Membrane Electrode Assembly Manufacturing Automation Technology for the Electrochemical Compression of Hydrogen

Cooperative Research and Development Final Report

CRADA Number: CRD-18-00744

NREL Technical Contact: Scott Mauger

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Technical Report
NREL/TP-5900-95403
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Cooperative Research and Development Final Report

Report Date: November 5, 2024

In accordance with requirements set forth in the terms of the CRADA agreement, this document is the CRADA final report, including a list of subject inventions, to be forwarded to the DOE Office of Scientific and Technical Information as part of the commitment to the public to demonstrate results of federally funded research.

Parties to the Agreement: HyET Hydrogen USA LLC

CRADA Number: CRD-18-00744

CRADA Title: Membrane Electrode Assembly Manufacturing Automation Technology for the Electrochemical Compression of Hydrogen

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Sponsoring DOE Program Office(s):

Office of Energy Efficiency and Renewable Energy (EERE), Hydrogen Fuel Cell Technologies Office

Joint Work Statement Funding Table showing DOE commitment:

Estimated Costs	NREL Shared Resources a/k/a Government In-Kind
Year 1 Year 2 Year 2, Modification #1 Year 3a, Modification #2	$100,000.00 $50.000.00 $0.00 $0.00
Year 3b, Modification #3	$0.00
TOTALS	$150,000.00

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Executive Summary of CRADA Work:

Electrochemical compression has the possibility to outcompete mechanical compression for hydrogen end- use applications. While HyET has a compressor that can output JO kilograms (kg)/day (fully scalable from home-to-industrial application) at up to 700 bar, the energy demand and reliability require top-quality electrochemical hydrogen compression (EHC) membrane electrode assemblies (MEAs), preferably prepared by cost-effective high-capacity manufacturing. High pressure requires a special MEA design, deviating from typical proton exchange membrane fuel cell (PEMFC) MEAs with adapted catalyst layer substrates, asking for a modified coating process.

The National Renewable Energy Laboratory (NREL) will help HyET by developing an automated catalyst coating process fit for EHC MEA manufacturing. In addition, inline quality inspection methods will be developed/selected to improve the MBA quality as it is used for EHC stack assembly. In a joint effort, NREL and HyET will even design an automated manufacturing process for the EHC MEA and approach potential United States (US) suppliers of manufacturing equipment.

CRADA benefit to DOE, Participant, and US Taxpayer:

- Assists laboratory in achieving programmatic scope,
- Enhances the laboratory's core competencies, and/or
- Uses the laboratory's core competencies.

Summary of Research Results:

Purpose:

For the past 10 years, NREL has developed competencies and testing capabilities related to supporting the scale-up to high volume production of membranes, electrodes, and MEAs for fuel cell, electrolysis, and other electrochemical generation and storage technologies. In this project, NREL will bring to bear extensive testbeds, fabrication equipment, and expertise to assist Hy ET in the development of (a) inks and coatings relevant to high volume electrode production and (b) real-time inspection techniques capable of in-line quality control for HyET's electrochemical hydrogen compression (EHC) MEAs. In addition, NREL will assist HyET in the layout and specification of manufacturing equipment.

Task 1: Inks and Coating Process Development

NREL will bring to bear ink characterization tools, including rheology, dynamic light scattering, and zeta potential, and electrode fabrication equipment across several scales including a roll-to-roll coating line to evaluate and perform initial optimization of inks and coatings for EHC electrodes. NREL will perform parametric studies to understand the impact of formulation and process variables on the thickness and unifom1ity of electrode layers. NREL will provide electrode sheet materials to HyET for assembly into cells and in situ testing at HyET's facility.

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Throughout the project, wire wound (Mayer) rod, ultrasonic spray, and roll-to-roll slot die coating techniques were used. Mayer rod coating was used to generate gas diffusion electrodes (GDEs), catalyst coated membranes (CCMs), and electrode decals for membrane transfer.

Ultrasonic spray coating was used to generate GDEs and CCMs. Roll-to-roll slot die coating was used to generate GDE and decal samples.

Rod coating (shown in Figure 1) was the simplest of all the techniques used during the project. It can be reasonably scaled in a roll-to-roll environment but is not very common and provides far less control than slot die coating.

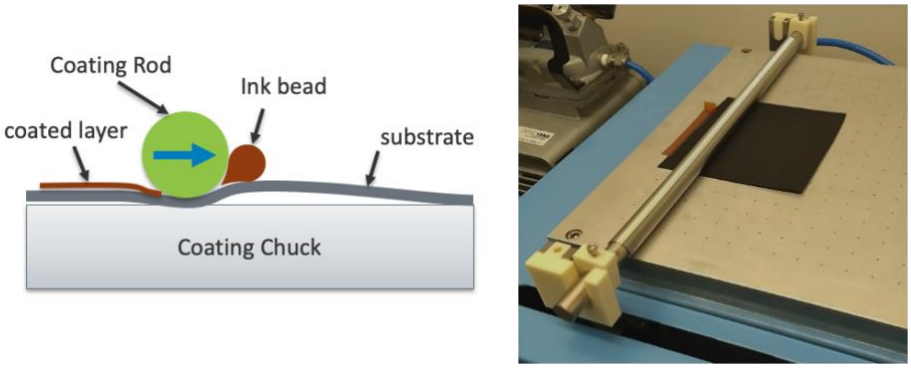

Figure 1 – rod coating cartoon (left) and rod coating tool (right)

Rod coating was used as a small-scale and rapid turnaround alternative to slot die coating as the ink experiences a similar shear environment immediately prior to drying. The inks used in rod coating are like those used with slot die coating, allowing for similar particle-polymer interactions in the liquid phase, whereas spray-coated inks usually must be lower in viscosity and thus concentration, which can change these interactions. Rod coating also mitigates the substrate flatness and fixturing requirements associated with the similar doctor-blade coating technique because the substrate is held down by the rod itself during film application. The loadings acquired by rod coating can be estimated by a simple geometric mass balance as shown in Figure. 2 where R_{wire} is the radius of the wound wire, Φ_{cat} is the platinum mass fraction in the ink, and ρ_{cat} is the density of the whole catalyst ink.

$$mg_{cat}cm^{-2} = R_{wire} \cdot \Phi_{cat} \cdot \rho_{cat} \cdot \left(1 - \frac{\pi}{4}\right)$$

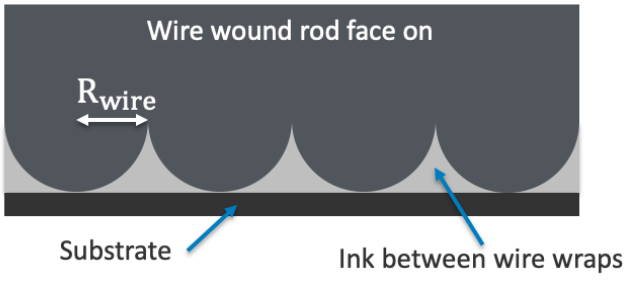

Figure 2 – Mass balance equation and associated diagram. R_{wire} is the radius of the wound wire, Φ_{cat} is the platinum mass fraction in the ink, and ρ_{cat} being the density of the whole catalyst ink.

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Masked rod coated CCMs were made by using wide rolls of 0.002 inch thick Kapton tape as a mask. Holes were punched in the tape of the desired coated area diameter and then the tape was placed onto the membrane and any bubbles were squeegeed out. The sample was then coated and dried as normal. The tape was then removed leaving a circularly coated CCM with sharp edges.

Ultrasonic spray coating, while not being a very scalable technique due to capital cost and production rates, is a very flexible technique. The coated area's geometry can be easily selected by using a mask to partially cover the substrate while leaving the area desired to be coated uncovered. A 48 KHz impact-type spray head (Figure 3) was used throughout the project to produce square and circular GDEs and CCMs. The use of an impact-style spray head allows us to spray coat the same ink used in rod and slot die coating and at much higher rates than standard ultrasonic spray heads. In the coating system, the spray head is held by a two-and-a-half axis CNC gantry, providing full X and Y movement and a small adjustment for spray head standoff distance. The substrate is held below the spray head gantry by a heated vacuum chuck and thin silicone gasket, which holds the mask in place and creates the seal for the vacuum chuck. This type of spray head utilizes a wide fanned-out stream of air to direct atomized ink down towards the substrate in a linear deposition pattern. This spray pattern is then repeatedly rastered over the substrate to produce an even coating. During spraying, the ink is effectively re-homogenized within the spray head due to the ultrasonic atomization and is deposited in relatively thin layers, which tends to result in a slightly different morphology than the bulk slot dies or rod coating techniques.

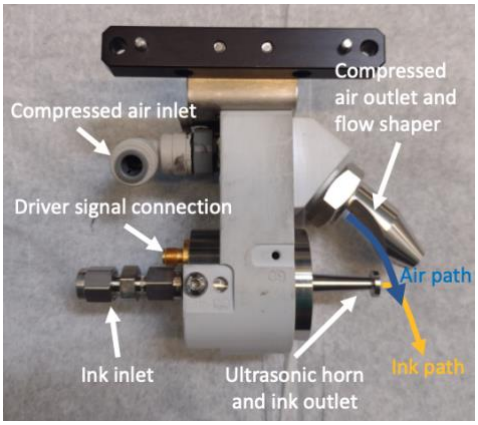

Figure 3 –Ultrasonic nozzle used at NREL with labeled parts and mass flow paths.

Predicting loadings for ultrasonically spray-coated samples can be difficult given that the spray pattern itself is not uniform but rather a gaussian distribution with more ink getting deposited near the center of the pattern. NREL used past spraying experience as a starting point and then adjusted until settings were found for our system that gave us the target loadings. To do this precisely, x-ray fluorescence (XRF) spectroscopy measurements were used to verify loading uniformity and indicate any needed modifications to the spray coating parameters. We ran our impact head at 200 mm/second linear travel speed and used an overlapping line raster. Each spraying line, shown in Figure 4 as green arrows, was spaced 3 mm apart. When the spray head starts and stops spraying, large irregular droplets can form and cause splattering. To prevent these droplets from getting on the substrate, spraying begins and ends roughly 30 mm from the edges of the substrate, shown as orange ovals in Figure 4. The head sprays for 0.5 seconds prior

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to starting to move to reach a steady-state spray pattern with uniform droplet formation. After spraying the line across and past the substrate, the head stops spraying and pauses to allow the sprayed area to mostly or completely dry. This drying pause prevents the air stream from the spray head from deforming the wet film from the previously sprayed line. The spray pattern the impact spray head produced was roughly an oval with a 35 mm major axis and a 10 mm minor axis. The major axis was perpendicular to the head's movement. This orientation and the 3 mm spray-line pattern spacing allowed the gaussian spray pattern to overlap and even out with each successive line. The orange ovals in Figure 4 are mostly overlapped.

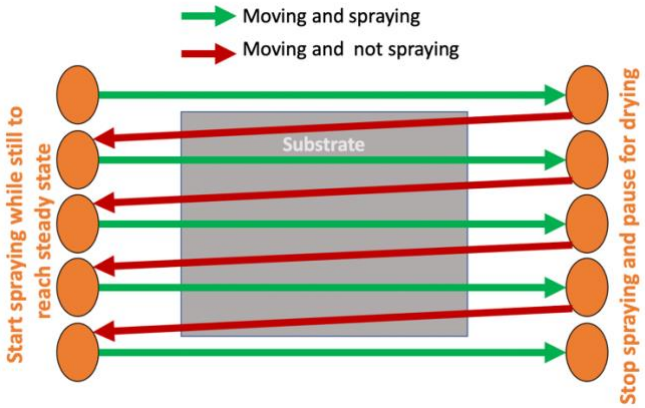

Figure 4 – Linear raster pattern for spray coating. Green arrows show movement paths while spraying. Red arrows show non-spraying movement. Orange ovals show stopping positions.

Slot die coating is a roll-to-roll technique that is highly scalable with good control and is widely used for continuous homogeneous coatings. Two precisely machined and mirror-finish ground plates of stainless-steel form the main body of the die. A flow path is determined by a shim with a flow path punched out of it, which is placed in-between the two plates as shown in Figure 5.

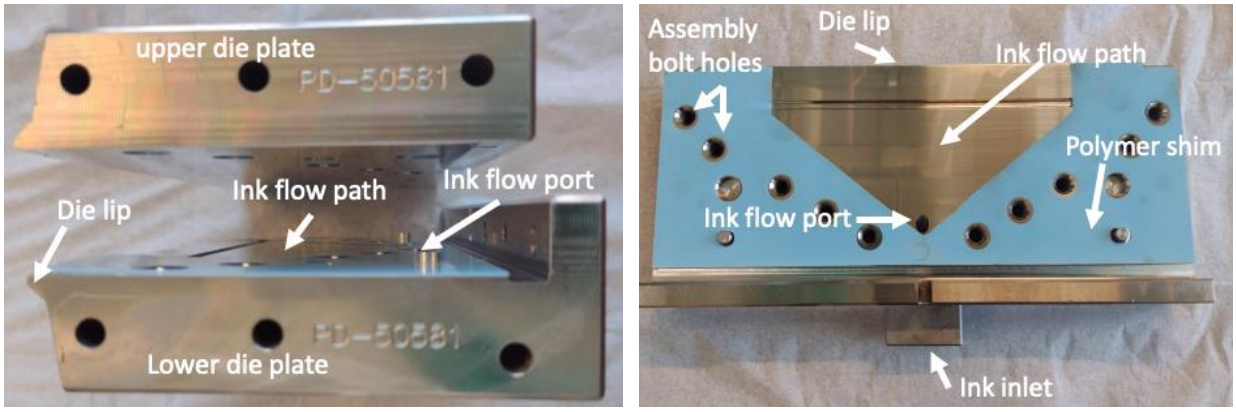

Figure 5 – Disassembled fixed lip slot die used at NREL with flow path shim in place.

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Ink Preparation

Two ink mixing techniques were used in this project: ultrasonic probe/bath and rotary high shear mixing. Only the early 2018 samples were prepared with ultrasonically mixed inks. All inks were made by weighing the components in their addition order within the final mixing container. Additions started with the catalyst material, followed by the deionized water, n-propanol, and then the D2020 nafion ionomer dispersion. Water was added immediately after the catalyst to avoid chemical reactions with the n-propanol or the solvents in ionomer dispersion. For ultrasonically mixed inks, direct probe sonication (Misonix Sonicators, model: s-4000 at 5 watts output) was first applied for 2 minutes with the main ink container held in an ice bath to prevent the platinum nanoparticles from sintering and reducing their surface area. The probe sonication was then followed by 40 minutes of bath sonication (Emerson, Bransonic M3800) in an ice water bath to again prevent platinum nanoparticle sintering. For rotary high shear mixed samples, the ink components were assembled in the same order as stated above but in a jar with ample head space to allow the vortex action and foaming to remain contained. High shear mixing was then applied within the jar using an Ultra Turrax T25 drive unit equipped with an 'S 25 N–18 G' mixing head from IKA for 20 minutes at 10,000 rpm. This process results in some foaming of the ink. To remedy this foaming, the inks were left covered and stirred at room temperature with a magnetic stir bar at ~300 rpm overnight for use the next day. The ink recipe was kept constant throughout the project except for the catalyst material itself and in special instances.

Decal Printing

Decal printing and transferring were first tested and tuned using rod coated decals. decals were coated on 0.010-inch-thick PTFE sheets. To perform the double-sided decal transfer to the HyET-provided membrane, a stack of several materials was used which follows the same general construction as those used, in fuel cell decal transfer processes at NREL as seen in Figure 6.

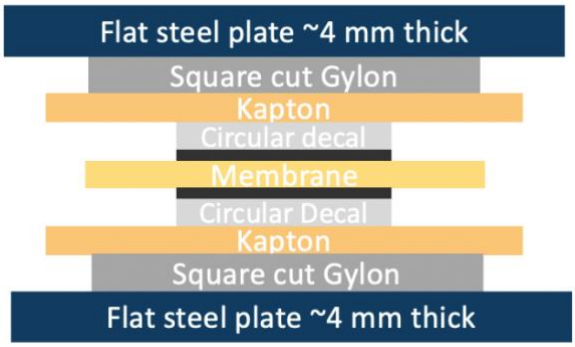

Figure 6 - Decal transfer stack for double sided CCM generation

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A flat steel plate was used as a base for the stack for handling and flatness requirements. Gylon gasket material (style 3540, 1.6 mm thick) is used to even out the load force like a surface area spring, and Kapton is used as a release layer between the Gylon and the decal. The stack is carefully aligned and is then put into a heated hydraulic press at 140 °C (near or roughly at the glass transition temperature of the nafion in the catalyst layer) with 1 kg of force applied per cm^2 of Gylon for 5 minutes to gently heat the stack to uniform temperature. After warming, 55 kg/cm^2 of force is immediately applied for 15 minutes. The pressure is then slowly released over ~2 minutes and the whole stack assembly is removed and left to cool on a rack (to minimize rapid conduction of heat) for 4 minutes. The stack is then carefully peeled apart to remove the CCM. While rod coating was used to tune and test this process with the HyET-provided membrane, the end goal was roll-to-roll slot die production testing of the technique for the larger 118 mm diameter electrode platform. For this, the standard turrax mixed ink recipe with a higher concentration of 7.5 wt% (with the same I:C and solvent ratio) was slot die coated onto the 0.010-inch-thick PTFE substrate. The higher solids loading ink was used to increase the ink viscosity and avoid the problems with the prior recipe.

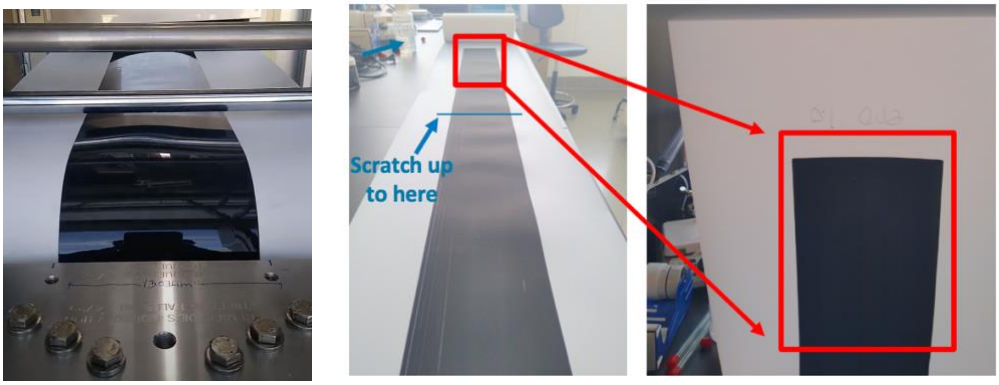

Figure 7 – (left) wet film of 0.1 mg_{Pt}/cm^2 decal immediately after being coated. (Center) 0.1 mg_{Pt}/cm^2 decal unrolled after coating and drying showing scratch from baffle and where/when the baffle was removed. (Right) unscratched section of the coating back lit to show uniformity.

With the higher concentration and thus viscosity, the catalyst ink dried, leaving a uniform coating as seen in Figure 7. An attempt was made to coat a 0.3 mg_{Pt}/cm^2 section as well. However, the flow rate required to achieve the loading target caused the ink bead at the die lip to drip. This is the well-known "weeping" limit of the coating window. A technique we have used in the past to prevent weeping is coated with the dye in a vertical position to effectively remove gravity from the force balance of the coating bead, as seen in Figure 8. With this modification, a uniform wet film was established, but a non-uniform dry film was produced.

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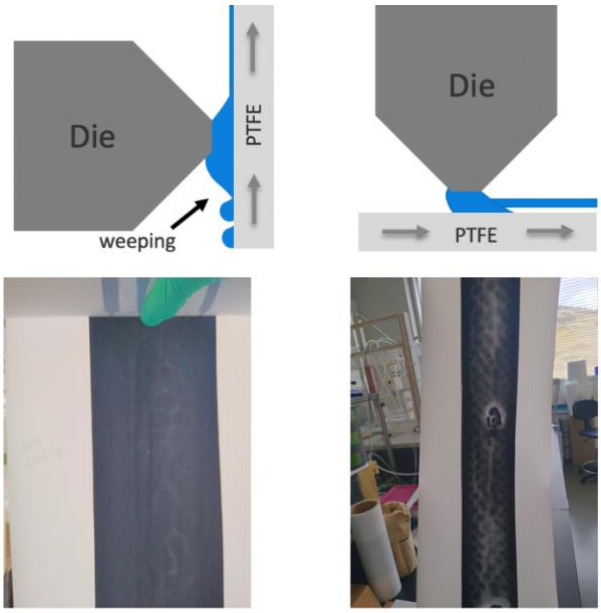

Figure 8 – (Top Left) slot die in standard horizontal position showing weeping. (Top right) die in vertical position without weeping. (Bottom both) 0.3 mg_{Pt}/cm^2 coated and dried section with back lighting showing non-uniformity.

The non-uniformity in the 0.3 mg_{Pt}/cm^2 section was likely due to the wet film thickness and the substrate curvature. The PTFE had a tendency when heated and under tension to curl toward the wet coating. This effectively created a trough for the wet film to settle into if it were thick (tall) enough to not have viscous forces hold it in place like it did in the 0.1 mg_{Pt}/cm^2 section. We also suspect that the 0.1 mg_{Pt}/cm^2 section was able to dry much more rapidly, which would help pin the wet film even while sitting on the curved substrate. This problem might be remedied by further increasing the concentration of the ink to increase the viscosity, though ultimately this mechanism will be limited by the ink pumping capabilities.

Task 2: Quality Inspection Development

NREL will bring to bear multiple testbeds for the development of real-time quality inspection techniques for electrodes, membranes, and MEA subassemblies for HyET's EHC MBA materials. NREL's activity focuses on areal inspection techniques, i.e., techniques that utilize optical or infrared imaging to provide the potential for 100% inspection of MEA material webs (rather than point measurements). These techniques enable detection of small discrete defects in MEA materials as well as determination of overall film or layer uniformity. If appropriate, NREL can utilize its industrial-style web-line to validate techniques with sheet or roll materials.

NREL measured membrane samples provided by HyET using linear camera scanning via an Epson Perfection V600 scanner with diffuse white light at a pixel density of 2400 pixels/inch (11 micron/pixel) in reflection and transmission modes (see Figure 9). Defect and foreign object detection were possible, though height location within the membrane and backer stack was difficult to consistently detect with currently viable in-line techniques that cover large areas (Figure 10). The most significant issue with the quality control effort for optical techniques was dust control. A glass encapsulation technique was developed to enable the quality control method

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validation, but this methodology is clearly not viable in a roll-to-roll setting. Nonetheless, the glass encapsulation was able to greatly reduce the amount of dust on the membrane. Fast optical imaging was repeated, and automated object detection algorithms were demonstrated on the scanned samples (Figure 11). As shown in Figure 12, defect density statistics were generated for each sample. Standard optical microscopy was used to image specific defects identified during the fast scanning in greater detail. Off-line laser confocal microscopy was also used to verify the linear-camera scanning results and was able to more accurately identify through-the-thickness location of suspected foreign objects (see Figure 13). It is believed that with proper in-line dust control and cleaning, optical techniques could be used to identify defects and dust particles on the order of 10 microns in size. The quality control team also produced membrane samples with pinholes of 25-, 100-, and 250-micron diameter via a custom NREL tool and provided the samples to HyET for MEA fabrication and testing of the impact of the pinholes.

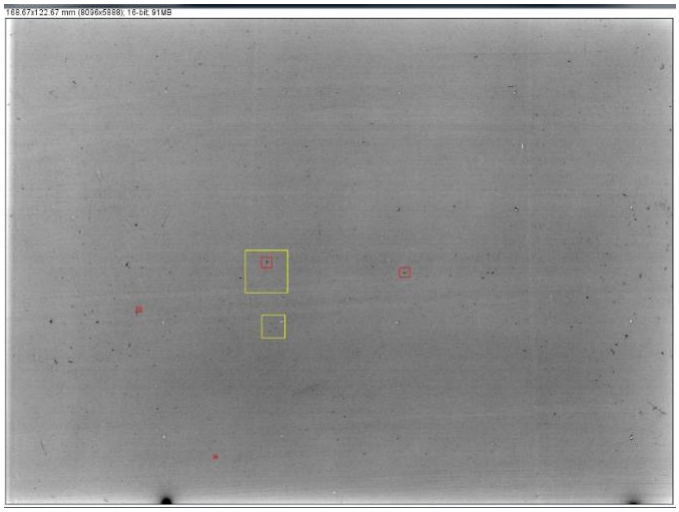

Figure 9: Large-area (169x123 mm) optical scanning image of a membrane sample, with the four largest identified objects marked in the small red boxes.

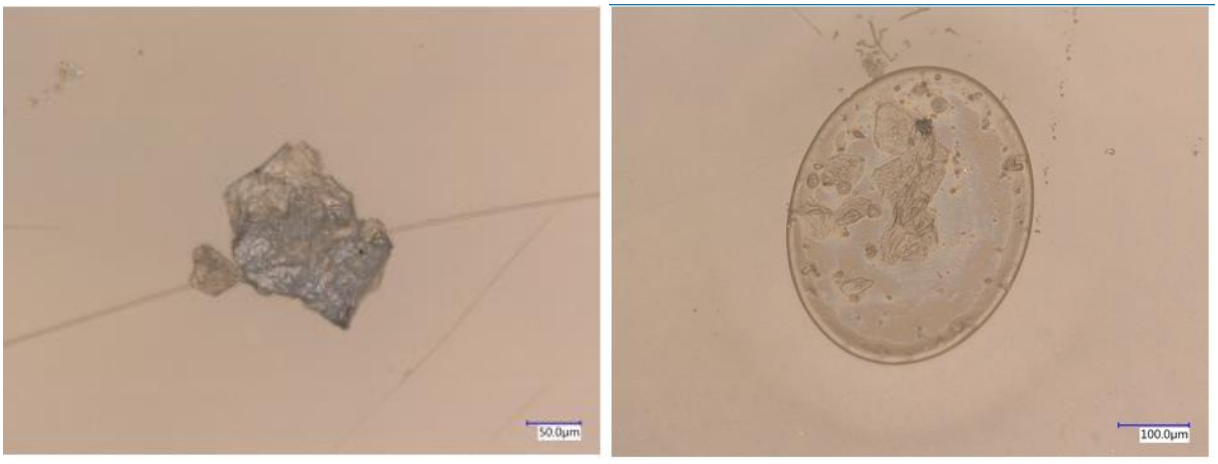

Figure 10: Representative objects seen during microscopy of membrane materials

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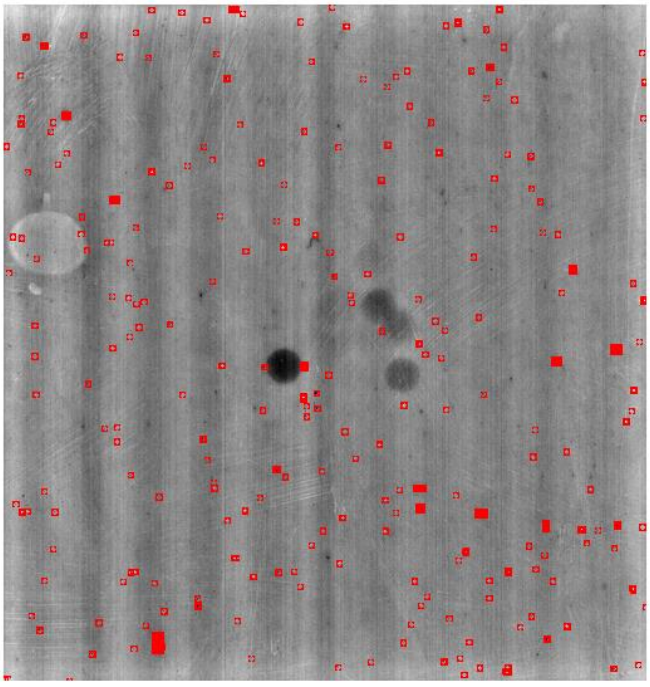

Figure 11. Distribution of optically detected objects for a glass-encapsulated membrane sample. A single rotation angle is shown.

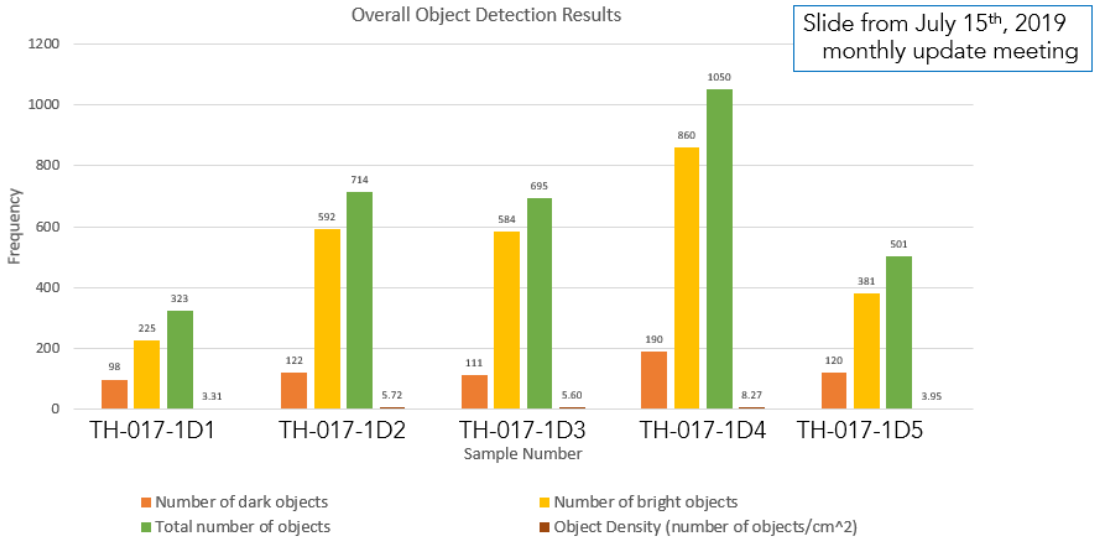

Figure 12. Summary of objects detected in five glass-encapsulated membrane samples.

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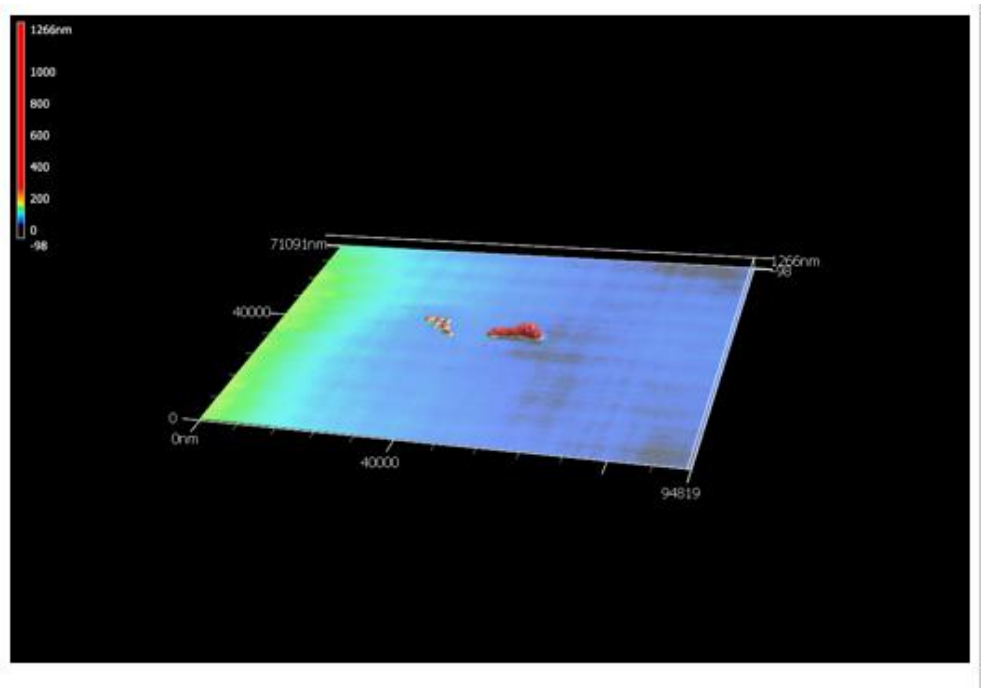

Figure 13. Objects observed during confocal microscopy of sample TH-017-1D4.

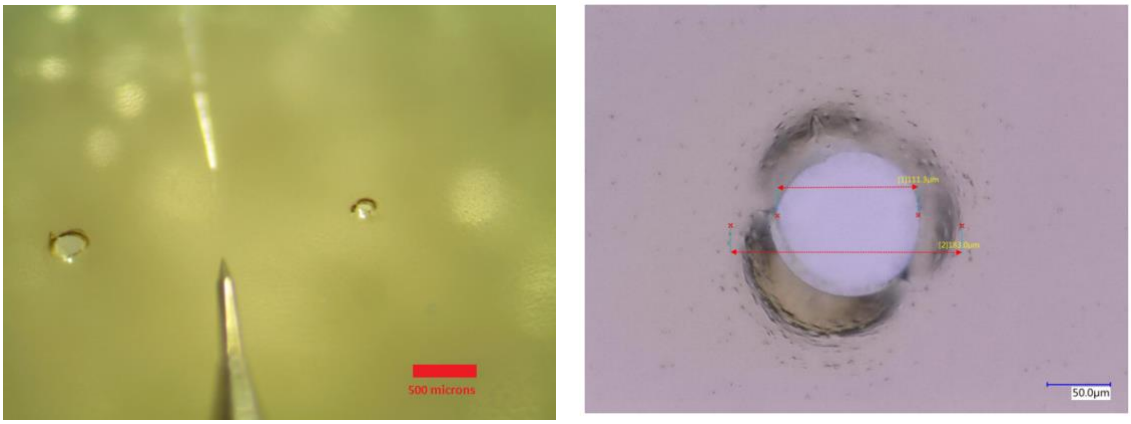

Figure 14. Left: Pinholing of membrane material. Final samples will have one pinhole per sample. Right: Digital microscope image of a pinhole made using a 120 µm tool, with an actual inner diameter of 111 µm.

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Task 3: EHC testing of R2R vs manual MEA based EHC single cell and stack (HyET)

HyET will manufacture the MEAs and perform the EHC cell testing, which will include standard diagnostics (as used for PEMFC) like shorting tests, IV-curve recording, electrochemical impedance spectroscopy EIS and cyclic voltammetry (CV analysis), hydrogen (H2) cross-over testing, H2 pumping at ambient pressure, and finally compression testing to maximum pressure. Duration testing to evaluate the performance stability will conclude the membrane evaluation test program. Performance of the manual versus automated manufactured MEAs will be compared.

Cell materials fabricated at NREL were provided to HyET. HyET assembled these materials into full MEAs. These were tested using a variety of electrochemical diagnostic methods. These tests were used to evaluate differences in performance, electrochemical surface area, and resistance as a function of catalyst type and catalyst layer fabrication method. These measurements also compared HyET's baseline fabrication process to NREL's processing methods. Examples of MEA testing results are shown in Figure 15. Figure 15a shows polarization curves of MEAs tested at 400 bar. These results show that in this study, catalyst type did not significantly impact performance, whereas coating method and substrate were more impactful. Electrochemical surface area (ECSA) values measured by cyclic voltammetry showed some difference in ECSA due to catalyst type and coating method (Figure 15b). Electrochemical impedance spectroscopy (EIS) measurements, in Figure 15c show that the coating method only has a noticeable impact on the charge transfer resistance. The other resistances do not seem to be greatly impacted by the coating method.

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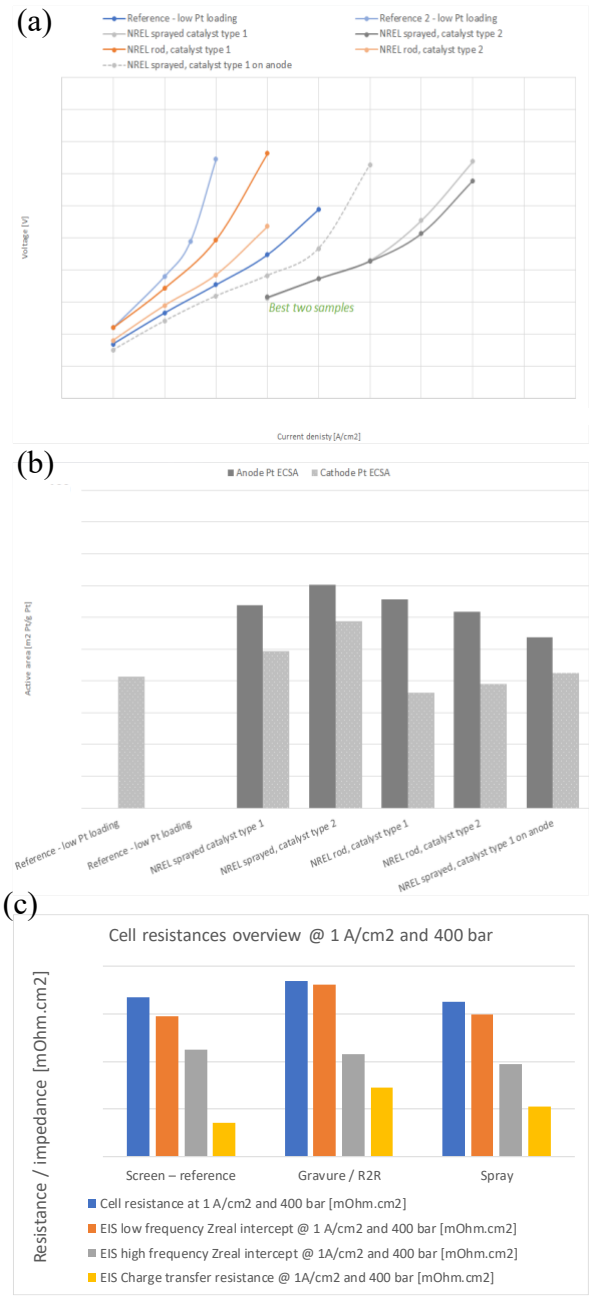

Figure 15. Electrochemical testing results for EHC MEAs prepared using different coating methods and catalyst: (a) polarization curves, (b) catalyst electrochemical surface area (ECSA), and (c) impedances.

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Task 4: Specification of a manufacturing Line (HyET + NREL)

Using its experience with R2R equipment and fabrication of MEA materials and based on its work and results in Tasks 1 and 2, NREL will assist HyET in the design and specification of a manufacturing line for the high-volume production of its MEAs. NREL will specifically provide inputs relative to electrode coating and quality inspection devices and integration. US suppliers of required production line equipment will be identified.

Based on the work conducted within this project, NREL provided information to HyET for different manufacturing processes and considerations based on project observations. The information provided reviewed the coating modalities explored within the project and provided information on relevant coating processes fundamentals, suppliers, and cell assembly processes. This information also provided notes on the pros and cons of the potential processes. NREL also provided detailed documentation of ink formulations and process conditions used in coating trials at NREL. For roll-to-roll coating NREL provided recommendations for equipment features such as web steering and roller diameters that are suitable to convey gas diffusion media, which is stiff and can break if bent around small-radius rollers. NREL also provided information on drying methods studied within the project and considerations for oven temperatures and oven residence times needed to obtain dried coatings. Information on equipment manufactures for coating machines, dies, and mixing/milling equipment was provided to HyET. For the processes studied, NREL provided details on materials used for coating and laminating processes. For the slot die coating process investigated within the project, NREL provided details on process limits for HyET to consider when developing a manufacturing process. For quality inspection, NREL provided details on the equipment used to inspect coated materials. The report also provided information on the best practices to ensure quality measurements.

Additional publications/peer review meeting reports:

https://www.hydrogen.energy.gov/docs/hydrogenprogramlibraries/pdfs/review18/h2006_ulsh_2018_p.pdf?sfvrsn=6f4ca19_1

https://www.hydrogen.energy.gov/docs/hydrogenprogramlibraries/pdfs/review19/h2006_ulsh_2019_p.pdf?sfvrsn=9d6bedda_1

https://www.hydrogen.energy.gov/docs/hydrogenprogramlibraries/pdfs/review20/h2006_ulsh_2020_p.pdf?sfvrsn=69b9d8ec_1

References:

None.

Subject Inventions Listing:

None.

ROI#:

None.

This report is available at no cost from NREL at www.nrel.gov/publications.