

Advanced Manufacturing Processes for Gigawatt-Scale Proton Exchange Membrane Water Electrolyzers

June 6th, 2023

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Project P197

DE-EE0009237

Project Goal

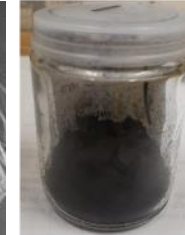
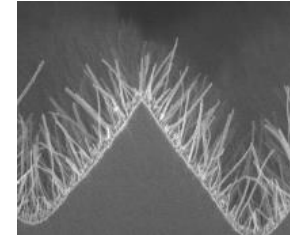
Develop Processes for Validated Gigawatt-Scale PEM Electrolysis Components

Project Goals

1. **Develop manufacturing processes** for PEM water electrolysis OER catalyst, electrode, and thrifted catalyst coated membranes suitable for GW/year scale.
2. **Scale up** developed processes to GW/year scale and **produce** components (2 MW equivalent).
3. **Assess and validate** resultant components against efficiency, durability, power density, and low Ir content metrics in megawatt-capable stacks relevant for GW/year deployment scale.

Project Components

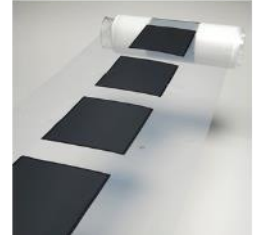
NSTF OER Powder Catalyst



NSTF OER Electrodes



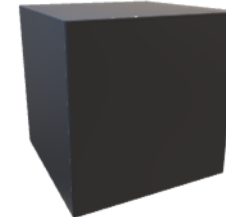
Thrifted WE CCMs



1 GW/yr Scale

2g/m², 35kW/m²

77kg Iridium



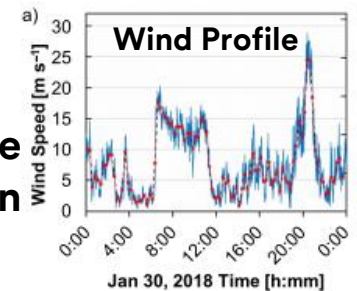
Cube, 13.6 cm per side

29k m² Electrode, CCM



~Surface Area of Mt. Fuji

Assess:
Variable Renewable Integration



Validate:
Plug Power MW-Capable Stack
(100kW test)



Project Addresses Key Cost and Manufacturing Barriers

Timeline

Project Start: 1/1/2021

Project End: 9/30/2024*

*Includes 6mo no-cost extension

Barriers

F. Capital Cost

K. Manufacturing

Budget

Total DOE Project Value: \$6.069MM*

Total Funding Spent: \$3.520MM*

Cost Share Percentage: 23.25%

*Includes DOE, contractor cost share and FFRDC funds as of 2/28/2023

Technical Targets

Catalyst/Electrode Fab. Rate*: 3.75GW/year

CCM Produced*: 2 MW

Cell Voltage at 2A/cm²: 1.725V

Electrode Loading: 0.2 mg/cm²

Decay Rate: 4 μ V/hr

Stack Power Density: 7.6 W/cm²

*Based on 3.45W/cm²

Partners

Giner (N. Macauley)

Plug Power (C. Mittelsteadt, Z. Green)

NREL (P. Rupnowski, E. Padgett, S. Mauger)

ORNL (D. Cullen)

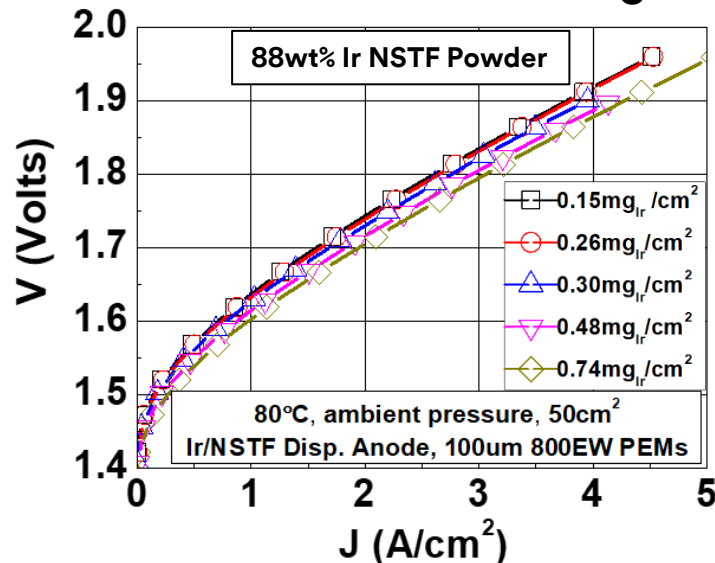
Stringent Material and Processes Requirements for GW/year Scale

• Industry Needs for PEM Electrolysis At Scale (*Gigawatts per year*)

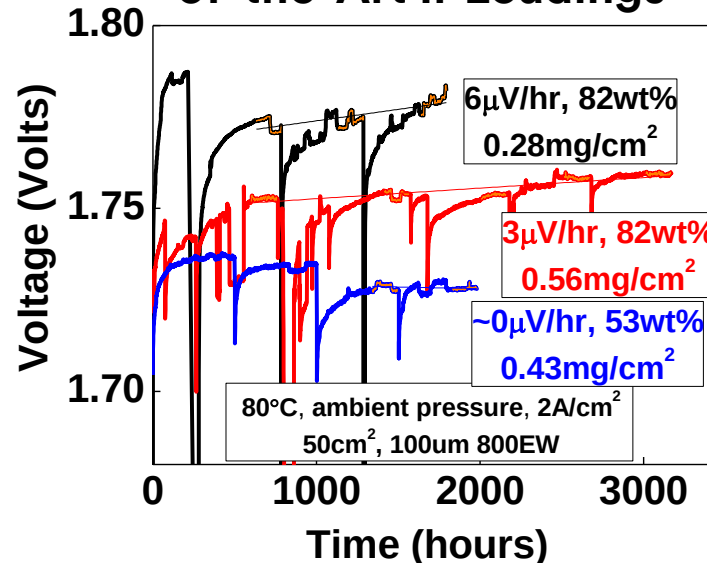
1. Advanced components which meet efficiency, durability **and** power requirements ***with low Ir content***.
2. High volume capacity and reduced manufacturing costs (lower CapEx).
3. High uniformity, reliability, reproducibility, and quality.

Project addresses industry needs by development of advanced fabrication processes capable of producing reproducible and uniform state-of-the-art PEMWE components at GW/year scale.

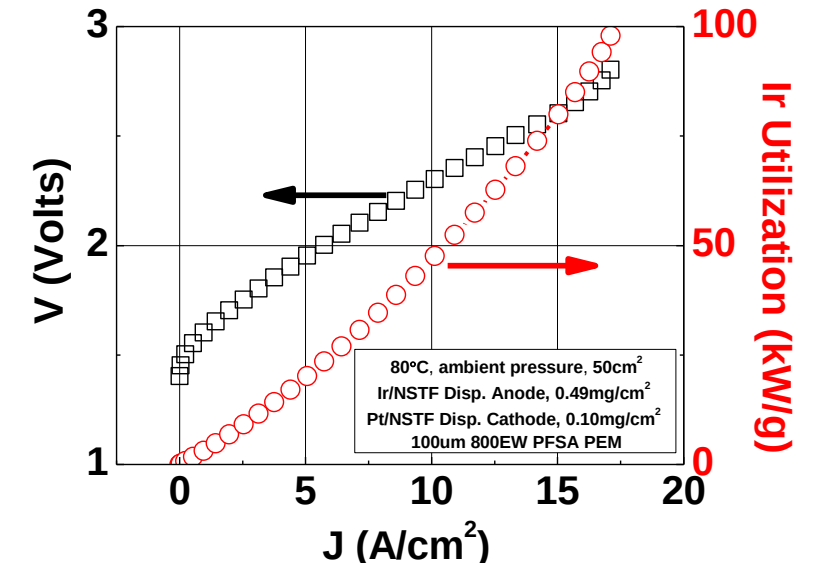
High Performance with Ultra-Low Ir Loadings



High Durability at State-of-the-Art Ir Loadings



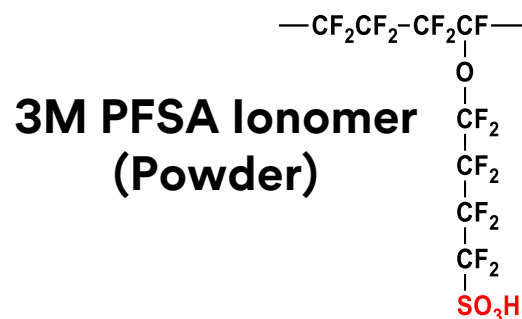
High Power Capability



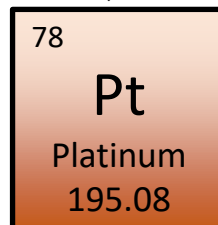
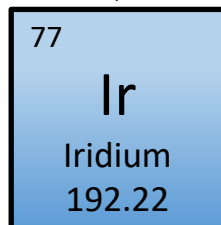
Project Approach

Develop High-Volume Roll to Roll Manufacturing for Scale

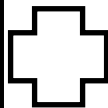
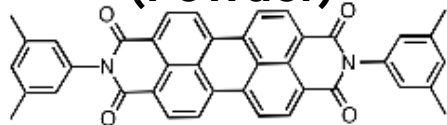
Input Raw Materials



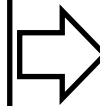
Catalyst Precursors (Metal Bars)



Catalyst Support Precursor (Powder)



High Volume Roll to Roll Manufacturing



Materials Process Focus

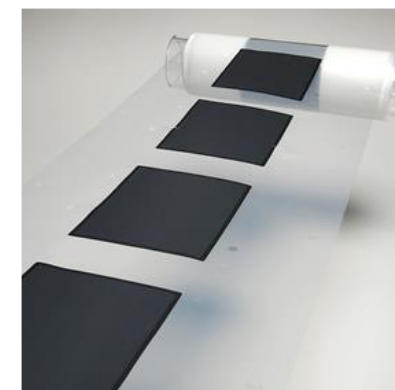
NSTF OER Powder Catalysts



NSTF OER Electrodes



Thrifty Catalyst Coated Membranes



- Identify feasible improvements to key process steps (BP1), implement in BP2, validate in BP3.

Project Performance and Durability Targets Achieved

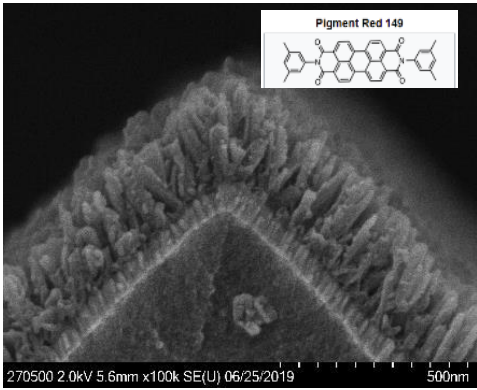
Project Target	Target Value	'23 Status	
Catalyst/Electrode Fabrication Rate (GW/year)	≥ 3.75	1.5 ¹	Mfg. Targets
CCM Produced (MW)	≥ 2	Not started (BP3)	
Cell Voltage @ 2A/cm ²	$\leq 1.725^3$	1.704 ²	Perf., Durability, Loading
Electrode Loading (mg/cm ²)	$\leq 0.20^3$	0.20 ²	
Decay Rate (μV/hr)	$\leq 4^3$	0 ²	At-Scale Validation
Stack Power Density (W/cm ²)	≥ 7.6	Not started (BP2)	
1. Fabrication rates based on demonstrated rates extrapolated to one machine/year per process step, and are based on process model incorporating capacity factors. 2. Laboratory CCM with 0.20mg/cm ² 78wt% Ir/NSTF powder OER catalyst/electrode, 0.08mg/cm ² Pt/NSTF dispersed catalyst/electrode, 3M 800EW 100 micron membrane. 50cm ² cell, 80°C, 2A/cm ² . Wind VRE Protocol. 3. Project target assessed with 50cm ² cell, 80°C, 2A/cm ² . 3M 800EW 100 micron membrane, project wind variable renewable energy (VRE) protocol. Performance and durability milestones in stack derated to 1.735V and 5μV/hr.			

- Catalyst/electrode manufacturing steps demonstrated at equivalent to $\geq 1.5\text{GW/year}$ – *on track*
 - Fabrication rate limited by two steps – upgrades occurring now.
- Performance and durability targets exceeded at target loading (0.20mg_{Ir}/cm²) using current processes
 - Ultimate project target requires demonstration with production-scale materials made at project target rate (BP3).
- Stack validation testing at Giner and Plug Power ramping now.

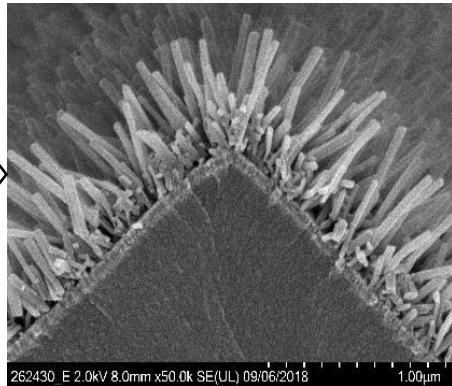
Accomplishments and Progress

3 of 5 Process Steps Demonstrated in Excess of Project Target Rate

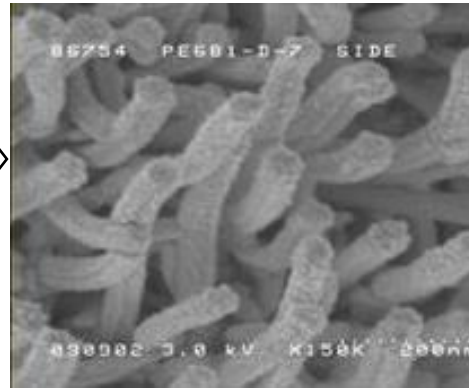
1. Precursor Deposition 1.5 GW/year



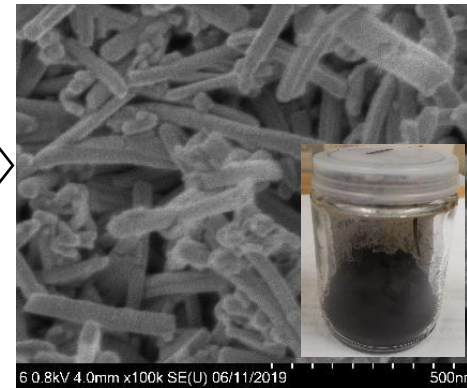
2. Support Growth > 3.75GW/year



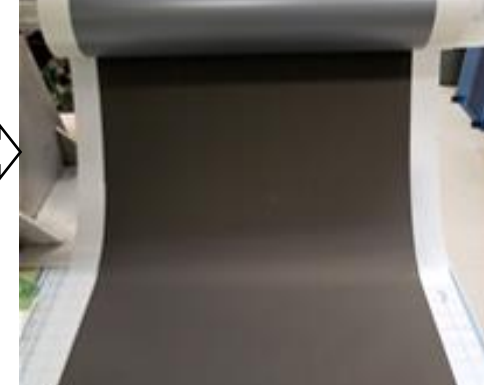
3. Catalyst Deposition 1.5GW/year



4. Catalyst Powder > 3.75GW/year



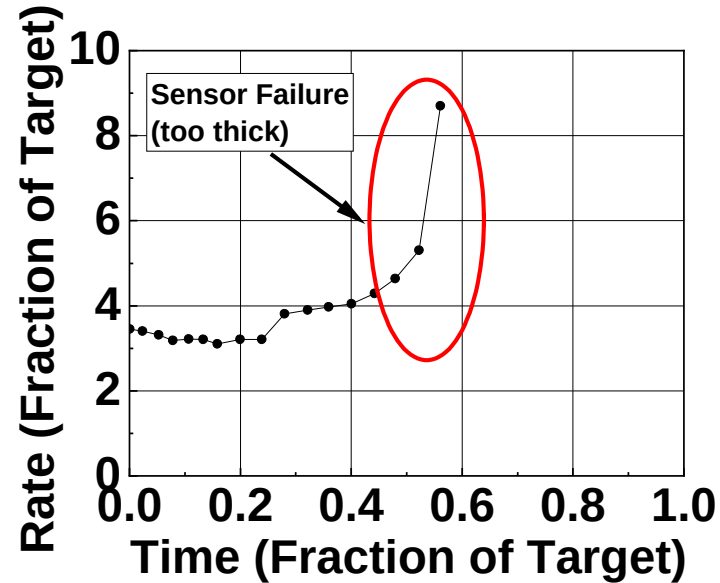
5. Electrode Coating > 3.75GW/year



- Overall catalyst/electrode process consists of 5 high level process steps
- 3 of 5 process steps demonstrated above project target rate.
- Key process focus of budget period 2 has been design and implementation of new precursor and catalyst deposition steps to enable achievement of project targets.

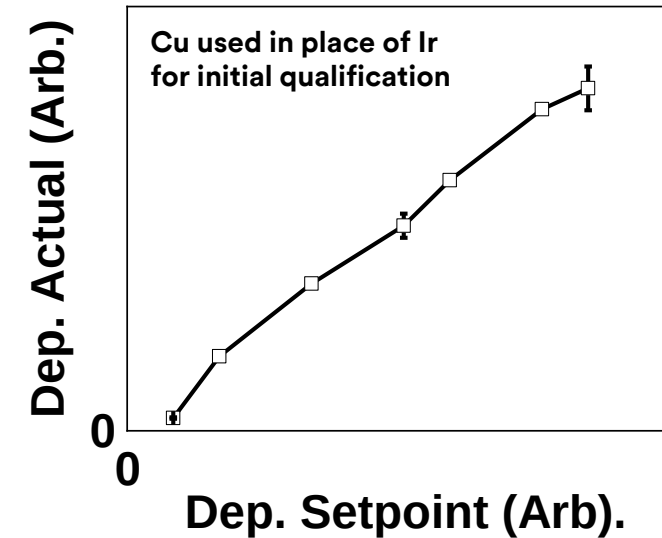
New Deposition Modules Commissioning Suggests Targets Feasible

Precursor Deposition Rate, Stability Test



- New module design has been completed.
- Module rate *estimated* to exceed target for rate and likely meets stability requirement.
- Installation complete; commissioning in progress

Catalyst Deposition Rate, Uniformity Test



- New module specs confirmed and unit installed.
- Cu test deposition (cheaper Ir analog) indicates good control and uniformity.
- Commissioning with Ir in progress

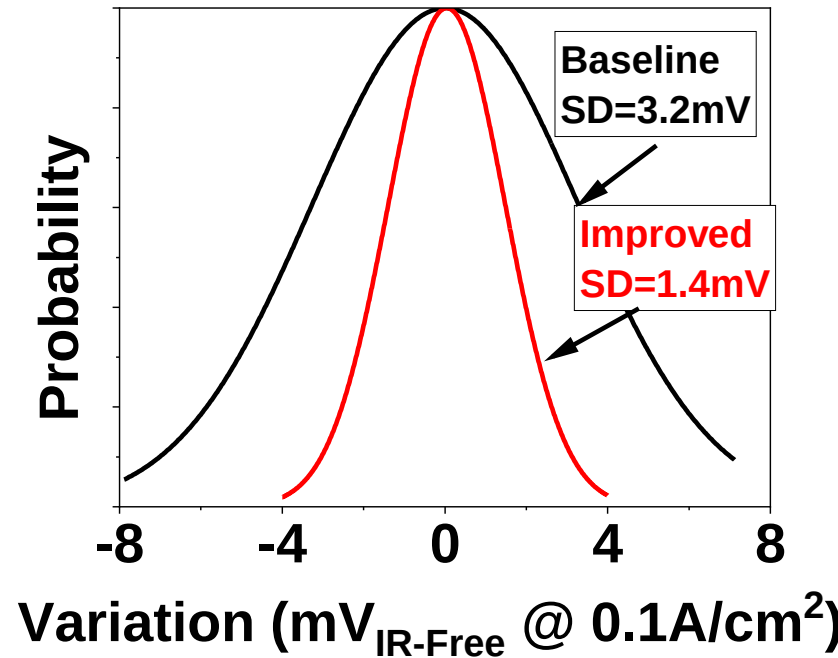
On track towards meeting requirements for BP2 GNG – 2GW/year equivalent

Improved Catalyst Process Yields Highly Reproducible Activity

Catalyst Powder



Total Uncontrolled Kinetic Variability Through All Process Steps



Uncontrolled Variability Sources

1. Catalyst production
2. Ink, electrode, CCM formation (lab).
3. PEMWE cell testing

- Catalyst process development has significantly improved catalyst consistency.
- Kinetic activity assessed from numerous lots produced with developmental processes.
- Overall variability (all sources) in kinetic cell voltage reduced from 3.2 to 1.4mV (-56%).

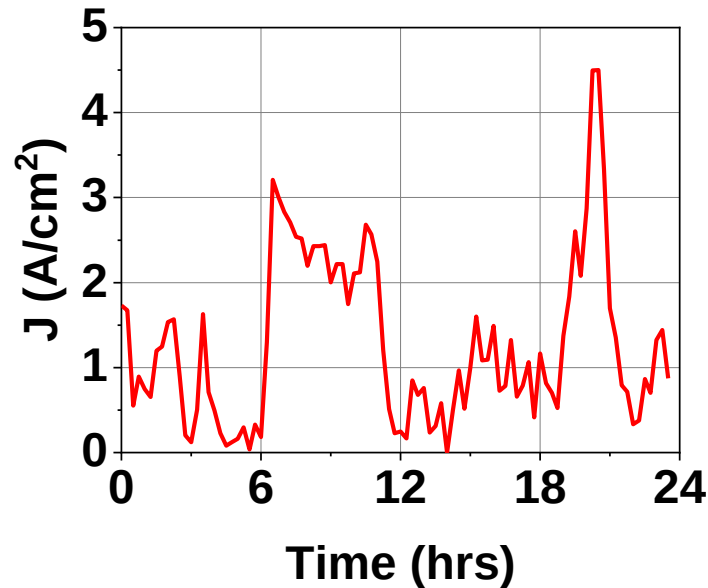
Project Materials Assessed Against Three Accelerated Stress Tests

1. Project Durability Target Protocol

Variable Renewable Energy (“VRE”)

500hrs x (24hr wind simulation, 0-4.5A/cm²)

Diag. pol. curves every 100 hours



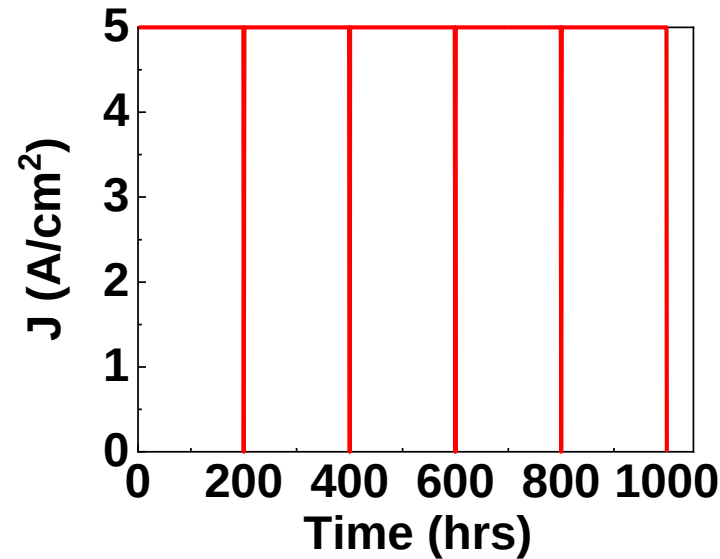
- Simulates direct integration to wind turbine; from Alia et al. (2019), scaled to 4.5A/cm².

2. High Power Steady State

Diagnostic/Comparative

1000+ hours at 5A/cm²

Diag. pol. curves every 200 hours



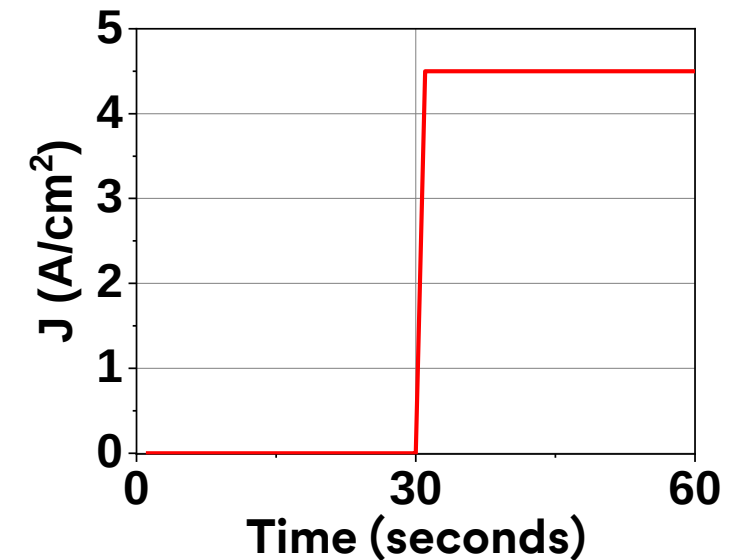
- Very limited cycling.
- >2x higher average power.

3. Highly Accelerated Stress Test

Rapid On/Off Cycling (“HAST”)

30k cycles (30s 4.5A/cm², 30s off); 500hrs

Diag. pol. curves every 100 hours



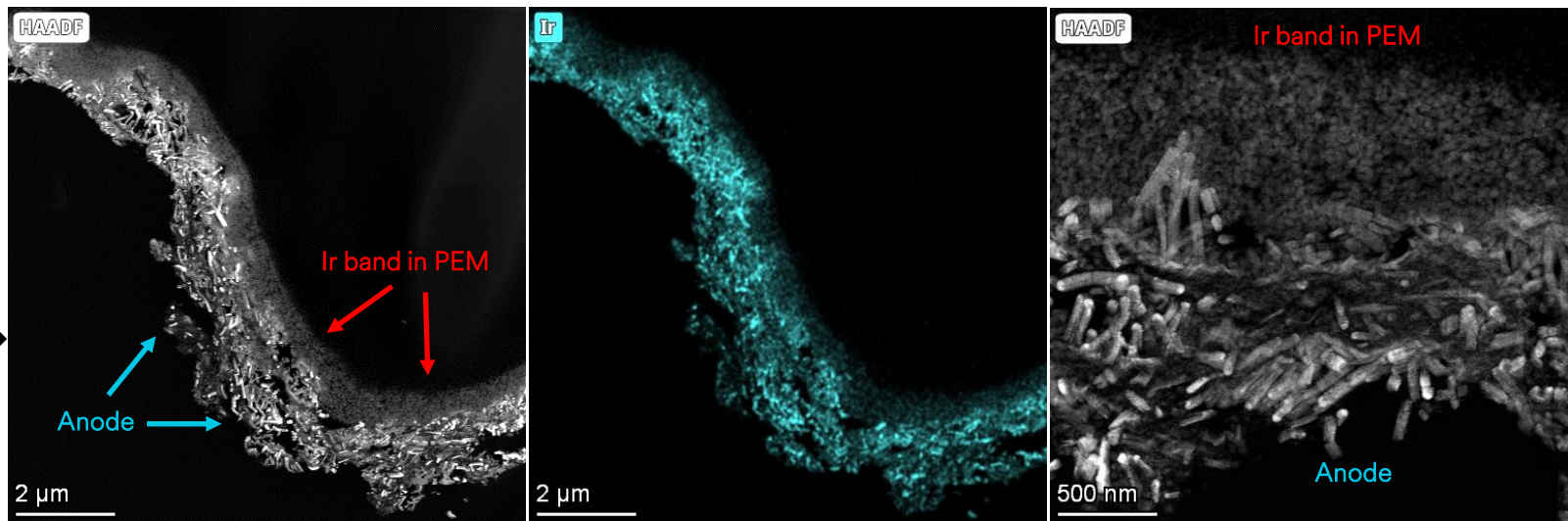
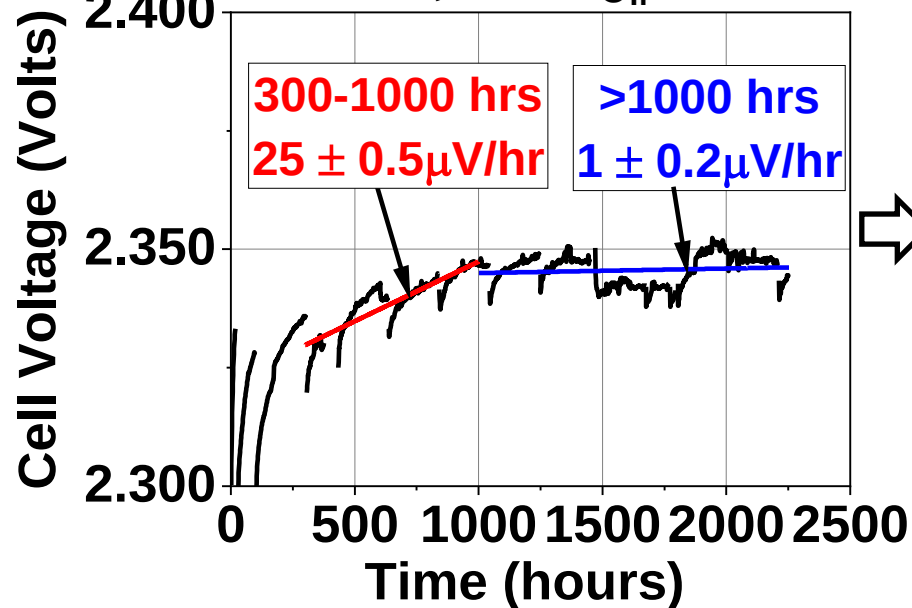
- (Ideally) enable larger degradation and elucidate differences more quickly.

Common conditions (3M): 80°C cell, ambient pressure, 75mL/min, 50cm² cells

Catalyst Structure Maintained After 2k Hours at 10A/cm²

Steady State Durability

10A/cm², 0.45mg_{Ir}/cm²



Ir loading (mg/cm ²)	Anode (Ir _x O _{100-x})	Membrane (Ir _x O _{100-x})	%Ir in membrane	Ir loading in membrane (mg/cm ²)
0.86	32	20	10	0.08
0.45	30	20	15	0.07
0.25	31	27	44	0.11

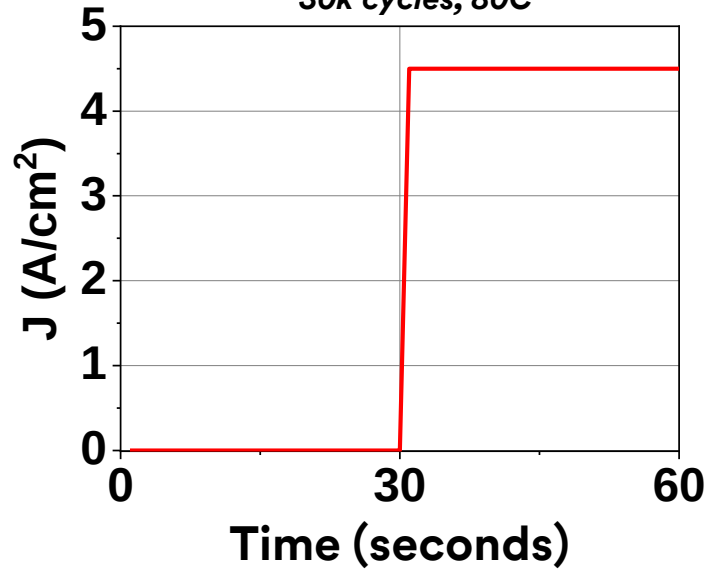
- HAADF STEM + EDS (ORNL) utilized to understand post-test catalyst and electrode structure
- Electrode structure and catalyst structure largely maintained with some Ir dissolution into PEM observed.
- Estimated Ir loading of band in PEM remained fairly constant with catalyst loading.

Anode: 0.45 mg_{Ir}/cm² 88wt% Ir/NSTF. Cathode: 0.09mg_{Pt}/cm² Pt/NSTF. PEM: 3M PFSA 800EW 98 μm.
PTL/GDL: Pt-coated sinter/Freudenberg H2C2. 80°C cell, ambient pressure, 50cm² cell.

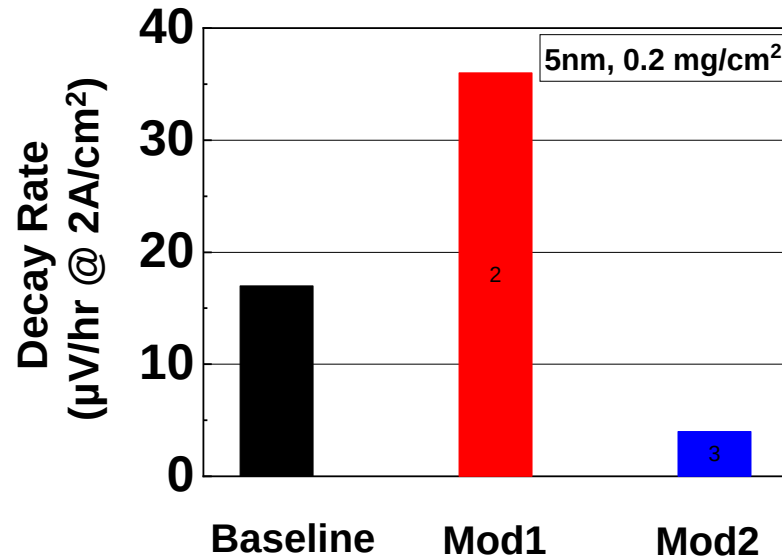
Catalyst Modification Yields Significant Durability Enhancement

HAST Protocol

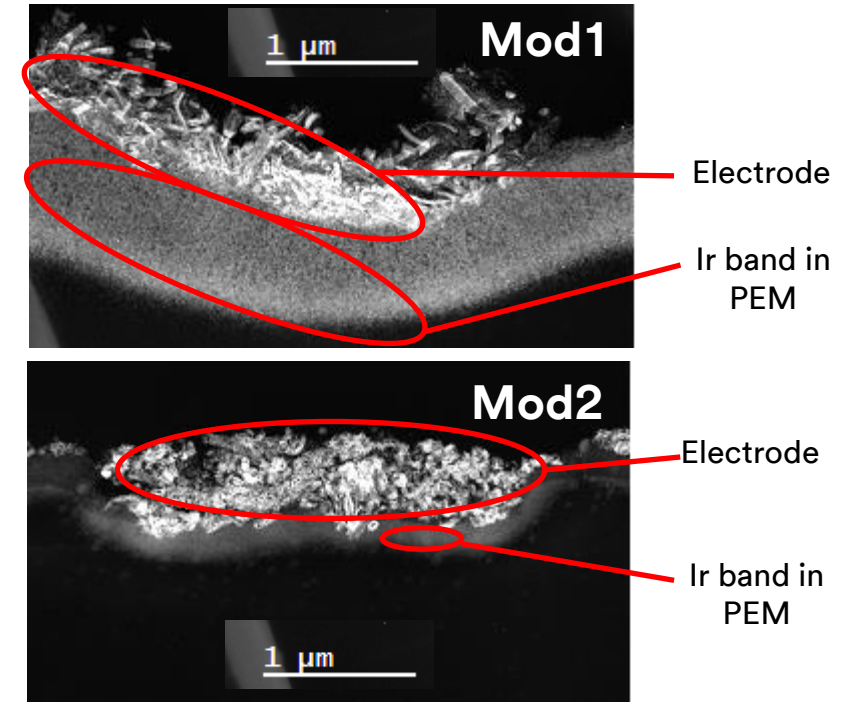
30k cycles, 80C



Catalyst Modification



TEM of Modified Catalysts

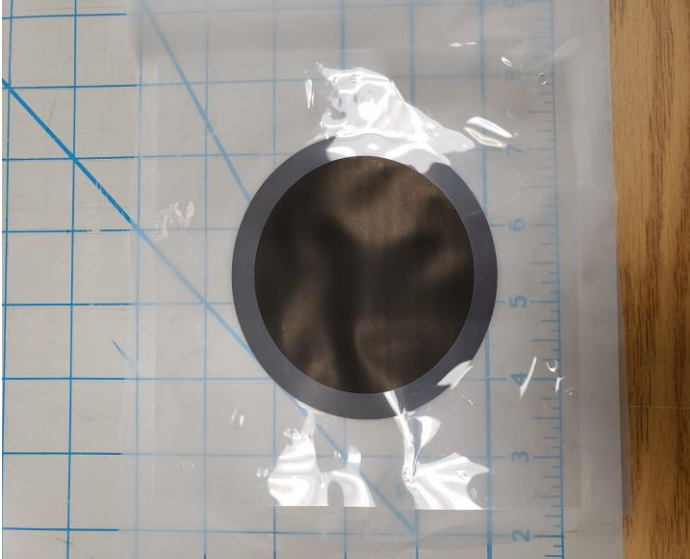


- HAST used to study impact of modifications to project catalyst.
- Mod1: 50% less stable than baseline Ir catalyst.
- Mod2: 3x more stable than baseline.
- Mod2 catalyst resulted in less Ir deposited into PEM, suggesting higher dissolution stability.

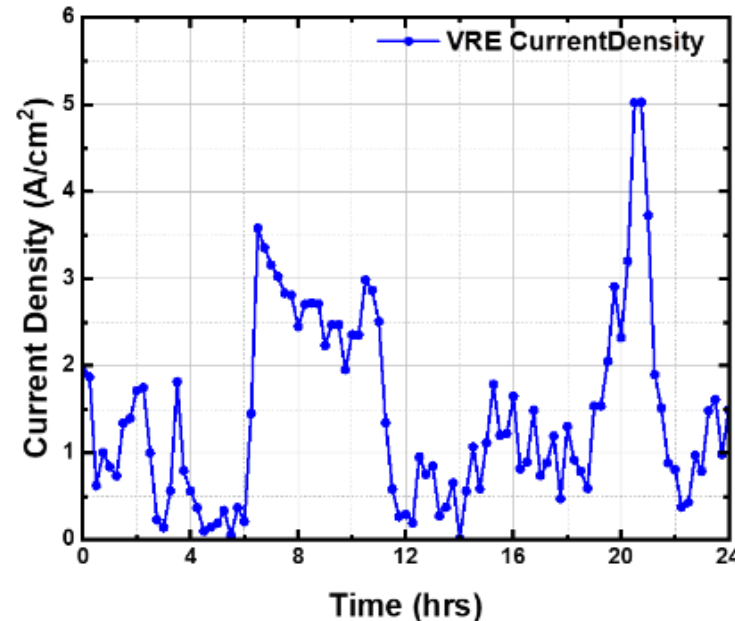
Anode: As Stated. Cathode: 0.09mg_{Pt}/cm² Pt/NSTF. PEM: 3M PFSA 800EW 98 µm. PTL/GDL: Platinized Sinter/Freudenberg H2C2. HAST: 80°C cell, ambient pressure, 50cm² cell, 30k cycles (0-4.5A/cm²)

Thrifty CCM Demonstrates High Durability Under VRE Protocol

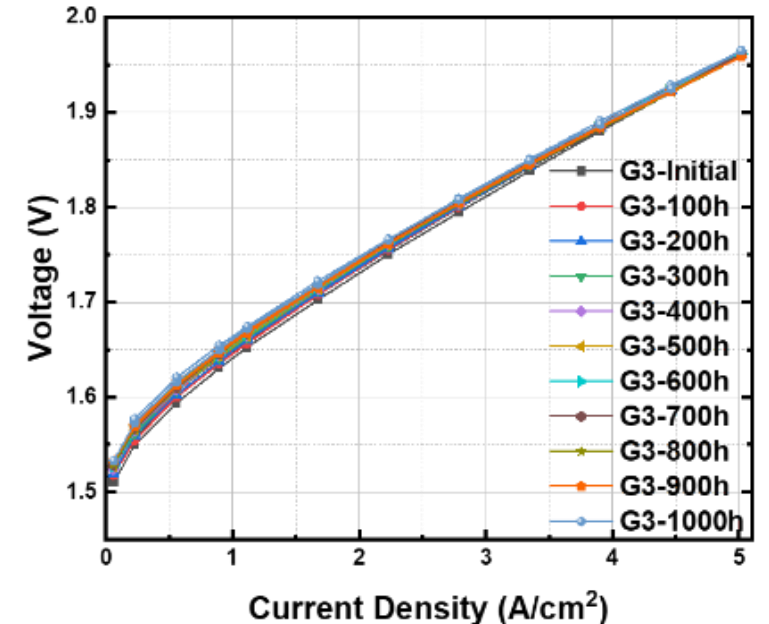
Thrifty CCM: CCM w/ Adhesively-Bonded Film Frame



Variable Renewable Energy (VRE) Protocol



Durability Test 80C, 300psi Differential Pressure



- Motivations:

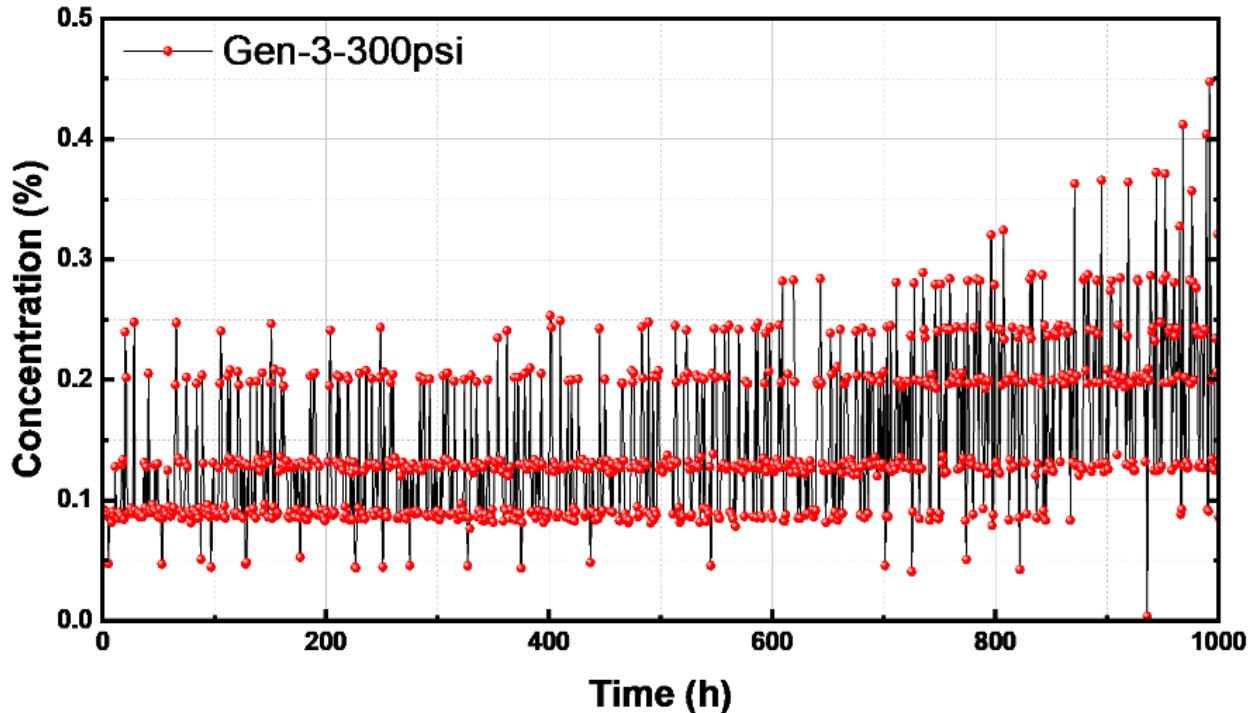
1. Reduce usage of expensive active materials.
2. Confirm material stability of adhesive and film.

- This year: Design iterations to improve mechanical durability.
- Downselect design tested for 1000 hours under 300psi differential pressure, 80°C, project VRE protocol.
- Steady state decay rate of 1 μ V/hr at 2.2A/cm² over 500 hours

Anode: 0.25 mg_{Ir}/cm² 88wt% Ir/NSTF. **Cathode:** 0.09mg_{Pt}/cm² Pt/NSTF. **PEM:** 3M PFSA 800EW 98 μ m.
PTL/GDL: Giner PTL/Freudenberg H2C2. 80°C cell, ambient pressure, 45cm² cell.

Thrifty CCM Failed After 1000 Hours Due to Crossover

Hydrogen Crossover During VRE Test



- Crossover mitigation effective with 300psi differential pressure; slight increase after 800 hours.
- After 1k hour test completed, abrupt crossover failure occurred -*coincident with idle*.

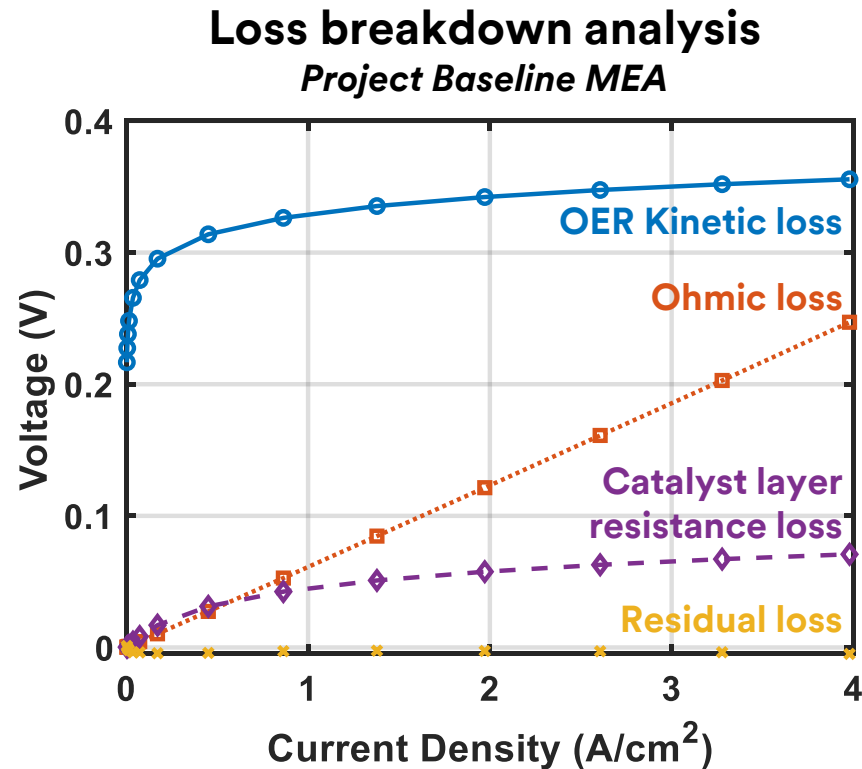
Thrifty CCM Appears Intact After 1000 hr Test



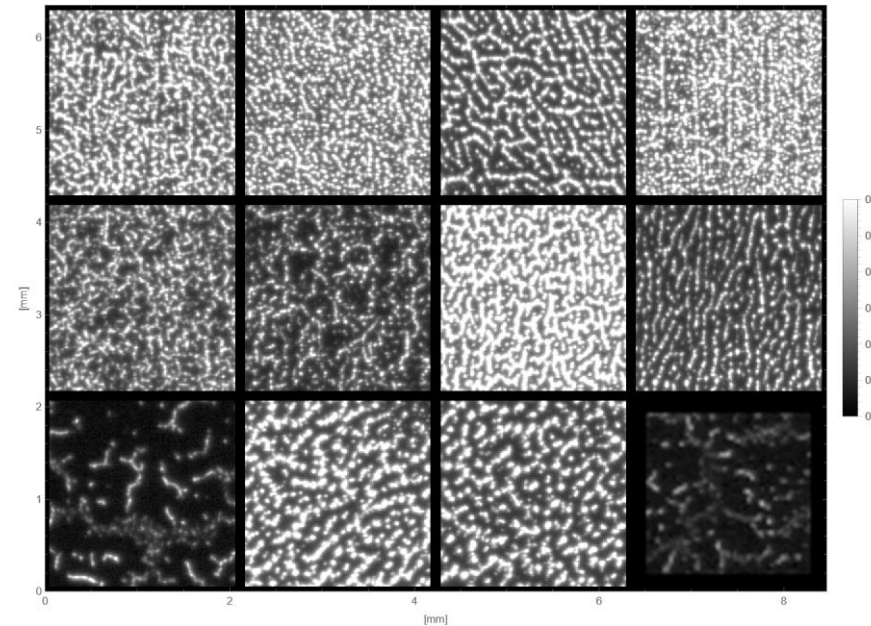
- No obvious failure of CCM or interfaces
- Film frame appears stable
- Adhesive remains well bonded to CCM.

CCM appears intact - not yet determined.

Studies Initiated to Address Catalyst Layer Resistance



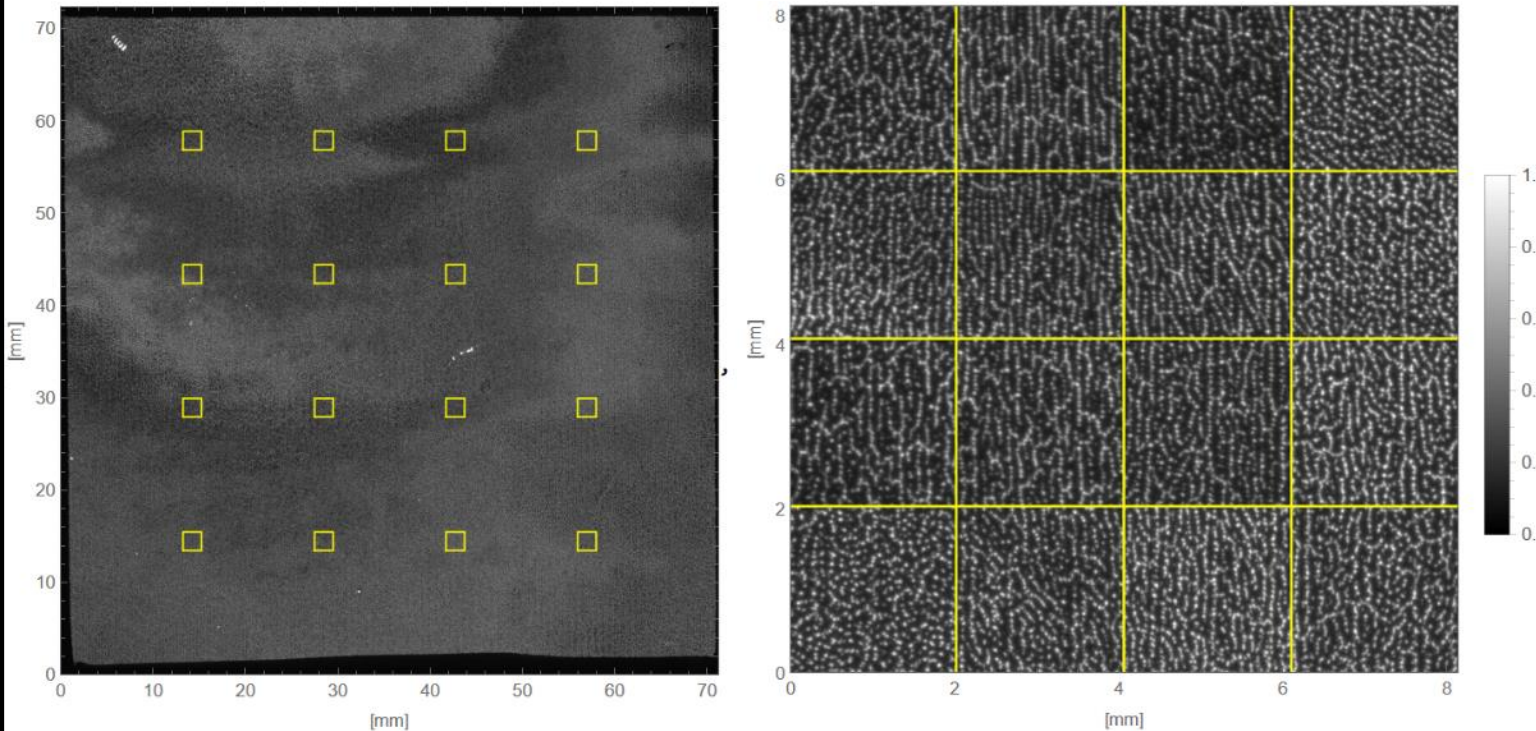
Electrode Structures by Hybrid Imaging
Various Lab Electrodes on Substrate



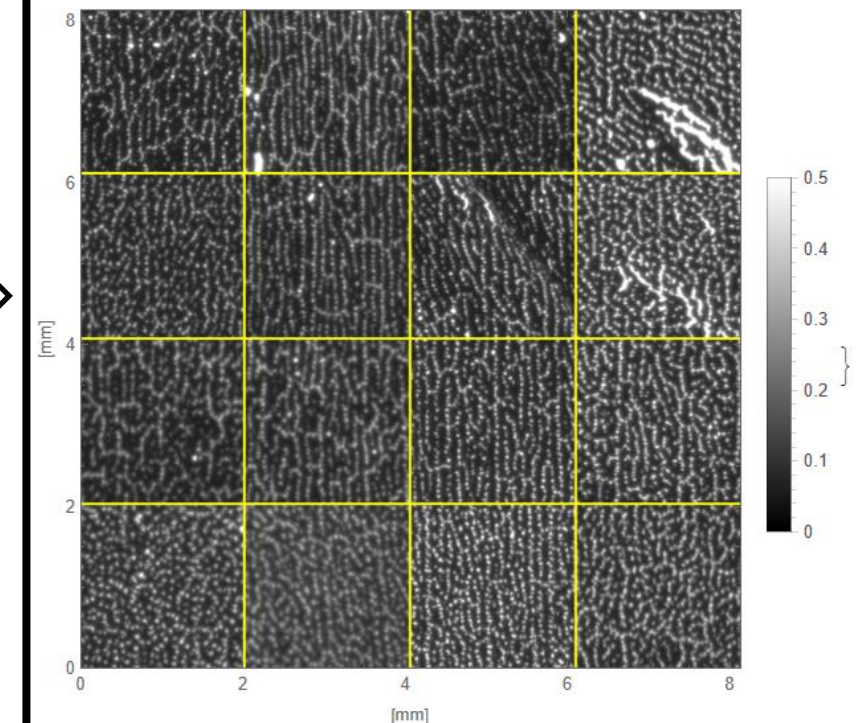
- Studies *last year* revealed:
 - Large catalyst layer resistance (CLR) identified as significant source of performance loss.
 - New electrode inspection technique revealed electrode microcracks, which could influence CLR.
- Studies initiated to better understand and address catalyst layer resistance, due to performance and potential durability concerns.

Inspection Development Reveals Electrode Microcracks in CCMs

Electrode Structure On Substrate



Electrode Structure on CCM

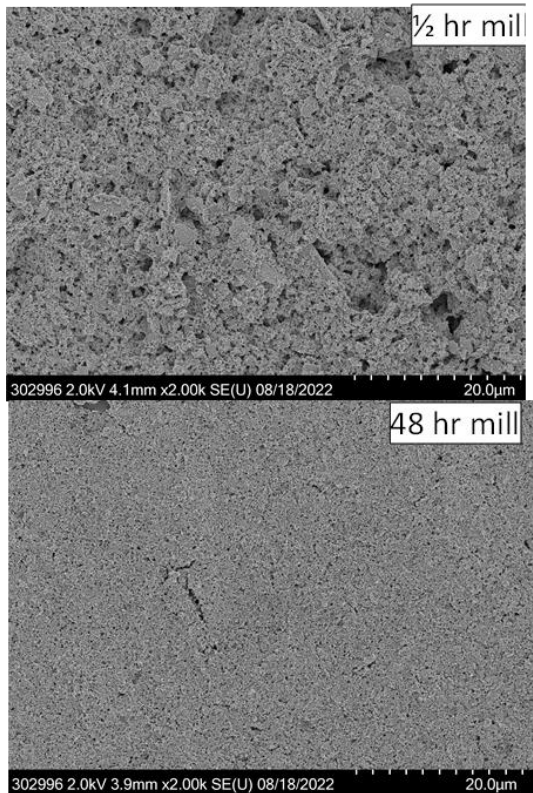


- Electrode microcrack networks and other large-scale discolorations observed on substrate also observed on CCMs (even after high pressure, high T lamination).
- Microcracks in CCM electrodes heightens potential performance, durability concerns

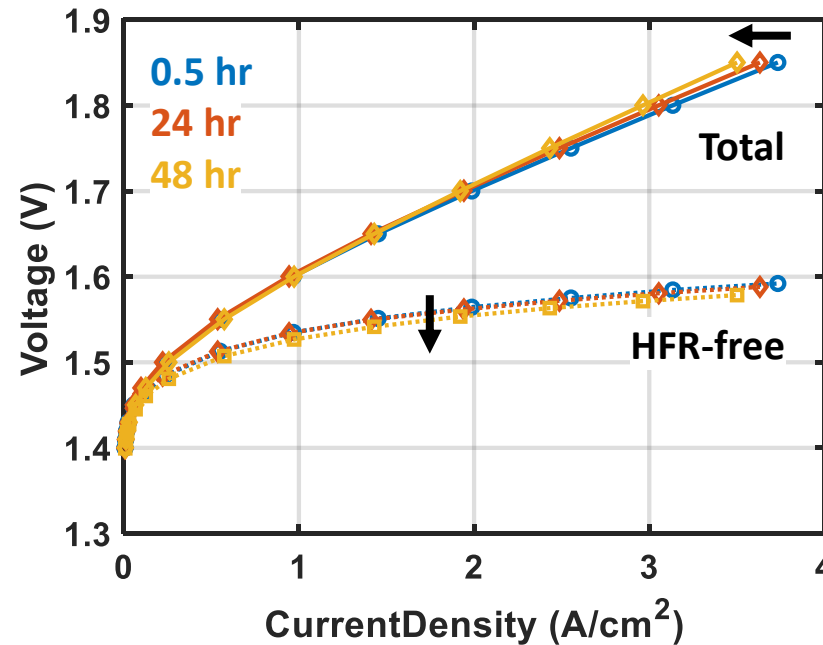
Ink Milling Influences Electrode Structure and Catalyst Utilization

Structure vs. Ink Mill Time

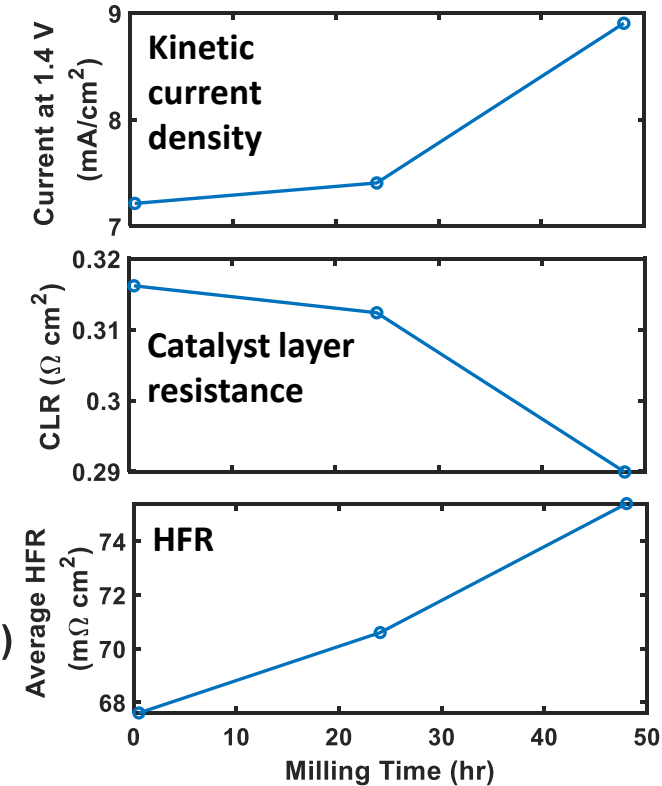
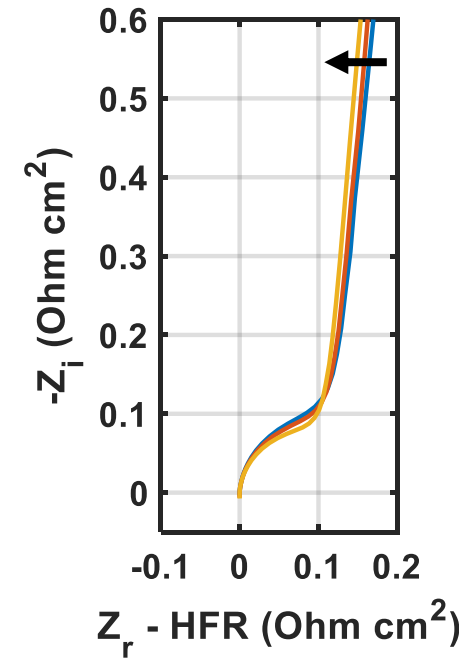
Top-down SEM



Cell Performance vs. Mill Time



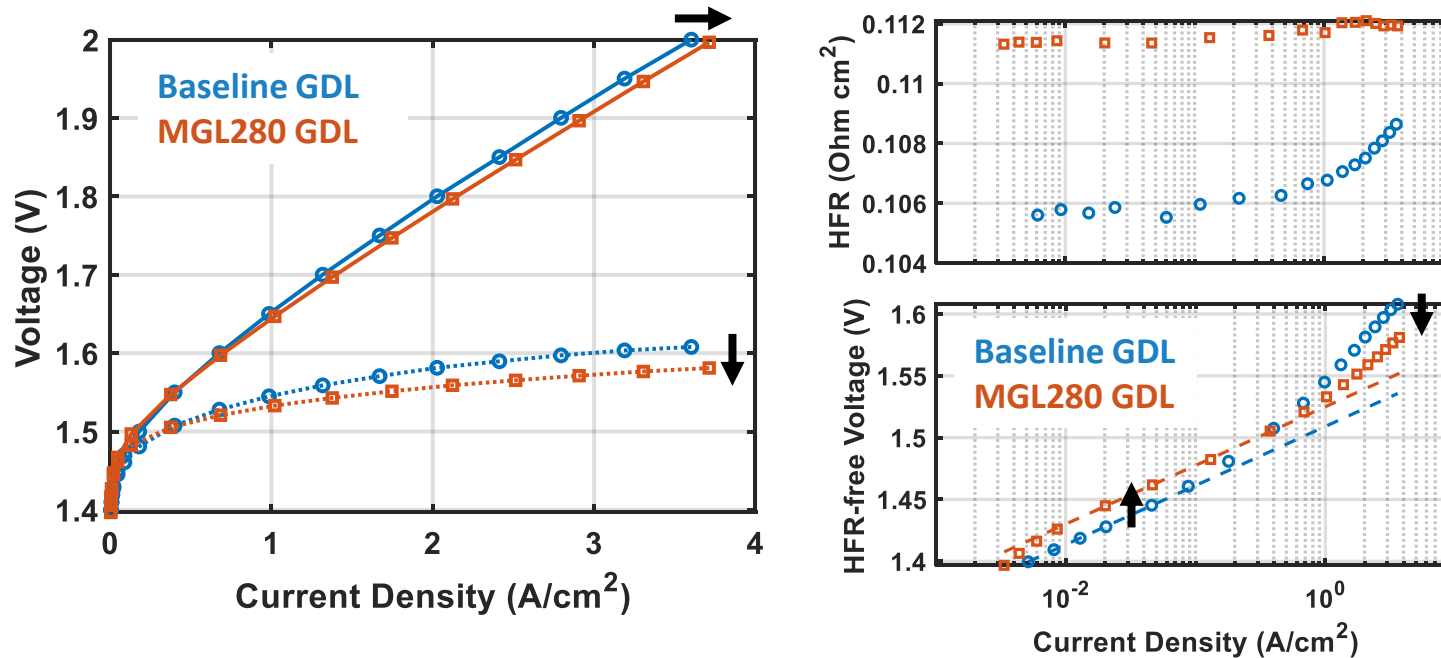
Loss Breakdown Analysis



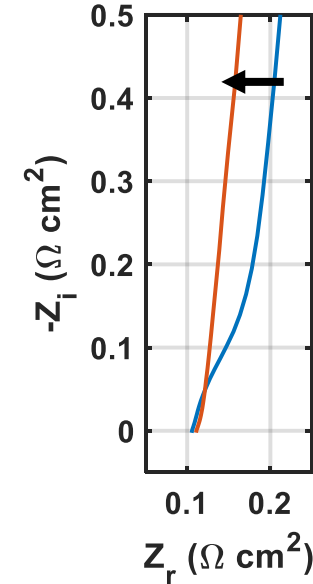
- Minor performance impact from increased milling time:
 - Benefits: Slightly improved kinetics, slightly decreased CLR
 - Negative: Slightly increased ohmic resistance

Cathode GDL Had Surprising Impact on Anode Kinetics, CLR

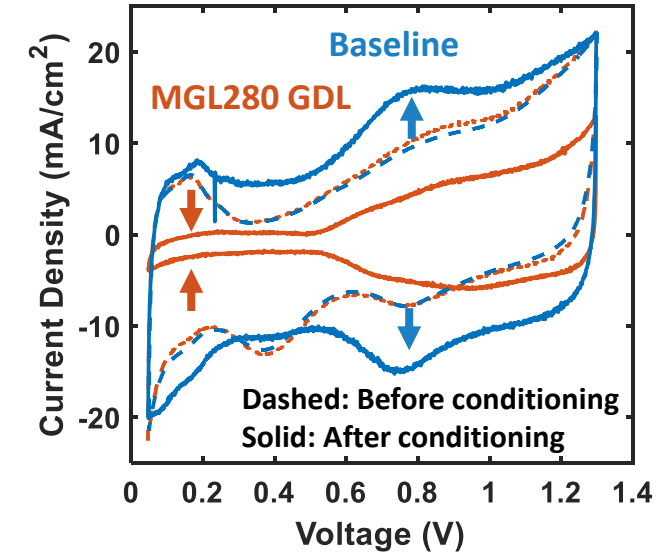
Performance, HFR, Tafel Analysis



Impedance



Voltammetry



- Replacement of project baseline GDL with MGL280 (H2New baseline) had significant effects.
 - Lower apparent CLR via impedance and HFR-free voltage.
 - Lower activity
- CVs imply that a different anode catalyst surface forms during conditioning.
- Result suggests possible potential complex interplay between components may strongly influence OER catalyst utilization.

Team spans Industry and National Laboratories

Collaborations

- **3M - Component Process Development, Durability, Inspection Development**
 - A. Steinbach (PI), G. Thoma, H. Dukes, F. Sun, J. Bender, P. Crain, J. Drumm
M. Burch, C. Rock, A. Marcella, J. Phipps, E. Iverson, M. Kuznia
- **National Renewable Energy Laboratory – Electrode and QC Development; Durability**
 - P. Rupnowski, S. Alia, S. Mauger, E. Padgett, J. Pfeilsticker
- **Giner, Inc. – Sub-scale Component Performance Validation; Durability**
 - H. Xu, S. Ding, and N. Macauley
- **Oak Ridge National Laboratory– Microscopy of Developmental and Durability Components**
 - D. Cullen, M. Zachman, S. Reeves
- **Plug Power – Performance Validation, Stack Testing**
 - C. Mittelsteadt, Z. Green

Response to Reviewers' Comments

- This project was not reviewed last year.

Project on Track; Challenges Addressable

Remaining Challenges	Future Work
2 of 5 process steps (deposition) are not capable of meeting the BP2 and project rate target.	Accelerate commissioning of new deposition modules; demonstrate at 2GW/year (BP2 GNG).
Electrodes are non-uniform and have appreciable ionic resistance, potentially impacting performance and durability.	Implement/demonstrate improved milling process at pilot scale. Further studies to optimize cell construction for maximum catalyst utilization and performance.
3M exited from PFAS manufacturing; impacts project electrode, thrifed CCM technologies. 3M to Exit PFAS Manufacturing by the End of 2025 - Dec 20, 2022	Project modification options being considered in near-term; meeting with DOE pending.
Stack power density target not yet demonstrated.	Anticipate demonstrating in Giner short stacks in Q2; Plug Power stack in BP3.

Any proposed future work is subject to change based on funding levels

Summary

- Project catalyst process development on track
 - New modules, addressing rate limiting steps, designed, installed and being commissioned.
 - Catalyst activity uniformity has improved 56%
- Project catalyst/electrodes are highly durable under variety of aggressive protocols
 - Low decay rates and good compositional stability at 10A/cm² (steady state)
 - Catalyst modification shows promise for excellent HAST (on/off) durability
- Improved understanding of factors influencing catalyst utilization in low loading electrodes.
 - Anode electrode homogeneity, cathode GDL
- Thrifted subgasketed CCM concept suggests good prospects for achieving targets
 - 1000 hour stability in single cell testing at 300psi differential pressure
 - Low decay rates under project VRE (slow cyclic) durability
 - Unexpected crossover failure after off-nominal operation.