MILLION MILE FUEL CELL TRUCK
U.S. DEPARTMENT OF ENERGY

M2FCT: Million Mile Fuel Cell Truck Consortium

DOE Hydrogen Fuel Cell Technologies Office
2024 Annual Merit Review and Peer Evaluation Meeting
May 7, 2024

Co-Directors: Rod Borup (LANL), Adam Weber (LBNL)

Deputy Directors: Rajesh Ahluwalia (ANL), Rangachary Mukundan (LBNL), Debbie Myers (ANL), K.C. Neyerlin (NREL)

DOE AOP project award: WBS 1.5.0.402

M2FCT Consortium - Overview

Timeline

- Project start date: 10/01/2020
- Original Project end date: 09/30/2025
- FOA support will continue through upcoming new awards

Partners/Collaborations

- DOE DE-FOA-0002044: (Projects ending)
- DOE DE-FOA-EE0009244:
  - Cummins, Plug Power, UT Knoxville
- DE-FOA-0002446:
  - Raytheon, NeoGraf, GM, TreadStone, Plug Power
  - Caterpillar, Eaton, R&D Dynamics, Mahle
- DOE DE-FOA-0002792
  - GM, Raytheon, U Hawaii
- DOE DE-FOA-0002920
  - GM, SUNY Buffalo, UC Irvine
- DOE DE-FOA-0002922
  - ~ 15 new projects
  - Some to be supported by M2FCT

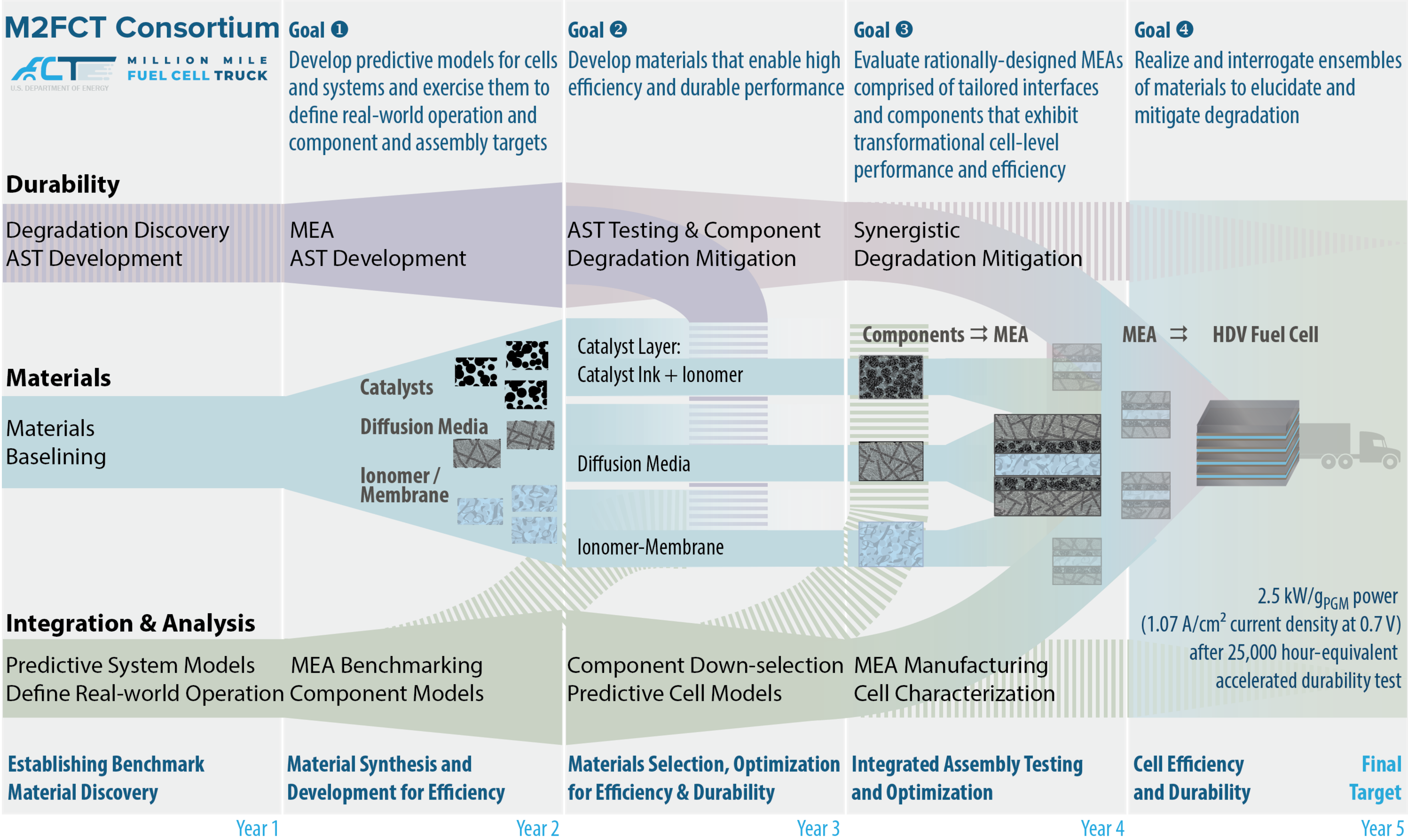
Fuel Cell Manufacturing	Fuel Cell Supply Chain
37. Ballard Power System	42. AvCarb Material Solutions
38. General Motors	43. Ballard Power Systems
39. Nuvera Fuel Cells	44. Cabot Corporation
40. Plug Power	45. Ionomr Innovations
41. Robert Bosch	46. Materic
	47. Pajarito Powder
	48. pH Matter
	49. Robert Bosch
	50. Robert Bosch
	51. Saueressig

Budget

- FY24 project funding: $10M
  - Expansion of FOA Support for upcoming DOE Awards

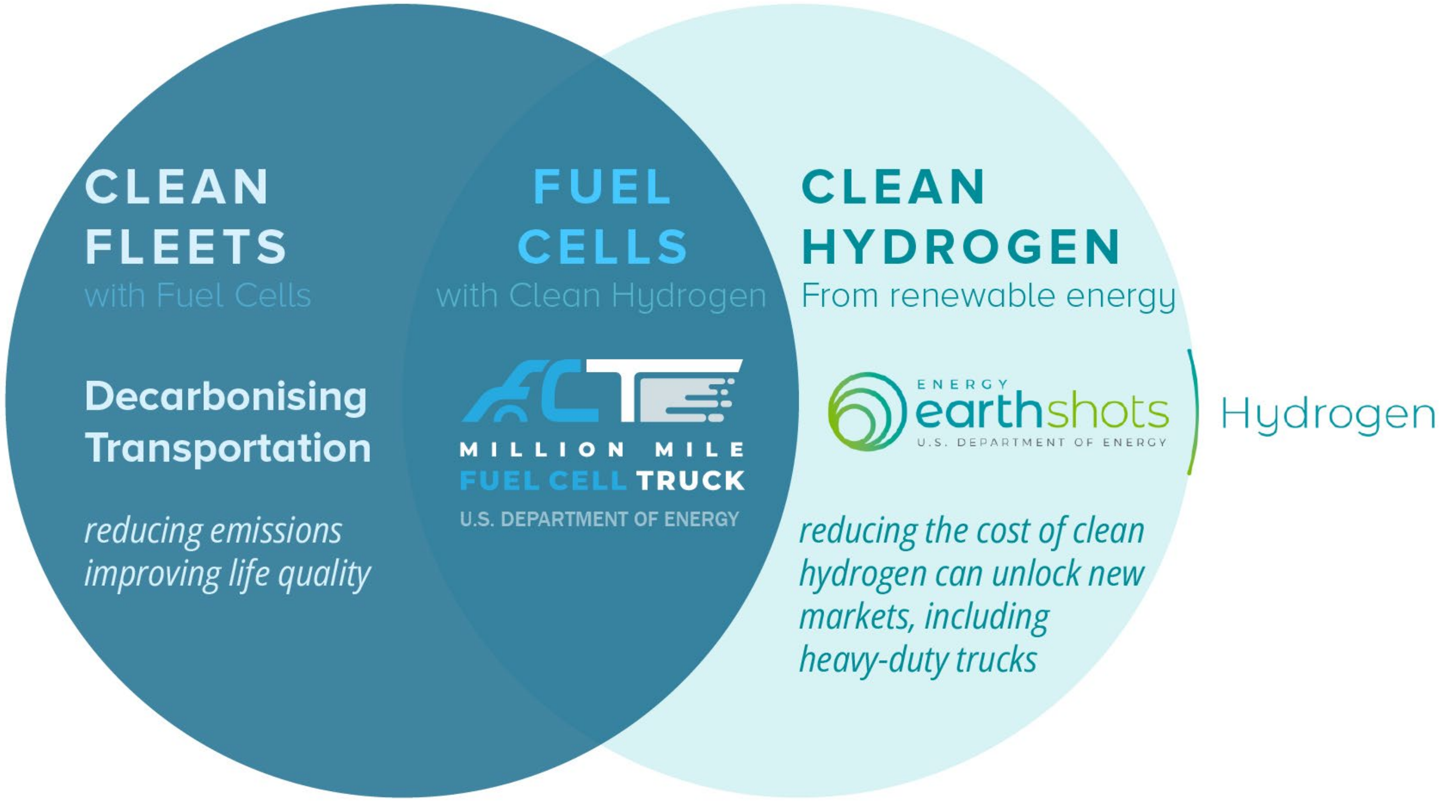
Barriers and Targets

- Cell durability
  - 25,000 hours (2025), 30,000 hours (2030)
- Peak efficiency
  - 68% (2025), 72% (2030)
- Fuel-cell system cost
  - $80/kW (2025), $60/kW (2030)
- **Overall Target:** 2.5 kW/g_{PGM} power - 750 mW/cm^2 (1.07 A/cm^2 current density at 0.7 V) - after 25,000 hour-equivalent accelerated durability test
- **2024 Target :** >1.3 A/cm^2 at 0.7 V and > 0.45 A/cm^2 at 0.8 V after catalyst AST (total PGM loading 0.3 mg/cm^2) with at least one 10 g batch of M2FCT-developed catalyst


M2FCT Consortium
MILLION MILE FUEL CELL TRUCK
U.S. DEPARTMENT OF ENERGY
Goal 1
Develop predictive models for cells and systems and exercise them to define real-world operation and component and assembly targets
Goal 2
Develop materials that enable high efficiency and durable performance
Goal 3
Evaluate rationally-designed MEAs comprised of tailored interfaces and components that exhibit transformational cell-level performance and efficiency
Goal 4
Realize and interrogate ensembles of materials to elucidate and mitigate degradation
Durability
Degradation Discovery
AST Development
MEA
AST Development
AST Testing & Component
Degradation Mitigation
Synergistic
Degradation Mitigation
Materials
Materials
Baselining
Catalysts
Diffusion Media
Ionomer / Membrane
Catalyst Layer:
Catalyst Ink + Ionomer
Diffusion Media
Ionomer-Membrane
Components ⇉ MEA
MEA ⇉ HDV Fuel Cell
Integration & Analysis
Predictive System Models
Define Real-world Operation
MEA Benchmarking
Component Models
Component Down-selection
Predictive Cell Models
MEA Manufacturing
Cell Characterization
2.5 kW/g$_{PGM}$ power
(1.07 A/cm^2 current density at 0.7 V)
after 25,000 hour-equivalent
accelerated durability test
Establishing Benchmark
Material Discovery
Material Synthesis and
Development for Efficiency
Materials Selection, Optimization
for Efficiency & Durability
Integrated Assembly Testing
and Optimization
Cell Efficiency
and Durability
Final
Target
Year 1
Year 2
Year 3
Year 4
Year 5

M2FCT Vision

Enabling decarbonization of heavy duty transportation by clean hydrogen and associated market liftoff



Million Mile Fuel Cell Truck consortium
To advance efficiency and durability of PEM fuel cells at a pre-competitive level to enable their commercialization for heavy-duty applications

millionmilefuelcelltruck.org

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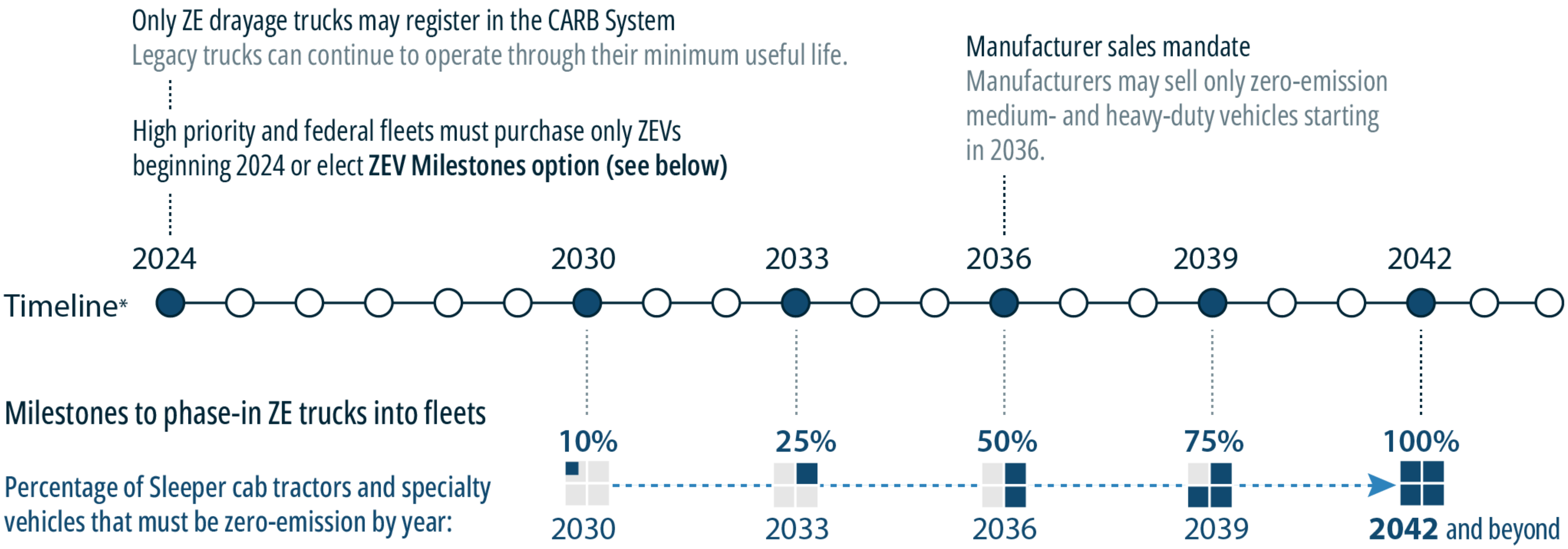
U.S. DEPARTMENT OF ENERGY

Zero Emission Trucks: Regulations for **large-scale transition**

Help enable adoption of FCETs via improved materials, designs, and understanding leading to better performing and longer durable fuel cells

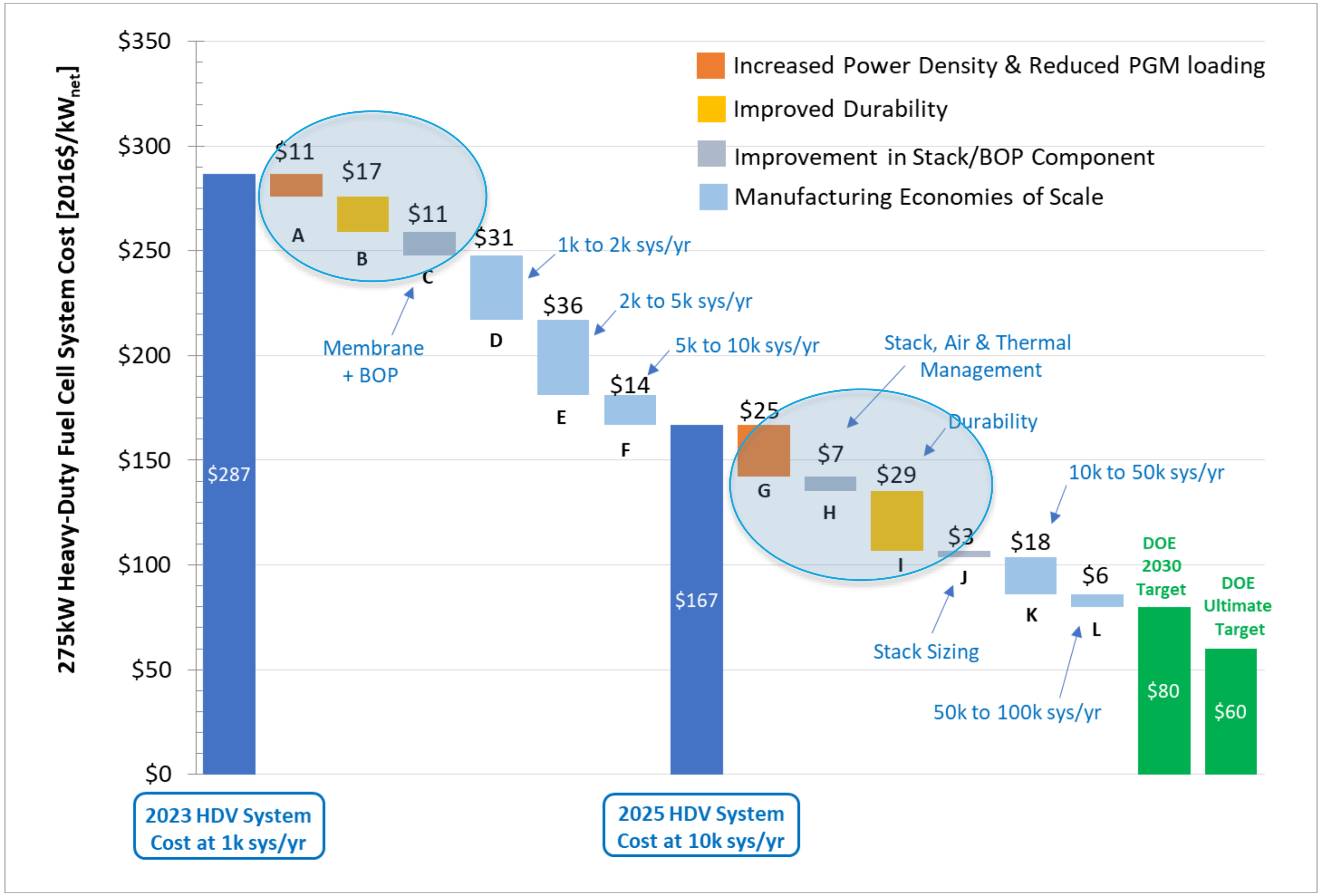
FCETs replace diesel and will drastically improve local air quality in frontline communities



Source: California Air Resources Board (CARB) | *Selected components are highlighted. Does not capture the entire components or regulation details.

M2FCT Focus Areas

From FC353

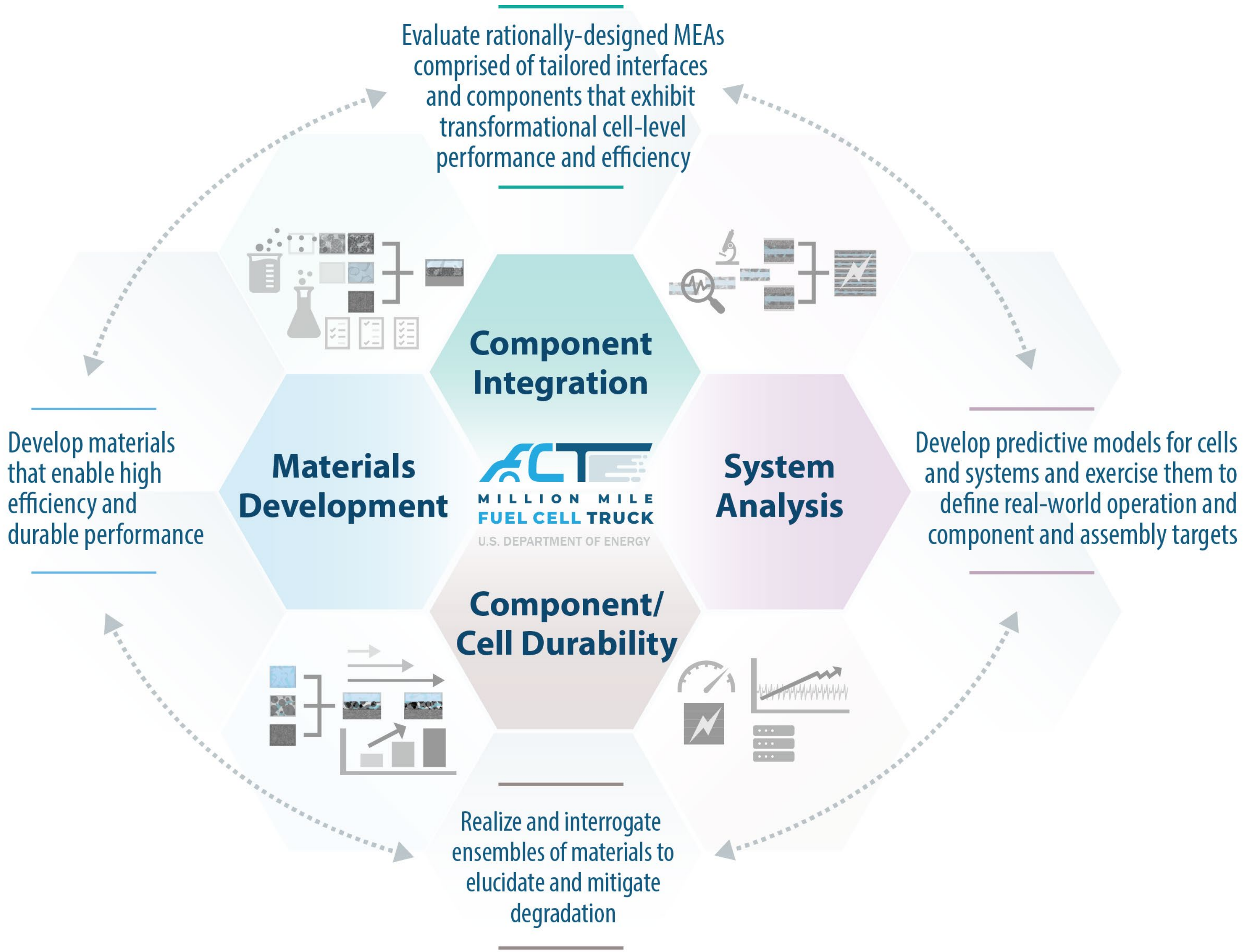


- Other than increased production volume, the largest cost reductions are from increased power density, reduced Pt loading, or improvement in durability through lower ECSA loss at EOL or removal of components that are currently needed to make the system more durable

M2FCT Approach

Million Mile Fuel Cell Truck (M2FCT) aims to tackle challenges through a "team-of-teams" approach featuring main teams in analysis, durability, integration, materials development.

By coming together as sets of dynamic teams, the integrated consortium will provide rapid feedback, idea development, and information exchange, resulting in an effort that is more than the sum of its parts.



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Task 1. Analysis	
	1. System analysis
Task 2. Materials development	
	1. Catalyst and support synthesis and characterization
	2. Membrane and ionomer synthesis and characterization
	3. Material scale-up
Task 3. Integration	
	1. Novel integration schemes and architectures for performance and durability
	2. Integrated assembly performance and models
	3. Multicomponent pre-assembly interaction exploration (diagnostics and models)
	4. Novel material integration into assemblies
Task 4. Durability	
	1. AST development and testing results
	2. Lifetime prediction and correlation
	3. Degradation mechanism discovery
	4. Mitigation strategies for improved component durability

Safety

- Project NOT required to submit a safety plan for review by the Hydrogen Safety Panel
- Every National Lab has a rigorous DOE approved Safety Procedure which is regularly reviewed and monitored by cognizant DOE Field Offices.
  - **ANL**: WPC (Work Planning and Control) which does use ISM (Integrated Safety Management)
  - **LBNL:** WPC (Work Planning and Control) which does use ISM (Integrated Safety Management)
  - **LANL:** ISM (Integrated Safety Management) by an Integrated Work Management (IWM) process
  - **NREL:** WPC (Work Planning and Control) and ISM (Integrated Safety Management)
  - **ORNL:** Research Hazard Analysis and Control (RHAC) System

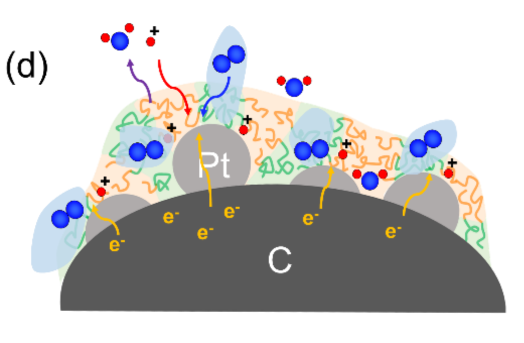
- Integrated Safety Management Process can be described:
  - Define the scope of work
    - What are you going to do?
    - Work planning should consider what might go wrong.
  - Identify and analyze hazards associated with the work
    - What are the hazards to people, environment, and equipment?
    - Anticipate human errors and put defenses in place.
  - Develop and implement hazard controls
    - How can the hazards be eliminated/mitigated?
  - Perform work within controls
    - Follow the plan that incorporates the controls and be aware of any new hazards
  - Provide feedback and continuous improvement
    - Make sure people who need to know are aware of successes and issues.

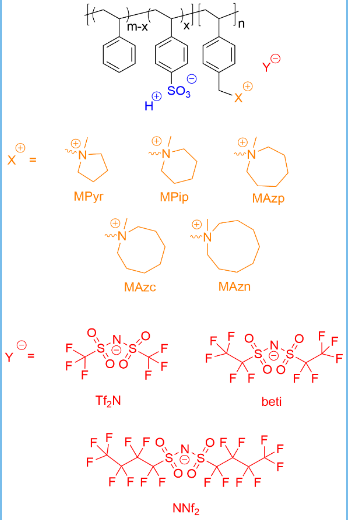
Discretionary Projects

Specific academic and non-primary lab call for projects to add into M2FCT

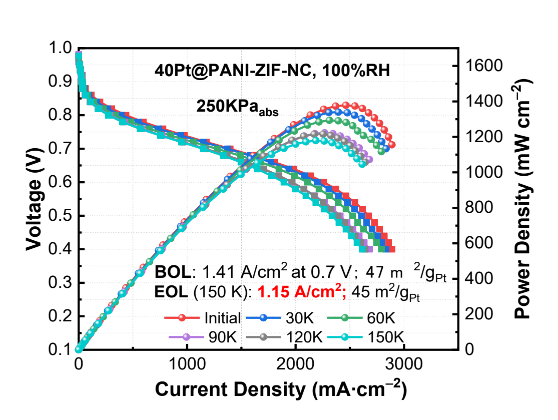
Material Development

- Advanced PILBCP Ionomers

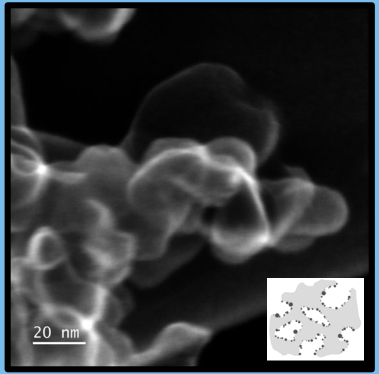


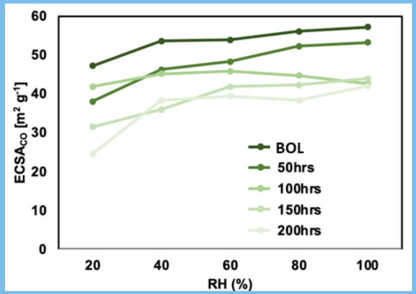


- Hybrid Carbon Support to Simultaneously Enhance Catalyst and Support Stability for PGM Cathodes in MEAs



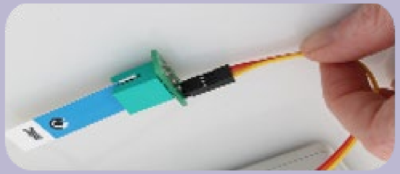
- Materials and Design Solutions for PEMFC Durability



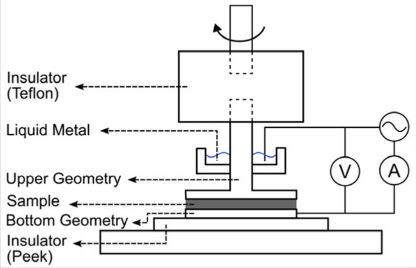


Diagnostic Development

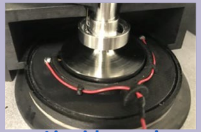
- On-line F-ion detection



- Rheo-Impedance



Schematic of the rheo-impedance set-up



Liquid metal

Insulated bottom plate

Insulated top plate

M2FCT Partners: National Labs, Universities, Industry

Mission

Advance efficiency and durability, and lower cost, of PEMFCs for HDV applications

Support various FOA Projects

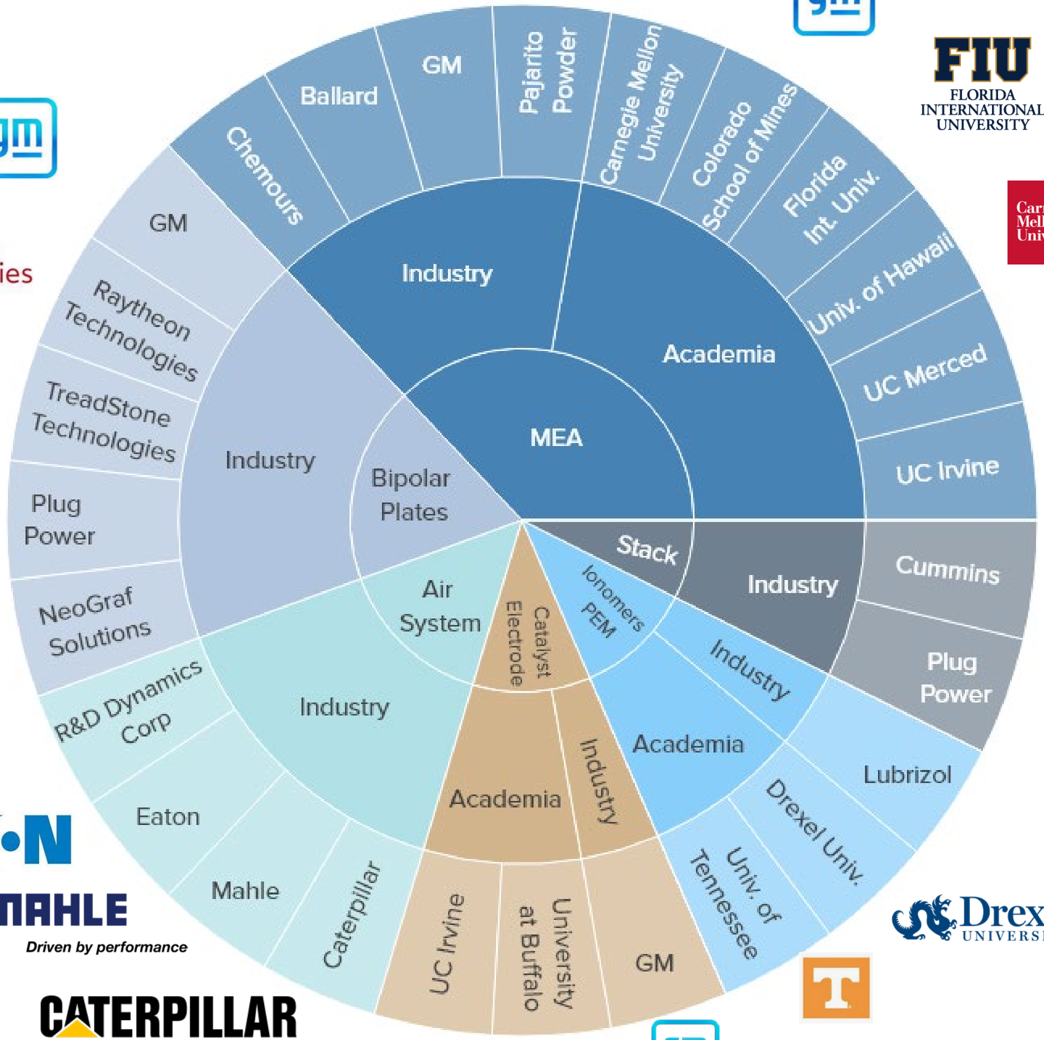
MEAs
GM
Raytheon
U Hawaii

Catalyst/CLs
GM
SUNY Buffalo
UC Irvine

Fuel Cell Manufacturing
37. Ballard Power Systems
38. General Motors
39. Nuvera Fuel Cells
40. Plug Power
41. Robert Bosch

Fuel Cell Supply Chain
42. AvCarb Material Solutions
43. Ballard Power Systems
44. Cabot Corporation
45. Ionomr Innovations
46. Materic
47. Pajarito Powder
48. pH Matter
49. Robert Bosch
50. Robert Bosch
51. Saueressig

M2FCT Consortium Partners and Projects



Main Laboratories: Los Alamos National Laboratory, Berkeley Lab, Oak Ridge National Laboratory, NREL National Renewable Energy Laboratory, Argonne National Laboratory

Affiliate Laboratories: Pacific Northwest National Laboratory, Brookhaven National Laboratory, NIST National Institute of Standards and Technology

FY24 Milestones

Milestone Name / Description and Criteria	Quarter/ Milestone Type	Status
Complete OCV tests of at least two state-of-the-art (SOA) reinforce and stabilized membranes with FER measurements. Analyze the degradation of SOA from the OCV tests plus HDV duty cycle and determine the operating conditions and design parameters to achieve 25,000-h lifetime.	**Q1 QPM**	Experimental complete, modeling in progress Slide 59
Define one or more "figure of merit" for benchmarking cell performance and/or degradation rate and other metrics, for tracking consortium progress, and for evaluating non-national lab M2FCT materials.	**Q1 QPM**	Complete See slide 23-24
Measure the baseline (Umicore PT500550) catalyst degradation rates at two different loadings (0.4 mg/cm^2 and 0.25 mg/cm^2) during the 50%RH MEA AST for 500 hours and determine acceleration factors based on modelled degradation rates during HDV drive cycle. Project performance out to 25,000-hour target using durability modeling based on ECSA and MA losses. Establish catalyst end of test (EOT) target for MEA AST corresponding to heavy duty lifetime of 25,000 hours.	**Q2 QPM**	Complete See slide 55-57
Project the performance and durability of a M2FCT's ORR-developed catalysts relative to the baseline metrics of 603 mW/cm^2 EOL power density, 25,000-h lifetime and 1.35 kW/g_{Pt} Pt utilization. Modeling projection will include changes based on ECSA, MA and Co loss.	**Q2 QPM**	Complete

FY24 Milestones

Milestone Name / Description and Criteria	Quarter/ Milestone Type	Status
Achieve current density of 1.3 A/cm2 at 0.7 V and 0.45 A/cm2 at 0.8 V after catalyst AST (total PGM loading 0.3 mg/cm2, 250 kPa, 85% RH, H_2/15% O_2) with at least one 10 g batch of M2FCT-developed catalyst. Characterize by STEM, BET and other available techniques to provide metal particle size distributions, Pt to transition metal ratios, and inter-atomic spacings for scaled up catalyst batches. Improve quantification of degree of ordering of inter-metallics, comparing different characterization techniques, and verify ordering with TEM.	**Q3 QPM**	In progress Small batch 24
Complete the scenario analysis for both 175 & 275 kW_e fuel cell systems and 0.6, 0.65 and 0.7 V EOL cell voltages at rated power for Class-8 long haul FC electric trucks.	**Q3 QPM**	In progress
Correlate the total fluoride emission rate (FER) from a 500-hour PFSA membrane OCV test to the membrane failure model and develop a target for a total FER from the 500-hour test. Model to be developed with M2FCT data from parametric study. Compare the MEA AST H_2/air data and determine the acceleration factors for electrode and membrane degradation. Develop and utilize methods for understanding HC membrane degradation mechanisms, including effluent water analysis for molecular fragments of membrane materials toward the goal of developing a metric equivalent to FER for HC membranes. Utilize triple-quad and quadrupole time-of-flight mass spectrometry to analyze the effluent of a single cell to determine identity and quantity of molecular fragments from HC membrane-based cells and PFSA membrane-based cells.	**Q3 QPM**	OCV tests complete; FER correlation in progress HC work in progress and defining MEA AST

FY24 Milestones

Milestone Name / Description and Criteria	Quarter Milestone Type	Status
Determine the improvement in performance, durability, and cost of fuel cell system with a top-performing M2FCT catalyst relative to a system using the baseline annealed Pt/C cathode catalyst and relative to the consortium targets, including \$80/kW cost and 2.5 kW/g_{PGM} at 0.7 V after 25,000 h with <0.3 mg/cm^2 total PGM loading.	**Q4 QPM**	In progress
Integrate scaled M2FCT catalyst with other SOA materials including GDLs and membranes into MEA. Optimize the catalyst layer/ink formulation and integrated MEA, including GDL, using multiple CL fabrication processes (e.g., Rod coating). Complete MEA durability test of 500 hrs and assess EOL performance. Project performance improvements required to meet 2.5 kW/g_{PGM} power (1.07 A/cm^2 current density) at 0.7 V after 25,000 hrs, as projected using M2FCT-developed modeling methodology.	**Q4 QPM**	In progress
Test at least three types of hydrocarbon (HC) membranes with different chemistry and evaluate their degradation factors; identify HC membrane degradation mechanisms, including those leading to abrupt failures. Assess effect of radical scavengers on HC membrane degradation and lifetime. Develop, fabricate and evaluate the performance and durability of M2FCT-developed membranes/ionomers (non-fluorinated). **Membrane:** Synthesize and characterize sulfonated hydrocarbon PEM with target IEC of 1.8-2.2 meq./g, conductivity of 0.1 S/cm, molecular weight (Mn), 10,000 g/mol). **Ionomer:** Synthesize and characterize sulfonated hydrocarbon ionomers with IEC of ~2.0 meq./g (10 ml of 5 wt% solution).	**Q4 QPM**	Preliminary results Met Ionomer See slide 32

FY24 Milestones

Milestone Name / Description and Criteria	Quarter Milestone Type	Status
MSI Recruitment Initiative Engage in activities that support recruitment of students and alumni of Minority Serving Institutions, including national lab staff going to MSIs/HBCUs/TCUs, hosting visits from professors from those institutions to a national lab, and other related activities. There will at least be 12 engagements a year at these institutions. Include at least 4 MSI students in M2FCT R&D activities. Provide at least one MSI student/PD with an industrial internship related to M2FCT activities.	**Q4 Annual**	~ 90% complete 12/12 and 3/4
Finalize M2FCT MEA AST and publish it on the M2FCT web site. Disseminate the final AST through presentations to the International Durability and AST working groups. The published M2FCT AST will have EOT targets for both catalyst and membrane degradation to correlate to 25,00 hours of simulated HDV operation.	**Q4 Annual**	In progress see slide 53-54

MILLION MILE
FUEL CELL TRUCK
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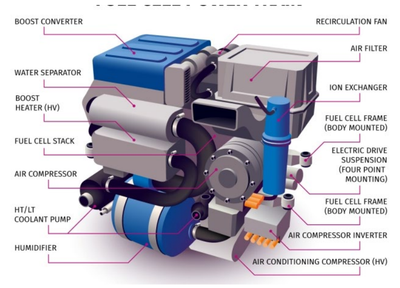
System Analysis

System Analysis Approach

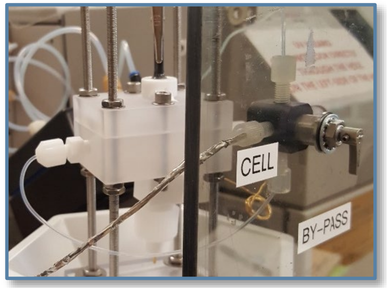
FY2024 Approach: Evaluate system with M2FCT's ordered intermetallic ORR catalyst on ZIF-8 support ($L1_0$-PtCo/C_{1100}ZIF-8, 0.3 mg/cm^2 total Pt loading) that has shown multiple advantages in lab tests



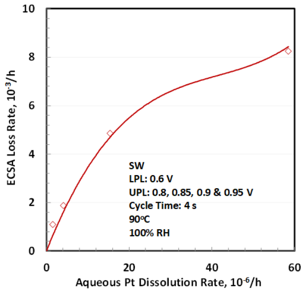
Define system configuration

Identify Class-8 truck duty cycle

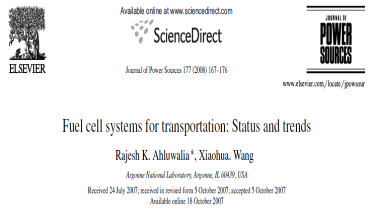
Baseline materials on ASTs



Characterize degradation mechanisms



Measure degradation rate in H_2/ N_2 or H_2/air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Fuel cell systems for transportation: Status and trends

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Run simulations

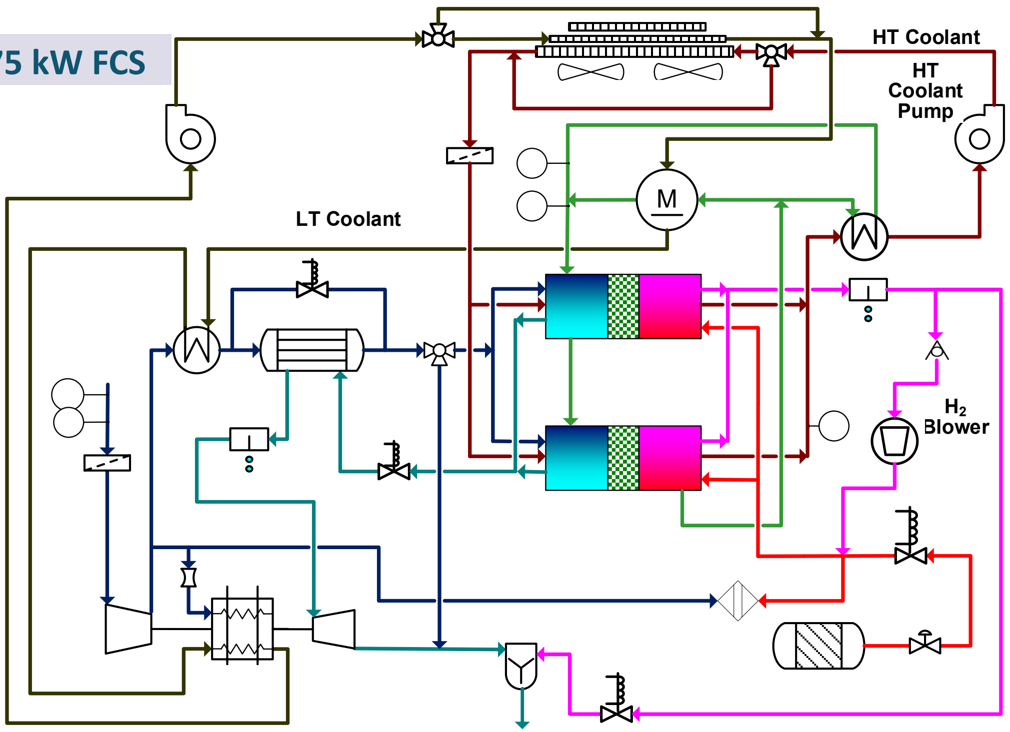
M2FCT Reference Fuel Cell Systems for Class-8 Heavy-Duty Trucks

FCH-175
- 175 kW net, 240 kW stack at EOL
- Two stacks
- 183-kWh ESS

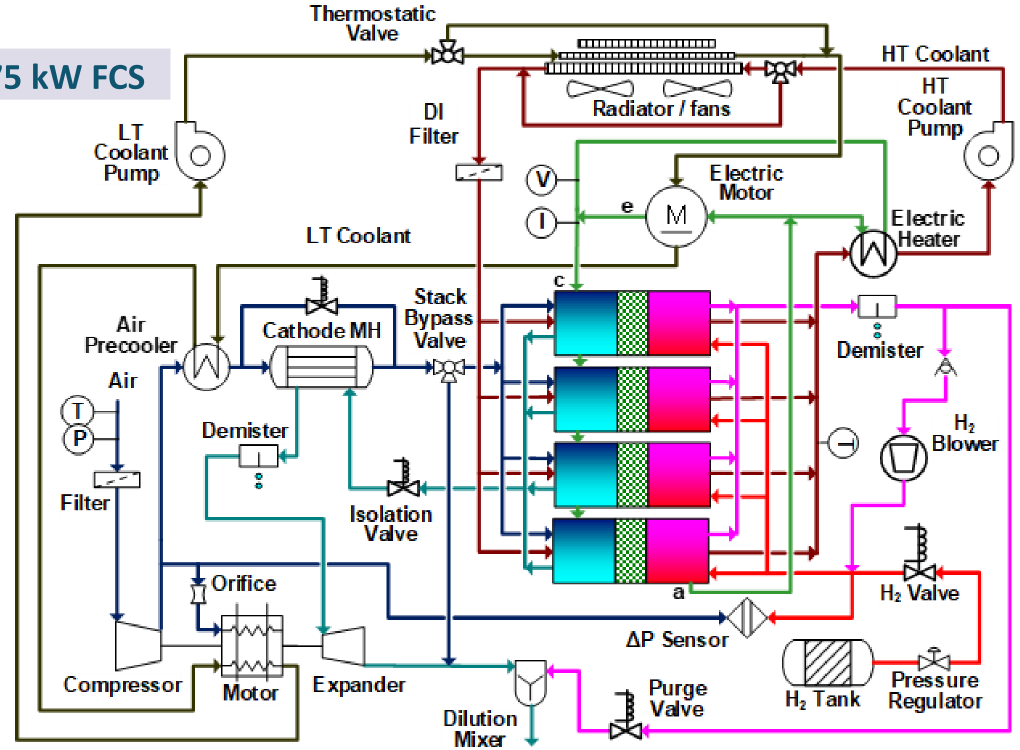
FCH-275
- 275 kW net, 360 kW stack at EOL
- Four stacks
- 106-kWh ESS

- Electrodes
  Cathode: a-Pt/C, 0.4 mg_{Pt}/cm^2, 50 wt.% Pt
  Anode: Pt/C w IrO_2, 0.05 mg_{Pt}/cm^2
- Membrane: 14 μm, chemically stabilized, mechanically reinforced
- Air system with expander
- Anode system with recirculation blower
- Cathode humidifier: Cross-flow with high flux WVT membrane
- Rated power conditions at EOL: 2.5 atm, 90°C, 700 mV

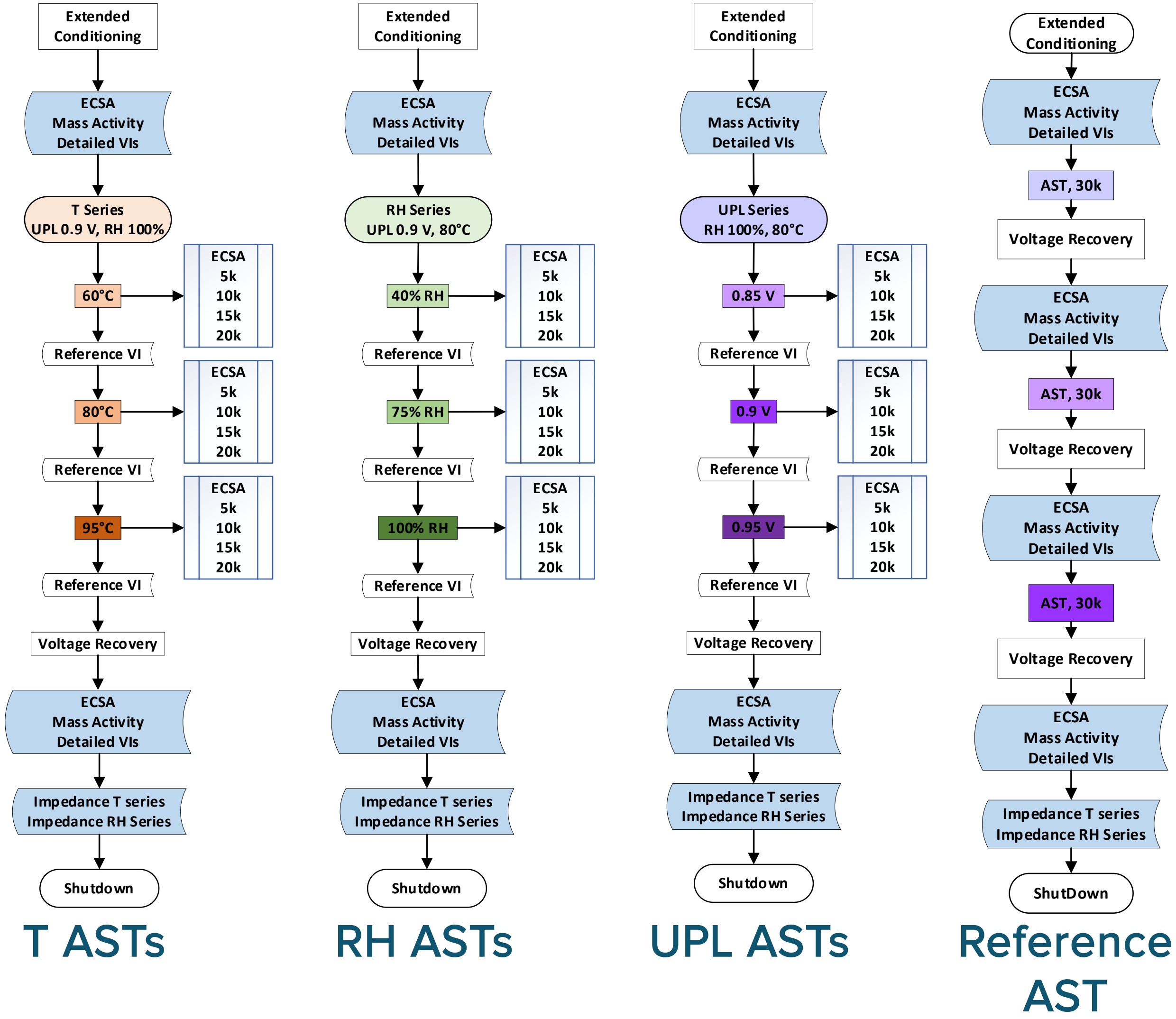
175 kW FCS



275 kW FCS



Developed and Implemented Test Protocol for Formulating Durability Model



Modeling

ECSA, ECSA Loss Rate, ORR Kinetics, O2 Transport in CCL

Reference Catalyst AST

- 5-cm^2 differential cell
- H_2/N_2, 80°C, 100% RH
- 0.6 V LPL, 0.9 V UPL
- 6 s/cycle

Detailed Polarization Curves

Test MEA

Cathode

- Catalyst: $L1_0$-PtCo/ C_{1100}ZIF-8, 30 mol% Co, 4.2 nm particle size
- Electrode: metal wt% = 37%, I/C = 0.6, 0.25 mg/cm^2 Pt loading

Membrane

- NC700, 15 µm thick

Anode

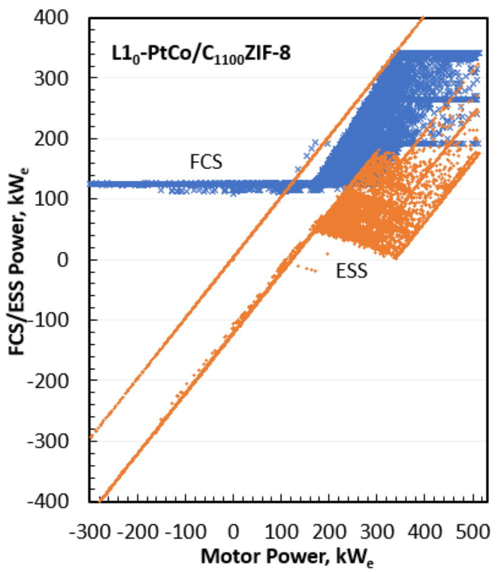
- Catalyst: Pt/C, 0.05 mg/cm^2 Pt loading

Pol Curves		P, atm	T, °C	X(O_2), %	RH, %
H_2/O_2	Mass Activity	1.5	80	100	100
H_2/Air Pol Curves	P Series	2.5, 1.5, 1.0	80	10	100
	T Series	1.5	95, 80, 60	10	100
	RH Series	1.5	80	10	100, 75, 40
	X(O_2) Series	1.5	80	21, 15, 10, 5	100
Ref. H_2/Air Pol Curves	Reference VI	1.5	80	10	100

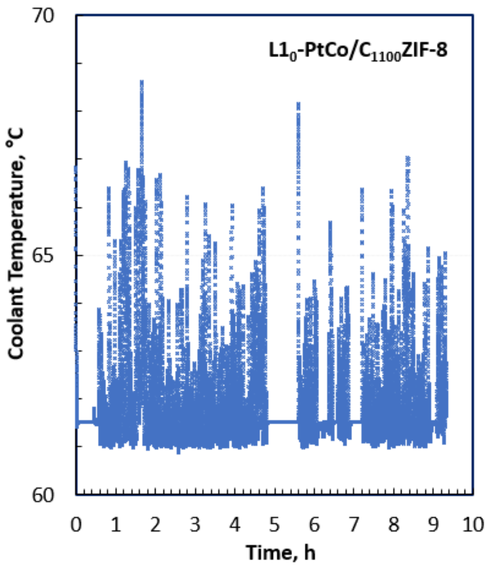
Step	Time
Extended conditioning	138 h
2 sets of detailed VI measurements before & after test series	69 h
3 reference VI measurements after every 20K cycles	9 h
12 ECSA measurements after every 5K cycles	1 h
60k AST cycles	100 h
1 voltage recovery	35 h
Total test time including conditioning step	352 h (15 d)

Mitigation Strategies to Reach 25,000-h Electrode Lifetime

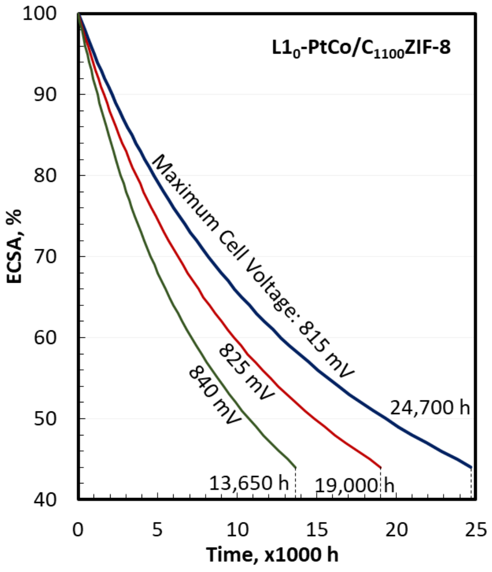
1. Distribute load to FCS & ESS



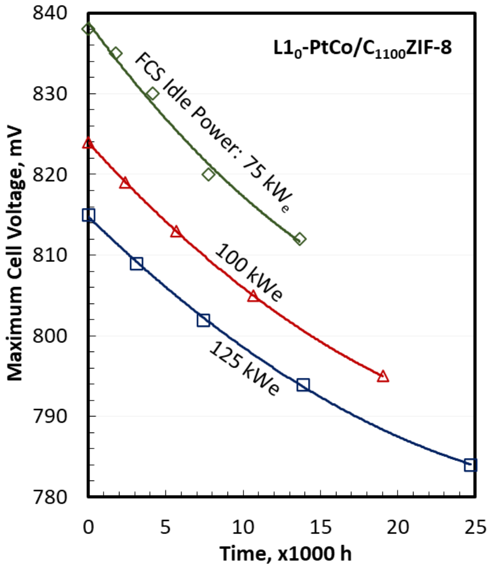
2. Control MEA temperature



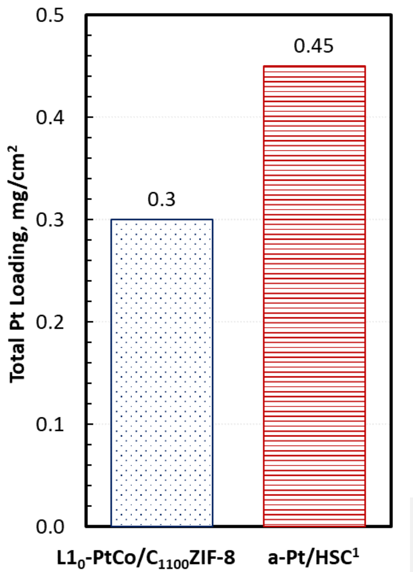
3. Clip cell voltage



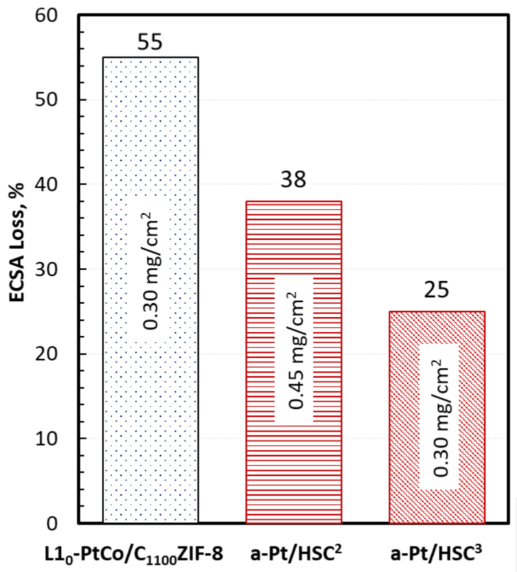
4. Maintain constant idle power



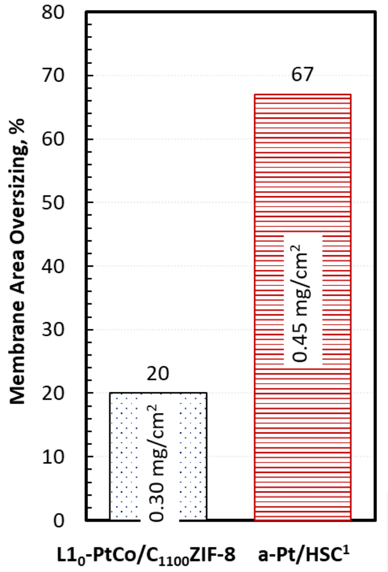
5. Overload catalyst



6. Limit ECSA loss



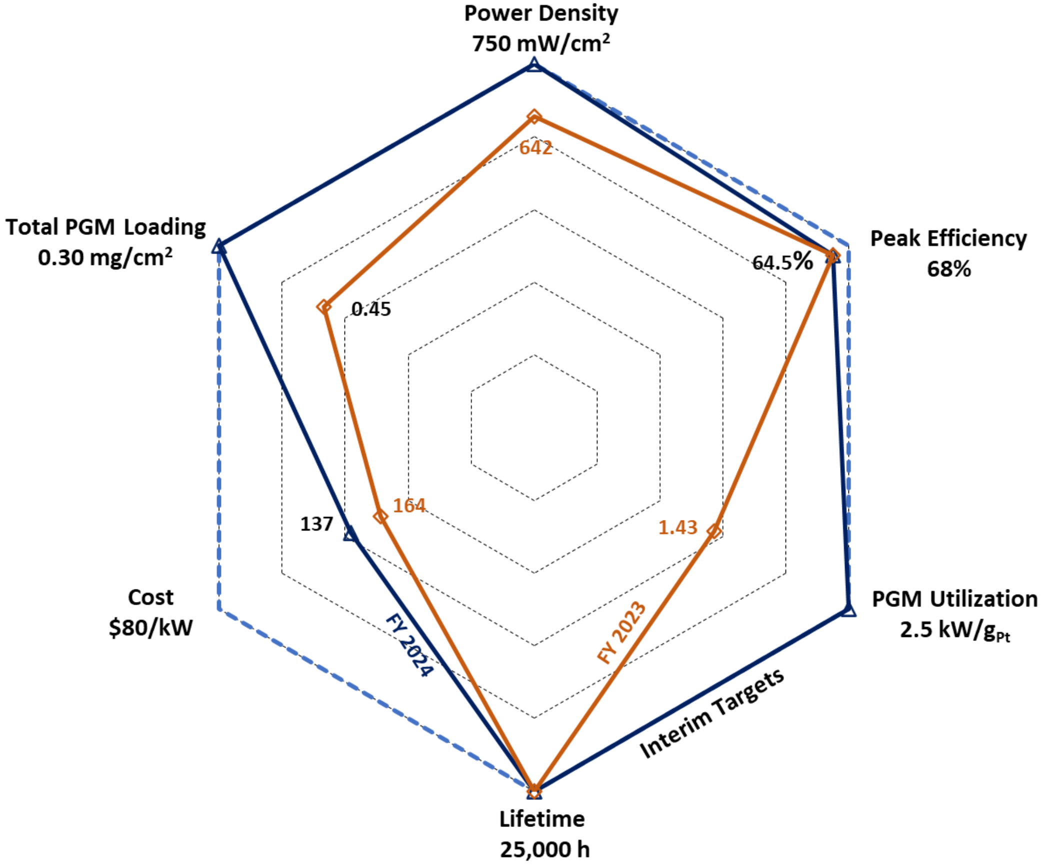
7. Oversize membrane area



Catalyst	L1$_0$-PtCo	a-Pt[1]	a-Pt[2]	a-Pt[3]
Catalyst Support	C$_{1100}$ZIF-8	HSC	HSC	HSC
Total mg-Pt/cm²	0.30	0.45	0.45	0.30
ECSA Loss, %	55	50	38	25
EOL PD, mW/cm²	750	642	750	750
Membrane Oversizing, %	20	67	44	28
Lifetime, h	25,000	25,000	17,440	4,700

Fuel Cells for Heavy Duty Trucks – FY2024 Status

These results indicate that the performance and durability targets may be met by applying system mitigation strategies to M2FCT's $L1_0$-PtCo/C_{1100}ZIF-8 ORR catalyst. However, further work is required to reduce the experimental uncertainties, refine the model, and validate results in larger integral cells.



Projected cost at 100,000 units/year manufacturing rate using 2023 cost correlations from Jennie Huya-Kouadio, Strategic Analysis

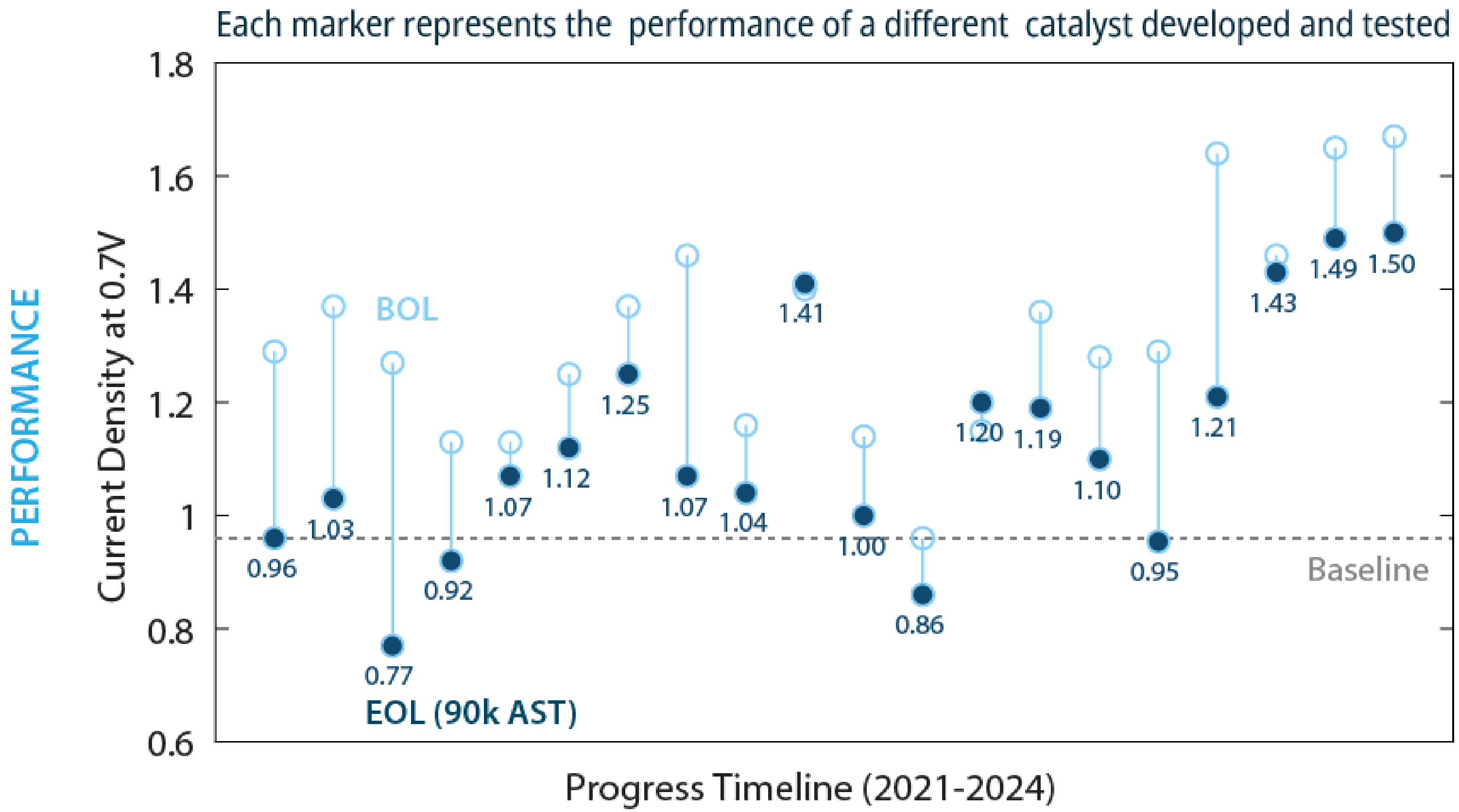
Next Steps

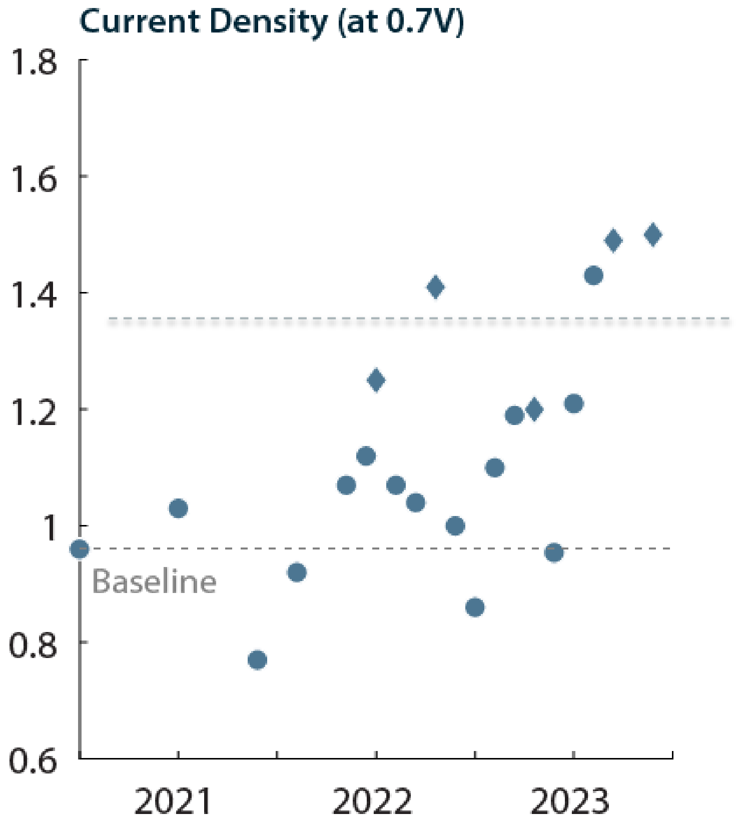
1. Obtain confirmatory data on differential cell hardware and reduce error bars
2. Further refine and validate the performance and aging model
3. Continue to investigate flooding of aged electrode at high current density and low temperature
4. Improve model using TEM and X-ray characterization data
5. Explore operating conditions to increase turndown and improve operability at 800-825 mV
6. Initiate aqueous dissolution experiments
7. Explicitly incorporate the effect of Co leaching on ORR kinetics and O_2 transport
8. Expand system model to consider membrane and catalyst support lifetime
9. Collaborate with SA to conduct cost analysis

MILLION MILE
FUEL CELL TRUCK
U.S. DEPARTMENT OF ENERGY

Materials Development

M2FCT Catalyst Materials Development





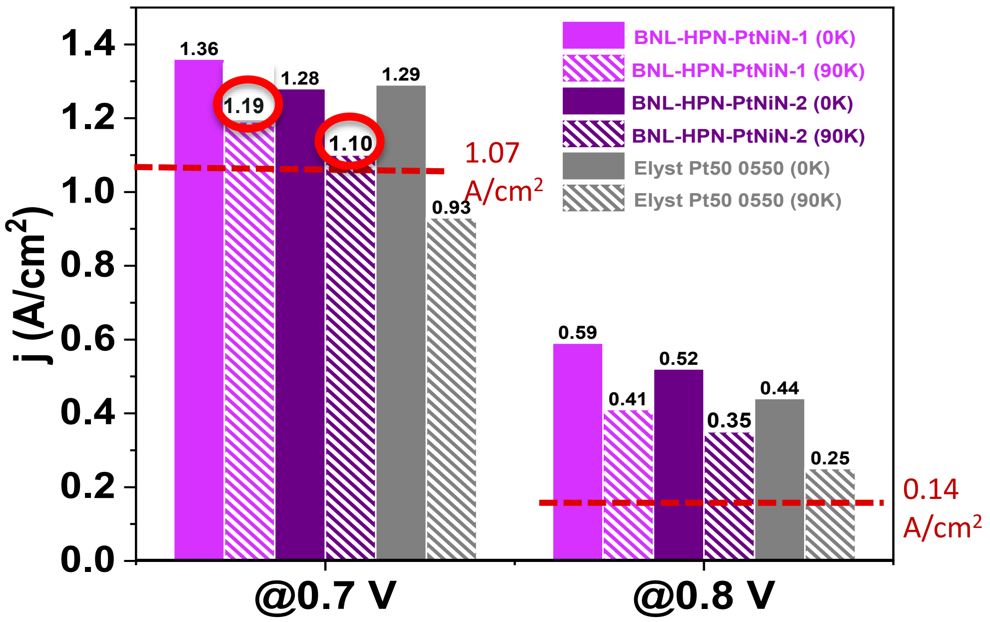
Overall M2FCT Goal: 2.5 kW/g_{PGM} power - 750 mW/cm^2 (1.07 A/cm^2 current density at 0.7 V) - after 25,000 hour-equivalent accelerated durability test

FY2024 Milestone: Achieve current density of 1.3 A/cm^2 at 0.7 V and 0.45 A/cm^2 at 0.8 V after catalyst AST (total PGM loading 0.3 mg/cm^2) with at least one 10 g batch of M2FCT-developed catalyst

M2FCT Catalysts Down-selected for Scale-up

High-Pressure Nitrided PtNi Intermetallic on Ketjen Black ((HPN)-Int-PtNiN/KB)

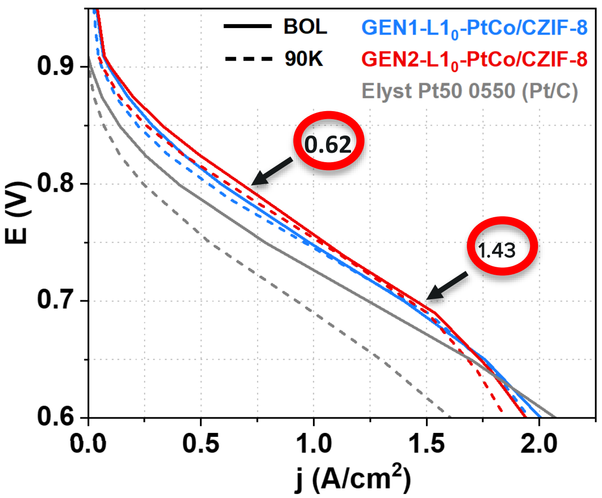
- Int-PtNiN/KB was first synthesized by ambient-pressure nitriding (APN) (560°C & 8h), then doped with more nitrogen by HPN at 800 psi at 500°C for 10h & 5h

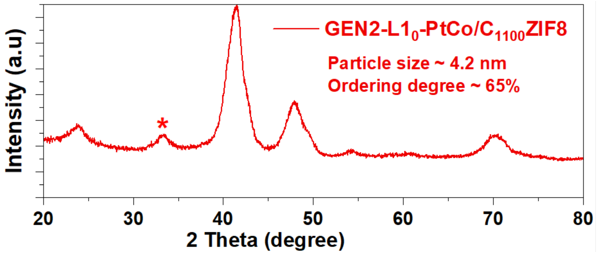


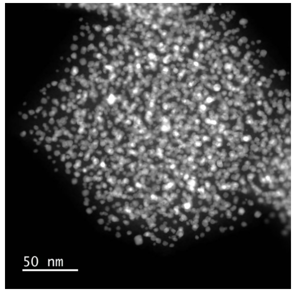
- Both HPN-Int-PtNiN/KB-1 (10h) and HPN-Int-PtNiN/KB-2 (5h) met the M2FCT catalyst target (1.07 > A/cm² @0.7 V after 90K catalyst AST cycles)
- BNL-HPN-PtNiN-1 had the higher performance of 1.19 A/cm² (longer annealing time)

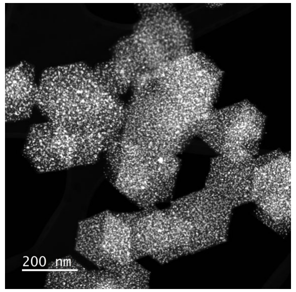
PtCo L1o Ordered Intermetallic on ZIF-Derived Carbon Support (GEN2-L10-PtCo/CZIF)

- L10-PtCo particles with high degree of ordering (e.g., 65%) deposited on high surface area carbon synthesized through carbonization of ZIF-8





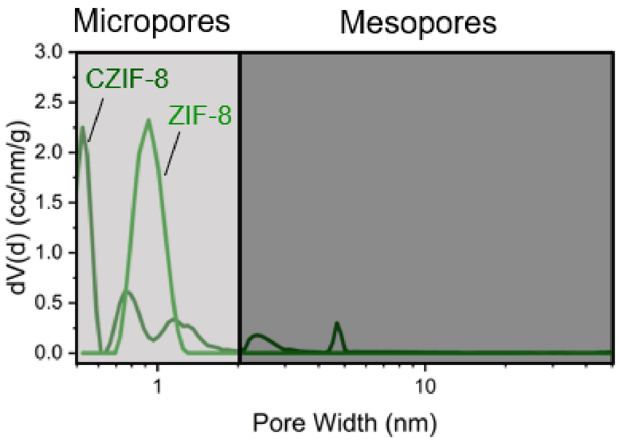


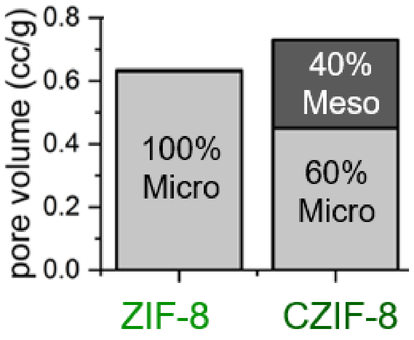


- GEN2-L10-PtCo/CZIF catalyst provides 3.3 kW/gPGM (1.43 A/cm²) @0.7 V after 90K catalyst AST cycles
- 90K performance @0.8 V of GEN2 catalyst is 17% higher than GEN1, 150% higher than M2FCT baseline

MEA test conditions: 250 kPa, 85% RH, 90ºC,. H_2/15% O_2, 5 cm² differential cell, NC700 membrane, A/C loading = 0.05/0.25 mg/cm²

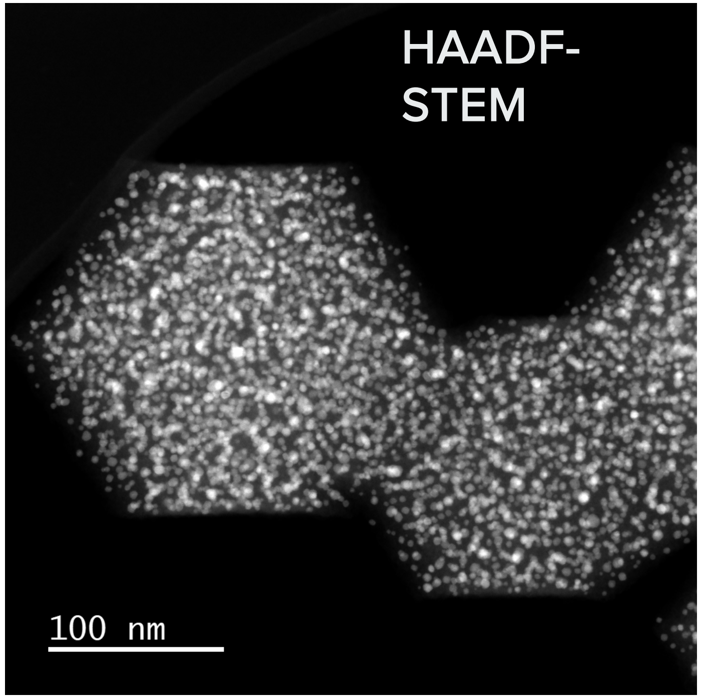
Continuing Development of New Supports

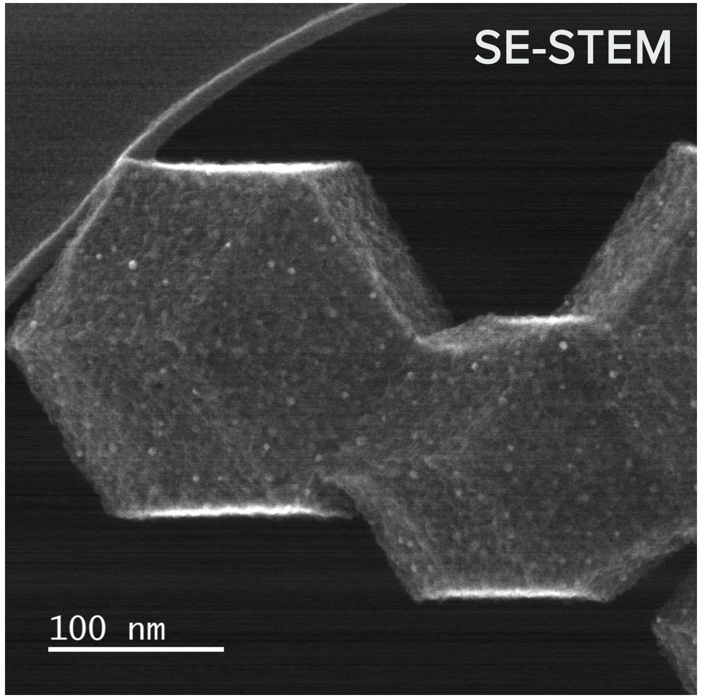


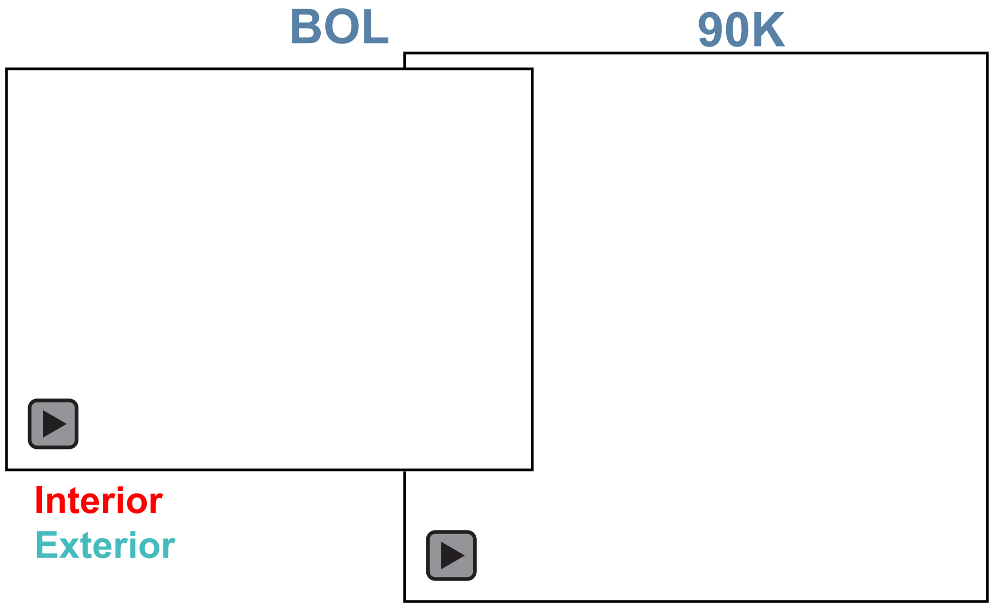


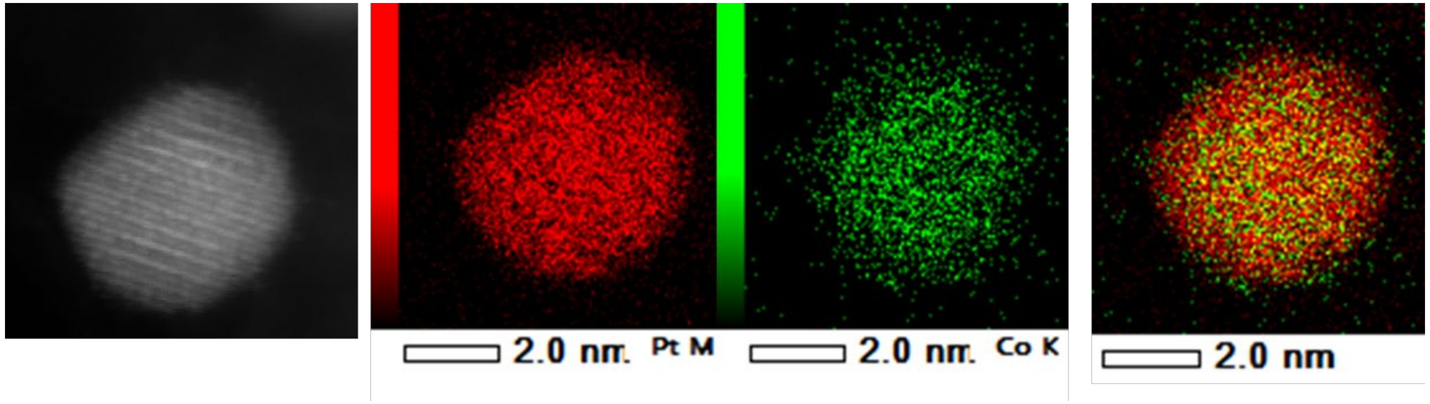
- CZIF-8 has excellent properties as ORR catalyst support:
  - Tunable balance of micropores and mesopores
  - Tunable particle size with narrow distribution
  - Abundant N doping for enhanced activity and durability

GEN2-L10-PtCo/CZIF8





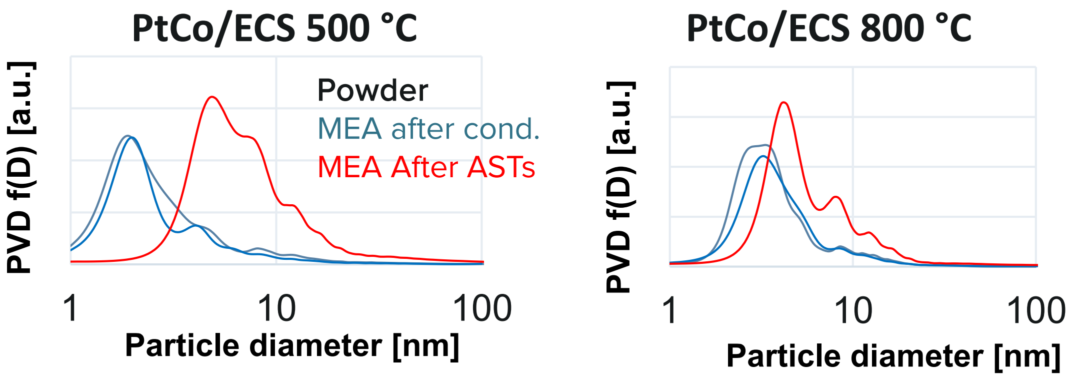


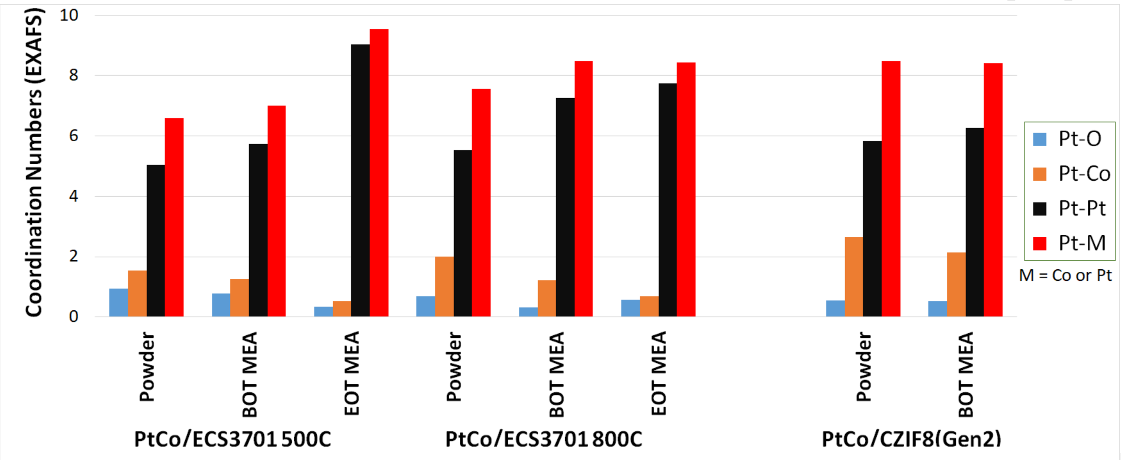


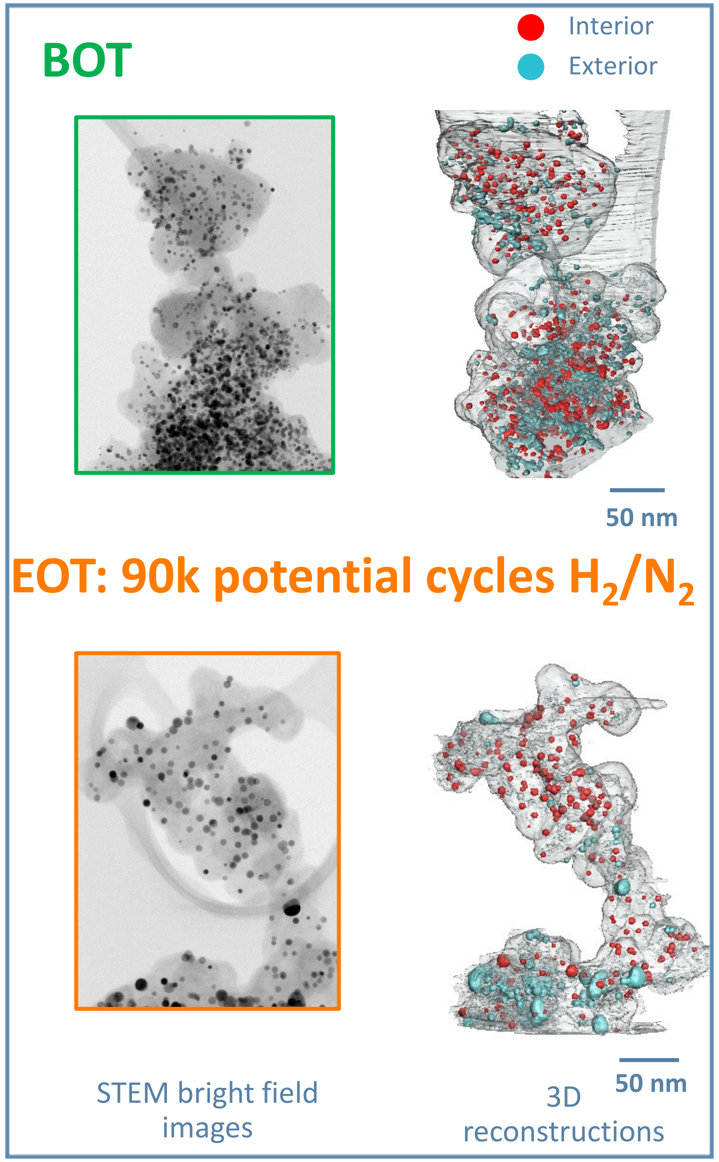
- >90% of PtCo particles located in CZIF8 interior – reduced degradation and reduced ionomer poisoning
- Uniform particle size distribution

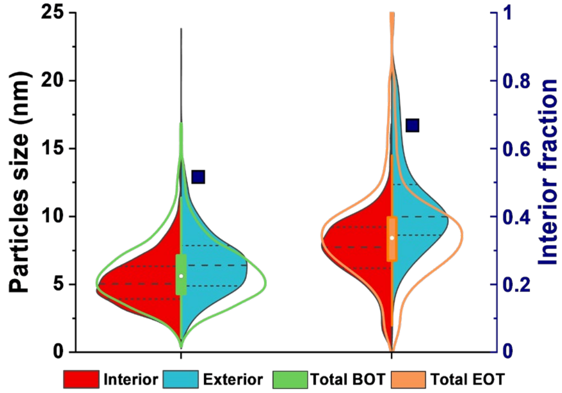
Advanced Catalyst Characterization

- X-ray characterization of M2FCT Intermetallic PtCo on ECS and ZIF-derived carbon
  - Increased annealing temperature enhances stability of Int-PtCo on ECS against particle growth
  - Int-PtCo on ZIF-8-derived C shows improved Co retention





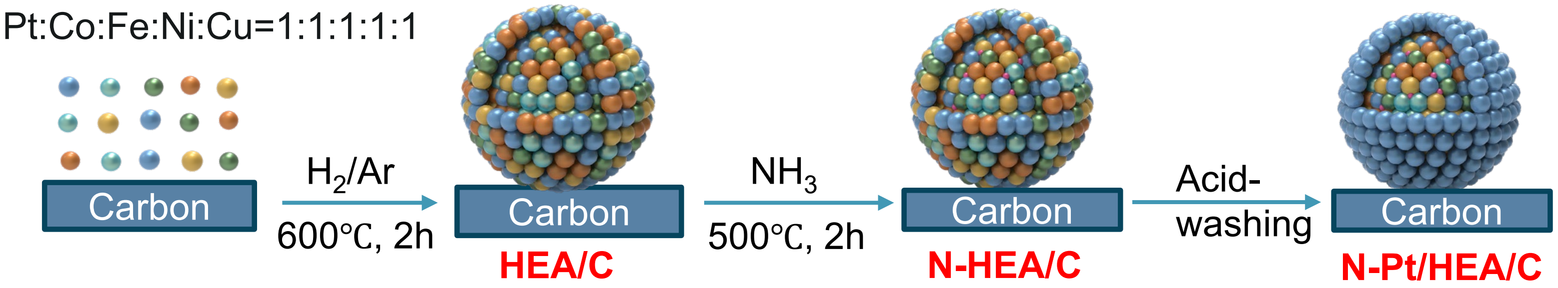




- Umicore Elyst Pt50 0550 : Higher interior fraction at BOT (53%) and EOT (63%).
- Particle coarsening affects both interior and exterior particles.
- Internal pores size distribution shift towards bigger pores

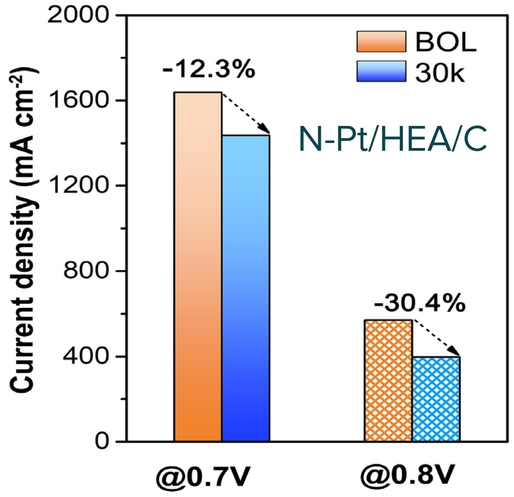
Continuing Development of New Catalysts and Supports

- High-Entropy Alloy (HEA) catalyst: PtCoNiFeCuN/C

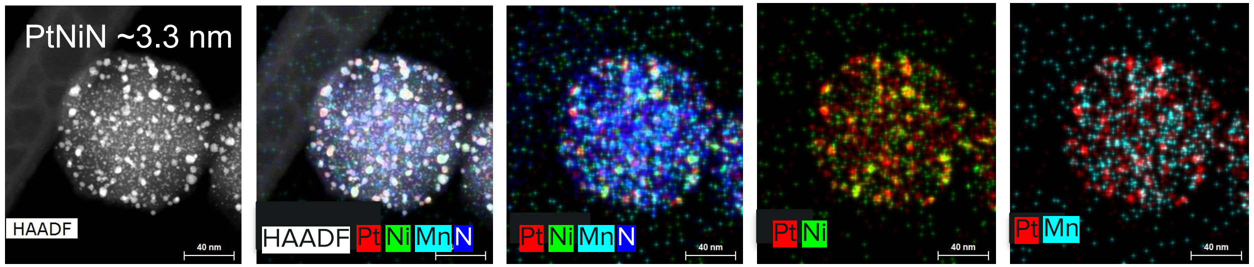


- **Distortion effect** (N dopants) : Improve activity
- **Pinning effect** (metal-nitrogen bonds) : Improve stability

Good MEA performance in IUPUI



- ZIF-based support for PtNiN: G. Wu (UB) provided ZIF-8-derived Mn-NC



RDE showed promising performance

Catalyst	MA@0.9V (A/mg)	ECSA (m^2/g)	SA@0.9V (mA/cm^2)
Int-PtNiN/Mn-NC	1.40	68.4	2.05

MEA testing is underway in UB

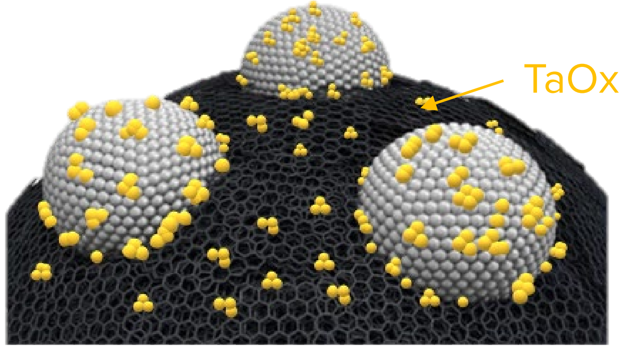
- Carbon with different pore size: pore size of 3.5, 5, 10, 30 nm (provided by N.E.Chemcat)
  - RDE showed good MA (1.22 A/mg) & high ECSA (157 m^2/g) for 3.5 nm-pore-carbon supported PtNiN
  - MEA testing is underway in LANL

X. Zhao, J. Xie, K. Sasaki, et al, *JACS,* **146** (2024) 3010-3022. DOI: 10.1021/jacs.3c08177

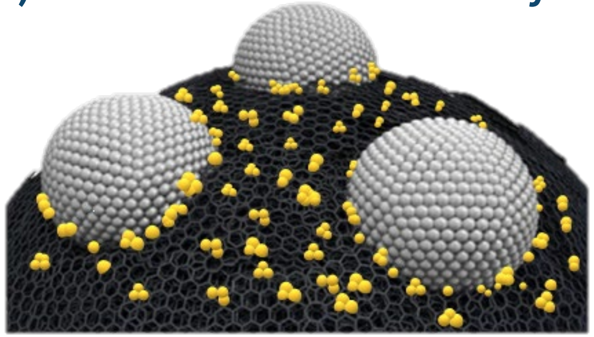
Tantalum oxide (TaOx) Modification of Pt/C

- Approach: Investigating electrochemical durability of Pt/C with MO_x nanoparticle incorporation to inhibit electrochemically-active surfaced area (ECSA) loss of Pt and PtCo

1) Inverse TaOx-Pt/C electrocatalyst

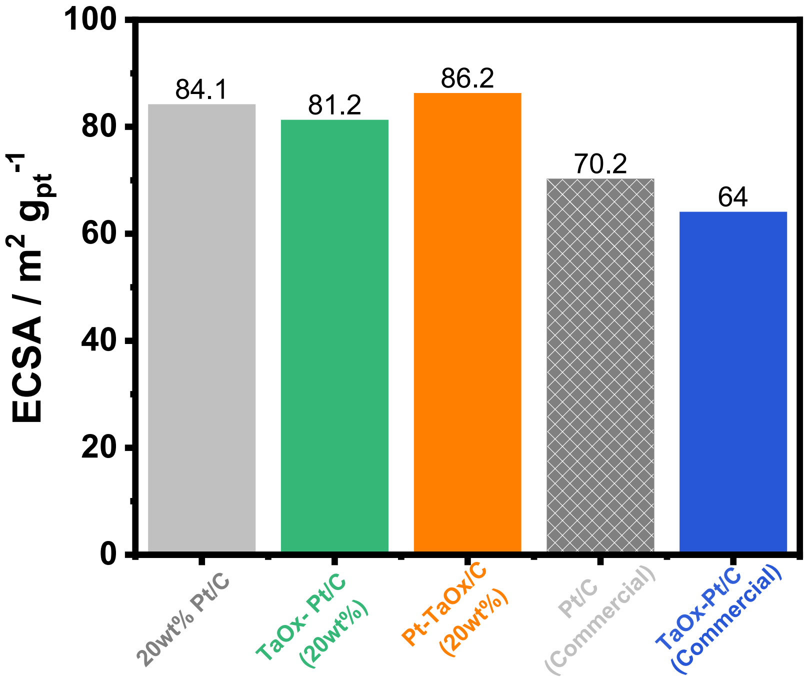


2) Pt-TaOx/C electrocatalyst

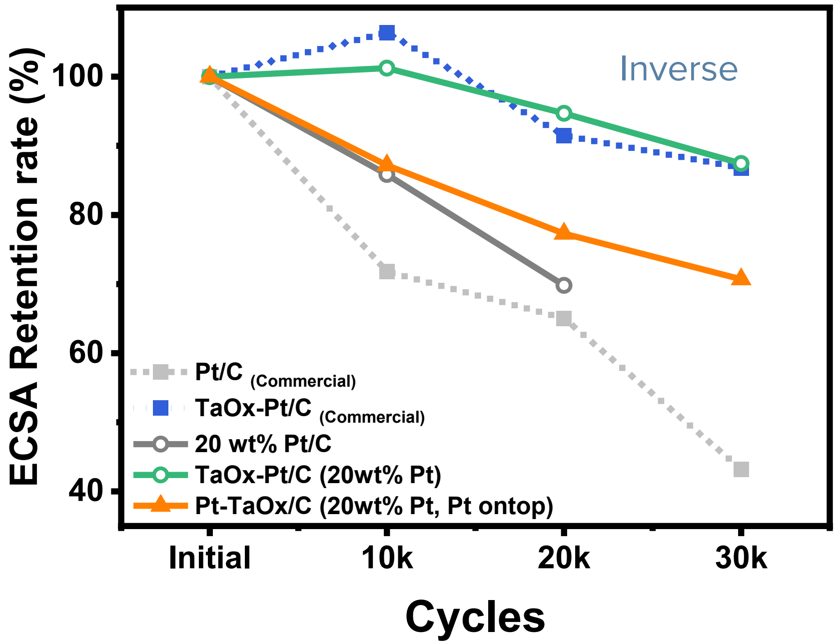


Figures adapted from [1]

Initial ECSA



Durability test



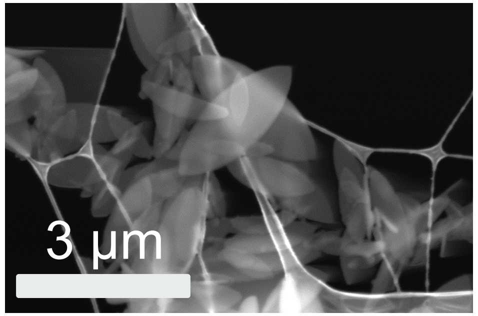
- Deposition sequence is the key parameter to affect ECSA and durability
  - Inverse TaOx-Pt/C slightly changes initial ECSA, but the durability (ESCA retention after stability test) is significantly improved

[1] Applied Catalysis B: Environmental 253 (2019): 187-195.

PtCo Ordered Intermetallic Catalyst Scaleup Efforts

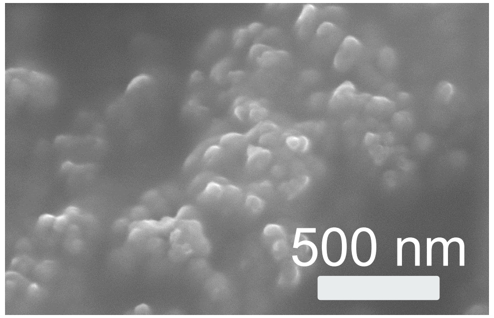
Q3 Milestone: Achieve current density of >1.3 A/cm^2 at 0.7 V and >0.45 A/cm^2 at 0.8 V after catalyst AST (total PGM loading 0.3 mg/cm^2, 250 kPa, 85% RH, H_2/15% O_2) with at least one 10 g batch of M2FCT-developed catalyst.*

Step 1: ZIF-8 Synthesis
Replace solvent-based ZIF-8 synthesis with ZIF-L to ZIF-8 transformation for large-scale ZIF-8 synthesis



ZIF-L (made at kg scale with water-based synthesis)

Ethanol wash at 60°C



ZIF-8

Step 2: Transforming ZIF-8 to CZIF-8
Replace tube furnace with rotary furnace for large-scale ZIF-8 carbonization

Step 3: Synthesis of PtCo/CZIF-8
Replace freeze-dryer with rotary evaporator for large-scale Pt and Co impregnation in CZIF-8

Step 4: Transformation to L1$_0$-PtCo/CZIF-8
Replace tube furnace with fluidized bed furnace or rotary furnace for atomic-level ordering of PtCo at >10 g scale

On track to deliver 10 g L1$_0$-PtCo/CZIF-8 batches to meet FY24 Q3 QPM

*Performance metric after AST chosen based on highest performing "small batch" catalysts shown in previous slide.

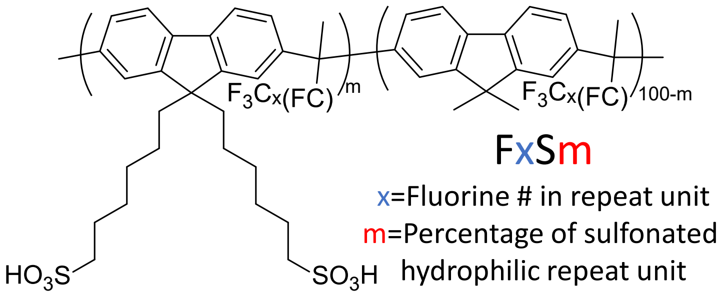
Nitrided Intermetallic PtNi Catalyst Scale-up Efforts

- The first BNL catalyst for scale-up is Int-PtNiN/C by atmospheric pressure nitriding (APN)
  - The scale-up synthesis is made in BNL. Use a large furnace having 3 heat-zones (60 cm) with a 3” O.D. quartz tube reactor for APN. The procedure is facile

  - Increment of scale-up quantity from 2 grams per batch
  - Optimize annealing temperature & cooling rate to enhance the intermetallic ordering in PtNiN
  - Selection of carbon support: The present carbon is KB EC-600J. We may also employ other carbon supports including ZIF-based carbon, mesoporous carbon with different pore size, etc.
- The scaled-up PtNiN catalyst will be delivered to M2FCT for evaluation in April to meet FY24 Q3 QPM

Hydrophobic Hydrocarbon Ionomers

Sulfonated polyfluorene ionomers

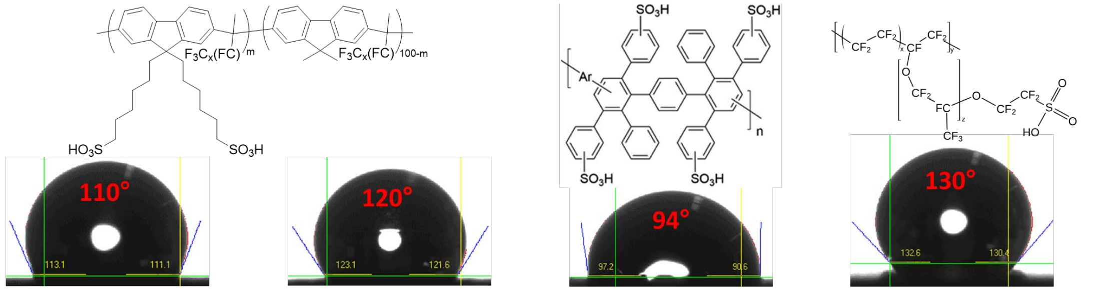


FxSm
x=Fluorine # in repeat unit
m=Percentage of sulfonated hydrophilic repeat unit

Ionomer	Titration IEC (meq g^{-1})	Molecular weight (kg mol^{-1}, M_n)	Water uptake (%)[a]	Conductivity (mS cm^{-1})[b]	
				Solution[c]	Membrane
F3S55	1.8	100	37	2.8	n/a
F5S40	1.4	17	19	2.5	n/a
F5S60	2.1	40	30	2.5	n/a
F7S45	1.6	12	19	2.3	n/a
F7S65	1.7	16	26	2.4	n/a
Recast Pemion[d]	3.8	n/a	72[e]	1.7	180 (320[e])

[a] Measured at 30 °C, 97% RH%. [b] Measured at 80 °C, 95% RH%. [c] Measured in 0.1 M dissolved in DMSO.
[d] Commercial Pemion dissolved in DMSO, recast to a 50 μm film. [e] *ChemSusChem* **2018**, *11*, 4033.

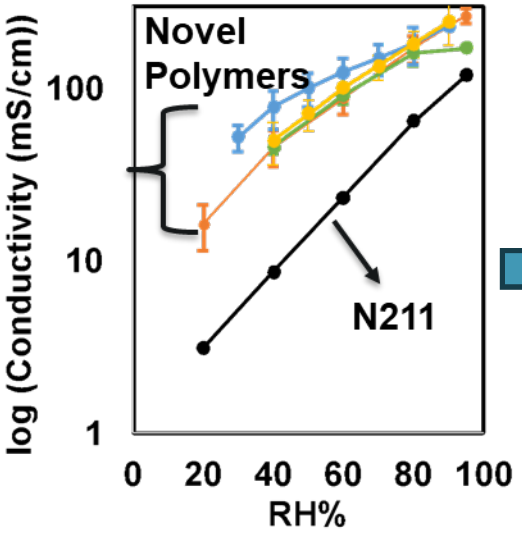
- Fluorene-based backbone with minimized interaction with electrocatalysts
- Oxidatively stable aryl ether-free structure
- Controlled ionomer hydrophobicity with a short fluoroalkyl chain (contact angle on GDL: up to 120° compared to 130° of Nafion)
- Good solubility in organic solvent/water for electrode fabrication (i.e., methanol:water 1:1)
- High proton conductivity (compared to recast Pemion™ as a standard)

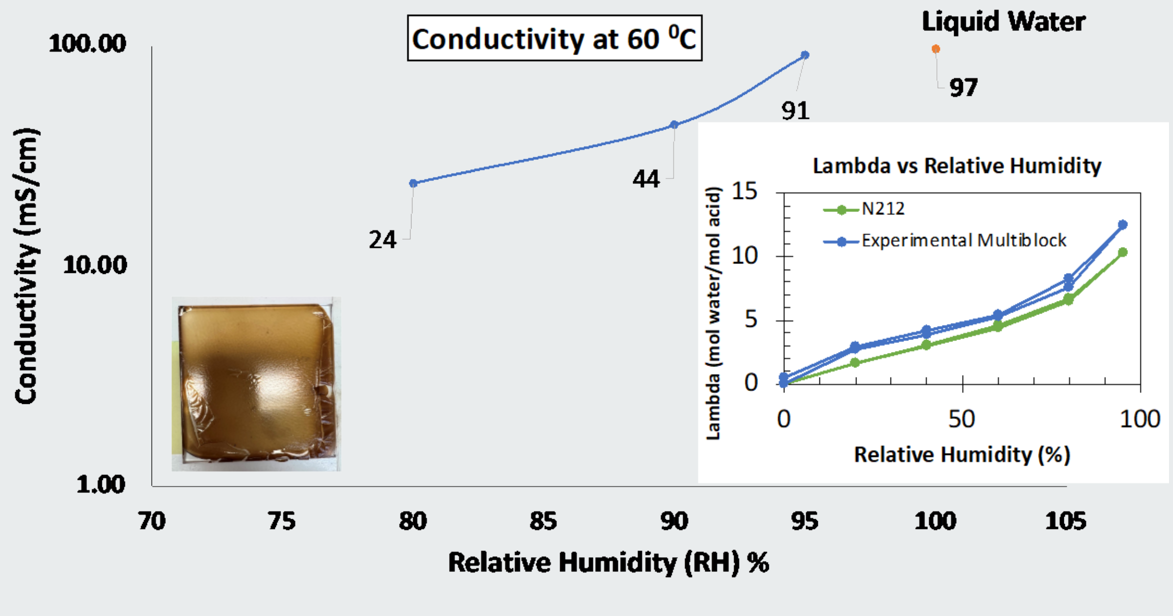


Ionomer	F3S55	F5S60	Pemion	Nafion D2020
IEC (meq/g)	2.0[a]	2.6[a]	3.8[b]	1.1[c]
F Percentage (%)	13	18	0	66
GDL	Avcarb MB-30	Avcarb MB-30	Avcarb MB-30	Avcarb MB-30
Ionomer Solvent	Methanol/DI water	Methanol/DI water	DMSO	NPA/DI Water
Catalyst	Pt/C	Pt/C	Pt/C	Pt/C

Novel Cation Conducting Polymer Electrolytes

Hydrocarbon and perfluoro or partially fluoro chemistry-based polymers





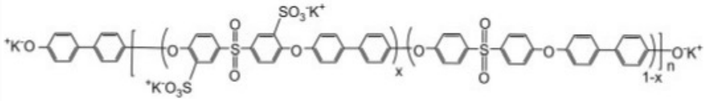
Multiblock of hydrocarbon-perfluoro material

- Earlier work showed novel polymers with significantly higher IEC and conductivity (5-10X improvement) than Nafion, but suffered from poor water stability

Hypothesis-improving mechanical properties and limit water swelling/dissolution through multiblock copolymer synthesis

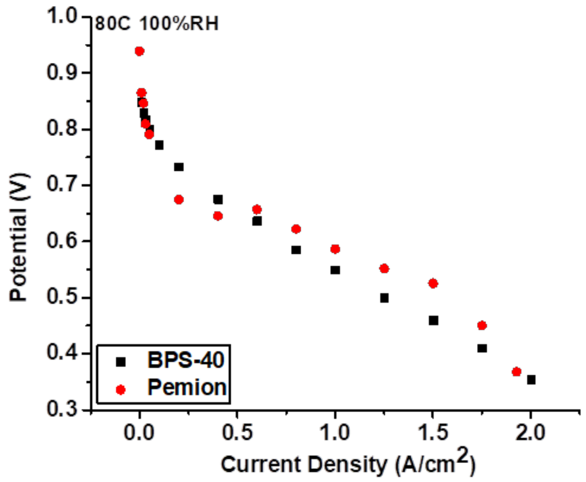
- Mechanical stability and water stability improved through multiblock synthesis between a perfluorinated oligomer and hydrocarbon-based oligomer and good conductivity obtained
- Increasing molecular weight of the oligomers seem to improve membrane formation, however, still one of the major challenges is fabricating tough, ductile films

Hydrocarbon based ionomers



Disulfonated biphenol based poly(arylene ether sulfone)

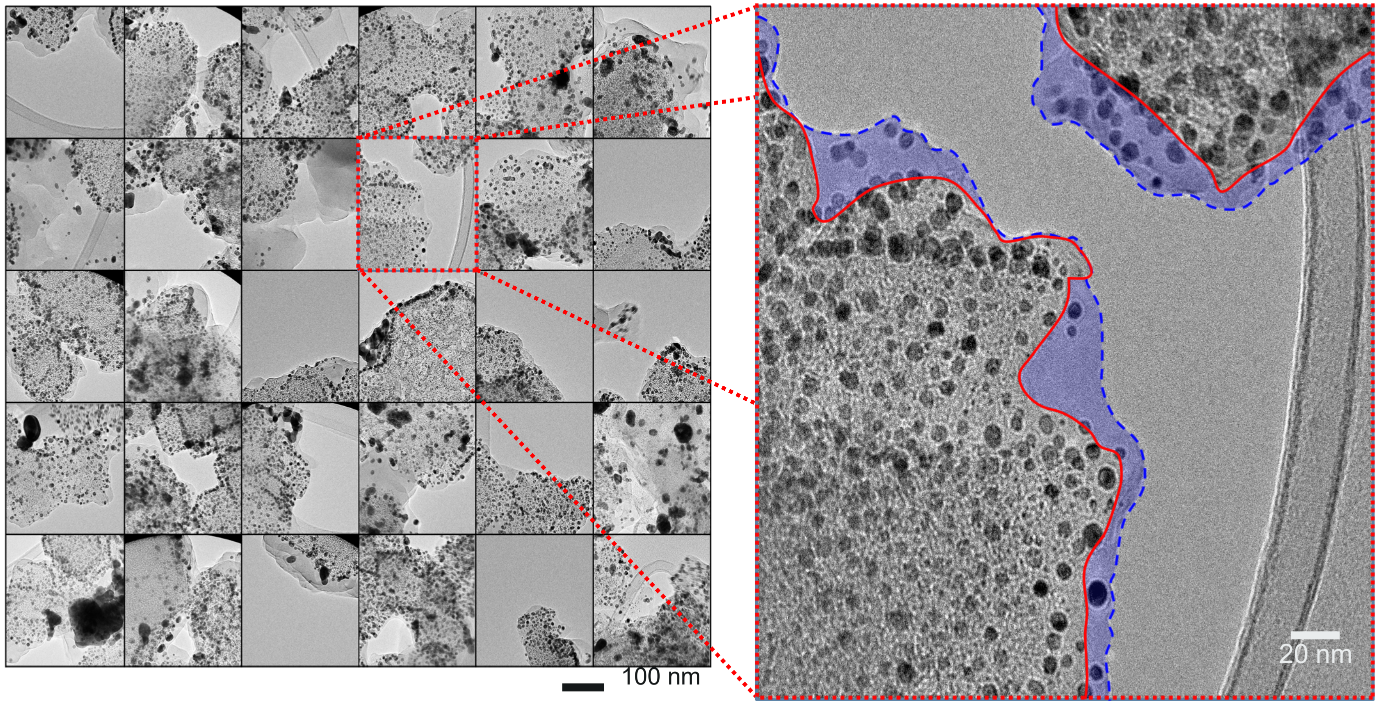
BPSH-XX where X= degree of disulfonation



- Performance of 40% sulfonated ionomer matched commercial hydrocarbon (PEMION) ionomer
- 'Green integration'- Electrode fabrication with alcohol/water system has been accomplished

Advanced Characterization

- Automated ionomer imaging across electrodes developed for spatially resolved information
  - Low electron dose to image near native state of ionomer
  - Pairing with data analysis tools will allow rapid statistical analysis
  - Demonstrated applicability to Pt/C electrodes by automatically acquiring a series of low-dose cryo-TEM images across an MEA

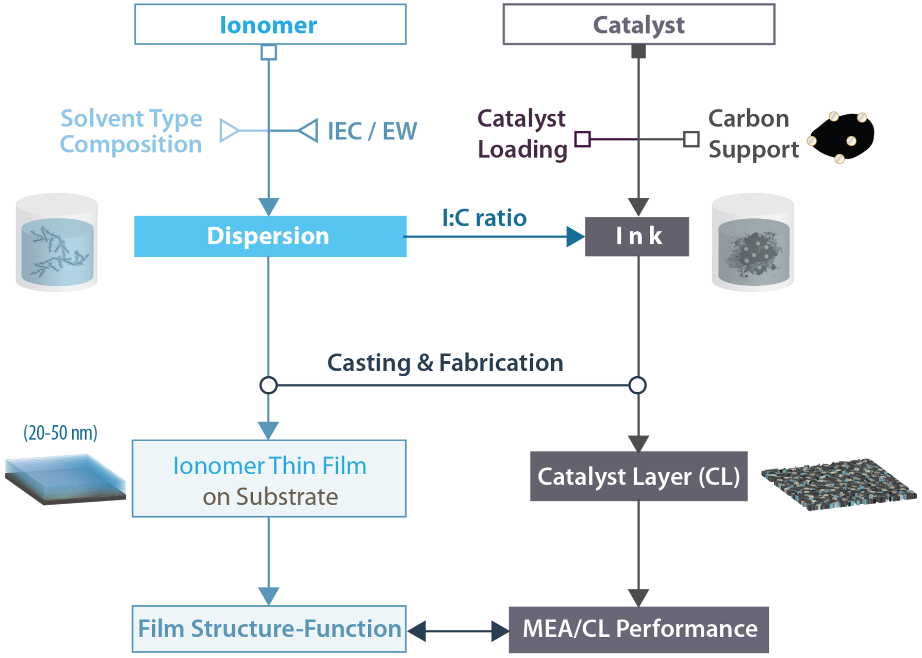


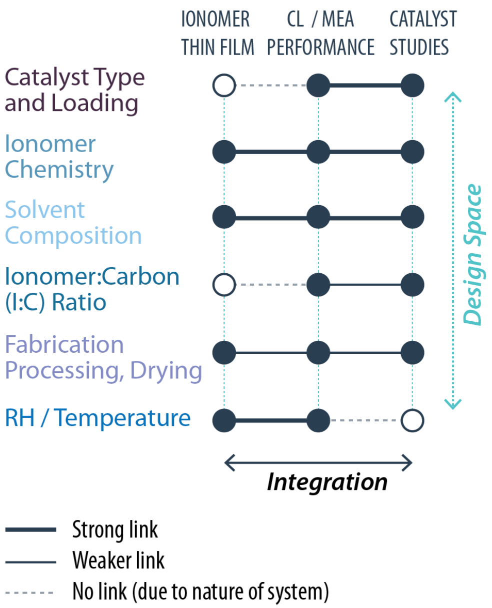
Integration

Ionomer-Ink-CL Integration: Connecting Design Variables

M2FCT employs an approach that bridges the properties of catalyst properties, thin film function and CL performance

- A systematic investigation of ionomer function, catalyst structure and properties, and CL properties & performance
- Goal is to identify the role of key variables (chemistry, I:C ratio, solvent composition) on measured properties to tie material function to the CL performance
  - Understand and baseline metadata needs
- The experimental efforts are supplemented by theory and modeling of inks and CLs

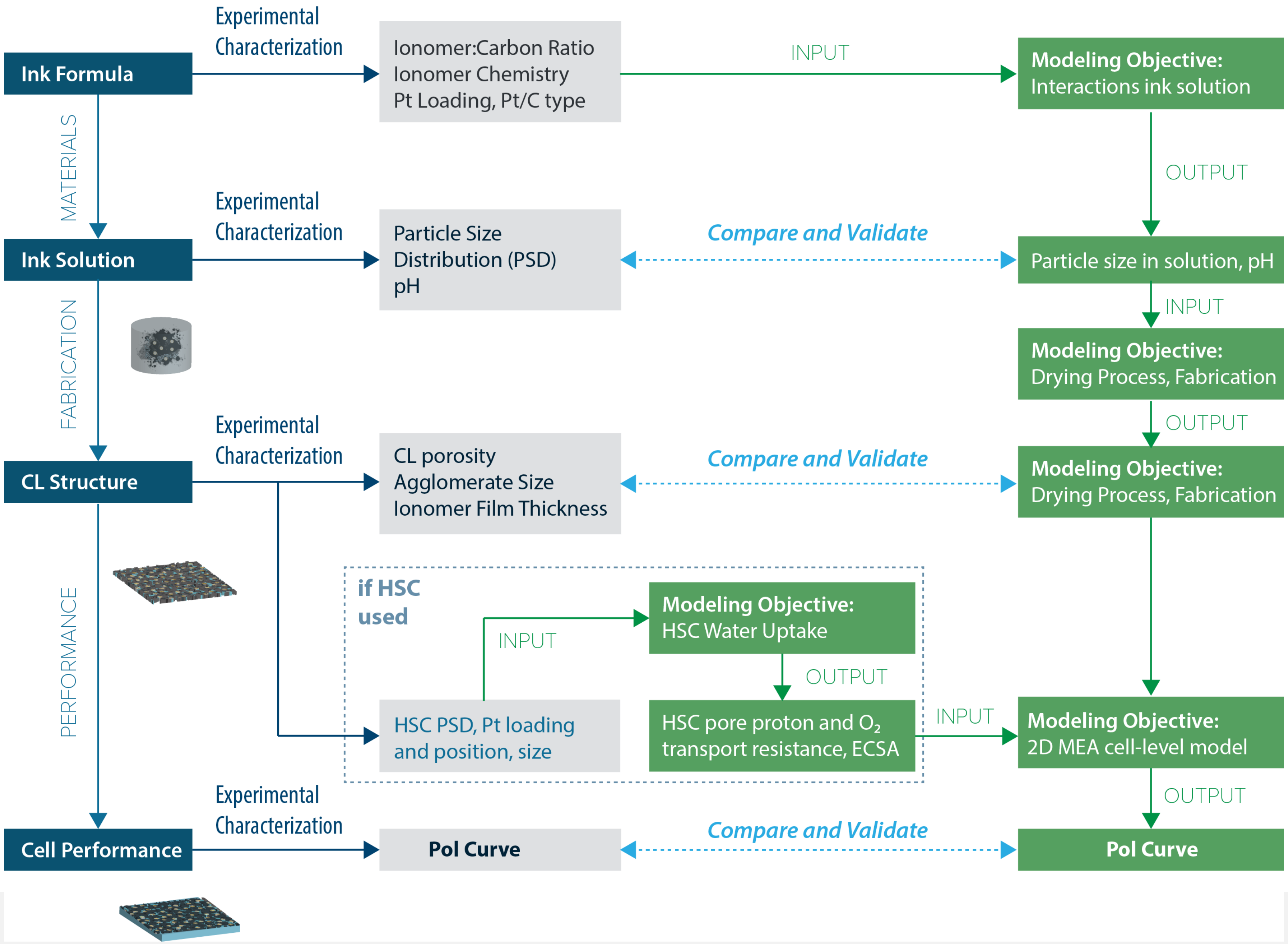




Material System		nPA:W	Catalyst (ANL)	MEA (NREL) | PtVu			MEA (NREL) | Pt 0550			MEA (NREL) | PtCo 0670			Thin Films (LBL)	Thin Films (LBL)
Component	Material		Pt 690 / 670	I:C	I:C	I:C	I:C	I:C	I:C	I:C	I:C	I:C	on Pt, on Si	on Pt, on Si
			PtCo 690 / 670	0.3	0.4	0.6	0.3	0.4	0.6	0.4	0.6	0.9	20, 50, 100 nm	20, 50, 100 nm
Catalyst	PtCo Um 0670 30 wt%													
Ionomer	HOPI (5006)	5:1	USAXS / WAXS			NREL	NREL		NREL				LBNL	GIXS, QCM, SE
Ionomer	HOPI (5006)	7:3	USAXS / WAXS	NREL		NREL			NREL	NREL	NREL	NREL	LBNL	GIXS, QCM, SE
Ionomer	HOPI (5006)	3:7	USAXS / WAXS	NREL		NREL						NREL	LBNL	GIXS, QCM, SE
Ionomer	D2020 (920 EW)	5:1	USAXS / WAXS		NREL	NREL							LBNL	GIXS, QCM, SE
Ionomer	D2020 (920 EW)	7:3		NREL									LBNL	GIXS, QCM, SE
Ionomer	D2020 (920 EW)	4:6	USAXS / WAXS		NREL	NREL								
Ionomer	D2020 (920 EW)	3:7	USAXS / WAXS	NREL		NREL	NREL	NREL	NREL	NREL	NREL	NREL	LBNL	GIXS, QCM, SE

Ionomer-Ink-CL Integration: Experiments and Modeling

M2FCT employs an approach that bridges the properties of catalyst properties, thin film function and CL performance



Nafion Adsorption in Catalyst Inks

Effects of Nafion Adsorption

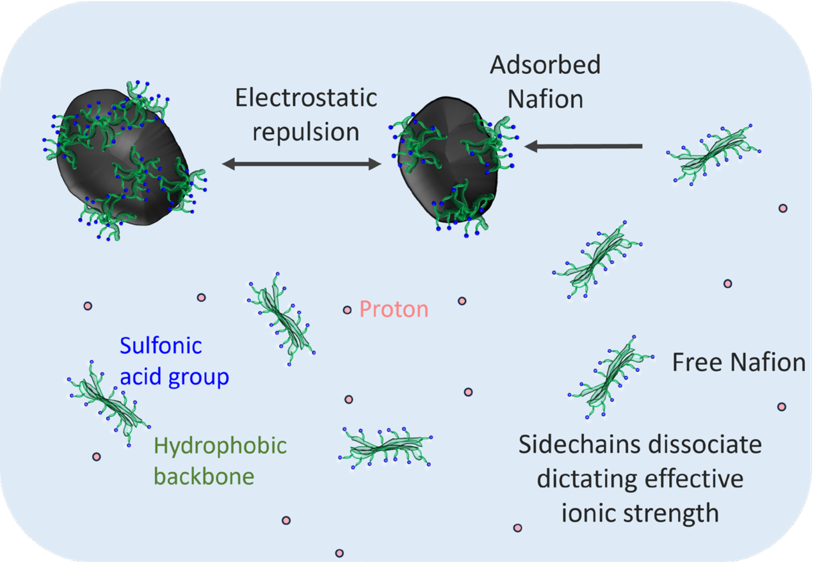
- Nafion adsorption affects
  - Ionomer distribution in CL
  - Interactions and aggregation in inks

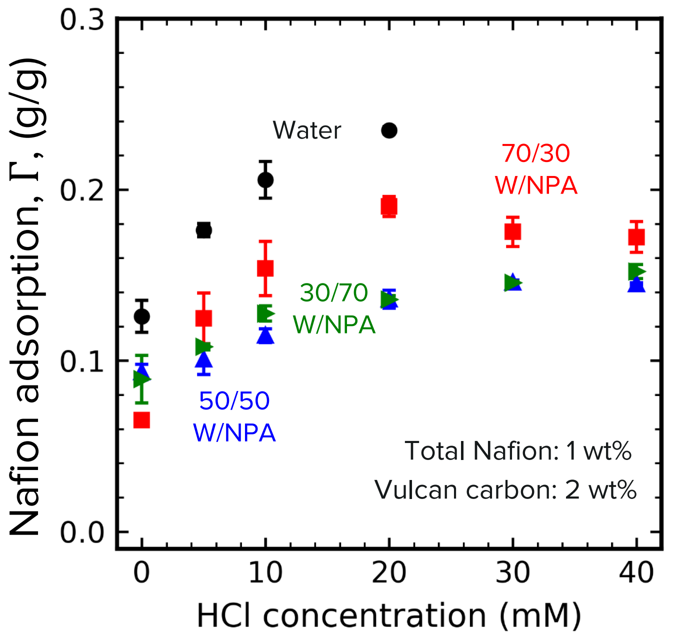
Solvent Dependent Adsorption

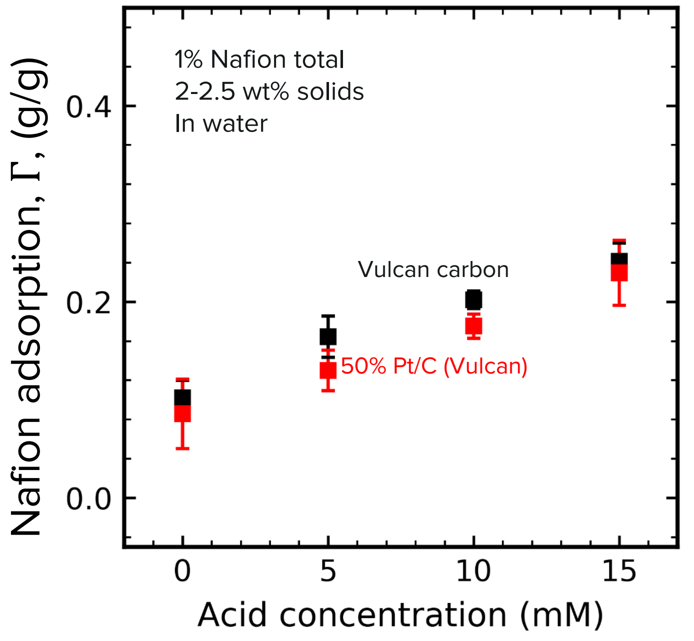
- Adsorption limited by electrostatic repulsion by charged Nafion sidechains
- Adding acid to inks reduces repulsion and increases adsorption
- Higher water content inks adsorb more Nafion

Particle Dependent Adsorption

- Nafion adsorbs to a similar extent on Vulcan carbon and 50 wt% Pt/C (Vulcan)
- Suggests interactions between Nafion-Vulcan carbon similar to Nafion-Pt



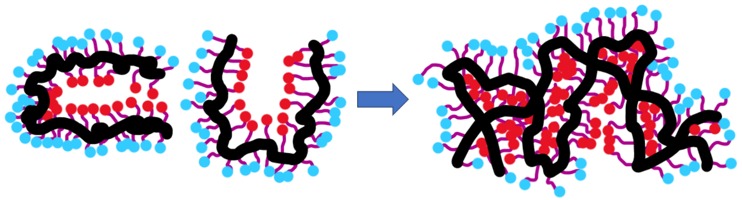




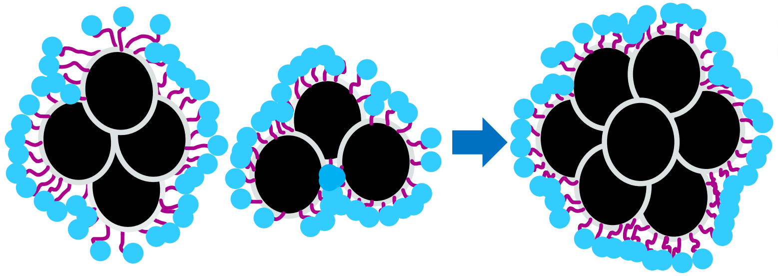
Modeling the Catalyst Layer Structure and Aggregation

Catalyst Layer inks are modeled using aggregation theory and verified experimentally

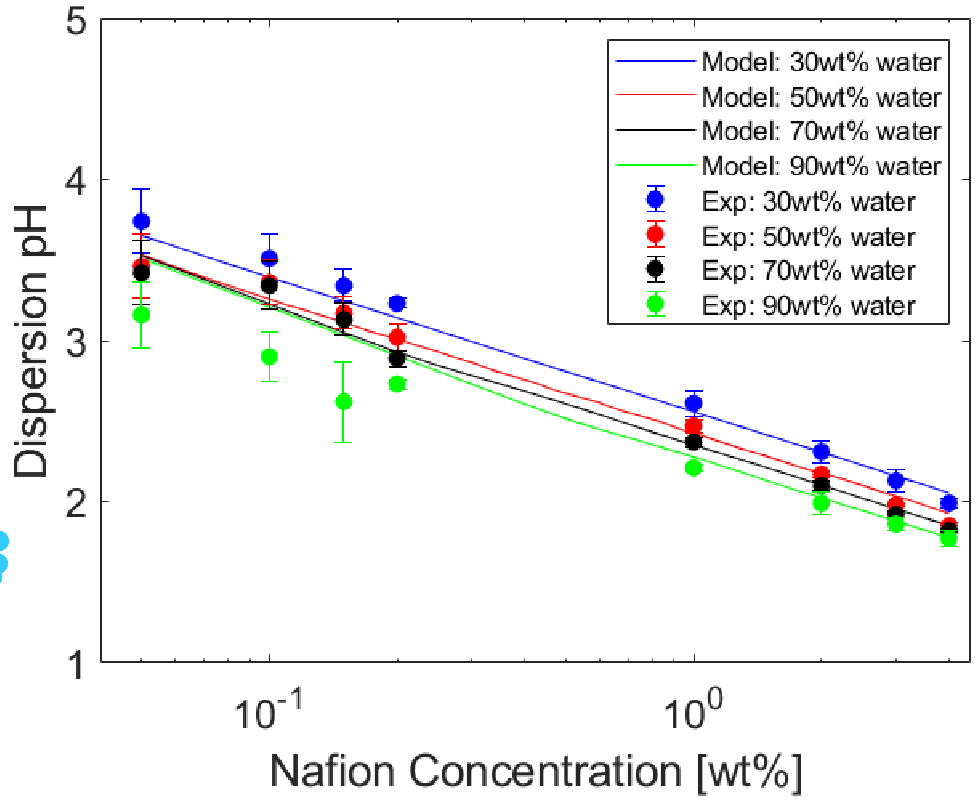
Nafion aggregation (buried charges red)



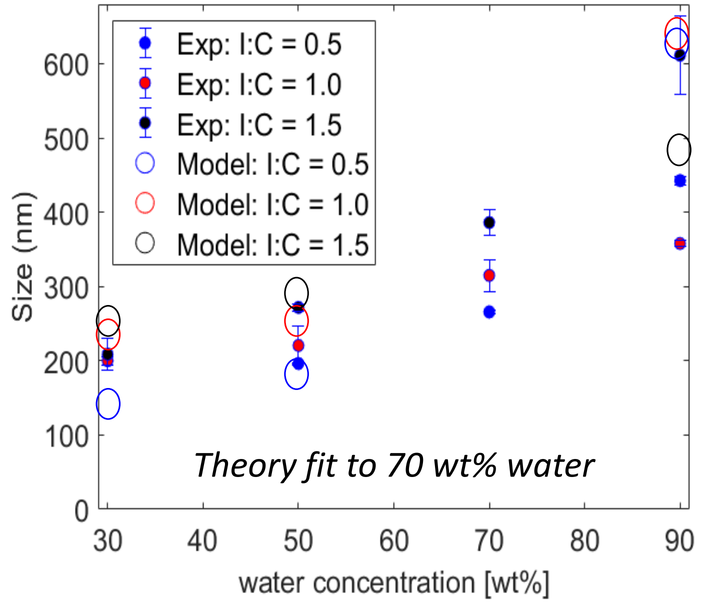
Carbon aggregation



Aggregation leads to further burying of charges on side chains



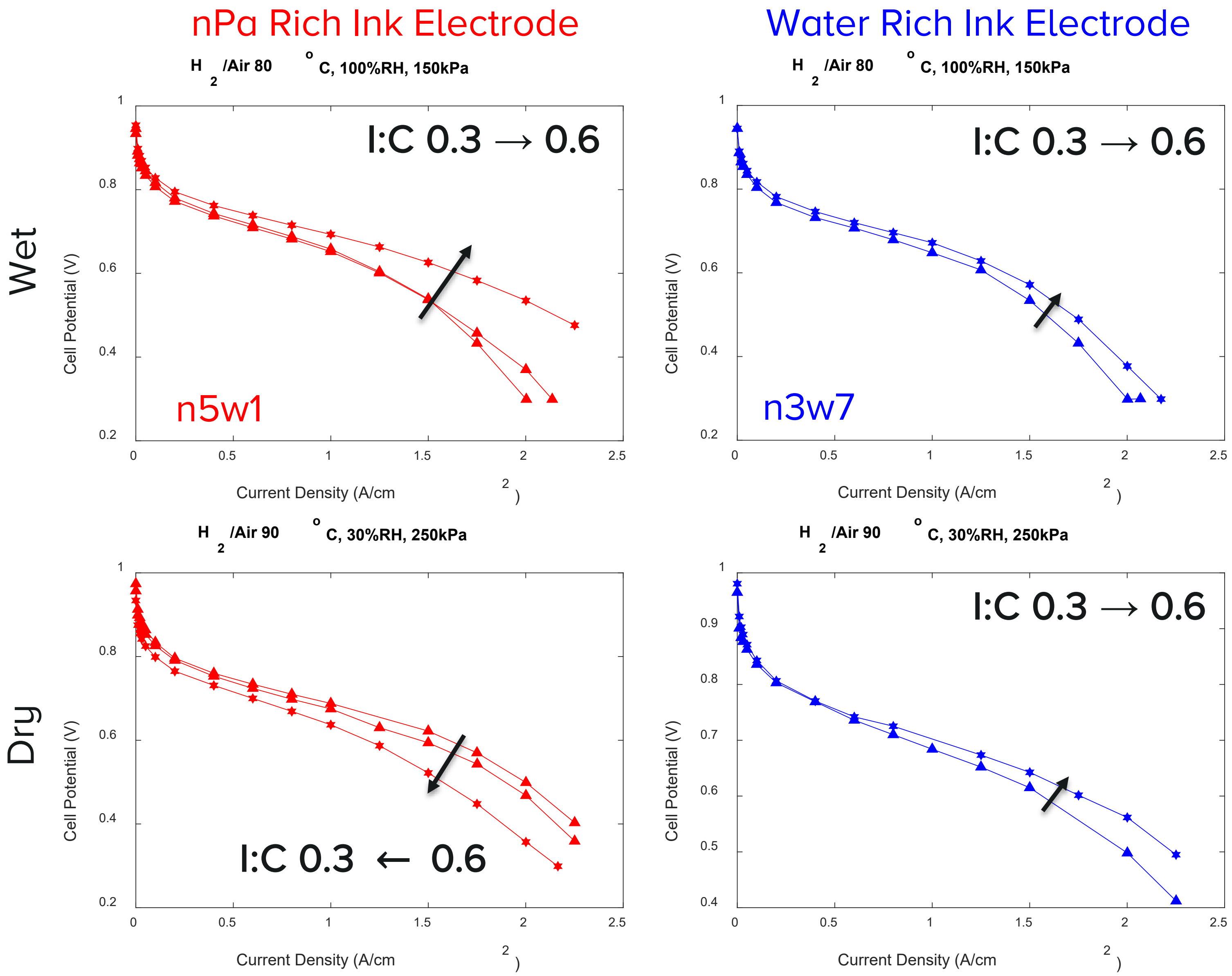
For Nafion dispersions, pH (from dissociated protons) is compared against experimental results (measured with pH probe)[1]



For Nafion + Carbon inks, DLS (Dynamic Light Scattering) is compared against reference[1]

1. Berlinger, S. A., et al. Inherent Acidity of Perfluorosulfonic Acid Ionomer Dispersions and Implications for Ink Aggregation. J. Phys. Chem. B 2018, 122 (31), 7790−7796.

I:C Effects on nPA- and H_2O-rich Pt/Vu Electrodes



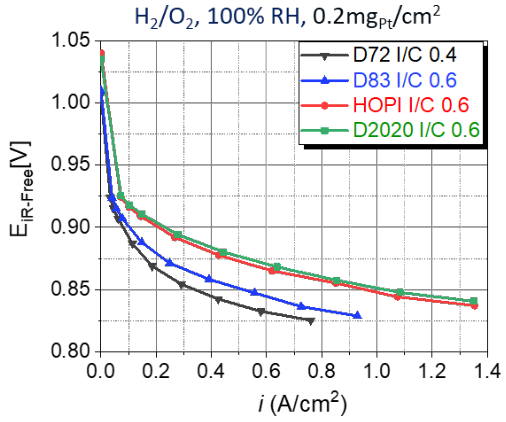
D2020-Pt/Vu Interactions lead to differences

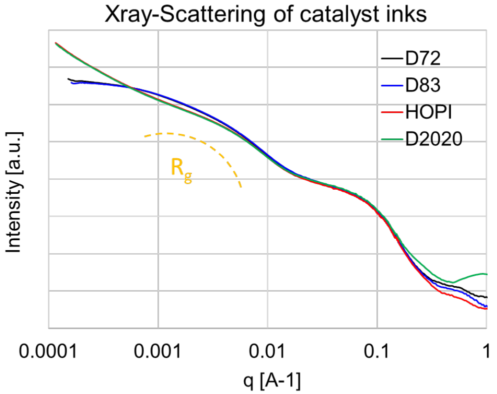
- Water-rich electrodes show smaller I:C effect compared to nPA-rich electrodes
- Under wet conditions where electrode is less dependent on ionomer for H+ conduction, both ink recipes show best activity w/ D2020 I:C 0.6
- When dry, more D2020 helps n3w7 electrodes, but hinder n5w1 performance
- EIS and coverage measures suggest more C coverage and ionomer aggregation for n5w1

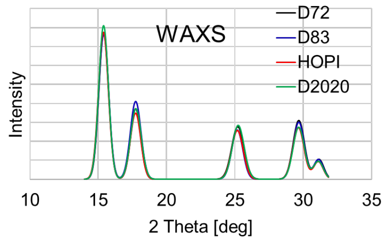
X-ray Scattering Characterization for the MEA Integration Study

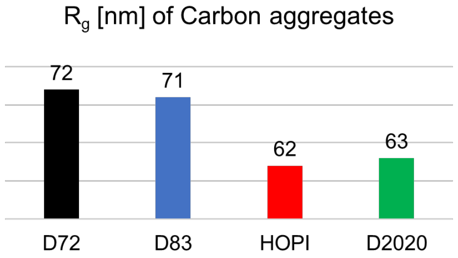
Agglomerate/aggregate size of various ink compositions has been correlated with the MEA integration data using USAXS-SAXS-WAXS at the Advanced Photon Source

PtCo/HSC (Umicore 0670)



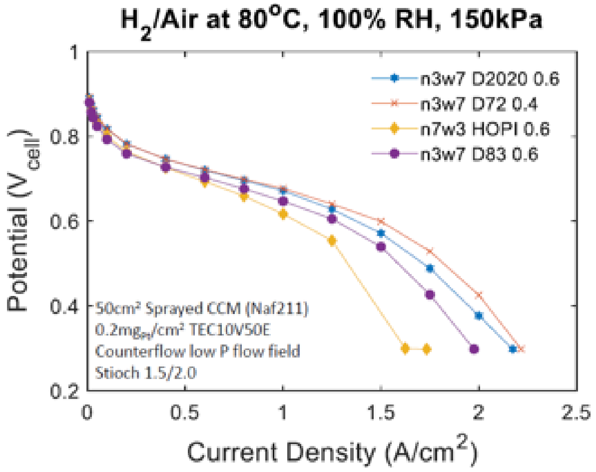


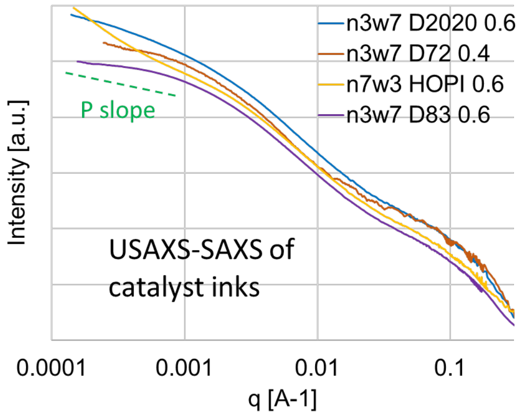




- D72 and D83 show poor ORR kinetic due to reduced Pt utilization from relatively bigger carbon aggregates
- ECSA and WAXS were unaffected by ionomer type

Pt/Vulcan

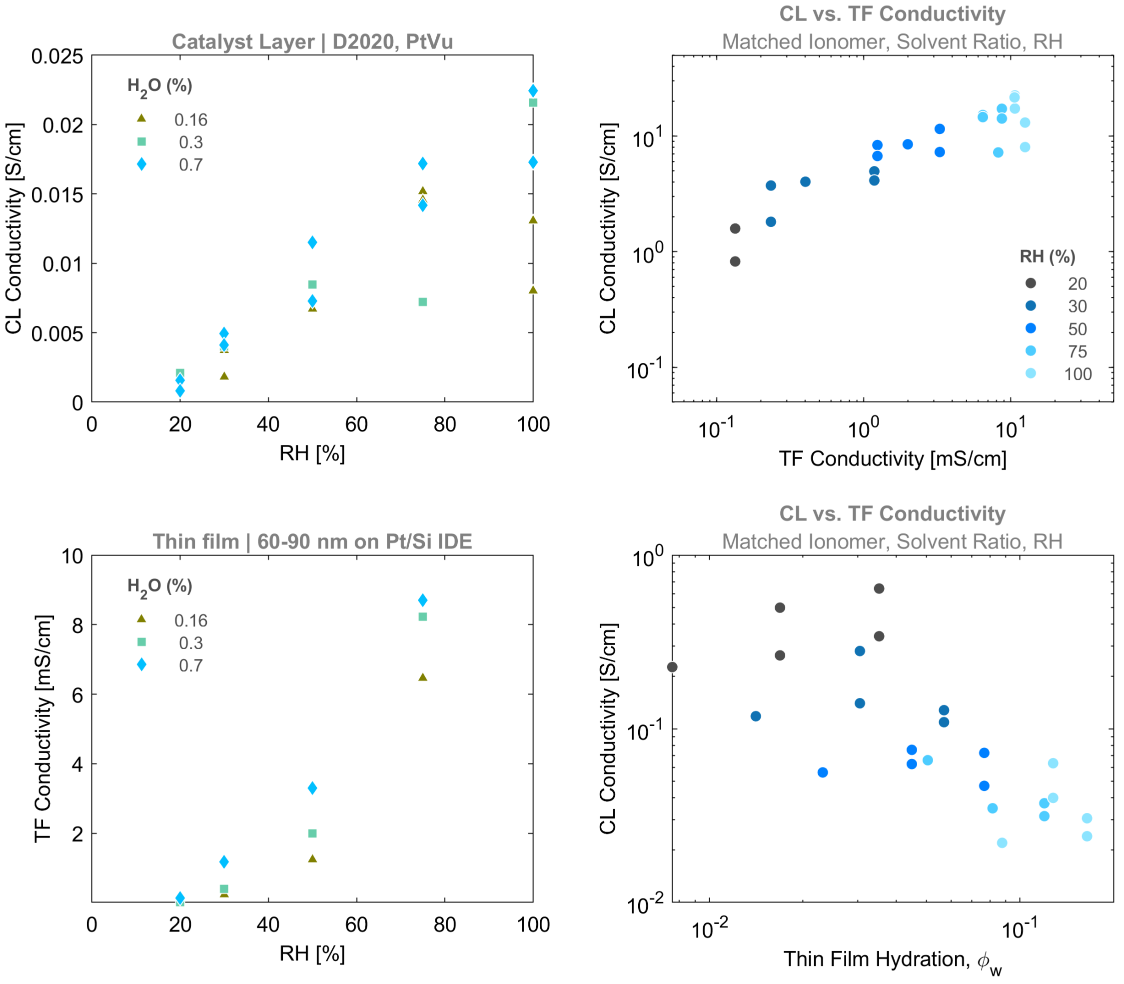




- HOPI shows the lowest performance among other ionomers, unlike HSC inks
- The slope (P) is related to the fractal dimension of the scattering structures, which is typical of carbon clusters → more agglomeration
  - P slope: HOPI>>>D2020, D72, D83

Linking Catalyst Layer and Thin Film Conductivity

- Both CL and TF conductivity increases with RH (as expected)
- There is an effect of solvent comparison: water-rich ionomers lead to higher conductivity
- CL conductivity and TF conductivity correlates fairly well
  - Thin Film hydration is a good proxy
  - First time CL and TF properties are correlated systematically



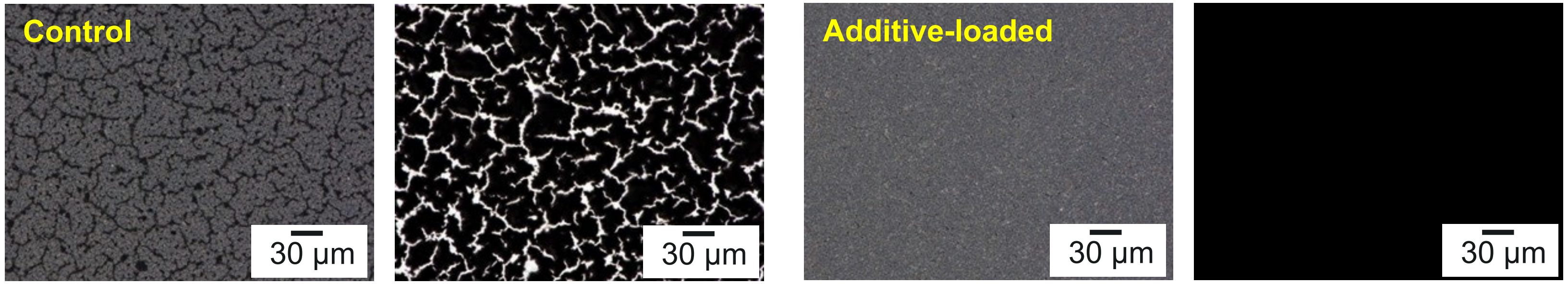
Correlations for D2020 and PtVu based electrodes

Scalable Fabrication of Defect–free Direct Membrane Coatings (DMCs)

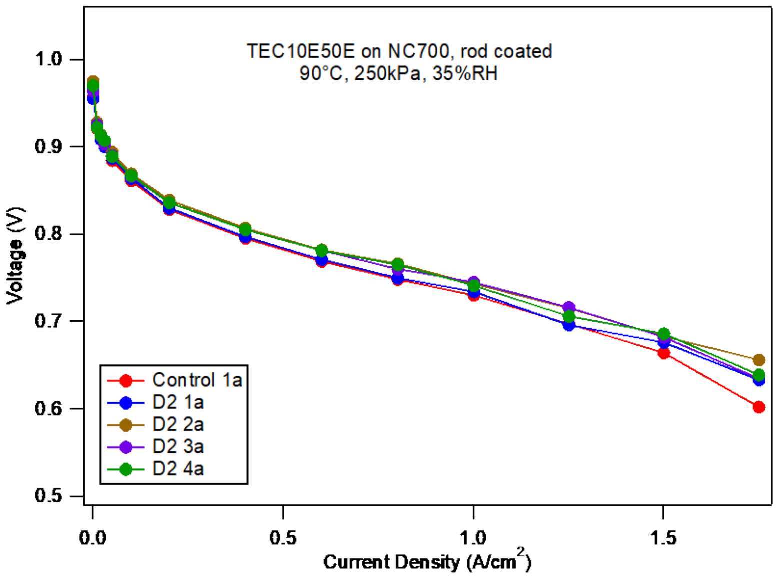
Catalyst	Ionomer	Pt Loading
Pt/HSC	Nafion	~0.2 mg/cm^2

Accomplishment: Manufacturing of DMCs without catalyst layer or membrane swelling.

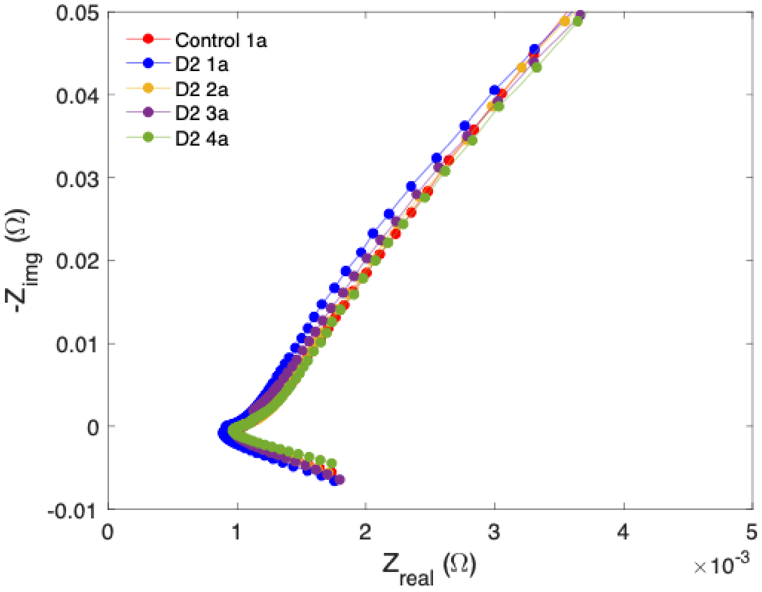
Achieved mitigation of scale-up defects through polymer additives.



90 °C, 250 kPa, 35% RH

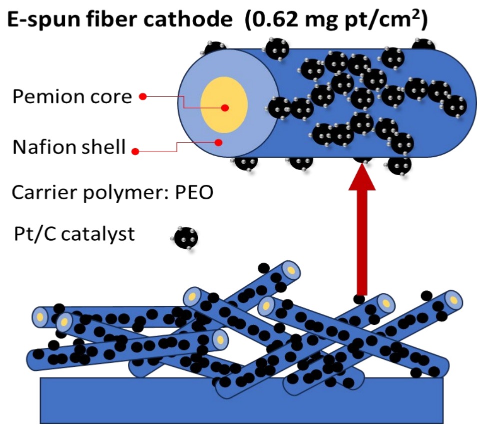


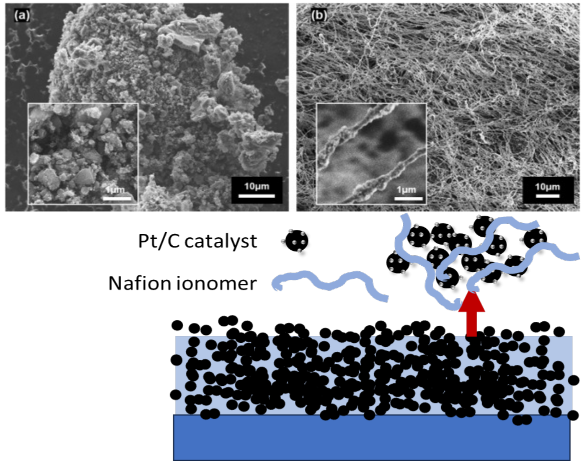
EIS 100% RH



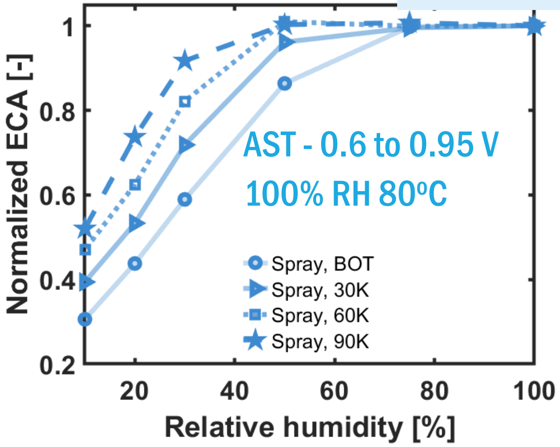
Ink formulation or fabrication procedure do not pose a risk to BOL performance

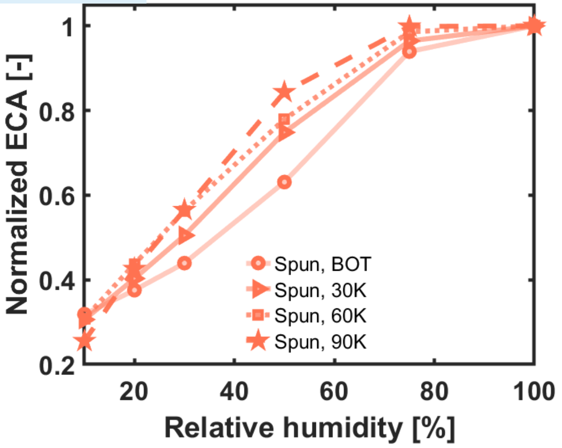
Modifying interfaces through electrospinning

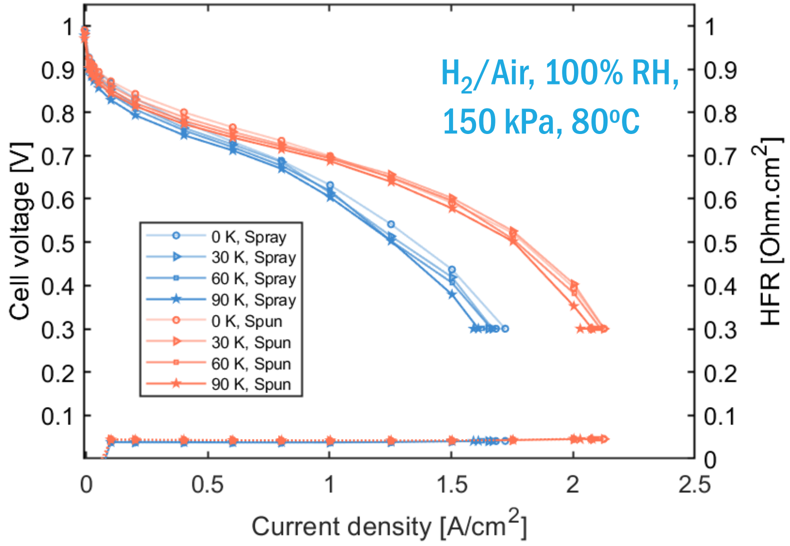


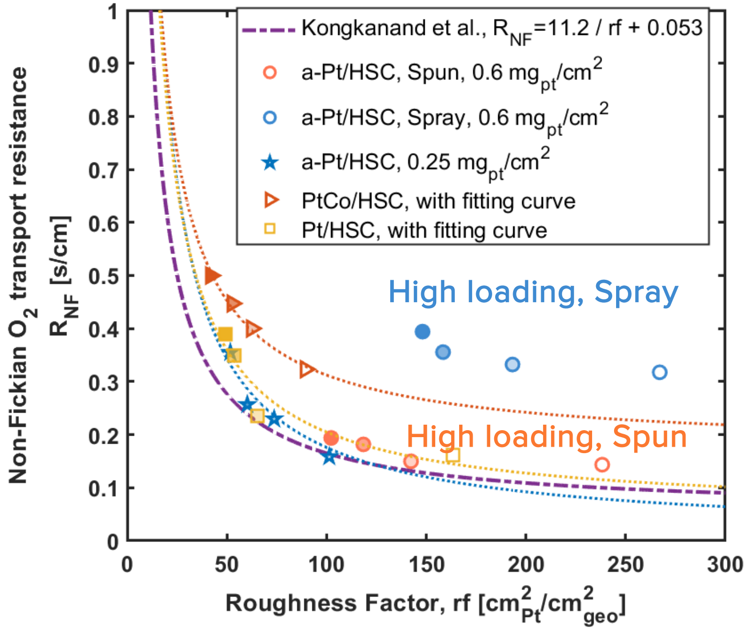


Pt accessibility



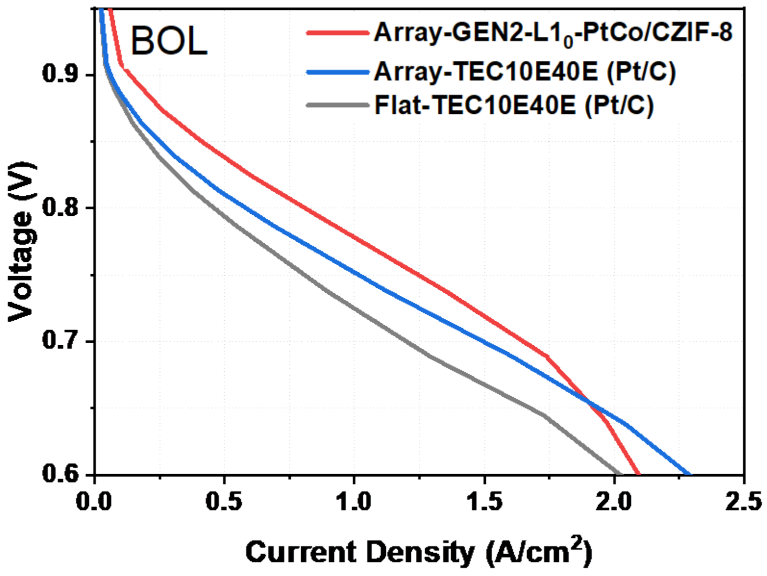


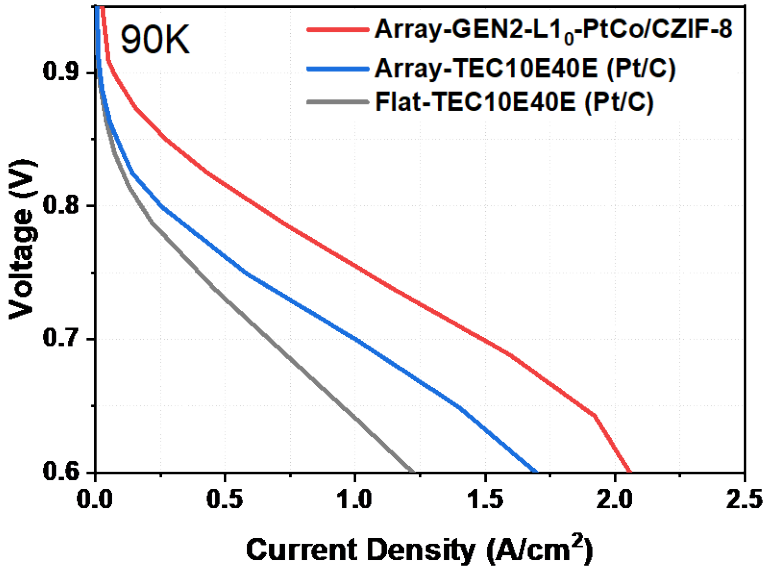


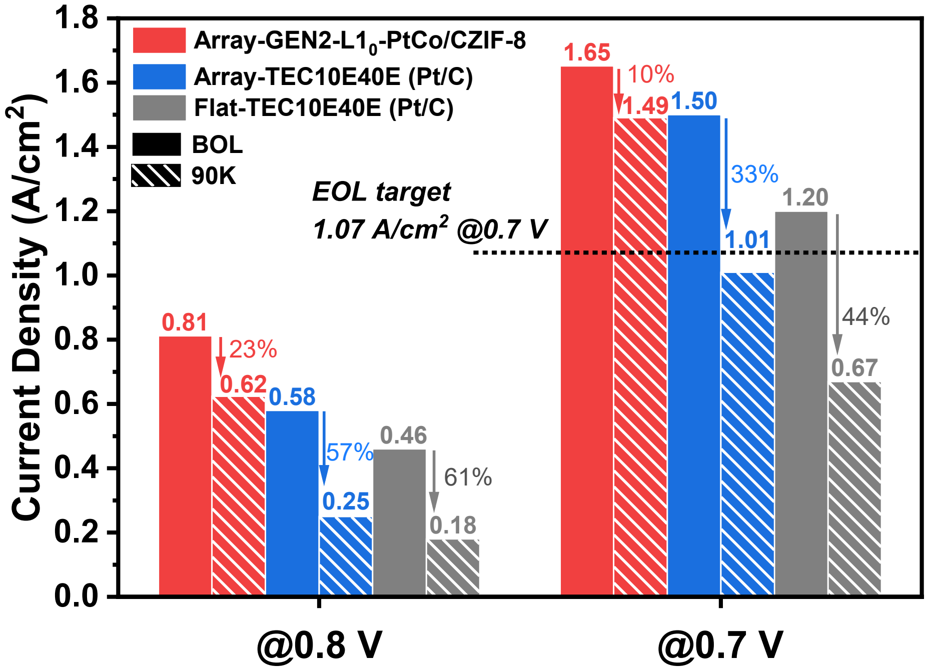


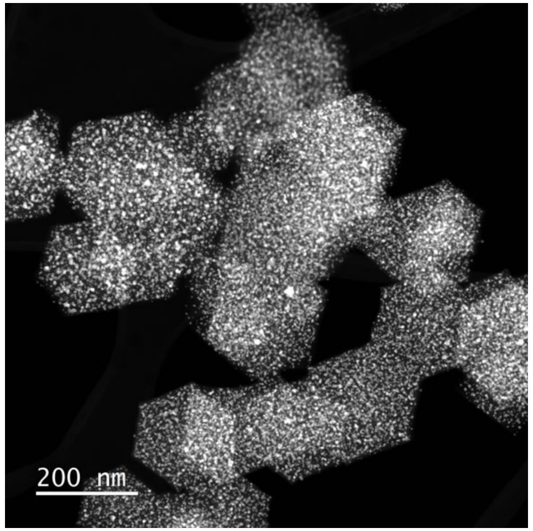
- Electrospun fibers yield no change in Pt accessibility over catalyst AST
  - Locked in ionomer/electrocatalyst interface
- Improved high current density transport due to lower non-Fickian resistance
  - Improved macroporosity

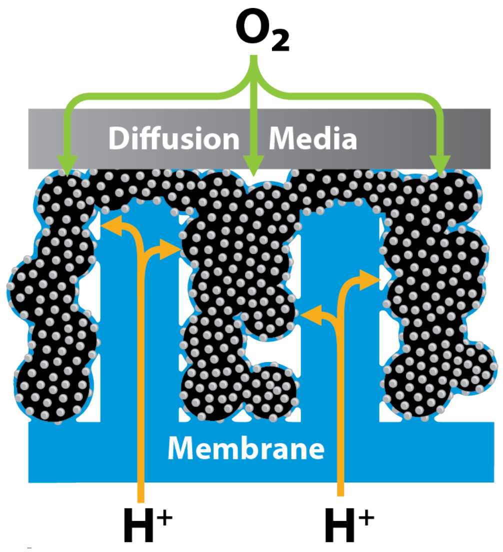
Advanced Catalysts in Advanced Electrodes











GEN2-L1$_0$-PtCo/CZIF-8 in array electrode provides 1.49 A/cm^2 (3.5 kW/g$_{PGM}$) at 0.7 V, 90K

Loadings: 0.05/0.25 mg$_{PGM}$/cm^2 (PtCo), 0.05/0.30 mg$_{PGM}$/cm^2 (Pt)
M2FCT conditions (250 kPa, 85% RH, 90°C, H_2/15% O_2), 5 cm^2 differential cell, NR-211

MILLION MILE
FUEL CELL TRUCK
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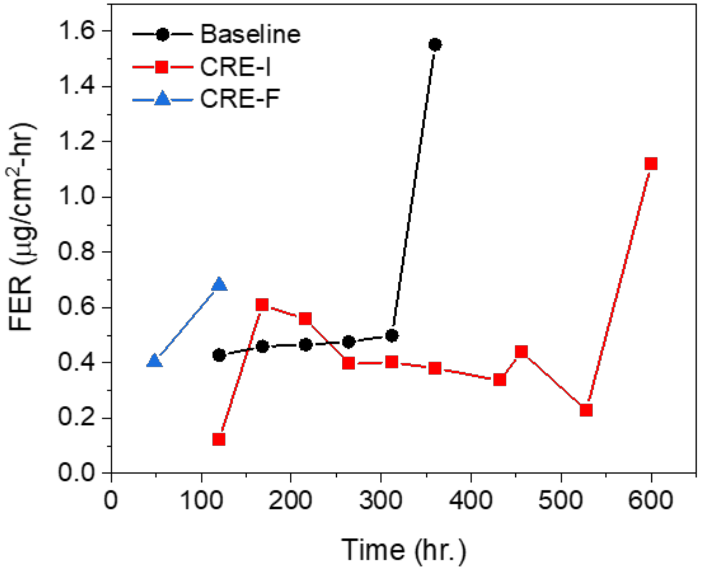
Durability

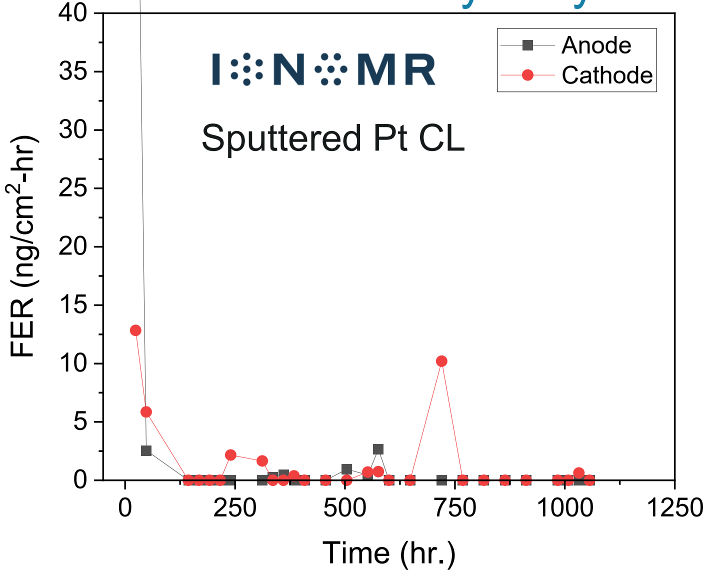
Catalyst Layer Ionomer Degradation

FER PFSA Membrane AST
(baseline and with additives)

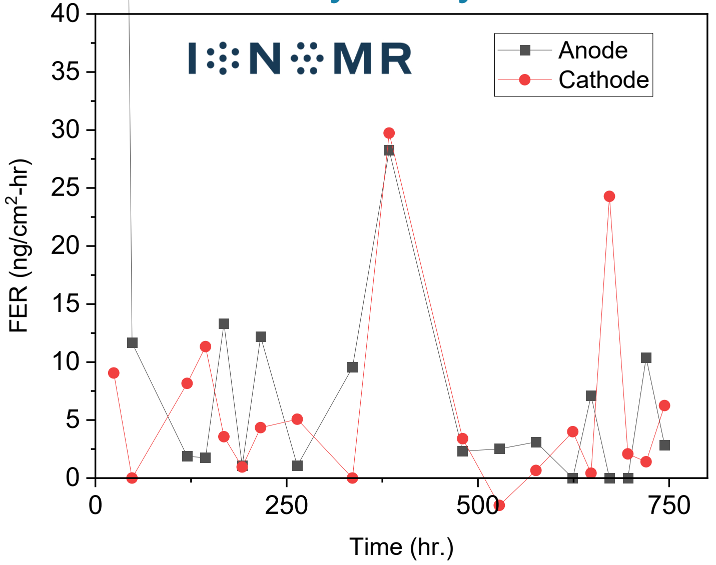


FER Hydrocarbon Membrane AST

No PFSA Catalyst layer



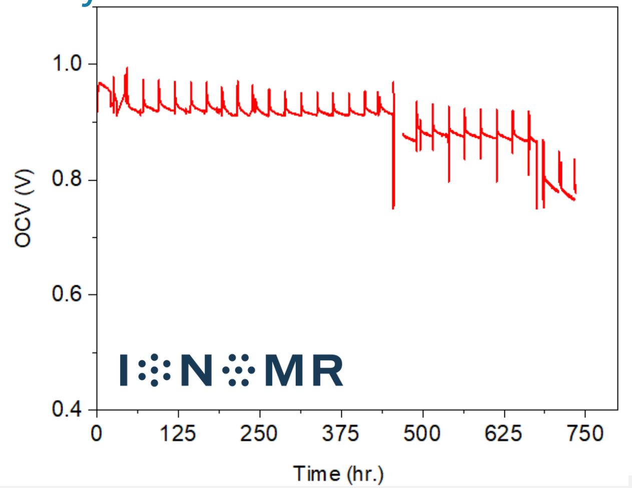
PFSA Catalyst Layer Ionomer



FER for ionomer free MEA was around 39.7 ng/hr

FER for PFSA CL/HC Membrane MEA was 221 ng/hr

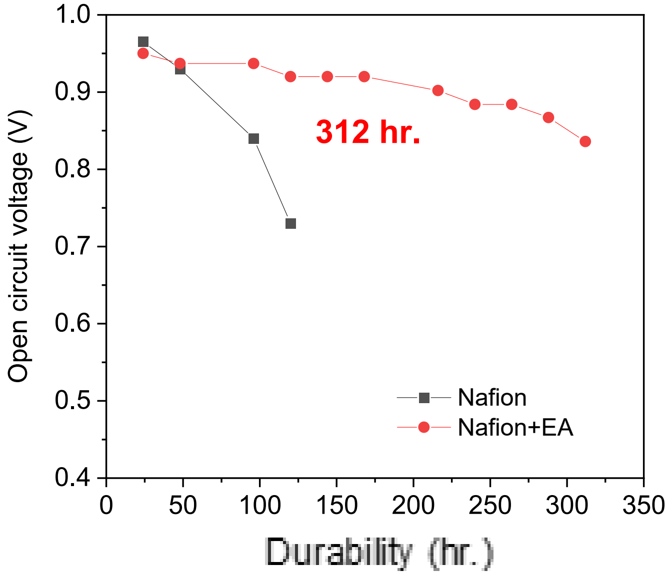
Hydrocarbon Membrane OCV



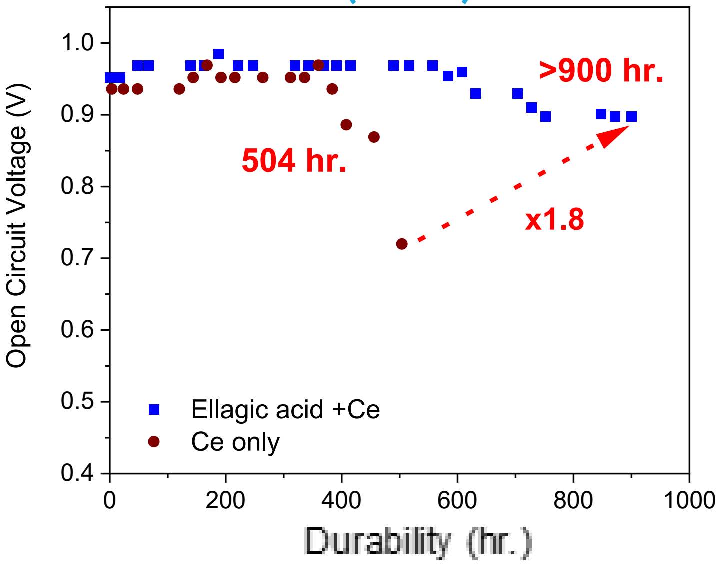
- Measurable ionomer degradation in catalyst layer (small)
- Total ionomer degradation in catalyst layer is about 0.4%
  - Negligible effect on durability
- PFSA Membrane Ionomer degradation is ~ 30-50%
- Also: HC Membranes can last 1000 hr in the OCV AST
  - Have to avoid mechanical stresses

OCV Tests show Ellagic Acid is a Strong Antioxidant for Nafion

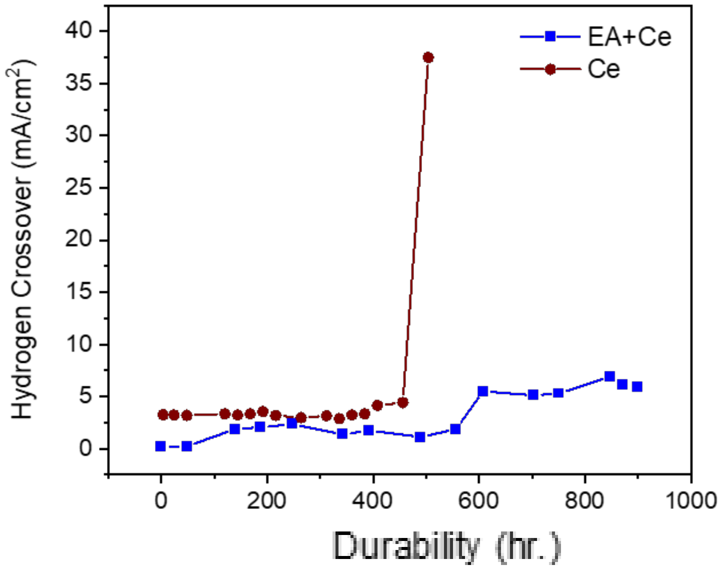
Baseline comparison with EA



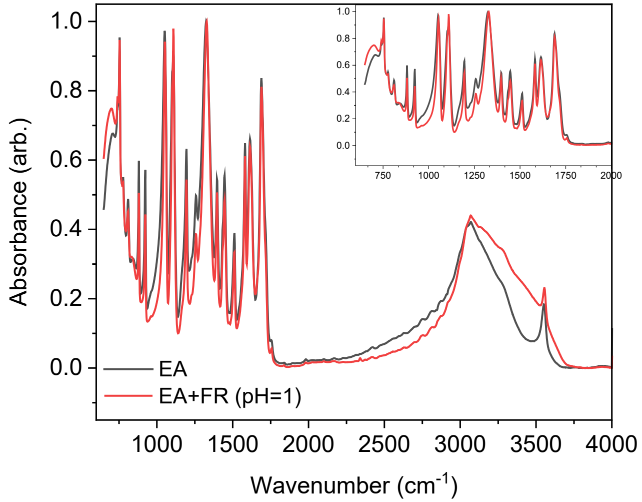
Ce compared with Ce+EA (OCV)



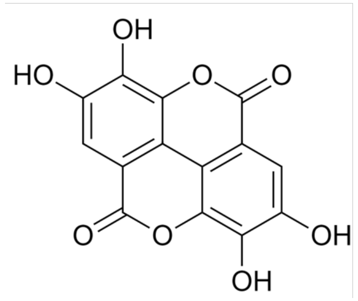
Ce compared with Ce+EA (Cross-over)



- Ellagic acid is an order of magnitude faster in hydroxyl scavenging than cerium
- EA can increase Nafion durability by 160%
- EA with cerium, further enhances durability by >1.8x
- Nafion incorporated with EA and cerium exceeded 900 hr of lifetime without failure



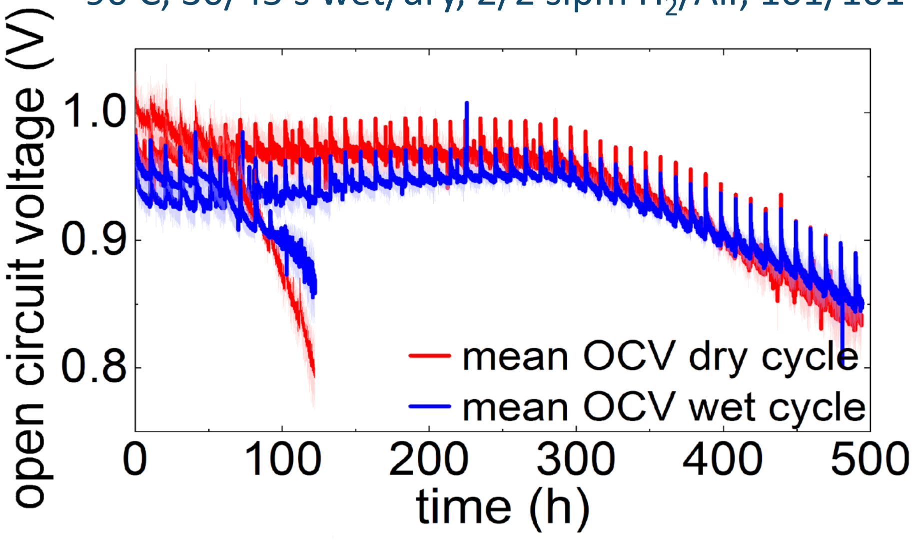
FTIR confirms Ellagic acid is a reversible antioxidant

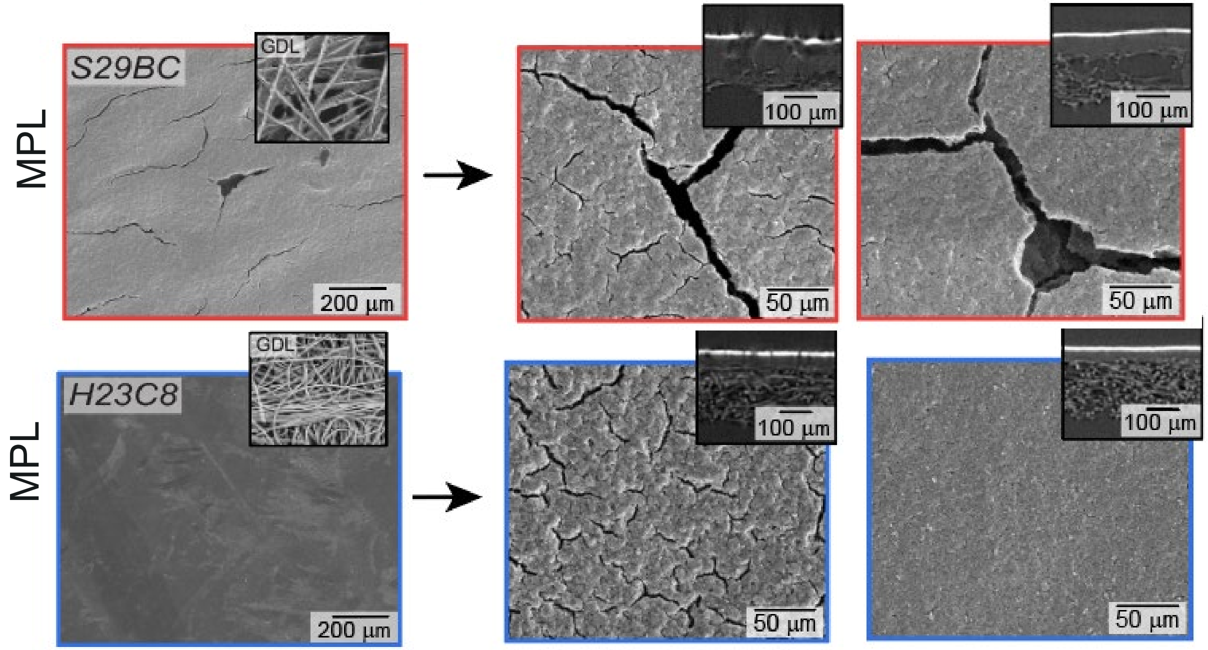


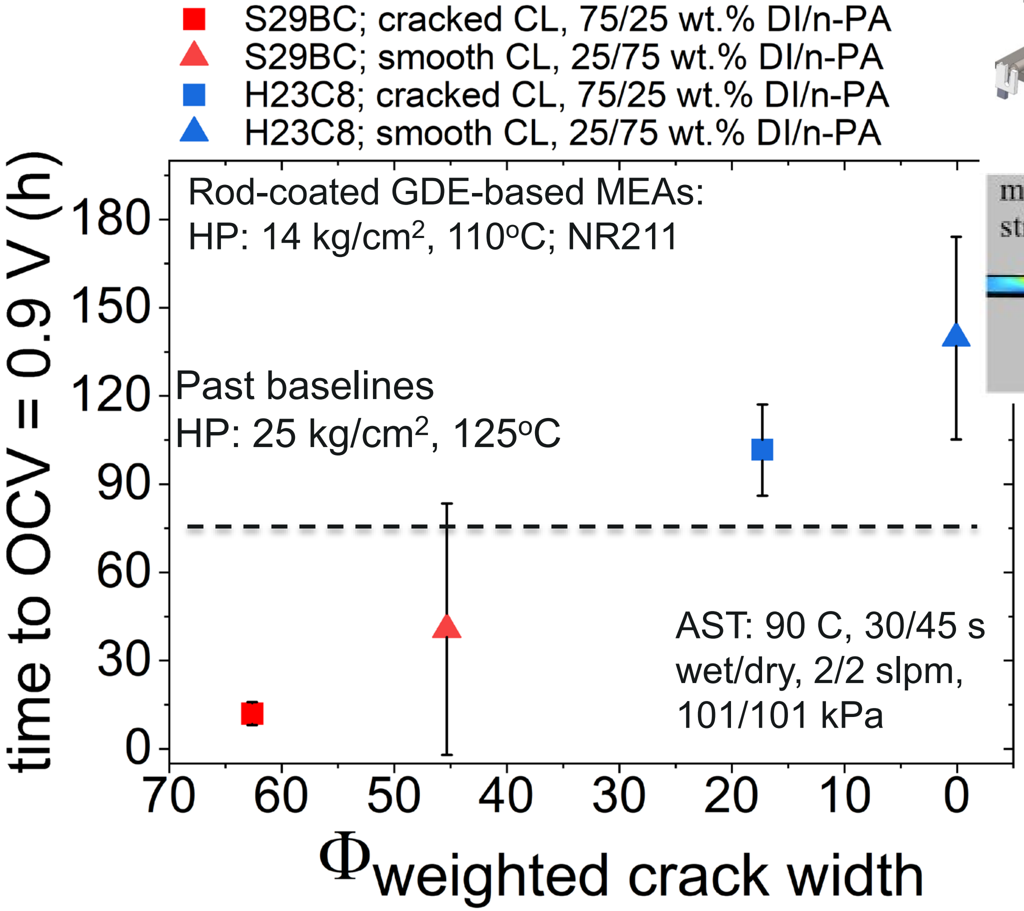
Agarwal, T., Adhikari, S., Babu, S.K., Prasad, A.K., Advani, S.G. and Borup, R.L., 2023. Biosourced Antioxidants for Chemical Durability Enhancement of Perfluorosulfonic Acid Membrane. Advanced Functional Materials, p.2308856.

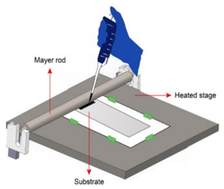
Benchmarking Membrane Durability - Cracked GDE Surfaces

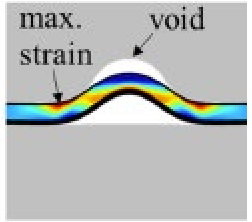
90 C, 30/45 s wet/dry, 2/2 slpm H_2/Air, 101/101 kPa











- Informs electrode morphology requirements
- $\Phi_{\text{weighted crack width}}$ = (areal crack coverage)(crack width)
- Microscale electrode cracks lead to early failures

Thin Film Anode Structure with Exceptional Reversal Tolerance

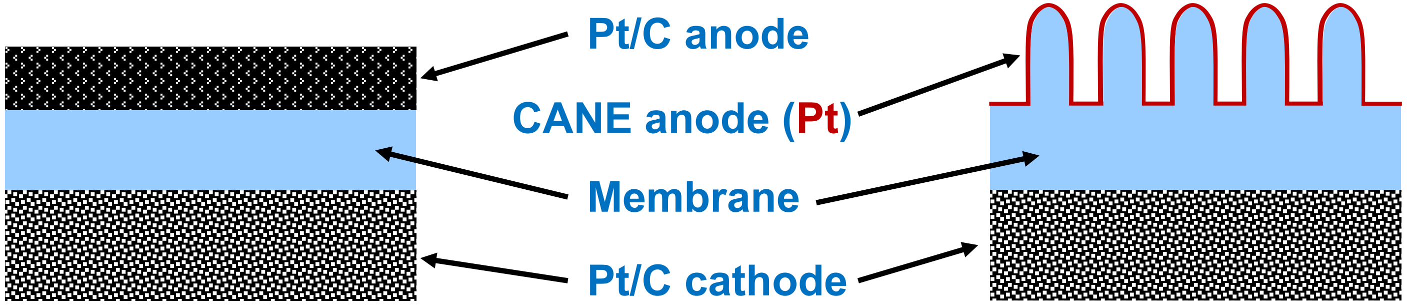
The Co-axial nanowire electrode (CANE) is composed of a platinum thin film supported by Nafion nanowires with high roughness, facilitating carbon-free electrodes

Reversal Tolerant Anode: The CANE electrode structure without OER catalyst outperforms state-of-the-art anode catalyst under a comprehensive anode reversal tolerance assessment

Development of Test Procedure: Developed and assessed an accelerated testing protocol to replicate both cell reversal conditions and regular fuel cell operations cyclically

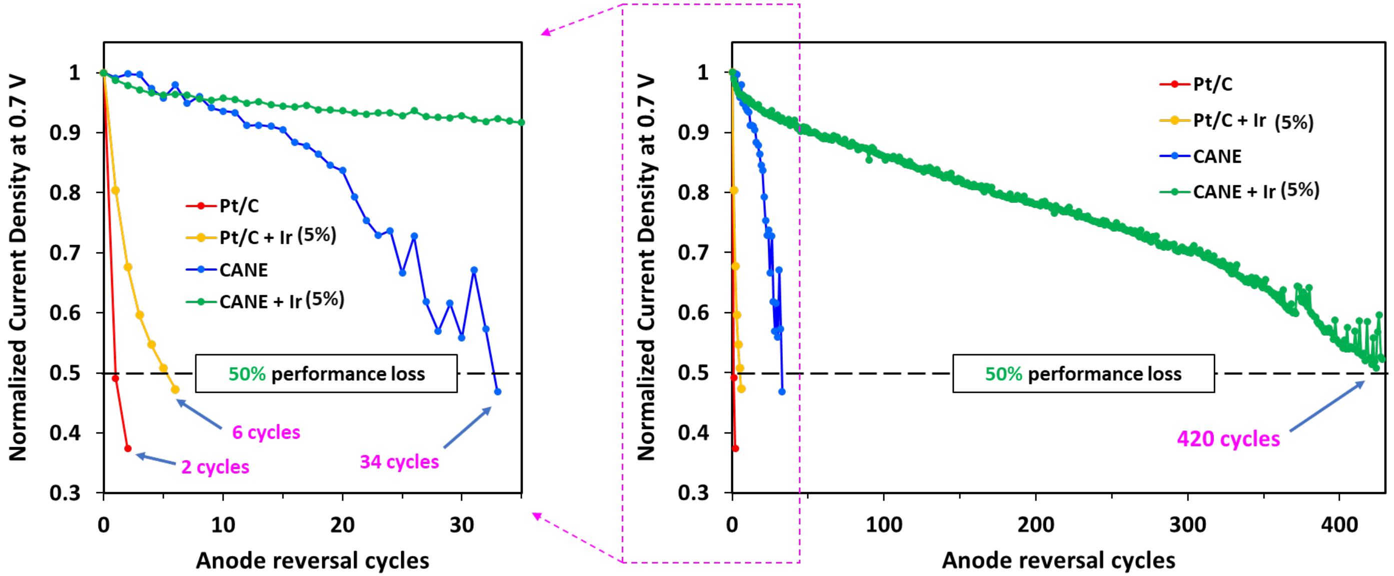
Durability Enhancement: CANE electrode with 5% (w/w_{Pt}) of OER catalyst (Ir) shows **85X** improvement over state-of-the-art anode (Pt/C + Ir)

Enhanced Durability and Reliability: Reversal tolerant CANE anode improves the overall durability and reliability

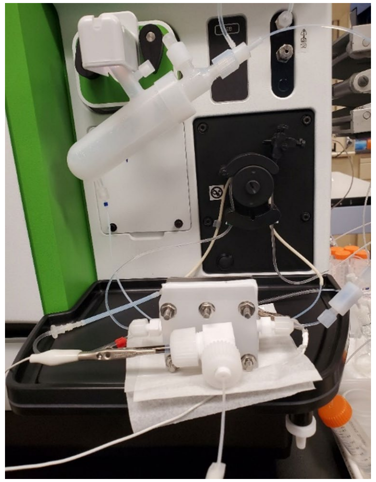


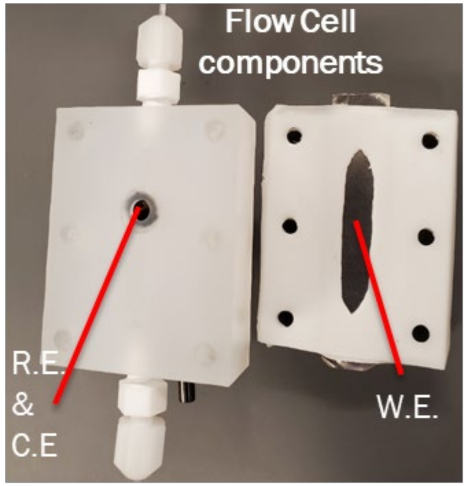
Schematic illustration of MEAs with Pt/C and CANE anodes

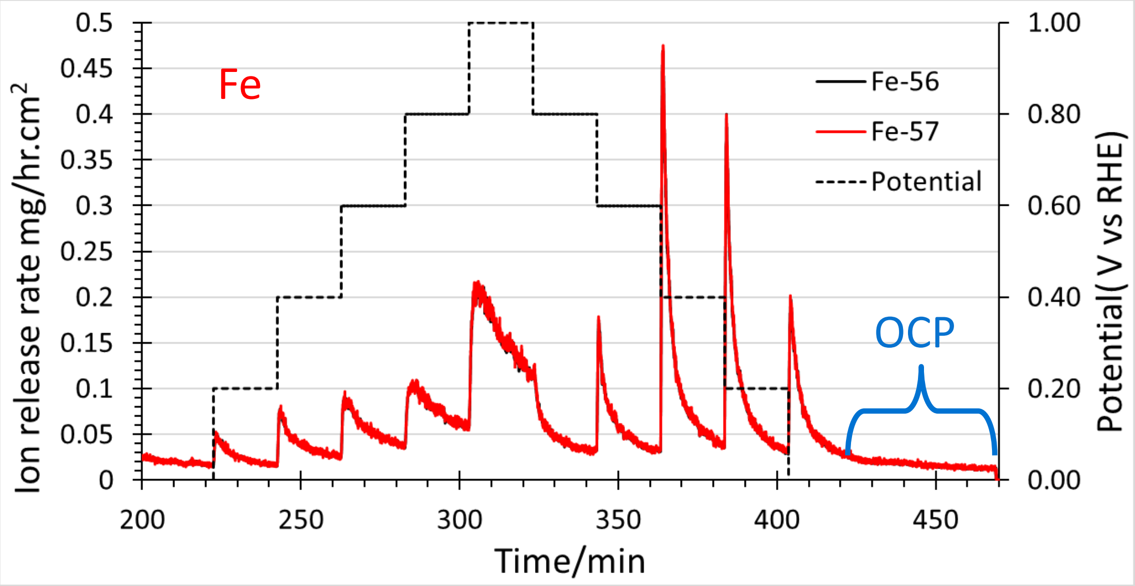
Current density @ 0.7V vs number of anode reversal cycles



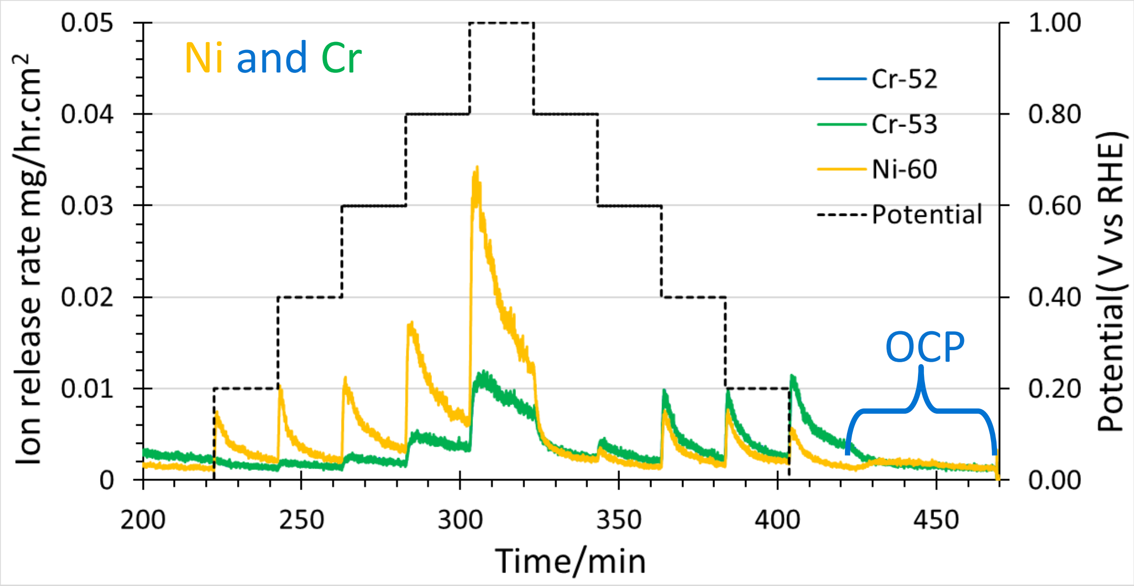
Time and Potential-Resolved Studies of Bipolar Plate Corrosion







- Cations in 0.1 M HClO4 electrolyte detected with ICP-MS coupled to outlet of electrochemical flow cell
- Fe, Cr, and Ni dissolution from 316L stainless steel working electrode determined as a function of potential and time to establish baseline for corrosion studies of coated stainless steel bipolar plates
- Fe dissolution rates:
  - Highest during cathodic-going potential steps – consistent with removal of passivating film at these potentials
  - >10x that of Cr and Ni



AST Development

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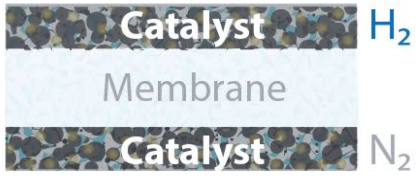
M2FCT MEA AST

COMPLETED

Catalyst AST

90,000 Cycles | 150 hours

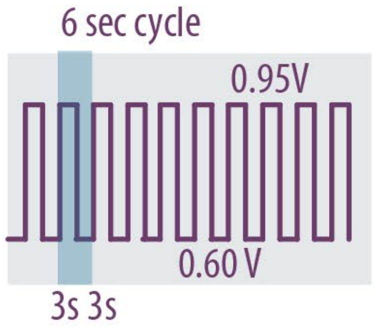


H_2/N_2 at 200/200 sccm

Temperature: 80˚C

Humidity: 100%/100%

Cycle: Square Wave
Upper Potential
Lower Potential



INTERIM

Membrane/MEA AST

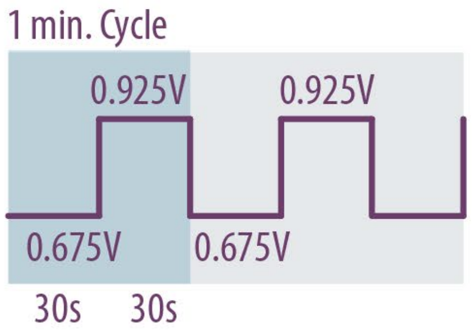
30,000 Cycles | 500 hours

H_2/Air at 1000/2500 sccm

Temperature: 90˚C — increase temperature from 80 to 90ºC for more acceleration

Humidity: 50%/50% — Lower inlet RH for higher memrane degradation

Cycle: Square Wave
Upper Potential
Lower Potential



Feedback from Industry & iDWG

PROPOSED

Membrane/MEA AST

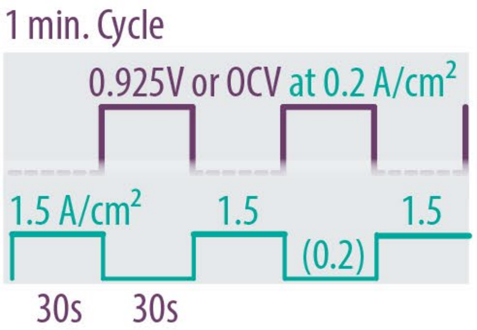
30,000 Cycles | 500 hours

H_2/Air at 1000/2500 sccm

Temperature: 90˚C

Humidity: 30%/30% — Further RH reduction for more aggresive MEA degradation

Cycle: Square Wave
Upper Potential
Upper Current Density — To maintain consistent stressor on membrane for accurately representing the chemical-mechanical MEA degradation in AST



Replace lower potential with upper current density

Notes: The protocol will be updated (if needed) and finalized. Website: m2fct.org • Contact: M2FCTSC@lbl.gov

M2FCT MEA AST

Heavy-Duty MEA AST Protocol:

PROTOCOL

Cycle	Square wave between 0.675 V (30s) and 0.925 V (30s); Single-cell 50 cm^2 [a]
Number	500 hours or 30,000 cycles
Cycle time	1 minute
Temperature	90°C
Relative Humidity	Anode/Cathode: 50%/50%
Fuel/Oxidant	Hydrogen/Air(H_2 at 1000 sccm and Air at 2500 sccm for a 50 cm^2 cell
Pressure	250 kPa

METRICS

Metric	**Frequency**	**Target**
Catalytic Mass Activity [b]	At BOT,[c] after 100h, 200h, 300h, 400h, 500h	TBD
ECSA/Cyclic Voltammetry [d]	At BOT, after 100h, 200h, 300h, 400h, 500h	TBD
Hydrogen Crossover [e]	At BOT, after 100h, 200h, 300h, 400h, 500h	TBD
Polarization curve [f]	At BOT, after 100h, 200h, 300h, 400h, 500h	TBD

a. 14-channel serpentine cell (Daniel R. Baker et al 2009 J. Electrochem. Soc. 156 B991) operated under counter flow conditions.

b. Mass activity in A/mg @ 150 kPa abs backpressure at 900 mV iR-corrected on H_2/O_2, 100% RH, 80°C, anode stoichiometry 2; cathode stoichiometry 9.5 (as per Gasteiger et al. Applied Catalysis B: Environmental, 56 (2005) 9-35).

Measured ORR current should be corrected for H_2 crossover and shorting.

c. BOT measured after a conditioning protocol comparable to the one reported in Kabir et al 2019 ACS Appl. Mater. Interfaces 11, 45016.

d. Sweep from 0.05 to 0.6 V at 20 mV/s, 30°C, 100% RH.

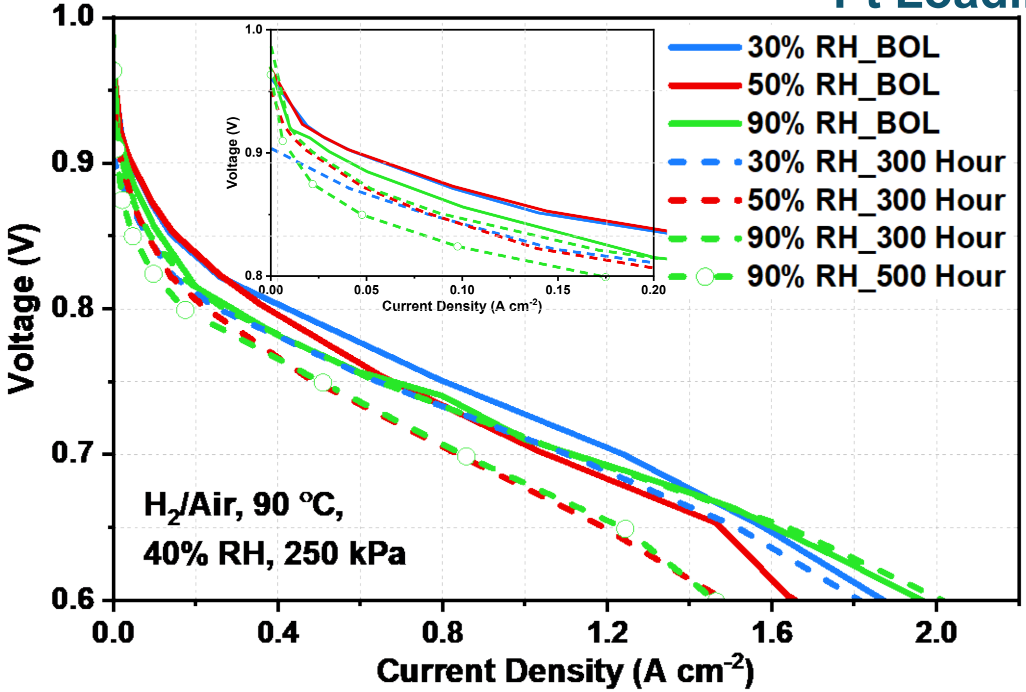
e. Crossover measured at T = 30 °C, RH = 100%, Pressure = 101.3 kPa, 2mV/s scan rate from 100 mV to 400mV. H_2 = 500 sccm, N_2 = 500 sccm.

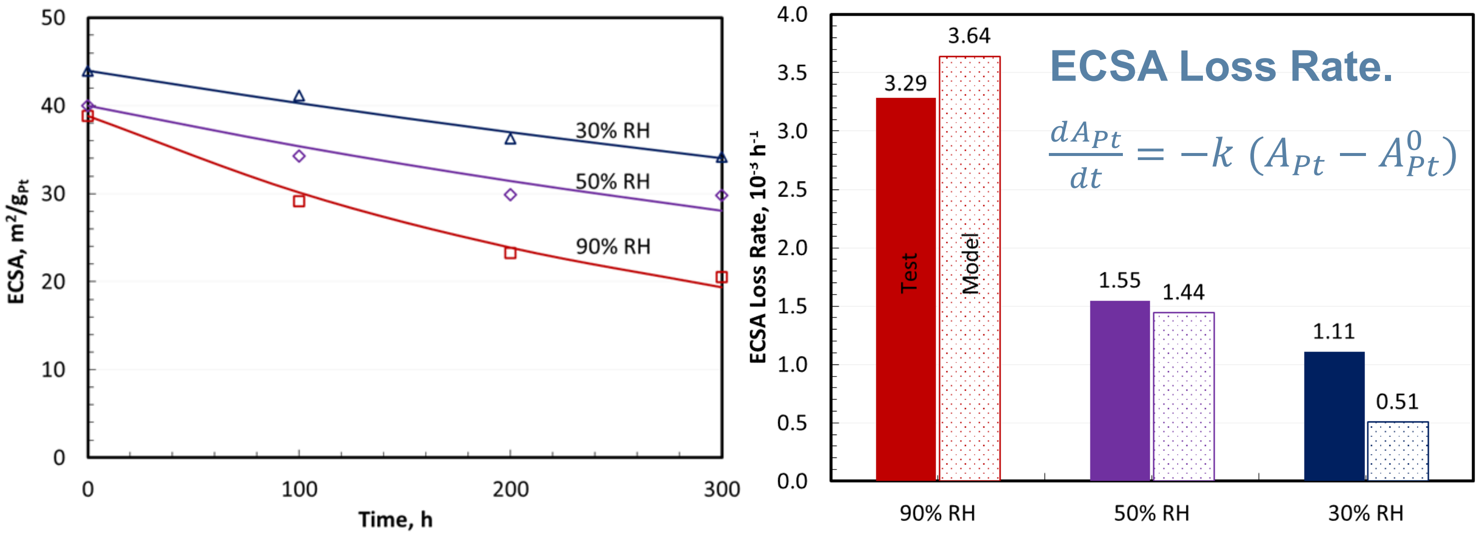
f. H_2/Air, 250 kPa abs backpressure, 90°C, 40% RH, cathode stoichiometry 1.5, anode stoichiometry 2; Recommend taking pol curves from high to low current densities at 0.01, 0.02, 0.03, 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, 1, 1.25, 1.5, 1.5 and 2 A/cm^2, 240s hold time at each data point

CO stripping is added at each interval to test the RH dependency of ECSA

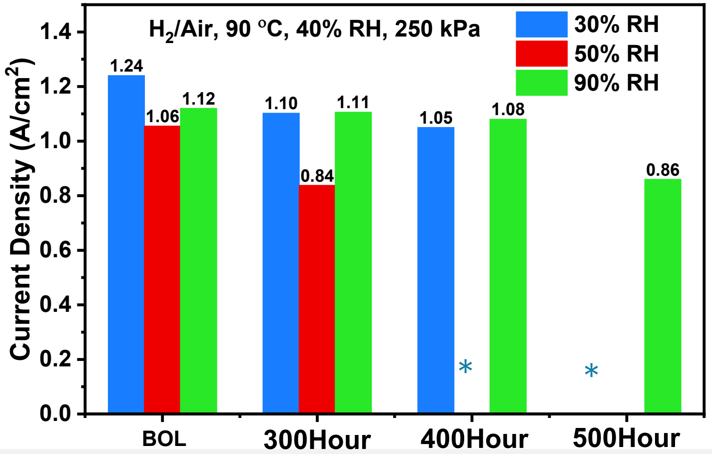
H_2/Air AST: Umicore Pt500550 catalyst and Nafion® HP membrane

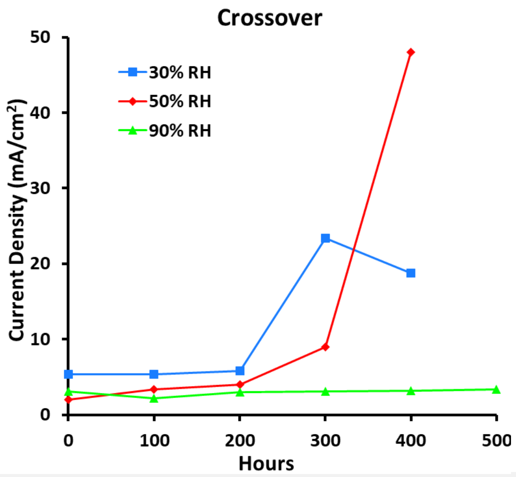
Pt Loading (c): 0.25 mg/cm²





$$\frac{dA_{Pt}}{dt} = -k\,(A_{Pt} - A^0_{Pt})$$

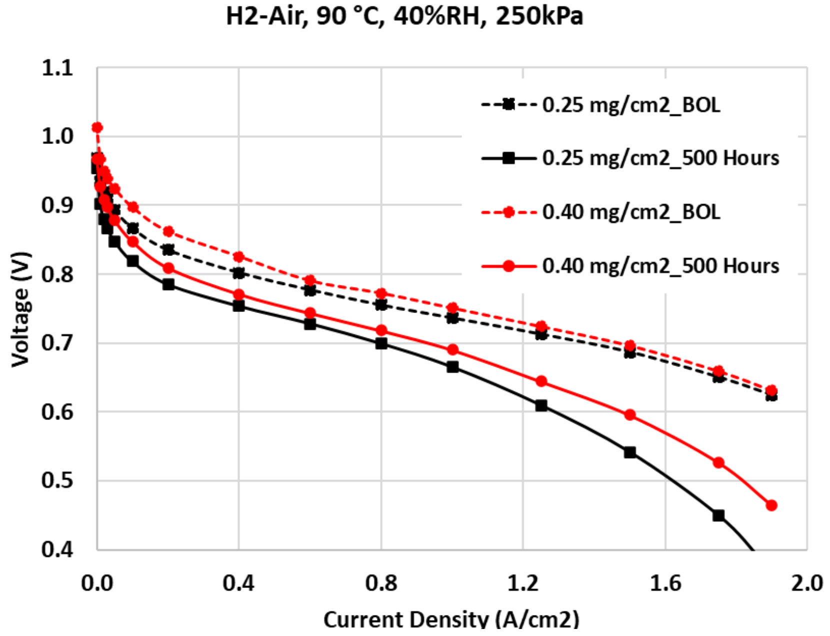




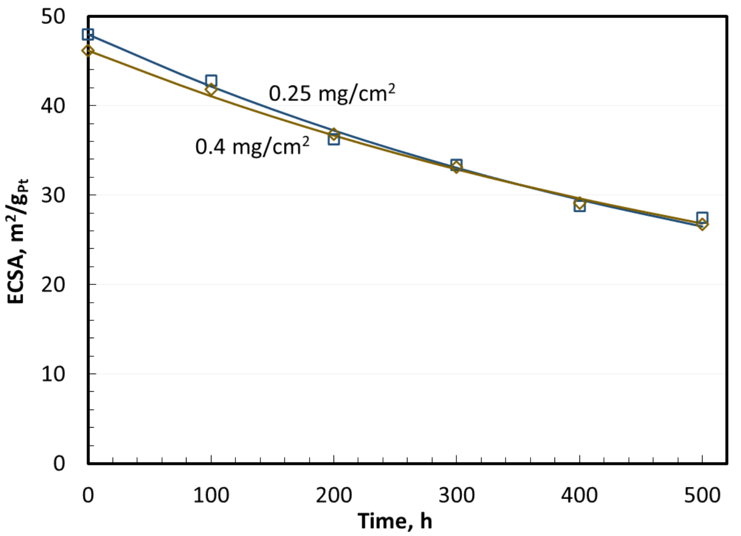
- With RH decrease, stress to catalyst is decreased: Decreased ECSA loss with lowering RH
- With RH decrease, stress to membrane is increased dramatically:
  - Higher crossover current
  - Lower OCV

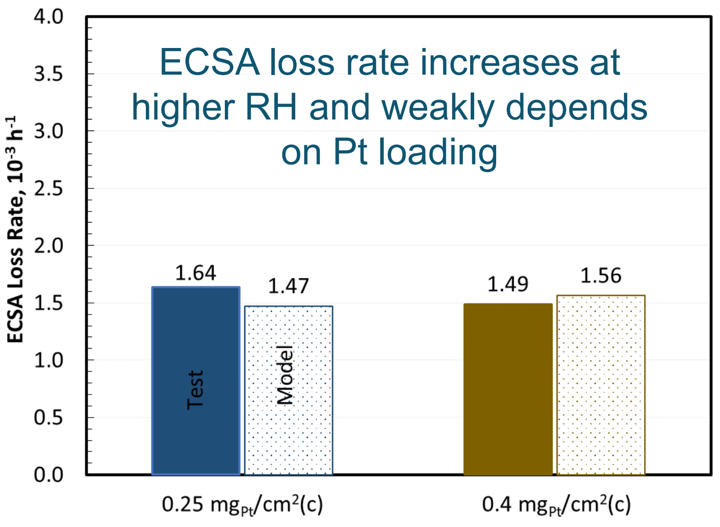
*Membrane failure stopped test < 500 hours

H_2/Air AST: Umicore Pt50 0550 catalyst and Nafion ®NC700

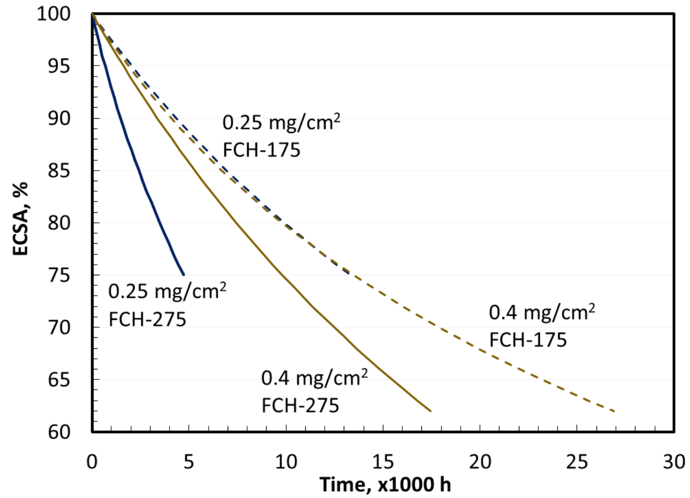


ECSA for MEA AST @ RH: 50%





FCS	Cathode Pt Loading	Idle Power	Voltage Clipping	Acceptable ECSA Loss	Membrane Oversizing	Electrode Lifetime
FCH-275	0.25 mg/cm^2	30 kW	835 mV	25%	28%	4,700 h
FCH-275	0.4 mg/cm^2	70 kW	815 mV	38%	44%	17,440 h
FCH-175	0.25 mg/cm^2	35 kW	815 mV	25%	28%	13,340 h
FCH-175	0.4 mg/cm^2	50 kW	815 mV	38%	44%	25,600 h

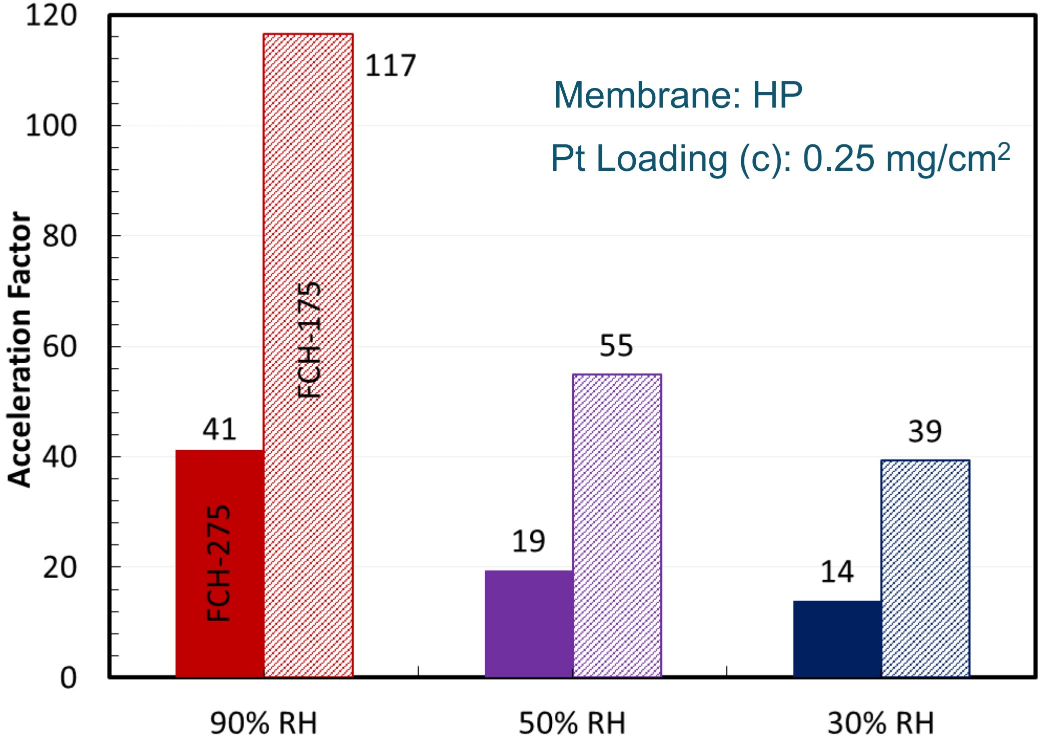


Electrode Lifetime: Duty cycle time after which the cell power density decreases to 750 mW/cm^2 at 0.7 V, 2.5 atm, 90°C, and 1.5 cathode stoichiometry with 14 mm, stabilized, reinforced membrane.

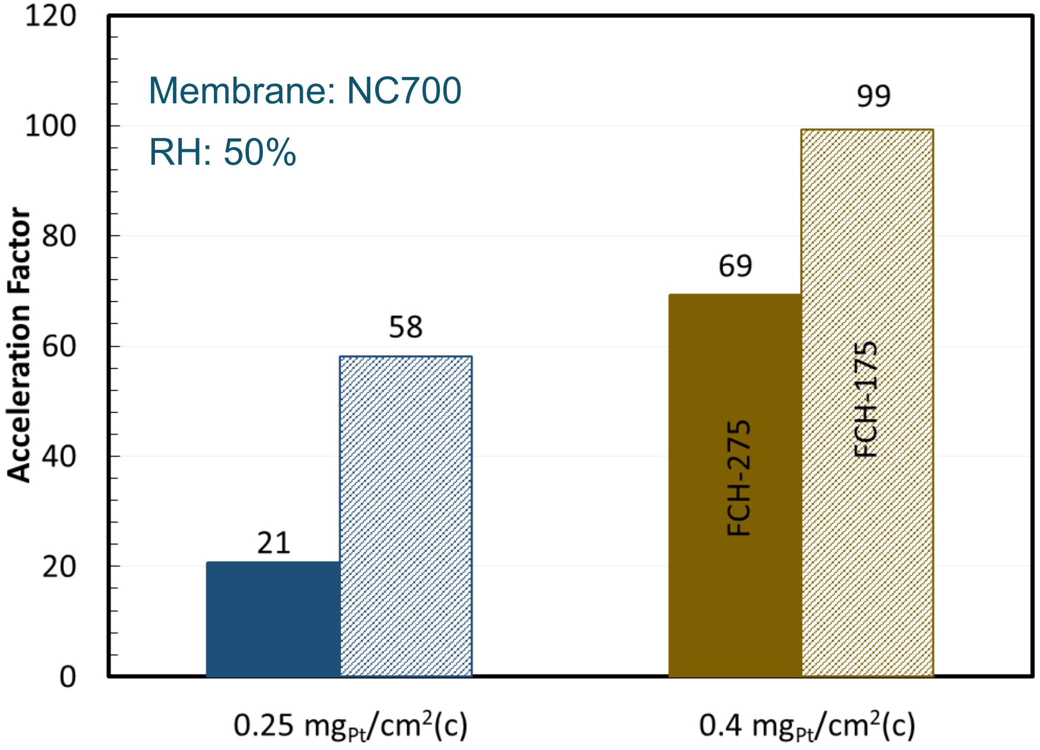
Acceleration Factor

- Acceleration Factor (AF): Ratio of electrode lifetime on Class 8 long-haul truck duty cycle to AST time for the same ECSA loss
  - Electrode acceleration factor is higher at 90% RH but the membrane is more stable
  - 50% RH may be a reasonable compromise for accelerating electrode and membrane degradation on MEA AST
  - Exploring 30%RH to further accelerate membrane degradation
  - Future work: Determine the acceleration factor for membrane stability

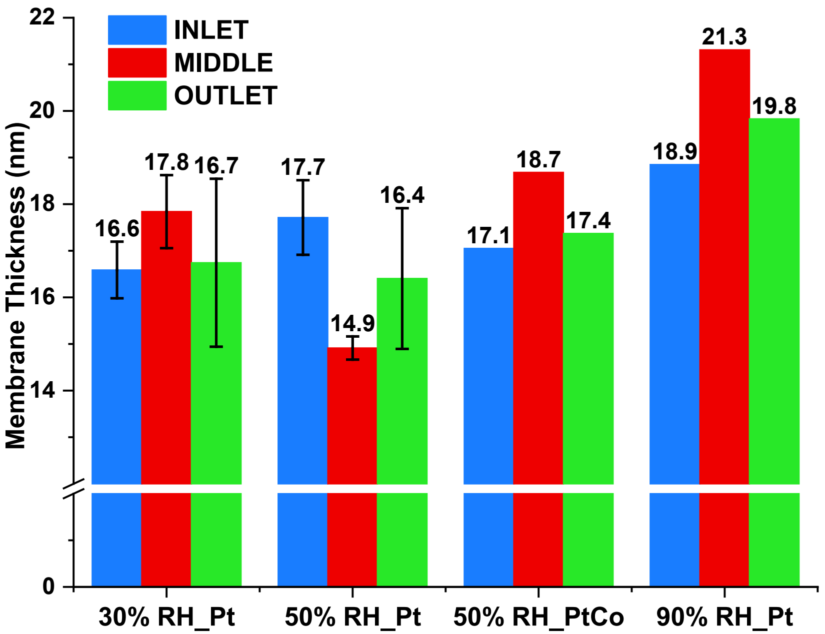
RH Effect on Acceleration Factor



Pt Loading Effect on Acceleration Factor

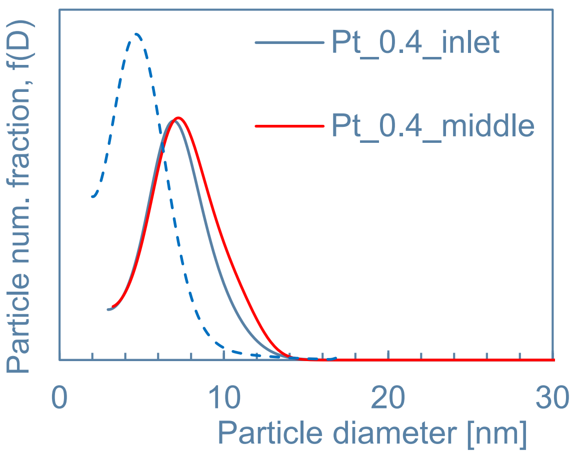


H_2/Air AST: Membrane and catalyst degradation EOT



- Membrane thinning for both 30% and 50% RH is more obvious than 90%RH
- Membrane thinning is more at the cathode side and at the inlets and outlets
- 50% RH shows similar Pt band location

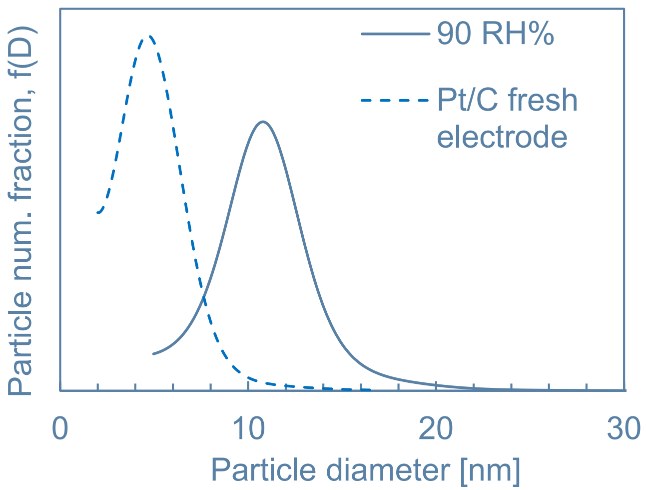
90 °C, 50% RH



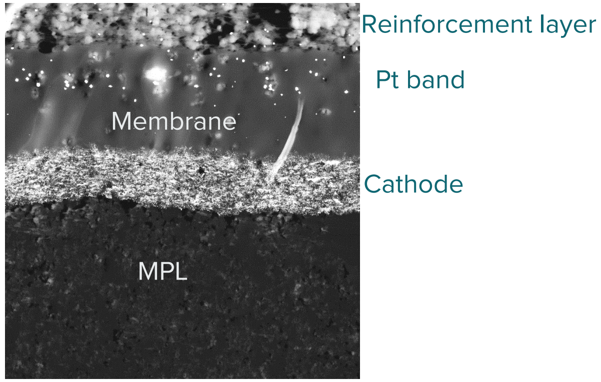
	Mean diameter [nm]
Pt_0.4_inlet	5.8
Pt_0.4_middle	6
Pt/C fresh	4.4

- Similar particle size growth between inlet and outlet at 50% RH

90 °C, 90% RH

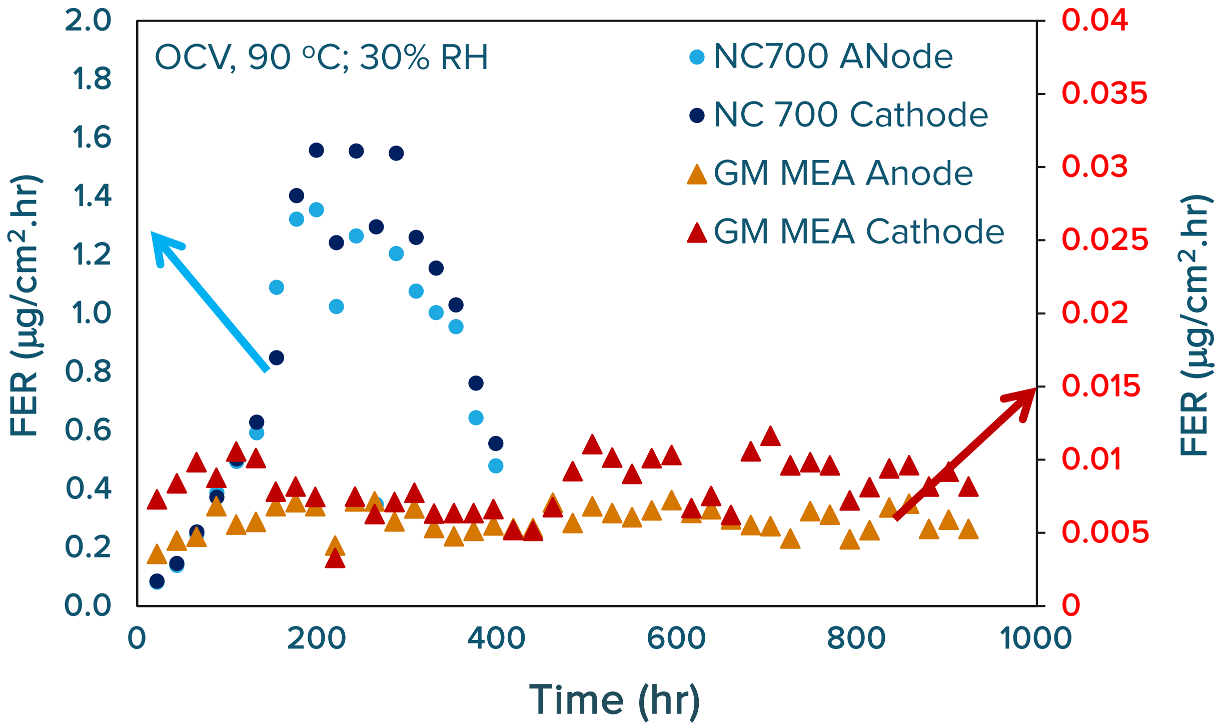


	Mean diameter [nm]
90%	7.2
Pt fresh electrode	4.4

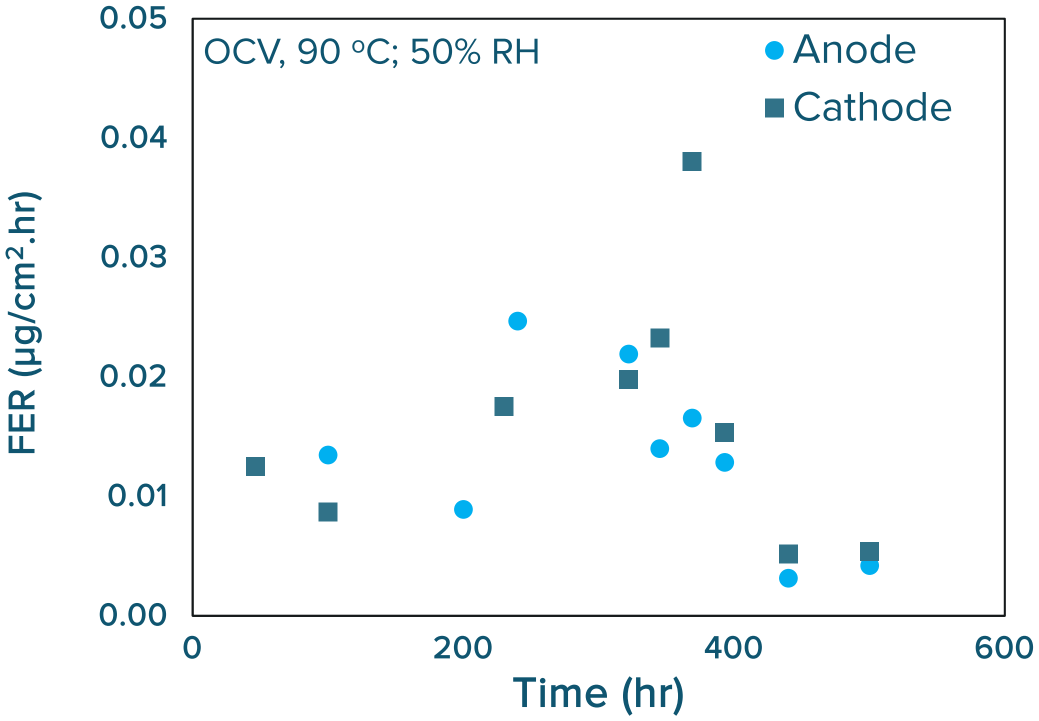


H_2/Air AST: Membrane Degradation - FER

Wide variation in FER during OCV tests with the two SOA membranes (30% RH)



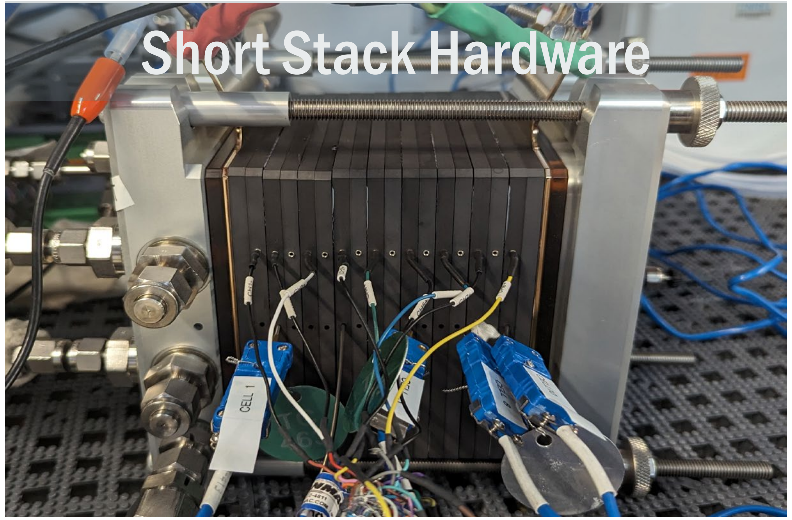
Low FER in MEA AST @ 50% RH from NC700 MEA



- Lower FER for 400-500 hours test mainly because the current was much lower under 0.675 V
- Proposed AST: 1.5A/cm2 fixed throughout the test with 30% inlet RH to increase membrane degradation. SOA membrane has FER ≈ 50 ng/cm²/hr in test

Accelerated Durability Test on 10-cell Short Stack

- **Performed MEA ADT on 10-cell short stack**
  - 30s at V_{avg} 0.675 and 0.925 (current cycle)
  - Rainbow stack w/ Direct Membrane Coating MEAs
    - Alternating cells **w/ additive** and **w/o additive**
- **Characterization every 100 hrs**
  - H_2/Air and H_2/O_2 Polarization Curves
  - Psuedo CVs
    - ECSA losses and H_2 crossover increasing



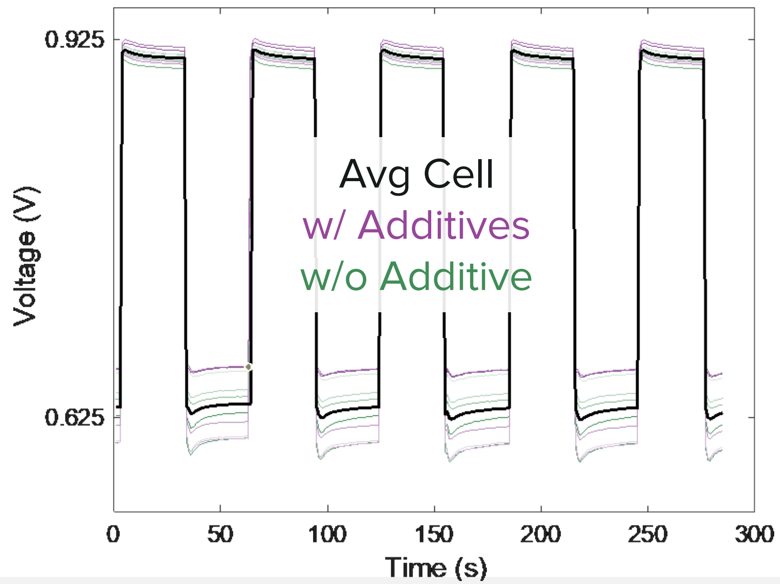
Cells	
1	2
3	4
5	6
7	8
9	10
Control	Additive

50cm² DMC electrodes (NC700)

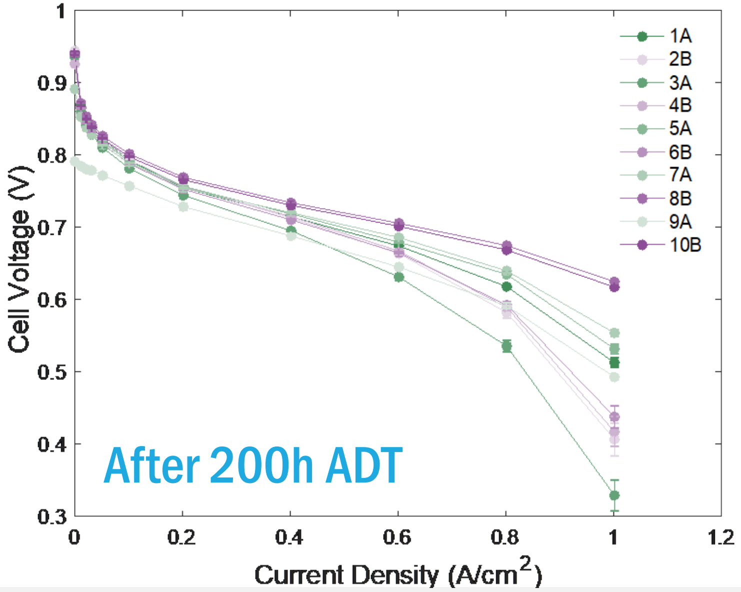
0.168 - 0.176 mg_{Pt}/cm^2 L_{cath}

Pt/C TEC10E50E D2020 I:C 1.0

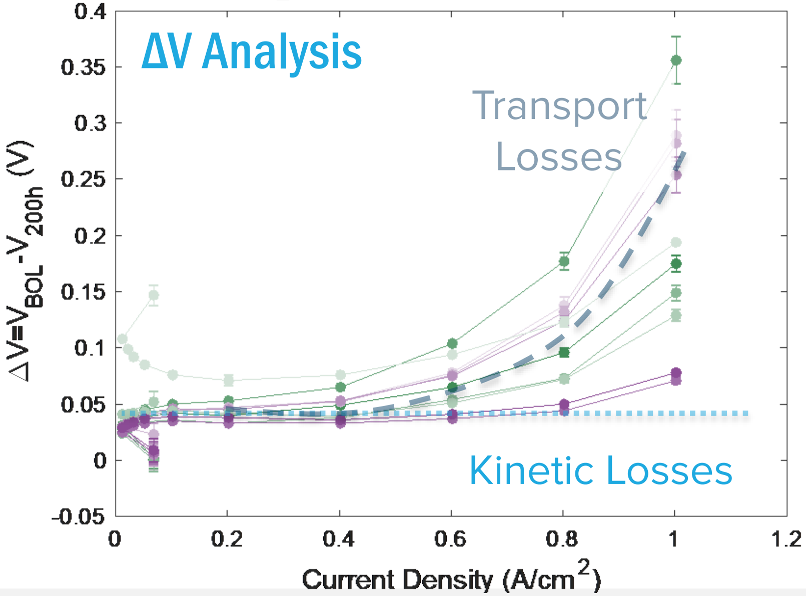
Short Stack ADT 90°C, 50% RH, 250kPa



H_2/Air 80°C, 100% RH, 150kPa



H_2/Air 80°C, 100RH, 150kPa



Van Cleve, T., et. al (*to be submitted*).

Reviewer Comments (Weaknesses)

- In general the comments were overwhelmingly positive showing that M2FCT is leading, advancing, and playing a critical role in the field. Below are some select comments that have been aggregated in topical themes and responded to
- The project has a very large scope and many participating PIs. Sometimes it may lose focus just because of the size and scope. Certain targets such as power output and PGM loadings are too safe, which may not be challenging enough to stimulate breakthrough and innovations.
  - The targets for FY24 are significantly more difficult to meet at: Achieve current density of [3]1.3 A/cm^2 at 0.7 V and [3]0.45 A/cm^2 at 0.8 V after catalyst AST. We do not stop improving materials such as catalysts just because a target is met. However, we do acknowledge that perhaps a higher target is needed and are discussing that with DOE.
- There is a multiplicity of high-risk concept materials under study. Hybridization approaches for the catalysts are lacking or were dismissed. High-risk electrode structures (CANE, etc.) are not verified for sensitivities to common electrode failure modes, such as flooding.
  - We are exploring the impact of nanowire size and spacing on robustness, but the recent focus has been on CANE for reversal tolerant anodes. Not clear what is meant by hybridization approaches for catalysts; if the request is related to novel supports like MOx; that is being explored but has not been highlighted by M2FCT as their performance was lower than the catalysts presented.
- Some of the work presented seemed to be unrelated side projects. A better explanation for the motivation or relevance of the advanced materials work would be beneficial.
  - Current materials do not meet longer term targets, especially the longer-term efficiency targets. M2FCT has intentionally mixed lower TRL work with higher TRL work for a combination of meeting near-term targets and meeting longer-term targets. Catalysts and ionomers with better performance, and better durability at higher temperatures was requested by other reviewers. That being said, we have entered a more convergent phase of scaleup and integration of materials and testing for performance and durability,
- There are too many ASTs.
  - There is no consensus on this point. Many comments have suggested that all ASTs should be single component/single mechanism ASTs
- There is too much focus on fundamentals and new concepts when the current need is an engineered solution that can compete with diesel trucks.
  - M2FCT tries to balance lower TRL with higher TRL activities and have been directed to accomplish both. M2FCT has significant activities related to Tech Transfer and support to industrial projects. Many engineered solutions tend to be proprietary, and thus not the purview of the National Labs.
- Comparison with real-life aged components may be increased.
  - M2FCT is collecting what real-life aged components we can get and correlating them to stressors and events. Unfortunately, and obviously, there are essentially no Heavy-Duty Fuel Cell trucks which have significant run time. There are three separate activities underway to validate the ASTs with real world stack testing data. AC Transit, Plug Power and EKPO. In addition, we are working on physics-based predictive models for lifetime operation

Reviewer Comments (Recommendations for additions/deletions to project scope)

- It is suggested that the project team deploy a systematic approach in identifying test protocols for durability validation for components to stack level. The consortium now has a study to define ASTs at MEA level with a subscale differential cell, which disengages the flow-field/bipolar plate effect. However, there is no test protocol to verify the stack design for durability. Obviously, stack condition is different from subscale cells. For example, change of the membrane's hydration state is quite different between stack-level and subscale cells. A systematic approach to define test protocols for stack-level validation is necessary. This could be proper for national labs and universities to lead.
  - M2FCT tries to be as systematic as possible as it develops durability data and new protocols. Full stack protocols are difficult as there is limited full stack testing capabilities, plus at a full stack level, operation is no longer OEM/stack independent as different developers use different geometries and flow-field designs. Using differential cells allows M2FCT to tailor flowrates, oxygen content and water content to simulate various locations within a stack which should be more independent of proprietary designs. M2FCT is looking at some short-stack ASTs but for the most part stack testing is beyond the scope of the consortium where we want to concentrate on understanding and mitigating mechanisms.
  - The M2FCT proposed MEA AST was discussed at the DWG (Durability Working Group); there was no consensus or agreement attained.
- It would be great if the consortium project could conduct a more thorough study on the heat rejection and management of the HD fuel cell system and take a more aggressive approach in improving existing or developing new materials that can increase the operation temperature of proton exchange membrane fuel cells. This is a major issue for HD truck design, which has not been sufficiently addressed in the project.
  - Temperature is a critical variable in durability testing - evaluating the effect of temperature on durability. The Cummins project is looking at high temperature operation for heat rejection. M2FCT is working to develop the most active and durable catalyst and ionomer materials, and that includes at higher temperatures. M2FCT has also participated in discussions related to novel heat rejection techniques for HDFC (SBIR Advanced Cooling Technologies Inc (ACT) PI: Ramy Abdelmaksoud)

Reviewer Comments (Recommendations for additions/deletions to project scope)

- In light of the polyfluoroalkyl substance (PFAS) regulatory discussions, it is recommended that the project look into perfluorosulfonic acid (PFSA) ionomer control and recycling strategies. The presented highlighted some work looking at hydrocarbon polymer materials; however, this technology is potentially decades away from being commercially viable. A more realistic approach to the PFAS regulation concerns is addressing control and recycling of these materials.
  - Recycling has not been included in M2FCT. DOE HFTO has announced funding of a $50M recycling consortium (H2CIRC) through DE-FOA-2922. M2FCT may interact with that new Recycle consortium, but clearly that consortium's role will be to develop these requested recycling strategies.
- It is recommended that the project further develop in-cell characterization highlighting MEA component deficiencies along degradation in ASTs (other than polarization curves, electrochemical surface area, and mass activity), specifically proton conductivity/mass transport in the cathodes. The team should develop explicit structure–property polarization curve relations for cathodes that remain true through the ASTs. Concepts should be weighed prior to disseminating.
  - M2FCT strives to use available techniques. We employ Impedance measurements and modeling (which incorporates both proton conductivity and mass transport), we analyze surface coverages of ionomer on catalysts, measure membrane durability by FER, cross-over and OCV. We do significant post-characterization with a wide array of characterization techniques (e.g. STEM, XRD).
- It would be a challenge, but if there is any way to show the impact of these other projects, or the summation thereof, on the spider plot, it would be great to see.
  - The systems analysis is highly integrated with the improvements made as shown in the catalyst efforts and durability, and these become reflected on the spider chart
- Something lacking from most project summaries is an objective gap and risk analysis to advance the technology to higher technology readiness levels (TRLs) and hopefully reach a commercial readiness level. With the cumulative brainpower and experience level of the M2FCT, this would be an invaluable service to help project principal investigators (PIs) advance their projects in the right directions, strengthen their future plans, and if need be, pivot in strategy to make the most of DOE funds. This may not be the current mandate of the M2FCT, but it would be invaluable.
  - This is a good comment and M2FCT is exploring additional figures of merit as well as a ranking scheme that we can put on the website for the different results and approaches

Diversity, Equity, Inclusion, Accessibility

Outreach and Workforce Development

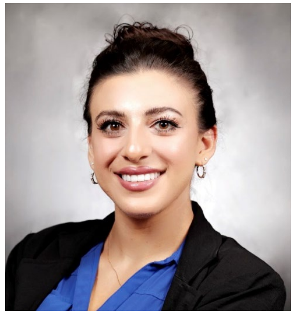
- M2FCT hosting UGS, GRA interns and DOE SCGSR Fellows to gain hand-on experience working with fuel cell systems and materials and learn about hydrogen technologies
- Multiple MSI students with M2FCT
- Defined multiple projects staffed and coordinated with MSIPP and M2FCT

MSI visits/recruitment

- Texas Tech: Growing Stems Meeting
- UT-Rio Grande Valley visit
- Visit to NMSU
- Visit/Tour by NTU and San Juan College
- ACS Meeting (Puerto Rico)
- Univ Puerto Rico (Mayaguez)
- Florida International
- United Tribes Technical College
- University of Arizona
- UT-San Antonio
- San Juan College
- Santa Fe Indian School

M2FCT/ MSI Collaboration: BPP corrosion with GM

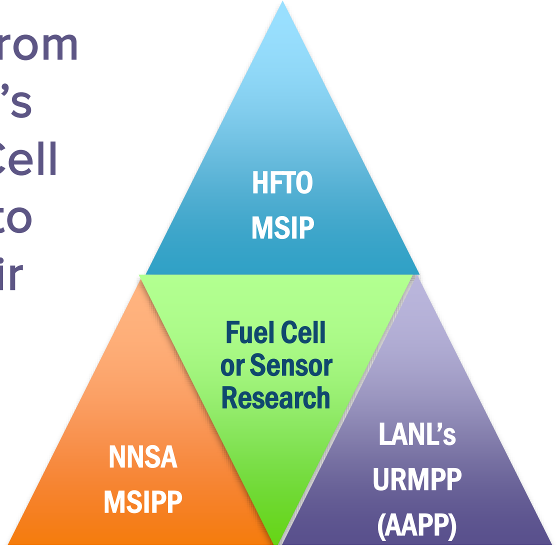
Lexy Murphy
B.S. Computer Engineering



Lyra Troy
M.S. Chemical Engineering

M2FCT working with Minority Serving Institution Partnership Program

Students from HBCU/HSI's use Fuel Cell Research to obtain their PhDs and Launch Careers



Note: A CBP or DEIA plan was not required

Collaborations: Non-FOA activities

Entity	Scope of collaboration
Pajarito Powder	Catalyst durability testing
EKPO	Durability and AST Development
AC Transit	Fuel Cell materials post operation in fuel cell buses for durability evaluation
3M Company	Development of novel materials. 3M provided, at no cost, with several critical components needed for synthesis of targeted cationic ionomers.
Hahn-schickard and Simon Fraser University (S. Holdcroft)	Hydrocarbon ionomers for thin film studies have been provided
Chemours	Membrane durability & characterization
Umicore	MEA Testing

Entity	Scope of collaboration
Ionomr	Hydrocarbon membranes durability testing, and samples being provided
IUPUI	Catalyst Testing, microscopy
Advanced Cooling Technologies Inc (ACT)	Discussions related to novel fuel cell heat rejection
pH Matter	Supplied catalyst powders and tested MEAs for microscopy evaluation
IMMORTAL	Durability discussions and presentations
Bar-Ilan University	Doped carbon supports
University of Delaware (Ajay Prasad, Suresh Advani)	Radical Scavenger Development
Purdue University	Modeling of catalyst structures

International Durability Working Group (i-DWG)

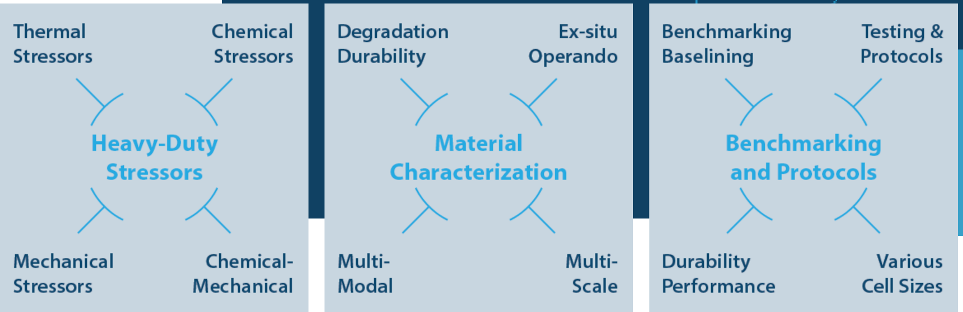
International Durability Working Group (iDWG)

12 Countries
from America, Europe, and Asia

64 Institutions
representing governments, universities, industry and labs

164 Researchers
facilitating data sharing, exchanging materials, promoting AST development

M2FCT | iDWG



with representation from the US, European Union (EU), Japan, and Korea to better coordinate international efforts currently underway to help commercialize fuel cells for trucks and heavy-duty applications.

https://millionmilefuelcelltruck.org/idwg

Future Work

M2FCT consortium aimed at delivering MEAs and components that meet 2.5 kW/gPGM power (1.07 A/cm^2 current density) at 0.7 V

- FOA Support
  - Continue and expand support for existing and future FOA projects
- Analysis
  - Refine models, characterization, and diagnostics for heavy-duty operating conditions
    - Incorporate various component durability in system analysis
- Discretionary
  - Hold new discretionary project call
- Outreach and communications
  - Continue AST working groups
  - Focus on website development and outreach including to MSIs

- Material Development/Characterization
  - Scale-up synthesis of best-of-class catalyst to 10 g batches and integrate into large active area MEAs
  - Improve catalyst activity and durability
    - Increasing ordering degree of Co with new platinum cobalt manganese catalyst to get increased catalyst stability
    - Understand the impact of Co leaching
    - Accurate measurements of catalyst state
    - Examine novel supports such as ZIF and MOFs
    - PtCo intermetallics melamine and melamine-based polymers
  - Ionomer, Membranes
    - Develop non-fluorinated hydrocarbon ionomers including optimizing MW, IEC, formability, etc.
    - Baseline critical material properties including transport properties, mechanical properties, and water uptake
    - Incorporate novel radical scavenger/additives
    - Evaluate effect of block length and ion-exchange capacity of multi-block co-polymers on ionomer properties and electrode structure

Future Work

- Integration
  - Novel electrode structures
    - Integrate new catalysts with advanced electrode designs to improve performance and durability
    - Pyramid like structures for CANE and Array electrodes and understand impact of ionomer swelling and manufacturing limitations
    - Catalyst-layer designs that are more membrane benign
    - Effects of structured cracks
  - Catalyst layer studies
    - Catalyst ink to structure formation models including process variables and mechanism discovery
    - Low-dose, cryo-electron microscopy imaging of ionomer distribution in epoxy-free electrodes with varying support and ionomer type
    - Understand ionomer impacts including local and layer distributions and coverage
  - Multi-scale cell models that account for carbon placement and CL structure
- Durability
  - AST development
    - Durability test of 50 cm^2 MEAs with M2FCT components under the newly developed M2FCT MEA AST
    - Collaborate with industry partners and analyze EOL stack MEAs
    - Determine acceleration factors for membrane lifetime including hydrocarbons
    - Determine applicability of AST with HDV FC stack
    - Understand potential overestimation of degradation mechanisms
  - Ionomer/membrane durability
    - Evaluate ionomer degradation/movement including hydrocarbons
    - Perform micro-electrode and micro-cavity-electrode studies to illustrate role of ionomer in catalyst durability
    - Perform fatigue testing of membranes and MEAs (before and after durability testing) to assess membrane state of health
    - Measure electrode wettability changes
  - Bipolar Plates
    - Examine degradation rates and mechanisms and impact of iron and other dissolution products on membrane lifetime
  - Modeling
    - Develop holistic MEA AST model including membrane chemical/mechanical degradation and Pt dissolution and movement
    - Improve physics-based EIS model to track various resistances

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

Summary

- Relevance/Objective
  - Optimize performance and durability of fuel-cell components and assemblies for heavy-duty applications
- Approach
  - Synergistic combination of modeling and experiments to develop materials, optimize component properties
- Technical Accomplishments
  - Analysis updated with performance and durability for new SOW M2FCT catalysts
    - Includes operating conditions, T, UPL, RH in modeling test matrix
    - ECSA, ECSA Loss Rate, ORR Kinetics, CCL O_2 Transport
  - Durability measurements at projected heavy-duty loadings
    - Developed more durable radical scavengers; evaluated catalyst layer ionomer degradation
    - Refined ASTs for Heavy-Duty Vehicle applications; defined acceleration factors
  - Integration
    - Connecting Ionomer-Ink-Catalyst Layer; experimental matrix and modeling
    - Demonstrated defect-free membrane coatings
  - Material Developments
    - Showing continuous improvement in catalyst performance and durability; scaling up of two M2FCT developed catalysts
    - Advanced ionomers and membranes for durable high-temperature operations
- Future Work
  - Develop the knowledge base and implement towards novel integration and improved durability and support FOA projects

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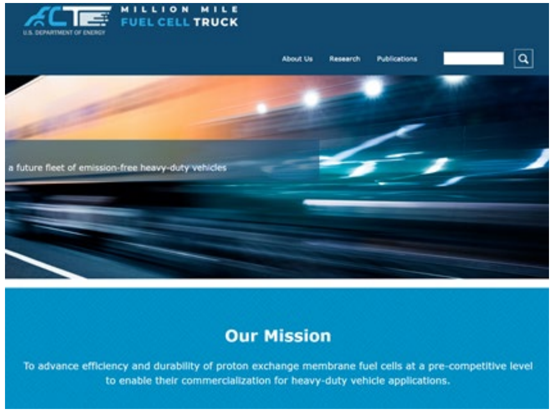
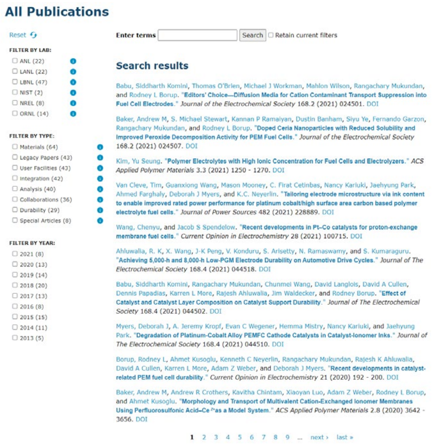
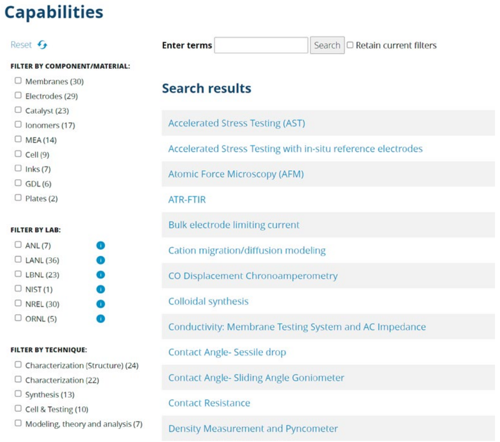
Main Page



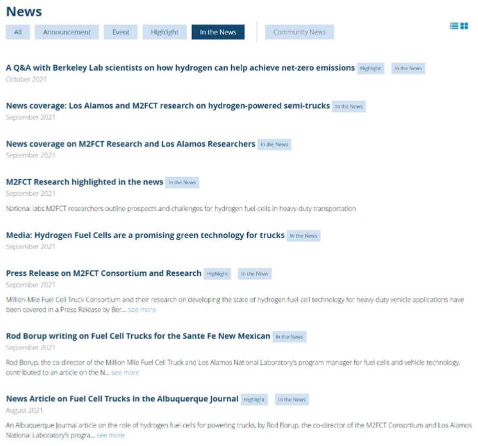
Publications



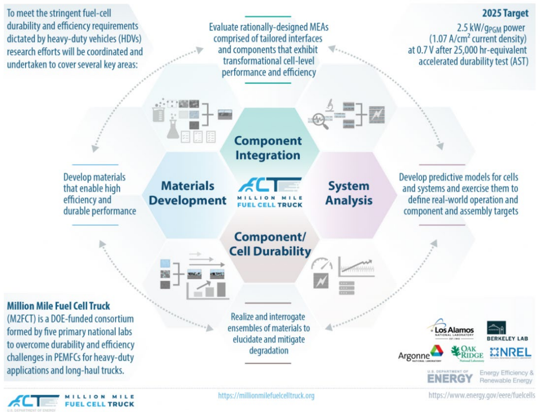
Capabilities



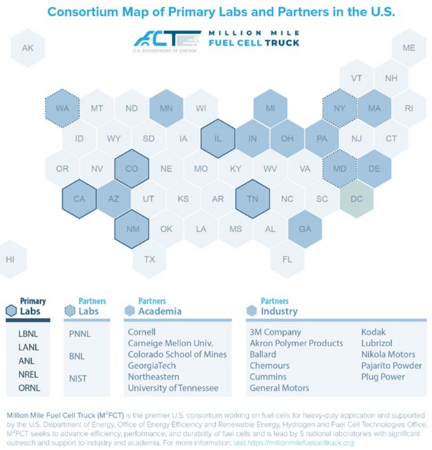
News



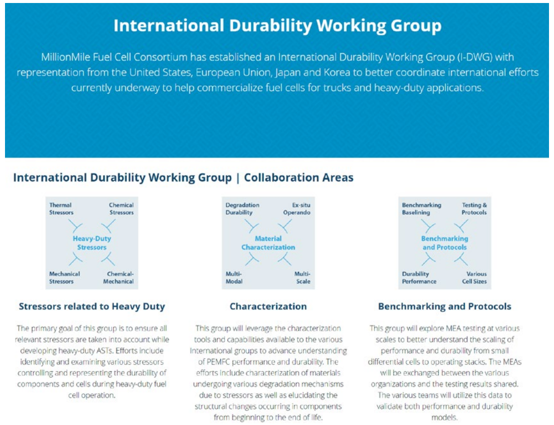
Research



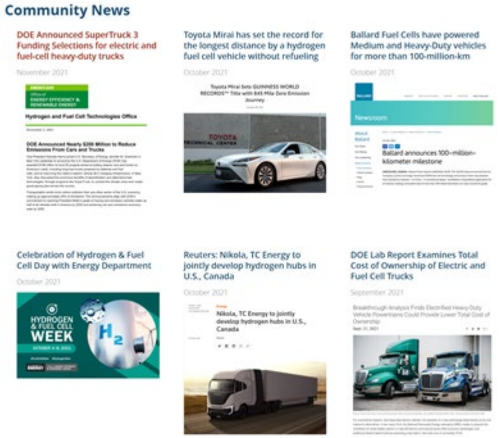
People and Partners



Outreach: Working Groups



Community News



Acknowledgements

DOE EERE Hydrogen and Fuel Cell Technologies Office

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