

Successful outcomes of the project included demonstration of durable electrode fabrication at 25% of the baseline catalyst loading, refinement of the process conditions and successful scale up of the process to Nel's larger electrode sizes, and resulting cost modeling demonstrating over $100/kW savings in component cost at scale. Based on the results, Nel is investing several million dollars to insource this process to the Wallingford facility and expand to 500 MW/year CCM capacity.

Project Overview

A critical challenge for the implementation of proton exchange membrane (PEM)-based water electrolyzers for the H2@Scale vision is the capital cost, which is largely driven by related factors of overdesign and highly manual legacy manufacturing methods. Typical electrolyzer cells have an order of magnitude higher membrane thickness and catalyst loading vs. fuel cells, in part because the manufacturing methods being used are not capable of producing uniform and reliable electrodes on thin membranes with low loading. Advanced electrode manufacturing is therefore the key enabler to reduce the cost of the electrolyzer stack, particularly roll-to-roll (R2R) manufacturing of catalyst coated layers (CCLs) and catalyst coated membranes (CCMs). The electrolyzer manufacturing process still relies on traditional sheet-to-sheet (S2S) processes, which requires intensive labor to make parts to meet the specification requirements. This project leverages fuel cell expertise (GM) with Proton's knowledge of electrolyzer components to develop the electrocatalyst ink formulations and coating processes using slot die (ORNL) and gravure (NREL) coatings, with proof of concept at the pilot-scale (Kodak).

Background:

Proton Energy Systems d/b/a Nel Hydrogen US is a leader in the PEM electrolysis market and has been serving customers for the last >20 years. Its global products range from benchtop to megawatt units that generate hydrogen from 0.2 to 7500 L/min. Nel's platforms are built on cell-stack components that were originally designed for oxygen generation to support life in critical environments, with >60,000 hour lifetimes demonstrated. Also, Nel has thousands of cells with >10 years in field service. At the same time, Nel has worked to continuously advance the technology, reducing membrane thickness by 30%, catalyst loading by 50%, and bipolar plate cost by 80%, while scaling to larger and larger units. Making the next technology leap to R2R manufacturing processes requires significant resources and a fundamental understanding of the catalyst-ionomer interaction, ink rheology, and catalyst layer properties, but also possesses the potential to decrease catalyst loading by >75% and membrane thickness by over 50% over Nel's state-of-the-art design. Reduction in CCM cost and labor also contributes substantially to Nel's overall cell-stack technology roadmap to drive electrolysis costs below $400/kW (see **Table 1** in the Project Goal section).

Ongoing research within the partnership provides strong confidence in the success of this approach. Both ONRL and NREL have been developing capabilities in R2R coating methods which have successfully been applied to electrochemical cells, and have already started a CRADA with Nel to evaluate the highest risk elements for electrolysis. GM has also been very active for the last 10 years in regards to developing manufacturing strategies to enable higher throughput manufacturing methods at a commercial scale. This progress has occurred in very close partnership with product design development. GM has demonstrated fully functional and durable fuel cell electrodes using both sheet coating methods and continuous R2R coating methods and has gained valuable experience in the relevant scale-up factors to enable robust, consistent, high-quality electrodes. Finally, the Kodak facility provides an ideal proving ground for translation of the fundamental material and process learnings from this project to relevant scale and equipment.

Project Goal:

The goal of this project is to establish R2R manufacturing of membrane electrode assembly components for PEM water electrolyzer through 1) improving the manufacturing speed rate by 100 times; 2) reducing the total PGM catalyst loading by 75%; 3) reducing the labor cost by 10 times; and 4) retaining a durability similar to baseline. The success of this project depends on the collaborative sharing of gained knowledge between the partner institutions and the integration of the development.

Table 1: Project goals vs. baseline and lab testing

Specification	Nel State of the Art	Nel Lab Demonstrated	Specific Project Target
CCM manufacturing	S2S	S2S	R2R
PGM catalyst loading, mg/cm^2	3	0.5	0.7
Membrane cost, % of baseline cell	40%	17%*	23%
Labor cost, % of baseline cell	27%	N/A*	2%
Total CCM cost, $ /kW	$250/kW	$70/kW*	$90/kW
Durability, mV/1000 hr	<5	<5	<5
Manufacturing Readiness Level (MRL)	9	2	5

* Estimated at high volume production

Key Accomplishments

- NREL performed studies on ink processing, rheology, and direct coating onto a membrane – a key learning was that Cowles blade and high shear mixing yield similar results.
- Nel was able to demonstrate performance parity with the benchmark MEA, while significantly reducing the PGM loading for both Pt (3x) and Ir (4x). This was accomplished with benchtop coating methods.
- Nel demonstrated the ability to effectively mitigate crossover on N115 membranes with an alternative approach.
- ORNL performed studies to understand the contributions to wrinkling from alcohols and water to support efforts to directly coat membrane material.
- Nel screened and down selected catalyst materials based on the following three criteria: i) ability to process well, ii) ability to reach desired performance at target loading, and iii) operational durability.
- The team transferred learnings from subscale, benchtop testing to the Kodak pilot coating line and showed comparable performance and durability in the 86 cm² cell architecture. Techniques explored include forward gravure, reverse gravure, and slot die coating. Direct deposition on to the membrane and deposition onto a substrate were both explored.
- Team demonstrated 3,000 h durability on an 86 cm² stack and scaled up to Nel's 680 cm² platform and demonstrated good beginning of life performance.
- NREL demonstrated in-line defect identification using visible light transmission.

Status:

Task 1 – Development of electrolyzer catalyst electrodes.

The purpose of this task is to understand whether we can obtain similar rheology for each ink dispersion technique in preparation for future expansion from lab scale to industrial scale. The bead mill is an ink dispersion method mainly used at lab scale, but there are problems such as the number of beads that would be needed for high volume ink processing and difficulty in recovering the ink from the surface of the beads. Turrax mixers are suitable for high volume dispersions, and Kodak, who is contributing to the upscaling study, uses a similar mechanism. A Tube Turrax mixer could provide an acceptable interim approach between these two options if the ink behavior can be shown to be similar. The different equipment can be seen in Figure 1.

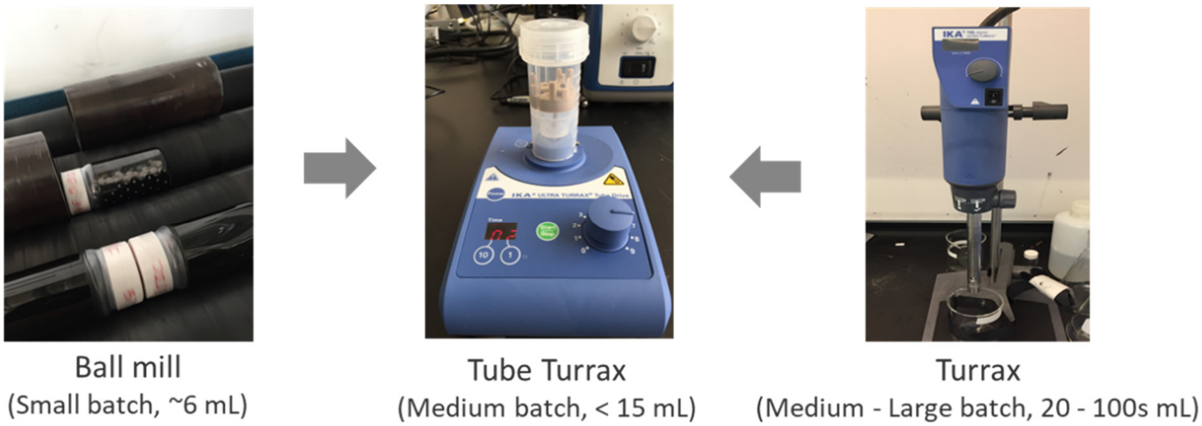


Figure 1: Ink mixing methods in use at NREL: bead mill (left), Tube Turrax (middle), and Turrax (right).

The cathode catalyst used was 46.6wt.% Pt on high surface area carbon (HSC). The inks were prepared by mixing desired amounts of all the components, Pt/HSC (at 3 and 6 wt%) and Aquivion ionomer solution, in a water—n-propanol mixture. Each solution was formulated in different volumes. In the case of bead mill, a total of 5 g of solution was prepared, 50 g of zirconia beads were added, and the solution was milled at a speed of 80 rpm . For Turrax mixing, a total of 20 g of solution was prepared, and shear mixed at 10,000 rpm in an ice bath. For Tube Turrax, a total of 15 g of solution was prepared and mixed at 6,000 rpm for 3 different times (1x, 2x, and 4x) for comparison. The results showed that all three methods produced inks with similar rheological properties, including when ink was scaled up in the Turrax mixer.

Proof of the direct coating concept: A Pt/HSC ink was coated onto the membrane with a Mayer rod. The 46.6wt.% Pt/HSC catalyst and Aquivion ionomer solution were used, with the same formulation as the 6% catalyst ink above. The formulated solution was coated on 50 µm and 175 µm membranes using a metering rod. The coated films were dried at 60˚C. Figure 2 shows the coating results that proved excellent coating properties without dewetting on both membranes. Microscope images also show that the catalyst ink is uniformly well coated. However, it was observed that membrane swelling occurred during the coating trials. This result suggests the need for a more detailed membrane swelling study as a function of ink formulation conditions.

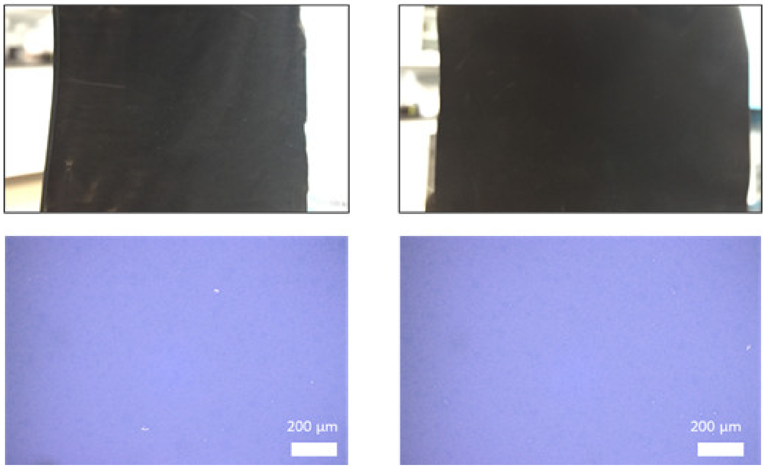


Figure 2: Photos and microscope images of Pt/HSC coated membranes: coated on 50 µm membrane (left column), and 175 µm membrane (right column).

A study on impact of ink properties on viscosity was performed. The following, in order of greatest significance, were found to have an impact: nPA content, then I:C ratio, and catalyst wt.%. The contour plots in Figure 3 also show the relationships between these variables, and the plots can be represented by the following quadratic regression model, which can be used for ink optimization in the future operating conditions.

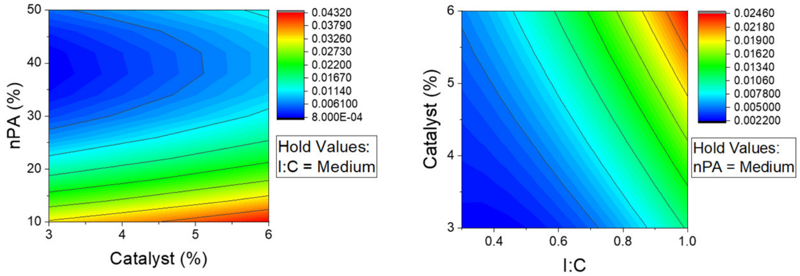


Figure 3: Contour plots of normalized viscosity values via catalyst and nPA (left) and I:C and catalyst (right).

Cathode coating on membrane: NREL performed lab-scale coating of platinum on carbon inks (50 wt% Pt/HSC) inks directly on membranes. Three inks corresponding to No. 1, 8, and 3 in Table 3 were selected from the formulated inks for repeat trials as listed again in Table 5. The ink was coated at a controlled rate of 35 mm/s using an auto applicator (QPI-AFA3800, Qualtech Products Industry) on a 127-μm-thick commercial membrane. Because each ink had a different wt.%, different US Mayer rods of #25, #30, and #35 were used for each of No. 1, 8, and 3 inks, respectively to get to similar loading. The membrane was fixed with tape to apply tension. Swelling of the membrane was observed immediately after the rod passed. The coated film was dried for 15 min at 80 ˚C in a hot-air oven. After drying, the bumpy surface on the membrane was relaxed, but not completely removed.

Table 2: Selected inks for coating trials

No.	Catalyst, wt%	nPA, %	I:C ratio
1	Low (-1.5%)	Medium	Low (-0.35)
8	Medium	High (+20%)	High (+0.35)
3	High (+1.5%)	High (+20%	Medium

Figure 4 shows the direct coated Pt/HSC inks on the membrane. The prepared CCMs visually looked similar to conventional sprayed CCMs and showed uniform distribution of catalyst.

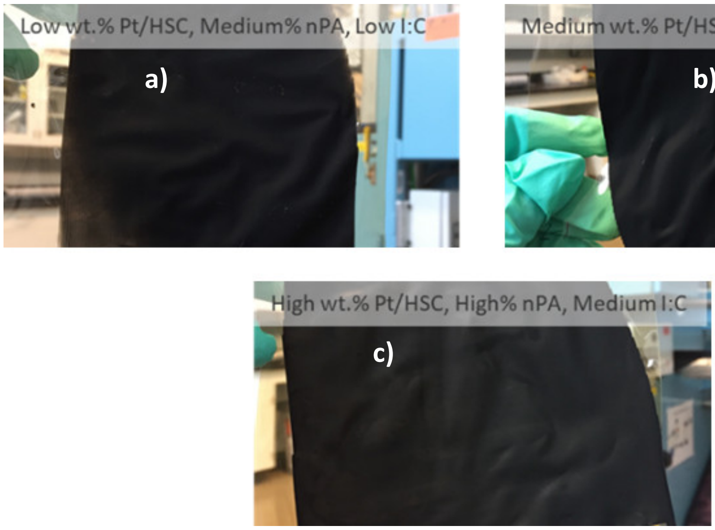


Figure 4: Photographs of the direct coated Pt/HSC inks on a commercial 127 µm membrane. Samples with low wt.% Pt/HSC dispersion (4a – top left), medium wt.% Pt/HSC dispersion (4b – top right), and high wt.% Pt/HSC dispersion (4c - bottom).

Microscopic analysis confirmed the morphology of the coating surface as shown in Figure 5a-c. Excellent coating surfaces without cracks or pinholes were obtained with low and high wt.% Pt/HSC inks (Figure 5a and 5c). However, when using medium wt.% Pt/HSC ink (Figure 5b), many cracks occurred on the surface.

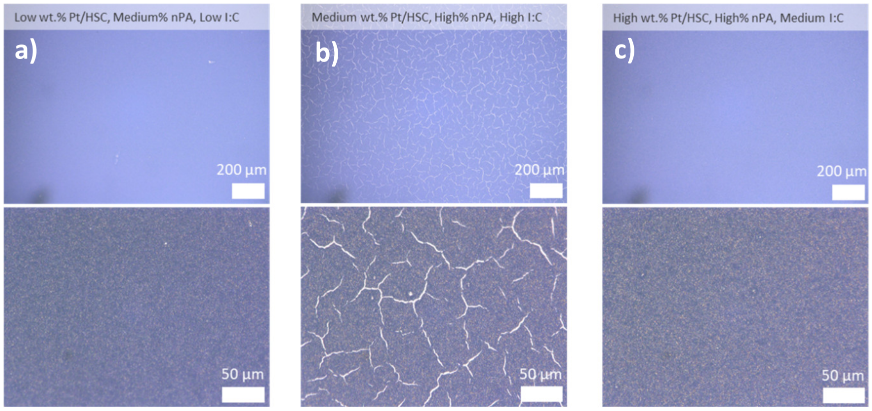


Figure 5: Microscopic images of the direct coated Pt/HSC inks on the membrane with low wt.% Pt/HSC dispersion (5a - left), medium wt.% Pt/HSC dispersion (5b - middle), and high wt.% Pt/HSC dispersion (5c - right). Top and bottom images show 200x and 1000x magnification, respectively.

This result is the same for the repeated experiments shown in Figure 6a, which improved as the I:C ratio dropped. Through this experiment, it can be seen that high I:C causes cracking.

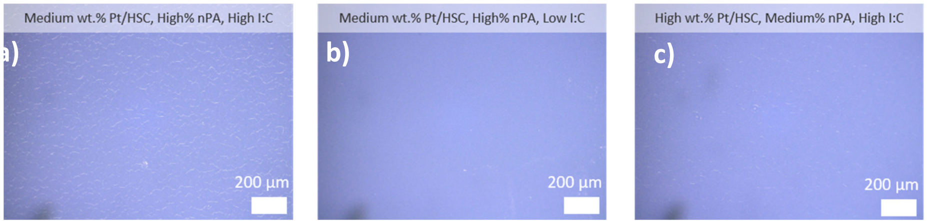
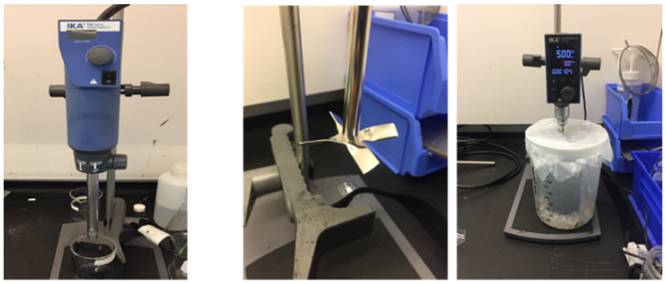


Figure 6: Microscopic images of the direct coated Pt/HSC inks on the membrane. Comparison of coating surfaces of inks with high I:C (6a – left) vs. low I:C (6b – middle) and high% nPA (6b – middle) vs. medium% nPA (6c – left). The re-coated ink composition as shown in Figure 6a is the same as shown in Figure 5b. Cracking also occured when I:C ratio was high for high catalyst wt.% ink.

Turrax vs. Cowles Blade: Kodak uses a Cowles blade/propeller type mixer, not the high shear mixing we have been using up to now. As shown in Figure 7, the mixing performance of the Cowles blade mixer owned by NREL and the Turrax mixer used previously was compared. The Cowles blade mixer has a low mixing speed of 500 rpm compared to the Turrax mixer at 10,000 rpm. In this experiment, we focused on finding a time condition that can achieve dispersion performance similar to high-speed mixing with low-speed mixing (Figure 8).


Turrax mixer
(10,000 rpm)

Cowles blade mixer
(500 rpm)

Figure 7: High speed Turrax mixer (left) and low speed Cowles blade mixer (right)

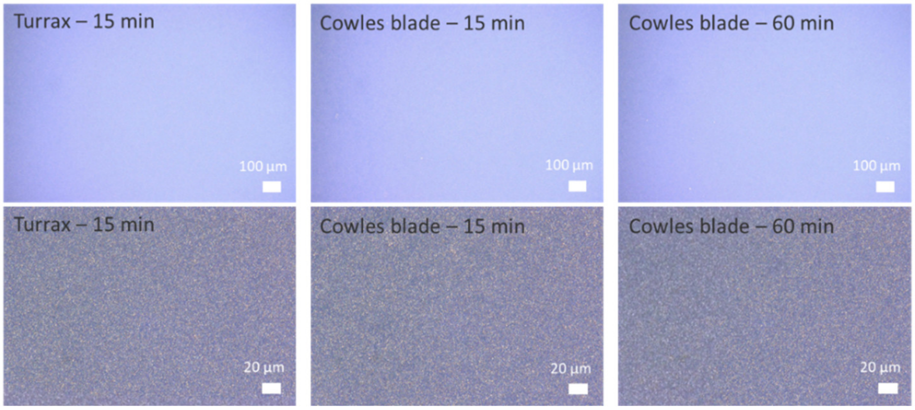


Figure 8: Optical microscopic images of the half-CCMs using differently mixed inks: 200x (top row) and 1000x (lower row) magnification

Vulcan carbon was selected as the catalyst support of interest for pilot coating at Kodak since the low surface area carbon (Vulcan) showed better performance than the high surface area carbon (HSC) from the *in-situ* testing at Nel Hydrogen. In ink formulation, pure Vulcan carbon powder without platinum was used to prevent catalyst loss. The water to 1-propanol (Water/nPA) ratio was controlled to 75:25. The ionomer to carbon (I/C) ratio was controlled to 0.45.

Silverson High Shear Mixing: We purchased a Silverson L5MA lab mixer equipped with several different mixing heads (Figure 9). This same mixing physics is able to scale to 100s of liters, so the learnings from the lab scale are directly transferable. After several iterations and building on the background knowledge of our partners, we optimized the mix speed and time. We then successfully tested decals made with this ink and our benchtop coating process to show comparable performance to the ball milling method.

Figure 9: Silverson Lab Mixer capable of mixing from 1 to 12,000 mL

Task 2 - Bench Scale Fabrication of CCLs

The Mayer Rod Coating (MRC) method was chosen to print electrode decals due to its ability to finely control decal thickness and ultimately, catalyst loading. The wide range of ink loadings that can be achieved with this method is governed by its ability to coat at speeds ranging from 1 to 39 m/min and its flexibility to coat with various Mayer rods. These Mayer rods have K bar numbers and the K bar number for each rod correlates with a specific flow of catalyst ink through the grooves of the coiled wire. Specifically, an increase in K bar number equates to a larger groove size and an increase in ink layer thickness and catalyst loading.

Before printing each decal, the bench coater was set to a speed of 7 m/min and the bench coater rod hooks were “homed” next to the substrate clip. A piece of Teflon® was cut and placed under the bench coater clip, to be used as the substrate. Earlier in the project, a K bar was selected, based on a series of loading tests, that would allow us to test values that would achieve program targets. After pipetting ink onto the coater, the K bar then travels across the substrate, coating the Teflon® sheet. See Figure 10 below. Once the decal air-dried, it was placed into a low-temp oven to cure and set the decal. This process was used for both the cathode and anode development.

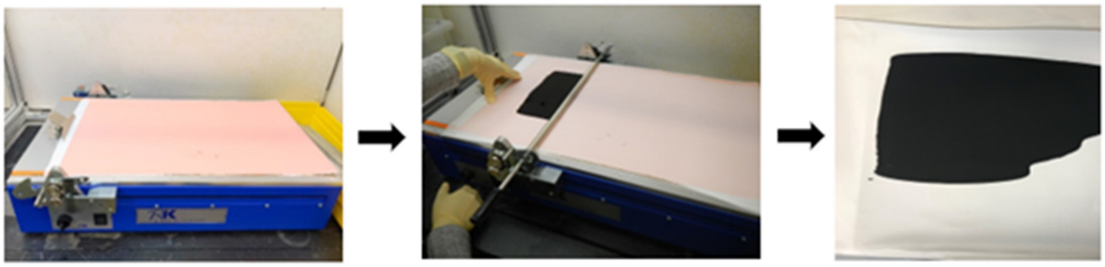
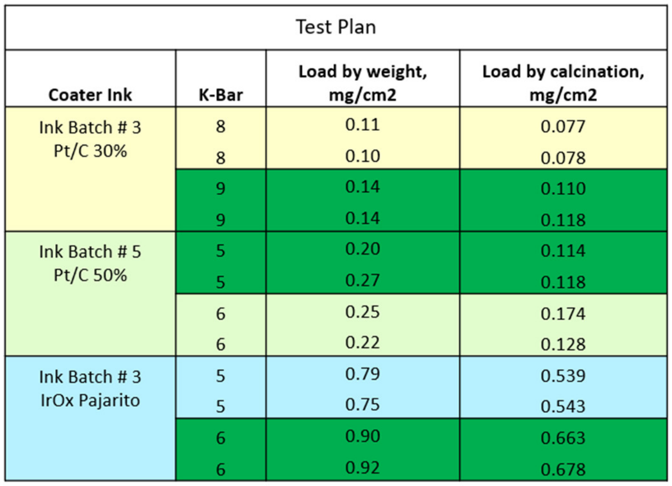


Figure 10: Bench coater setup and ink printing, as deposited on the substrate

A decal loading test matrix, shown in Figure 11, was developed to define the metal loading in mg/cm^2 of coated ink layers. This was needed to ensure metal loadings of the cathode and anode coater layers achieved the targets of 0.1 mg/cm^2 and 0.6 mg/cm^2, respectively. Printed decals were cut and calcinated at 600°C to compare loading measurements with a loading by weight technique. Surprisingly, the loading by calcination technique suggested that the actual decal loadings were lower than what was calculated using the loading by weight method. Higher K bars rod sizes were then used to increase the metal loadings of the ink coater layers. After repeating both loading determination techniques, K bars were selected for each ink that would achieve the optimal loading. As a result, one K bar was selected for IrOx printing to achieve a metal loading of 0.6 mg/cm^2, a lower K bar was selected for 50wt% Pt/C printing to achieve a metal loading of 0.1 mg/cm^2, and a higher K bar was selected to achieve a metal loading of 0.1 mg/cm^2.



Test Plan

Coater Ink	K-Bar	Load by weight, mg/cm2	Load by calcination, mg/cm2
Ink Batch # 3 Pt/C 30%	8	0.11	0.077
	8	0.10	0.078
	9	0.14	0.110
	9	0.14	0.118
Ink Batch # 5 Pt/C 50%	5	0.20	0.114
	5	0.27	0.118
	6	0.25	0.174
	6	0.22	0.128
Ink Batch # 3 IrOx Pajarito	5	0.79	0.539
	5	0.75	0.543
	6	0.90	0.663
	6	0.92	0.678

Figure 11: Test plan and results for metal loading evaluation of Meyer rod coated layers using two different techniques: loading by weight and loading by calcination

Pt is the best known catalyst for hydrogen evolution reaction (HER), and it requires only small overpotential even at high current densities. Pt black catalyst has been historically used as an HER catalyst on the cathode to maintain durability over tens of thousands of hours of operation. In this work, carbon-supported Pt catalysts are being evaluated for HER since supported catalysts have the potential to reduce the cathode catalyst loading through increased utilization. Typically nanoparticles have high surface energy due to a higher number of uncoordinated surface atoms relative to bulk atoms, which increase both Ostwald ripening and sintering of the nanoparticles. However, based on years of fuel cell experience, suppliers have improved the catalysts with proprietary manufacturing processes to mitigate the sintering effects. In this work, a 30 wt% Pt/C was evaluated. As shown in Figure 12, the performance is comparable with 50 wt% Pt/C. A benefit of the 30wt% material vs. the 50wt% material is that the carbon support used is partially graphitized. A graphitized carbon catalyst should be beneficial in mitigating carbon corrosion in electrolyzers, which may be experienced during start-ups and shutdowns. Screen printing is one widely used technique for manufacturing electrodes, and an attempt was made to print Pt/C cathodes. Initial screening of these catalysts shows that both the supported catalysts' cell voltages are better than the Pt black baseline at lower current densities. Reducing the catalyst loading from 0.46 to 0.37 mg-Pt/cm^2 significantly reduced the cell voltages for 50 wt% Pt/C, again confirming the performance sensitivity of the carbon-supported catalyst to catalyst loading/electrode thickness as seen in Task 2. The 30wt% Pt/C shows an increased polarization slope and needs optimization of the electrode. For the Pt/C electrodes, the electrode thickness is mainly driven by the carbon content and carbon density compared to Pt or ionomer.

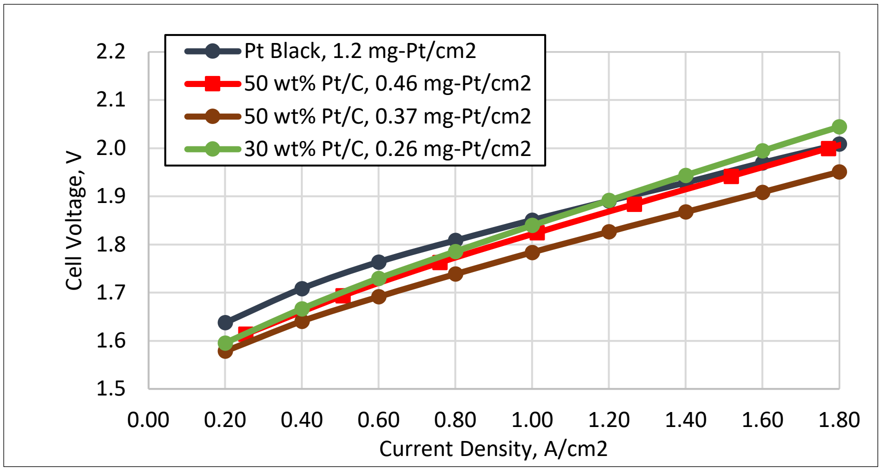


Figure 12: Polarization curves of CCMs with Pt black, 30, and 50 wt% Pt/C cathode catalysts. The active area of the MEA is 25 cm^2, and the test was conducted at 50°C cell temperature and ambient pressure.

IrOx is the best-known catalyst for oxygen evolution reaction (OER). It has a better trade-off between OER activity and durability when compared to alternatives such as RuOx. For the R2R work, a relatively high surface area (HAS) IrOx was evaluated. The surface area of the HAS catalyst is about 4 times higher than the baseline IrOx catalyst. As shown in Figure 13, the HAS IrOx shows good stability similar to the baseline during the 520 hours of test.

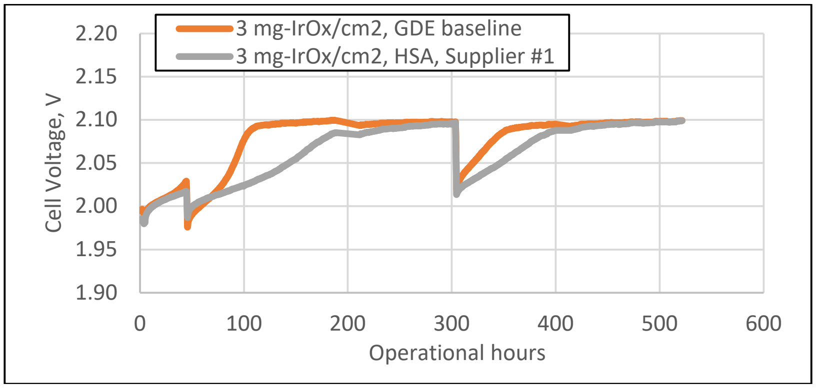


Figure 13: Short term stability test of the HAS IrOx at 1.86 A/cm^2. The active area of the MEA is 28 cm^2, and the test was conducted at 50 ^{o}C cell temperature and operated at a differential pressure of 400 psi.

To test the ability of the higher surface area material to perform well at lower catalyst loadings, an electrode with a 0.66 mg-IrOx/cm^2 loading, 4.5 times lower than baseline, was prepared by an airbrush technique by spraying the HSA catalyst ink on a porous transport layer (PTL) and an ionomer dispersion was also spray-coated on top of it. The MEA was prepared by assembling the airbrushed anode GDE and a standard cathode GDE with the 175 µm membrane. The cell voltage of HSA IrOx, as shown in Figure 14Figure 13, is 18 mV lower than the baseline at 2 A/cm^2. At the end of the second day of operation, the cell voltage of the HSA IrOx catalyst is similar to the baseline. Although the increase in cell voltage is not significant, it will be worth exploring the performance of the HSA IrOx with the rod coating method and determine the stability of the lower loaded electrode with operational time.

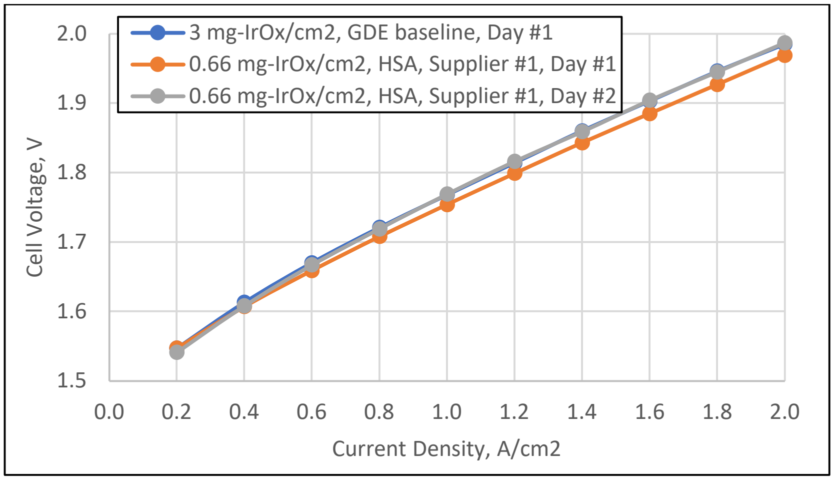


Figure 14: Polarization curves of MEA with HSA IrOx anode catalyst. A standard GDE with 3 mg-Pt/cm^2 was used as the cathode. The active area of the MEA is 25 cm^2, and the test was conducted at 50 ^{o}C cell temperature and ambient pressure.

The next step in CCM development was to fabricate a full CCM onto N115 membrane using the same down-selected OER and HER catalysts that were used for the N117 test. This allowed for a direct comparison between the different membrane thicknesses. Figure 15 reveals that both full CCMs on N115 demonstrated stable performance at approximately 1.95 V, even after un-intended shutdowns.

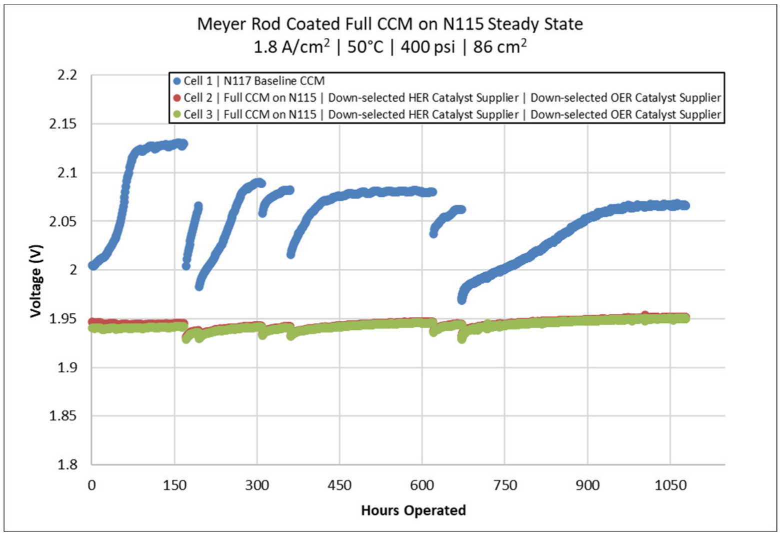


Figure 15: Performance of in-house laminated full CCMs on N115 using down-selected OER and HER catalysts benchmarked against a N117 baseline CCM.

The similarity in performance of both N115 full CCMs reflects the repeatability of the CCM lamination process. The performance of the full CCMs on N115 was more stable than the N117 baseline CCM. When compared to the full CCM on N117, the full CCMs on N115 show approximately a 60 mV improvement, which is expected from a thinner material.

Recombination Layer Development: Further recombination layer dispersion and ink development has been done to improve the effectiveness of preventing hydrogen crossover. Improvements have been made to both the formulation and processing of the recombination layer. The optimized dispersion, RC12, has been used for continued testing of the single layer and bilayer concepts. For the single layer concept, the recombination dispersion is mixed with the OER catalyst and laminated as one. Conversely, the recombination dispersion is laminated as a distinct layer underneath the OER catalyst ink for the bilayer concept. See schematics of each version in Figure 16.

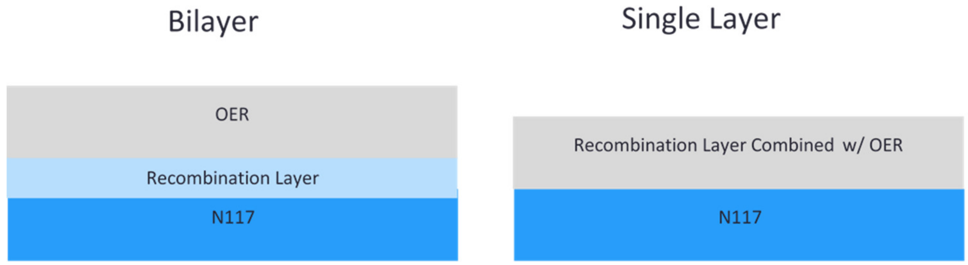


Figure 16: Recombination layer concepts: the bilayer (left) and single layer (right).

The most recent recombination layer work involved testing RC12 as a single layer on N117, a bilayer on N115, and a bilayer on N117. The extensive progress made on CCM fabrication has allowed for all of these recombination layers to be laminated as full CCMs, using down-selected catalyst inks for the appropriate electrodes.

Initial turndown testing, final turndown testing, and polarization curves were collected for 3 different stacks, as depicted in Figure 17. The results indicate that all RC12 recombination layers were effective in reducing hydrogen crossover when compared to an unmitigated MEA. At 1.86 A/cm^2, there was a 35%LEL for an unmitigated MEA, while none of the RC12 recombination layer stacks exceeded 20%LEL.

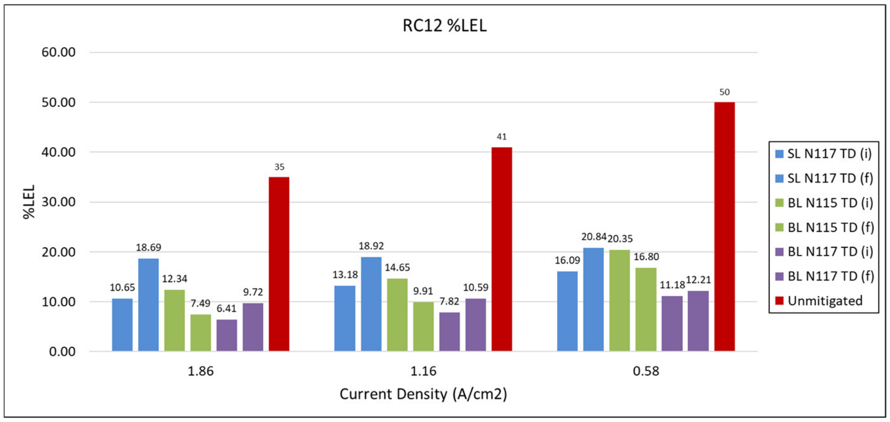


Figure 17: Hydrogen crossover levels (%LEL) of RC12 recombination layer tests during initial and final turndown testing of SL (single-layer) N117, BL (bilayer) on N115, and BL (bilayer) on N117, benchmarked against an unmitigated MEA

Testing was performed in order to explore changes in %LEL when using thinner membrane. The bilayer on N117 was most effective at lower current densities where it remained at approximately 12%LEL during both initial and final turndown. However, the bilayer on N117 showed a greater loss of effectiveness than the bilayer on N115 at higher current densities where the N117 test increased to 9.72%LEL during final turndown and the N115 test decreased to 7.49%LEL.

Task 3 - Task 3: CCM integration and testing

The baseline ink formulations, as identified within Tasks 1 and 2, were used to build a 3 cell stack. The stack also tested an option for a scalable gas crossover mitigation strategy. This cell stack ran for 1500 h and the data can be seen below in Figure 18.

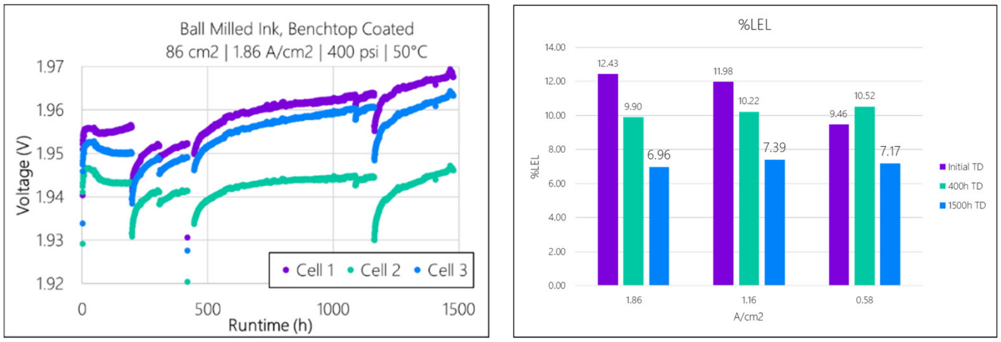


Figure 18: (Left) Cell voltage for 3 identical cells run at 1.86 A/cm², 400 psi H2 pressure, and 50°C. (right) % of the combustible limit of H2 in the O2 stream for the cell stack at different currents and different times during the test.

Throughout the test, the variation in cell voltage at full current was approximately 20 mV from cell to cell, which is within Nel's allowed range. The degradation rate also met the end of project goal of 500 hours with less than 10 mV/h decay. The linear degradation region from about 600 – 1100 h has slopes ranging from 4 to 10 µV/h for the three cells. However, another linear region from 1275 – 1500 h existed and the degradation rate ranged from 16 to 22 µV/h. While these rates are relatively low for an experimental, low loaded MEA, Nel will continue to work to understand and minimize the voltage degradation before integration into product. The combustible gas crossover mitigation was also part of this test with Nel's internal hydrogen permeation test performed at the start (t=0), after 400 h, and after 1500 h of runtime. At each step, the reduction in %LEL was slightly better than the previous step. This result shows that the layer is not losing effectiveness with runtime.

The next key durability stack involved decals made using high shear mixing. This stack reached the end of its planned runtime and all cells showed lower voltage at the end of test than at the beginning of the test – see Figure 19. Next it was time to test electrodes made using Kodak coating equipment. Cell 1 used cathodes from the Gravure trial (March 10th), Cell 2 used cathodes from the slot die trial (May 18th), and Cell 3 used a baseline benchtop coated cathode. All cells used baseline, benchtop coated anodes from the same batch to ensure the rest of the cell was consistent. The 1000 h durability test can be seen below in Figure 20. The results showed comparable performance between the benchtop cathode and the cathodes printed using the pilot coating equipment at Kodak. The degradation rate is slightly higher than the target of 10 µV/h, but this is still a very promising result. This test showed that both gravure and slot die printing are able to result in acceptable performance and durability.

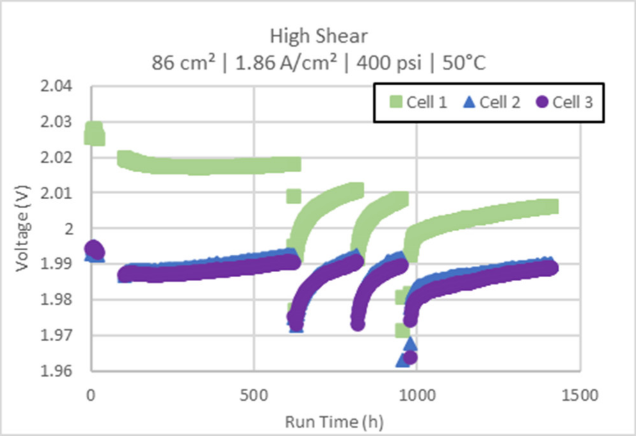


Figure 19: Cell voltage for 3 identical cells run at 1.86 A/cm², 400 psi H2 pressure, and 50°C.

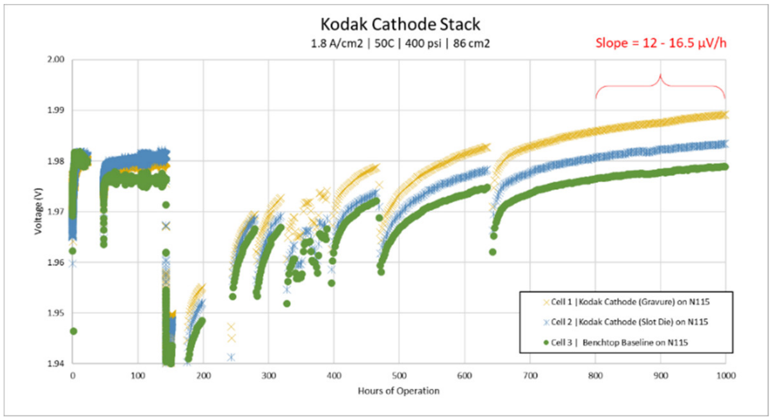


Figure 20: Durability test for pilot scale cathodes

The last key aspect to move from the benchtop to the Kodak coating equipment was the anode. A stack made of exclusively Kodak coated layers on N115 was built with nominally identical cathode layers and three different anode configurations, as noted in the legend in Figure 21. This stack reached >3,000 h and was able to demonstrate a degradation rate of ~1.5 µV/h with a low loaded electrode and even lower degradation rate with the average Ir loaded sample.

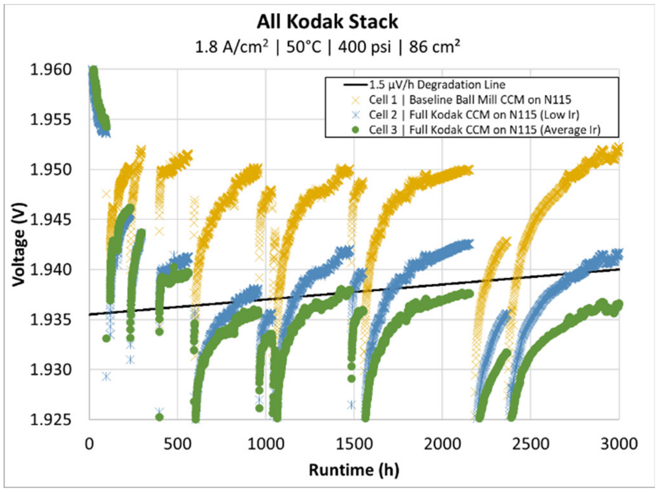


Figure 21: Durability data for a stack made from R2R coated layers.

Building on this success, focus shifted towards proving out the process at the 680 cm² active area scale. A cell stack was built with a commercial baseline MEA and a Nel R2R MEA. This stack ran for over 500 h and demonstrated similar performance between the commercial baseline and the Nel R2R MEA (see Figure 22).

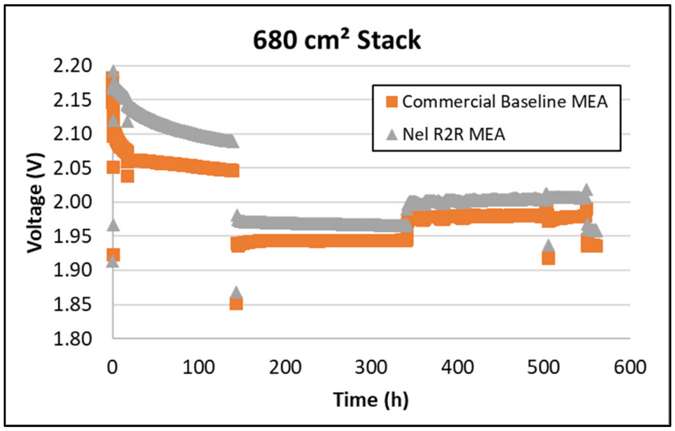


Figure 22: Cell performance data comparing a commercial baseline cell and R2R Cell in a 680 cm² stack.

In summary, cell stacks were tested for durability, mainly at the 86 cm² scale, moving from benchtop coating up through fully Kodak coated MEAs. The team then scaled up to the 680 cm² platform and showed performance parity with a commercial baseline MEA.

Task 4 - Task 4: Scale up and pilot trials

The first coating trial was performed in March 2021, testing several parameters including wet film thickness, engraving type, ink concentration, coating substrate (Teflon or N115), and line tension.

The different printing conditions were analyzed using the following metrics:

- Visual quality was determined using a qualitative rating system of 1 – 5.
- Platinum content, which ranged from 0.02 – 0.09 mg Pt / cm²
- General voltage performance compared to Nel's benchmark MEAs.

Sample 2a, 2b, 3, and 6 were determined to be the top priority samples and were tested for short term performance, which can be seen below in Figure 23.

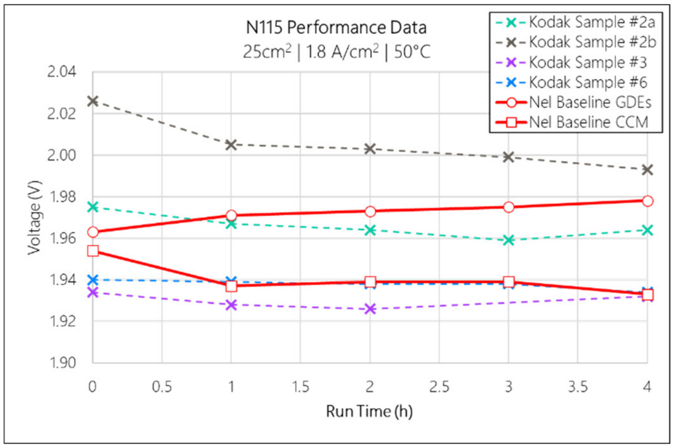


Figure 23: Short term voltage performance under standard conditions

Sample 2a and b were the same formula but coated on membrane vs. transfer substrate, samples 3 and 6 had a second pass of ink, and sample 6 was coated at faster speed. One noteworthy observation was that the visual quality correlated with the voltage performance, where better visual quality resulted in better overall cell performance.

Next, a slot die printing trial was performed in May 2021 at Kodak. Two batches of ink were prepared with different solids loadings. Ten coating runs were performed on Teflon substrate with a thickness of 5-mil. Different parameters like wet film thickness and in-line filtration were varied. The decal from trial #6 can be seen below in Figure 24. No obvious voids were observed using a backlit table. This trial also achieved the target loading for this program.

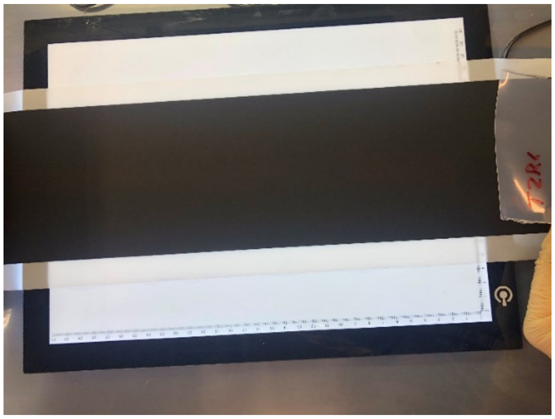


Figure 24: A back lit sample from trial #6.

Further analysis was performed using SEM to better judge the quality of the decal. All decals showed some non-uniform topography, but trial #6 was the most uniform.

In June 2021, we performed our third coating trial. This was the second trial using the gravure printer. Several parameters were tested including a custom cylinder, double pass alignment, and corona discharge treatment. In total, there were six printing trials which included an initial test on PET, a single pass, three attempts of aligning a double pass of ink, and a final trial of corona discharge treatment on a single pass.

The resulting prints of the six trials were analyzed using the following metrics:

- Visual quality was determined using a qualitative rating system of 1 – 5 (5 being the best).
- Voids / cm^2, which ranged from 1.30 – 45.61 voids / cm^2.
- Platinum content on select samples, which ranged from 0.015 – 0.091 mg Pt / cm^2 between the single and double pass.

Next, Nel conducted a 4th coating trial focused on the anode. The trial took place on In August 2021 and the primary variables tested were the wet film thickness and the impact of adding an in-line filter. The addition of an in-line filter resulted in poor quality decals, so most of the trials were run without a filter. The trial was able to achieve uniform coatings with target iridium loadings.

Now, with configurations providing satisfactory results at the sub-scale prints, the team focused on transitioning over to Kodak's digital pilot coater. A more narrow range of catalyst loadings was also targeted. The resulting cathode decals were defect free and wide enough to fit into Nel's 680 cm² active area (0.25 MW) cell stack platform. With the anode, some scale-up challenges were experienced resulting in less uniform decals at scale.

At Nel's direction, NREL put together a cost estimate of the project funds that would be required to modify NREL's existing roll-to-roll coating equipment to allow for gravure printing. Printing electrode patches would improve materials utilization, as the catalyst is printed in only the active area of the electrolyzer cell. Up to this point in the project, gravure coating had been used to coat the membrane in a single, large stripe, as shown in the top of Figure 25. That coating process results in a wide strip of electrode material with catalyst coated well outside the cell active area (red circle shown in the upper right), leading to much of the coated catalyst material not being utilized for electrochemical reactions. This coating process was useful for proving out the direct coating processes but is less desirable for manufacturing due to the low material utilization of the expensive Pt catalyst. Printing electrode patches that match the cell active area, as shown in the bottom of Figure 25, will result in 100% of the coated catalyst material being utilized for electrochemical reaction. Limiting the catalyst to the active area will also improve sealing of the cells for operation at high differential pressure.

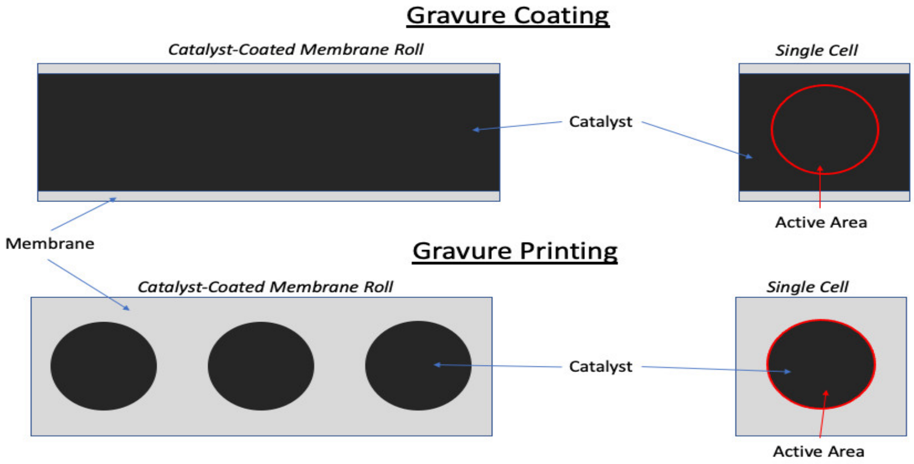


Figure 25: Different approaches to printing being explored at NREL

While ultimately moving to patch printing is optimal, NREL and Nel discussed the lack of progress in this area, due to supply chain challenges, and it was decided that it was best to abandon this effort for now and shift our focus to other matters, since the overall conversion to R2R is still a large benefit even with the excess catalyst.

NREL has been working on optical transmission imaging of half CCMs to assess catalyst layer loading uniformity. Pt/C catalyst layers on membrane created at Nel and NREL were imaged using a desk top scanner to measure light transmission. The same catalyst layers were also measured using x-ray fluorescence (XRF) spectroscopy. The average loading of the whole catalyst layer area was compared to the average transmission of the same area. These results are shown in Figure 26. It was observed that there is a strong correlation between XRF-measured loading and optical transmission.

The next step was to create high resolution images providing a loading map of the two catalyst layers. 80 local XRF loading measurements were obtained along two lines on each sample, as shown in Figure 27a. XRF measurement spots were then aligned spatially with the corresponding transmission images to obtain the transmission-to-loading correlation parameters

Figure 27b. Finally, the high-resolution loading maps of cathodes were obtained Figure 27c. Good correlation exists between the Pt loading and transmission values. This method has potential to become a valuable loading characterization technique for in-line monitoring of fabrication processes of PEMWE cathodes.

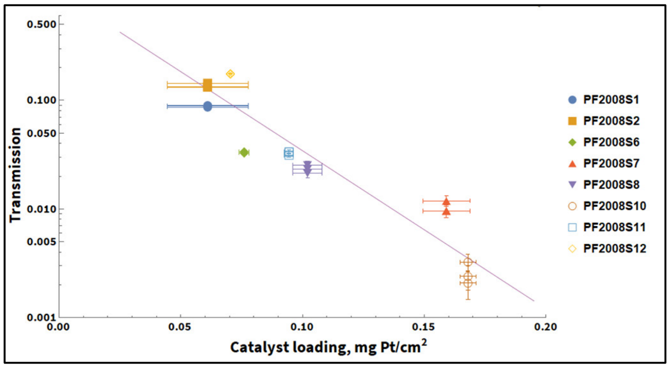


Figure 26: Correlation plot of XRF-measured catalyst loading and optical transmission.

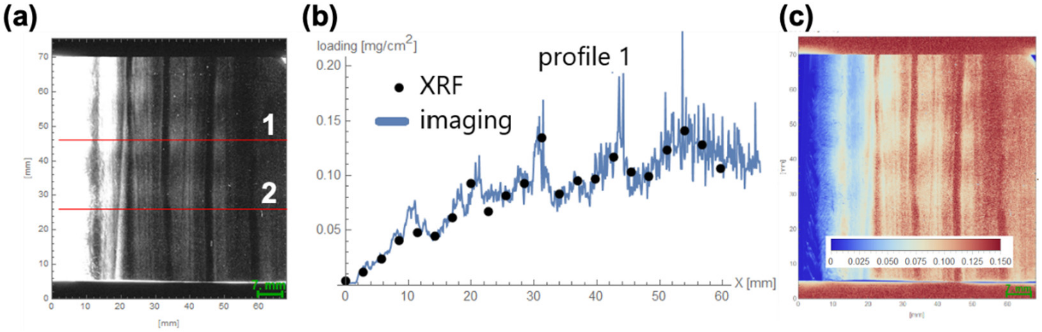


Figure 27: (a) Optical transmission image of half CCM. (b) Comparison of transmission and XRF-measured loading along line 1in (a). (c) High resolution spatial map of loading based on transmission-to-loading correlation generated from line scans.

NREL hypothesizes that the optical power of visible light transmitted through the catalyst layer of a material sample will correlate strongly with the catalyst areal loading, allowing the high spatial resolution of machine vision technology to be applied to loading measurement and defect detection. An initial study included nine cathode samples of varying catalyst loading and substrate type, and included scratch defects. These samples were prepared by Nel at one of their previous R2R pilot-coating trials at Kodak. To simulate machine vision scanning, the materials were scanned using a comparable desktop scanner, in both reflection and transmission modes, to compare the transmittance through these materials with the range of sensitivity of a typical machine vision system, and to learn how image results were affected by scanning mode. Figure 28 shows the transmission mode images for the measured samples.

Transmission mode scanning provided more coating structure information overall than reflection mode. Signal strength varied significantly with substrate type, with PTFE substrates significantly scattering or absorbing incident light. Typical coatings on PFSA and Kapton fell within the scanner sensitivity range, but a more sensitive system, and/or brighter light source, is likely necessary for high loadings and PTFE substrate materials. Loading qualitatively appears to correspond inversely to measured transmittance, and the technique is able to measure features including scratches, underlying striations, point defects, and large-area coating thickness variations. Overall, these experiments demonstrate that transmission-mode scanning would likely be a viable QC tool for loading quantification and defect detection.

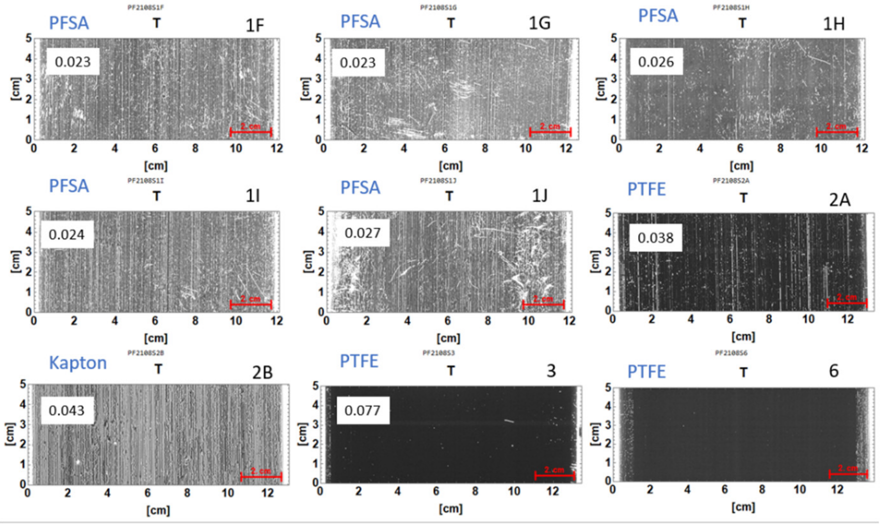


Figure 28: Optical transmittance scans of Nel electrolyzer cathode CCM and other materials. Defects and underlying striations running in the coating machine direction were observed. XRF-measured Pt loadings are reported in the inset text boxes.

Samples of approximately 7 ft in length were provided for webline evaluation. Two samples were measured in triplicate and confirmed the observed non-uniformities were real coating features (see Figure 29).

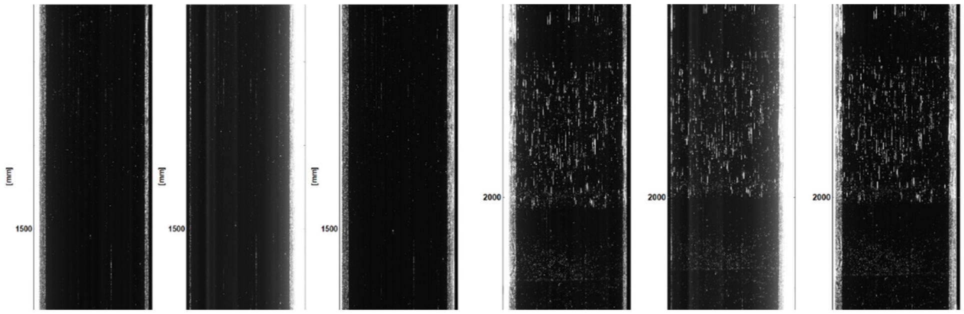


Figure 29: Images from transmission-mode webline optical scanning. (Left) Three scans of sample PF2108S6, showing very repeatable observed image features. (Right) A different section of the same sample, showing a region of varying density of possible defects.

Next, the region of possible defects was analyzed using automatic object detection. The highlighted objects were identified using image processing algorithms, with counts, intensities, and locations included in the outputs. The image processing was performed offline, but could be implemented in real time. An alert signal can then be sounded if the density of defects becomes too high. An example is shown in Figure 30.

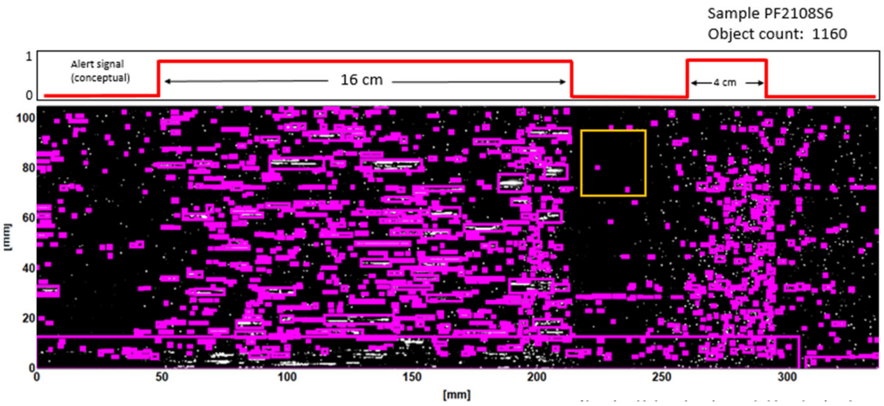


Figure 30: Defect identification using inline optical measurement.

Task 5 - Task 5: Cost analysis

Nel performed a total cost of ownership (TCO) analysis based on the final down selected configuration. The two biggest contributors to improving the TCO were moving from N117 to N115 and reducing the PGM content within the MEAs, having impacts on both OpEx and CapEx, respectively.

Task 6 - Program management and scale-up consulting

The technical work was completed over the project and all milestones have been achieved.

Plans for the next reporting period:

- N/A this is the final project report for the project.

Products: Nothing to report.

Participants & Other Collaborating Organizations:

Name	Organization	Project Role	Contribution	State, Country
Christopher Capuano	Nel	PI	Technical guidance, reporting, and project management	CT, USA
Andrew Motz	Nel	Co-PI	Technical Lead, research, data analysis, reporting, and project management	CT, USA
Kathy Ayers	Nel	Co-PI	Strategic advice & internal sponsor	CT, USA
Shaina Errico	Nel	Chemical Technician	Catalyst coating, cell assembly, electrochemical testing, and data crunching	CT, USA
Stephen Ekatan	Nel	Chemical Engineer	Process optimization, quality control, and scale-up	CT, USA
Allison Niedzwiecki	Nel	Chemical Engineer	Building cell, testing, evaluation, and data analysis	CT, USA
Judith Manco	Nel	Chemical Engineer	Ink preparation, catalyst coating, and electrochemical testing	CT, USA

Name	Organization	Project Role	Contribution	State, Country
Jennifer Glenn	Nel	Material Scientist	MEA material characterization	CT, USA
Michael Ulsh	NREL	Project Management	Lead, overall planning, executing, monitoring, and controlling	CO, USA
Scott Mauger	NREL	Technical Lead	Develop a project plan, determine the methodology used on the project	CO, USA
Jang-hoon Park	NREL	Research	Perform research tasks contributing to the overall project objectives	CO, USA
David Wood	ORNL	Project Management	Project oversight and management	TN, USA
Erin Creel	ORNL	Research	Ink and coating optimization	TN, USA
Craig Gittleman	GM	Lead	Project oversight and management	MI, USA
Matthew Fay	GM	Consultant	Consultation on ink composition and cost analysis	MI, USA
Swami Kumaraguru	GM	Consultant	Consultation on ink composition and cost analysis	MI, USA
Dan O'corr	Kodak	Lead	Pilot-scale coating	NY, USA
Brad Houghtaling	Independent	Consultant	Consultation on Ink formulation, coating, equipment, and accessories	NY, USA

Impact: Nothing to report

Changes/Problems: Nothing to report

Special Reporting Requirements: Nothing to report

References: N/A

Milestone Schedule

Milestone #	**Project Milestones**	**Type**	**Milestone Completion Date (Project Quarter)**				**Progress Notes**
			Planned	**Revised**	**Actual**	**Percent Complete**	
M1.1.1	Develop ink formulation for slot die and gravure printing with no visible voids and good adhesion.	Milestone	9/30/19	3/31/20	3/31/20	100%	NREL has established a regression model to tune the viscosity of the catalyst inks for direct coating on the membrane and ORNL has selected a high shear mixer for processing inks. The completion date is revised due to issues in procuring a membrane for the coating trials.
M2.2.1	Identify key coating parameters and optimize small-scale trial runs	Milestone	12/31/19	2/28/20	2/28/20	100%	Good uniformity of catalyst coating was demonstrated at NREL and Nel based on XRF and SEM, respectively. Nel also developed R2R compatible concept to mitigate hydrogen crossover through the membrane.
M1.2.1	Determine feasibility of reaching catalyst loading targets through demonstration of +/- 10%	Milestone	3/31/20		3/31/20	100%	Half-cell HER and OER samples were tested at 0.1 mg/cm^2 and 0.6 mg/cm^2 for a total of 0.7 mg/cm^2. Samples showed stable performance and comparable performance as compared to the baseline.
G/NG 3.1	**Validate target total PGM loadings of <0.7 mg/cm^2 & benchmark performance**	**Go/No-Go**	**6/30/20**		**6/30/20**	**100%**	Half-cell HER and OER samples were tested at 0.1 mg/cm^2 and 0.6 mg/cm^2 for a total of 0.7 mg/cm^2. Samples showed stable performance and comparable performance as compared to the baseline.
M1.3.1	Lock-in coating technique (slot-die/ gravure) & electrode concept (CCLs/ graded/direct coating on membrane)	Milestone	9/30/20		9/30/20	100%	Technique for initial trial will be gravure coating based on successful lab trials at NREL

Milestone #	**Project Milestones**	**Type**	**Milestone Completion Date (Project Quarter)**				**Progress Notes**
			Planned	**Revised**	**Actual**	**Percent Complete**	
M4.1.1	Transfer technology to R2R facility and demonstrate initial performance metrics for single electrode	Milestone	12/31/20		12/31/20	100%	Nel succesfully completed the first coating run at Kodak
M4.2.1	Demonstrate initial performance metrics of pilot scale R2R process for both electrodes	Milestone	3/31/21	7/30/21	8/4/2021	100%	Complete, both anode and cathode have been translated to the Kodak facility.
M4.3.1	**Establish acceptance criteria, defect identification & reduction, and demonstrate stable MEA performance**	**End of the project**	**6/30/21**	**12/31/21**	**6/30/2022**	**100%**	Complete, high quality MEAs that meet our commercial standards have been produced.

Budgetary Information:

Quarter	From	To	Estimated Federal Share of Outlays*	Actual Federal Share of Outlays	Estimated Recipient Share of Outlays*	Actual Recipient Share of Outlays	Total
1Q19	Start	9/30/2019		$93,293		$36,892	$ 130,186
2Q19	10/1/2019	12/31/2019		$126,839		$51,087	$177,926
3Q20	1/1/2019	3/31/2020		$142,733		$56,869	$199,602
4Q20	4/1/2020	6/30/2020		$110,649		$42,380	$153,029
5Q20	7/1/2020	9/30/2020		$76,489		$28,041	$104,530
6Q20	10/1/2020	12/31/2020		$76,253		$25,154	$101,407
7Q21	1/1/2021	3/31/2021		$172,165		$63,886	$236,051
8Q21	4/1/2021	6/30/2021		$200,726		$73,648	$274,375
9Q21	7/1/2021	9/30/2021		$202,065		$88,831	$290,896
10Q21	10/1/2021	12/31/2021		$180,039		$33,213	$213,252
11Q22	1/1/2022	3/31/2022		$1,239		$0	$1,239
11Q22	4/1/2022	6/30/2022		$17,507		$0	$0
Totals			$0	$1,399,999	$0	$500,000	$1,899,999

Project spending and estimate of future spending

*Updated quarterly – previous quarters should show actual expenditures; future quarters should show estimates. If spending for a given quarter is different than estimated, then the remaining quarter's estimates should be updated in a following table (Table 8, not included) to account for the difference. Total DOE and Cost Share amounts should be the same as the Award amount.