U.S. DEPARTMENT OF ENERGY

MILLION MILE FUEL CELL TRUCK

M2FCT: Million Mile Fuel Cell Truck Consortium

DOE Hydrogen Fuel Cell Technologies Office
2022 Annual Merit Review and Peer Evaluation Meeting
June 6–8, 2022

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

Deputy Directors: Rajesh Ahluwalia (ANL), Rangachary Mukundan (LANL), 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
- Project end date: 09/30/2025

Budget

- FY20 project funding: $10M
  - $1.5M Effort to Support FOAs
  - 5-year consortium with yearly milestones & Go/No-Go

Partners/Collaborations

- DOE DE-FOA-0002044:
  - GM, Nikola, Carnegie Mellon
- DOE DE-FOA-EE0009244:
  - 3M, Lubrizol, Nikola, UT Knoxville
  - Cummins, Plug Power
- Discretionary Project Additions
  - UCI, UCM, SUNY Buffalo, Drexel, FIU
- No-cost collaborations

Heavy-Duty Transportation (2025)

- Durability: 25,000 hour lifetime
- 68% peak efficiency
- $80/kW fuel cell system cost
- **Overall Target:** 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

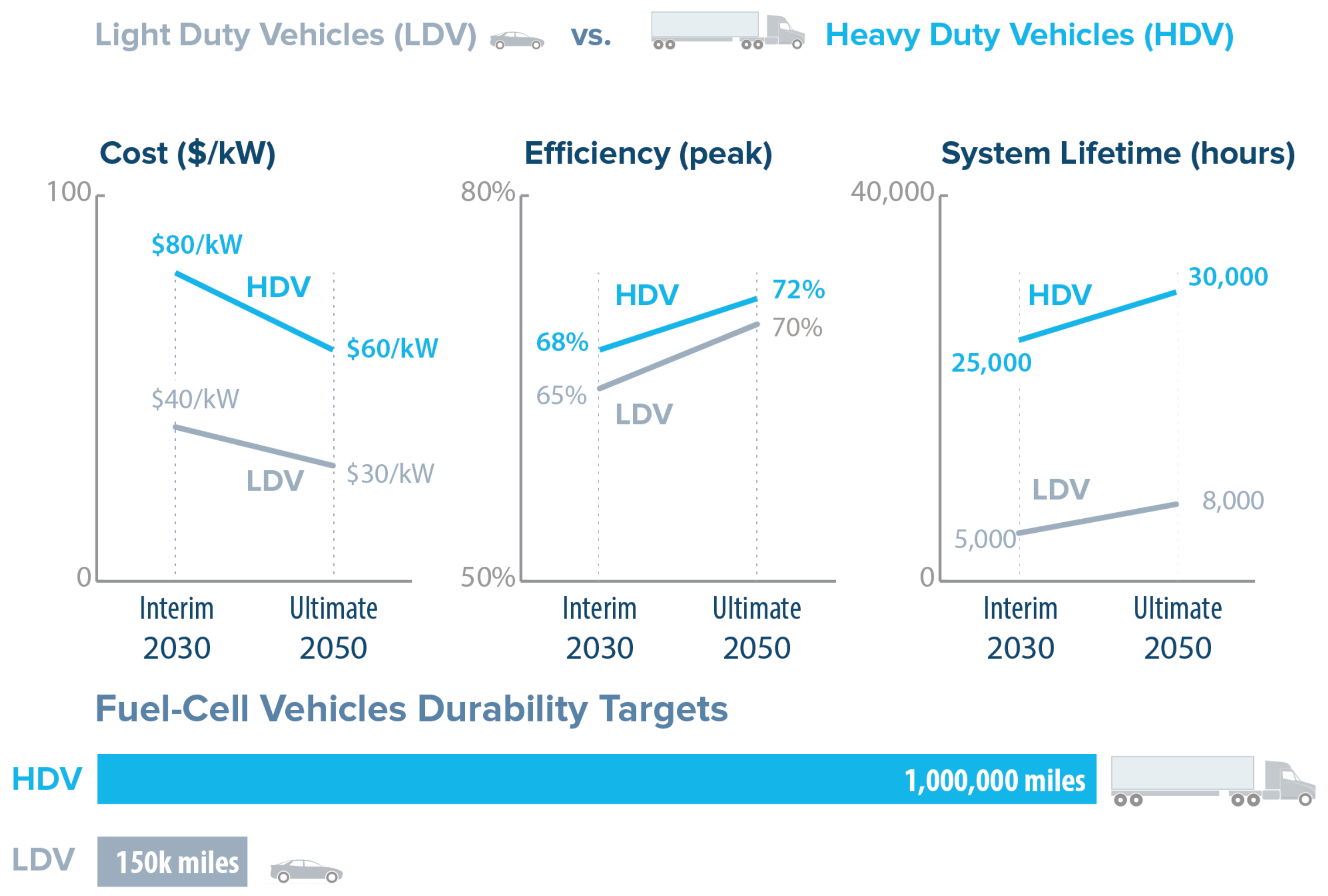
Heavy-Duty Transportation (2030)

- Durability: 30,000 hour lifetime
- 72% peak efficiency
- $60/kW fuel cell system cost

M2FCT focuses on fuel-cell trucks that demand a greater emphasis on system efficiency and longer lifetimes

The change in focus from light-duty (LDV) to heavy-duty vehicles (HDV) exacerbates durability and efficiency challenges for fuel cells, requiring material and system innovations that enable new classes of hydrogen vehicles that take advantage of the high efficiency, power density and scalability of this technology.

DOE Targets for Fuel-Cell Vehicles



Source: DOE HFTO Program Record #19006 | Cullen,...,Kusoglu, *Nature Energy* 6, 462–474 (2021)

Notes: Current target of $50/kW for LDV is based on 100,000 units/year. HDV Targets are for Class 8 Tractor-Trailers.
Ultimate targets are based on simple cost of ownership assumptions and reflects anticipated timeframe for market penetration.

M2FCT Partners: National Labs, Universities, Industry

"Team-of-teams" approach for rapid feedback, idea development, information exchange, resulting in an effort that is more than the sum of its parts

MEA Projects

gm, Carnegie Mellon University, NIKOLA

Membrane Projects

Lubrizol, 3M, NIKOLA

Stack Projects

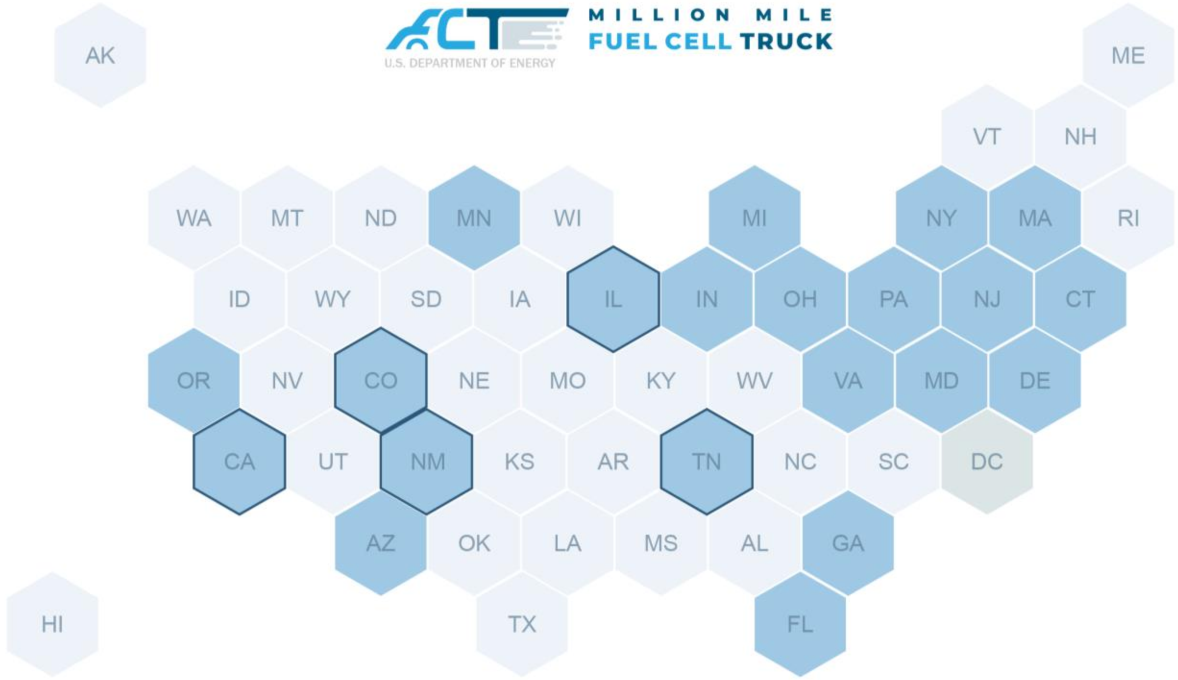
Cummins, PLUG POWER

Bipolar Plate Projects

Raytheon Technologies, NEOGRAF SOLUTIONS, TreadStone Technologies, Inc., gm, PLUG POWER

Air Management Projects

CATERPILLAR, EATON, R&D Dynamics Corporation, MAHLE Driven by performance



LABS

Primary Labs:
- LANL
- LBNL
- ANL
- NREL
- ORNL

Partners:
- PNNL
- BNL
- NIST

ACADEMIA

Partners:
- Cornell
- Carneige Mellon Univ.
- Colorado School of Mines
- Drexel University
- Florida International Univ.
- GeorgiaTech
- Northeastern
- UC Irvine
- UC Merced
- University at Buffalo
- University of Tennessee

INDUSTRY

Partners:
- 3M Company
- Akron Polymer Products
- Ballard
- Chemours
- Cummins
- Caterpillar
- Eaton
- General Motors
- Kodak
- Lubrizol
- Mahle
- Nikola Motors
- Pajarito Powder
- Plug Power
- NeoGraf Solutions
- R&D Dynamics Corp
- Raytheon Technologies
- Strategic Analysis
- TreadStone Technologies

Main Laboratories

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

Affiliate Laboratories

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

Discretionary Funds Project Partners

Drexel University, University of California Merced, Buffalo State The State University of New York, FIU Florida International University, UCI

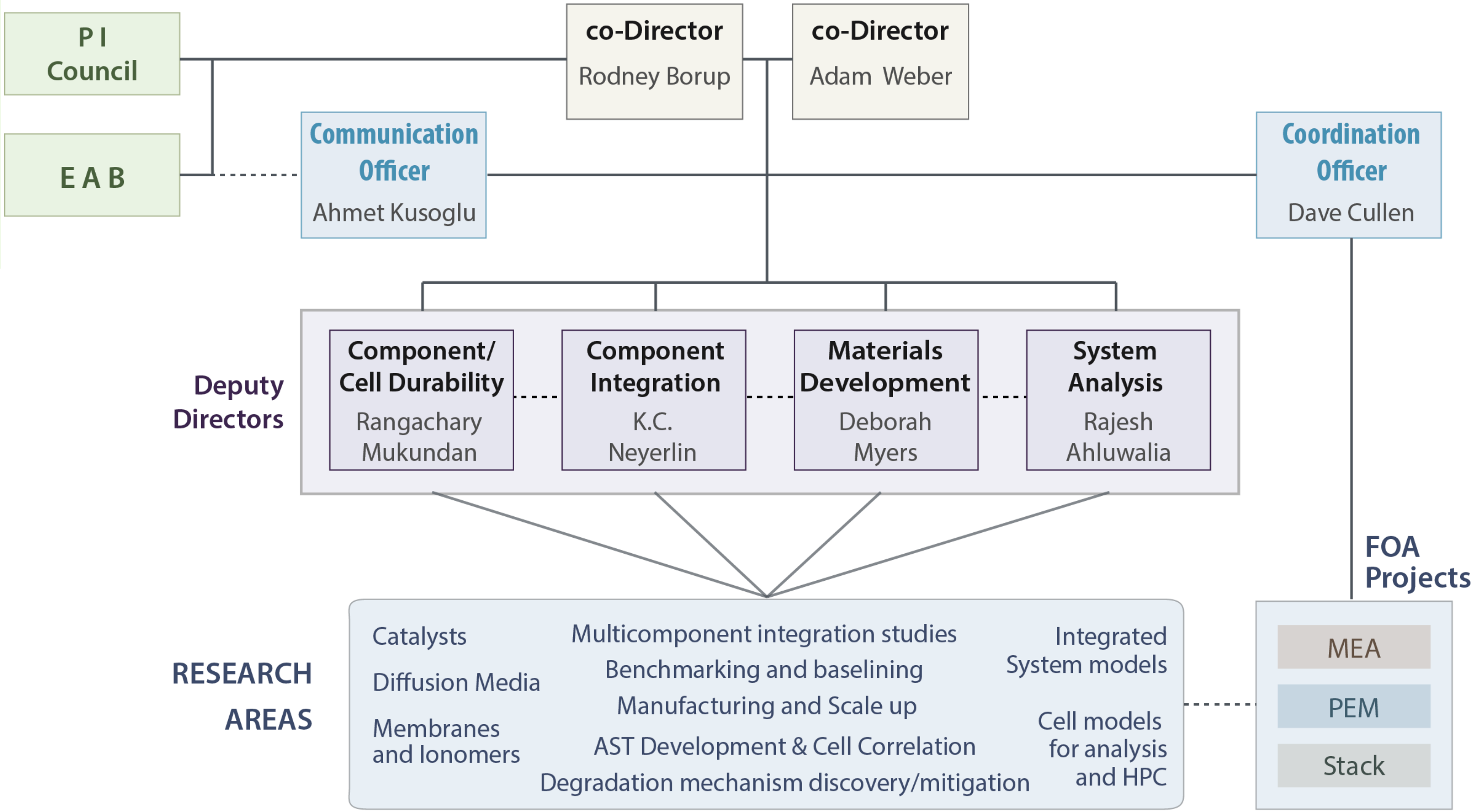
Organization Chart

EAB: External Advisory Board

Morgan Andreae – Cummins
Christian Appel – Nikola
Simon Cleghorn – W.L. Gore
Ken Howden – DOE (21st Cent. Truck)
JoAnn Miliken – Retired DOE
Mike Perry – Retired UTRC
Gary Robb – Retired GM & Hyzon

National Lab PIs

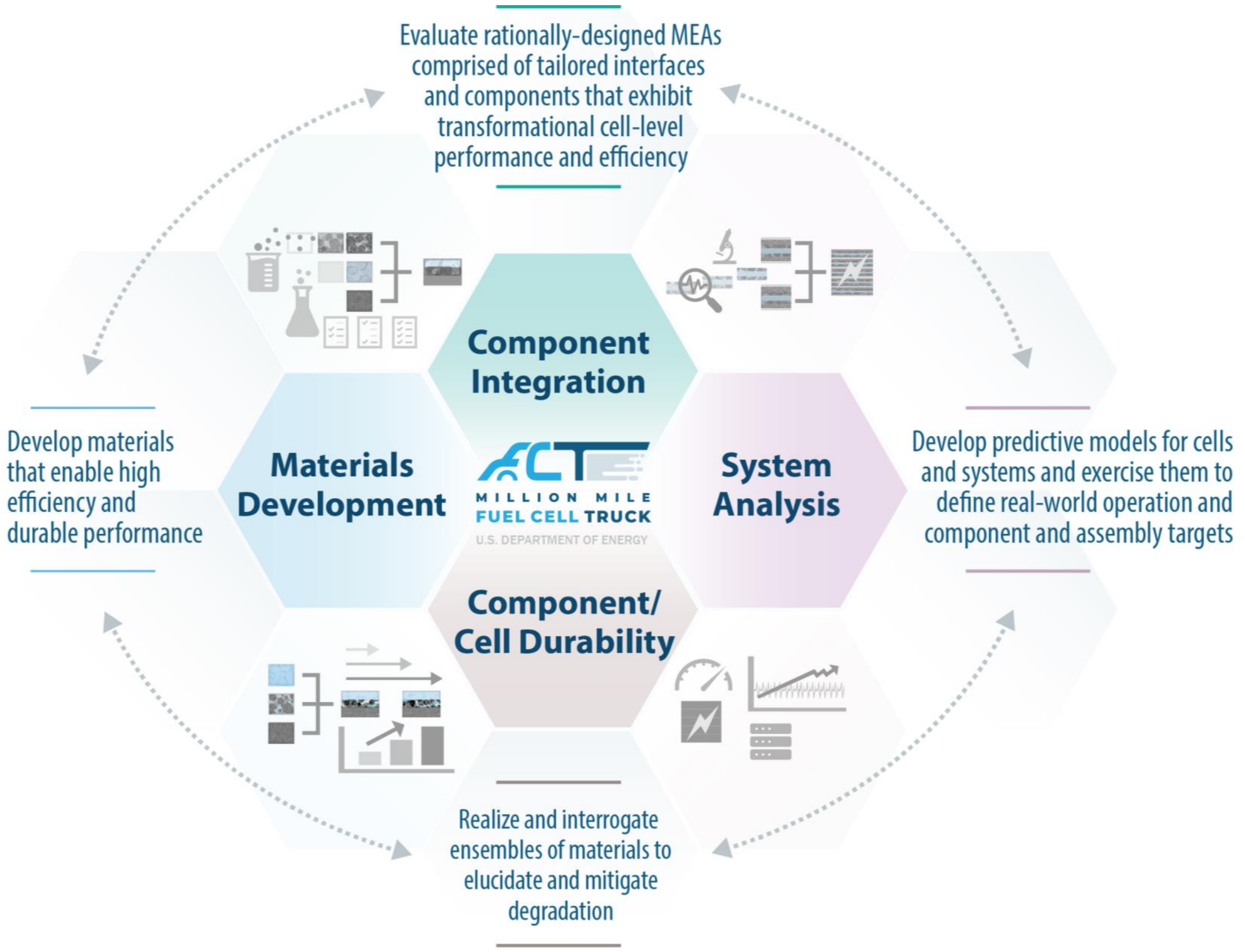
1. Rajesh Ahluwalia
2. Rod Borup
3. Dave Cullen
4. Dan Hussey
5. Hans Johansen
6. Yu Seung Kim
7. Ahmet Kusoglu
8. R. Mukundan
9. Debbie Myers
10. KC Neyerlin
11. Bryan Pivovar
12. Kotaro Sasaki
13. Alexey Serov
14. Yuyan Shao
15. Jacob Spendelow
16. Voja Stamenkovic
17. Mike Ulsh
18. Adam Weber



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.



M2FCT: *Originally* NOT Included

May be included in future FOAs as part of the M2FCT:

- Modular standardized stack and system designs to lower cost across heavy-duty applications
- Balance-of-plant components, including **air management** and low-cost power electronics
- **Advanced bipolar plates/coatings**
- Addressing unique, heavy duty application specify durability challenges, including saline contamination for marine, power density and ruggeridization for rail, and specific energy for aviation

NOT Included:

- PGM-free catalysis
- Fuel quality studies
- Fuel cells other than PEM (e.g., alkaline fuel cell technologies, solid oxide fuel cells)

<u>Bipolar Plates</u>

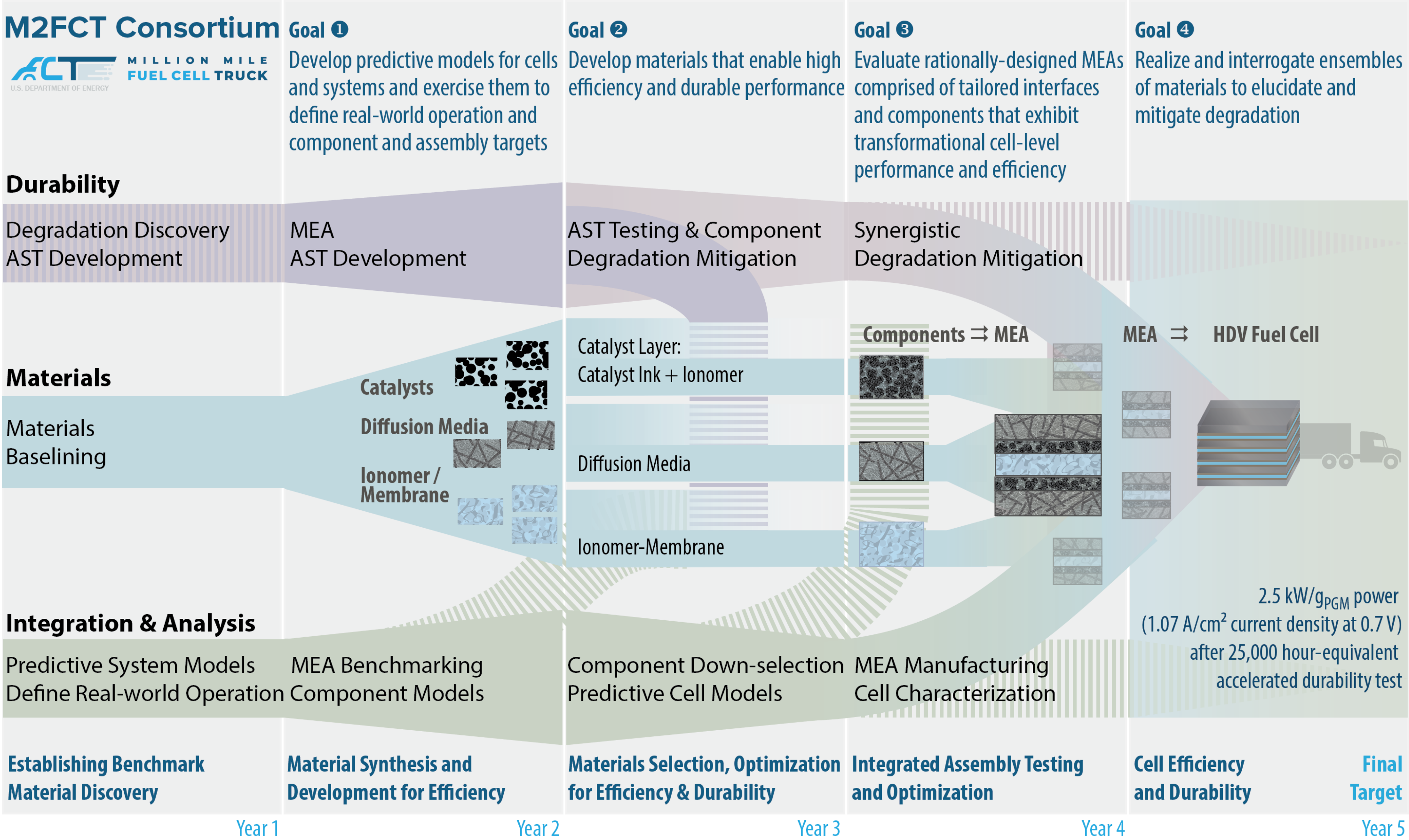
- M2FCT interactions and SOW related to BPP projects under discussion
- Durability AST projecting HDV life needed

Bipolar Plate Projects

Raytheon Technologies
NEOGRAF SOLUTIONS
TreadStone Technologies, Inc.
gm
PLUG POWER

Air Management Projects

CATERPILLAR
EATON
R&D Dynamics Corporation
MAHLE Driven by performance


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

FY22 Milestones

Milestone Name / Description and Criteria	Quarter	Responsible
Name: Analysis of Efficiency and Fuel Consumption Report of analysis results related to efficiency and fuel consumption over the entire vehicle lifetime with correlation to MEA degradation (LANL, ANL, NREL, ORNL)	Q1	Rajesh
Name: Catalyst dissolution models Convergence of catalyst dissolution models (LBNL, ANL)	Q1	Adam
Name: Drive Cycle Carbon Corrosion measurements Complete NDIR measurements for carbon corrosion for three different drive cycle conditions. Drive cycles will simulate different HDV power sizes (150 kW, 275 kW, 450 kW) (LANL, NREL)	Q2	Rod
Name: Catalyst material Go/No-Go (PNNL, ANL, LANL, NREL, BNL) Demonstrate ≥ State-of-the-Art (Defined by Year 1 Bench-Marking) at 0.8 V on hydrogen-air at 250 kPa, 100% RH, 80°C cell temperature after 90,000 catalyst AST cycles (or equivalent of M2FCT-developed AST) using an MEA with ≤0.3 mg/cm^2 Total PGM loading	Q2*	Debbie

FY22 Milestones

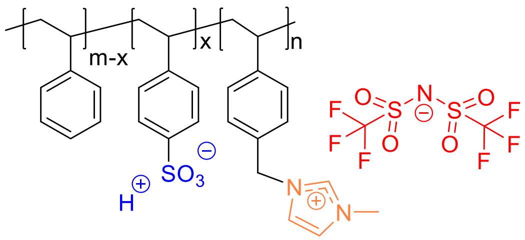
Milestone Name / Description and Criteria	Quarter	Responsible
Name: MEA Fabrication Demonstrate at least 10% improvement in current density at 0.7 V over baseline materials by varying MEA fabrication method, solvent ratio in catalyst-ionomer ink, ionomer type and content, etc. Total PGM loading constrained to 0.3 mg/cm2. MEA test conditions: 88 °C, 2.5 atm, SR: 1.5 cathode/2 anode, 40% RH inlet, simulated integral cell. (All)	Q3	KC
Name: Characterization of Ionomer adsorption Measurement of ionomer adsorption and binding energy (GIXS/QCM/ITC) and characterization of the size and shape of aggregated catalyst clusters in non-aqueous catalyst ink (USAX/WAXS/SAXS). (LANL, LBNL, ANL)	Q3	Ahmet
Name: New AST Protocol Development New AST protocol development for materials not currently covered (e.g., GDL, Ce migration, Metal BPP). (All)	Q4	Rod
Name: Refinement of AST Protocols Catalyst and membranes AST protocols refined for extrapolation to 25,000-hr performance at HDV operating conditions through modeling and testing. (All)	Q4	Mukund

* Can be extended to 24 (Q8) months as no-cost extension

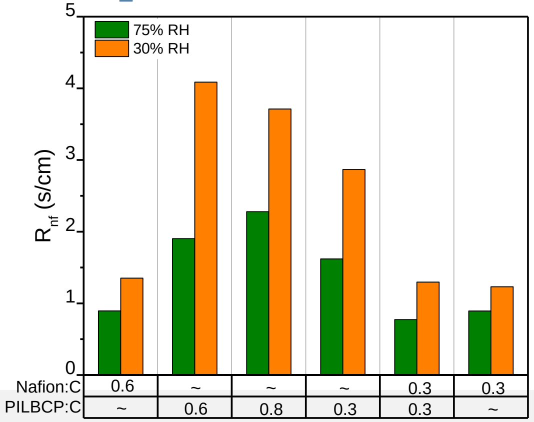
Advanced PILBCP Ionomer Composites for Durable Heavy Duty PEMFCs

Joshua Snyder (PI; Drexel), Yossef Elabd (TAMU), Rui Sun (TAMU)

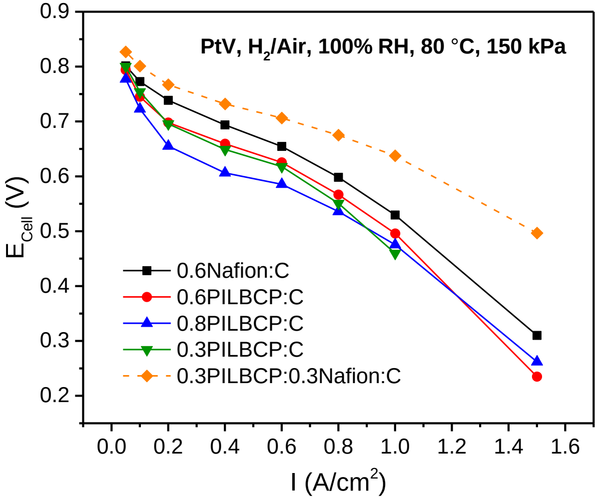
1st generation S-PILBCP



O_2 Transport Resistance



S-PILBCP



- Identification and synthesis of new S-PILBCP chemistries
- Open questions related to polymer structure at catalyst layer relevant thicknesses
- Clearer picture of mechanism of kinetic enhancement
- Batch scale-up
  - Validation of MEA performance for larger batch sizes
- Direct measure of impact of S-PILBCP on catalyst durability
  - In-situ ICP-MS
- Electrode structural characterization

(PILBCP: polymerized ionic liquid block co-polymer)

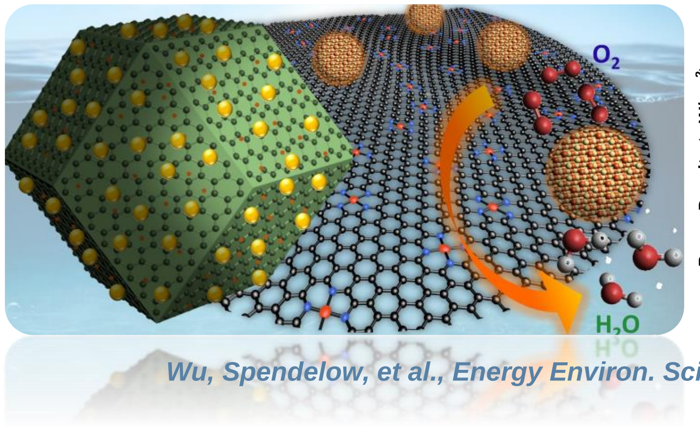
Integrating Highly Durable Carbon Supports and Intermetallic PtCo Catalysts for Heavy-Duty MEAs

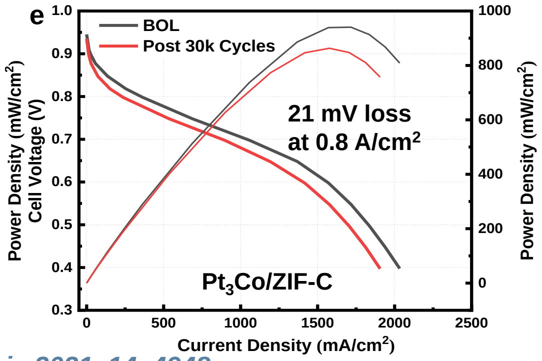
Buffalo State
The State University of New York

Gang Wu

- **Atomically dispersed single iron sites (M-N-C) for promoting Pt and Pt_3Co fuel cell catalysts**

~0.1 mgPt/cm^2 , 150 kPa





Wu, Spendelow, et al., Energy Environ. Sci., 2021, 14, 4948

Integrate the promising PGC/M-N-C support and the ordered PtCo intermetallic nanoparticles includes **four tasks**:

- (**1**) Engineering the PGC/M-N-C supports to uniformly dispersed Pt_3Co intermetallic nanoparticles concerning optimal carbon nanostructure, morphologies, degree of graphitization, and nitrogen dopants to strengthen metal-support interactions;
- (**2**) Controlling ordering structures, particle sizes, and Pt loadings (up to 40 wt.% against carbon) of Pt_3Co intermetallic alloy for optimal activity-stability trade-off;
- (**3**) Optimizing ink preparation and electrode structure for maximum durability and power density;
- (**4**) Evaluating catalyst and support stability for heavy-duty applications.

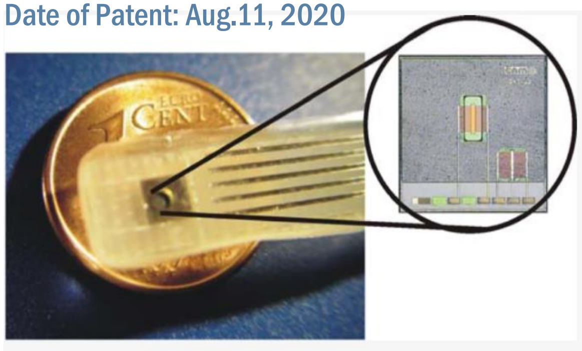
Real-Time Continuous Monitoring of Ionomer Degradation with Ion-Sensitive Field-Effect Transistor (ISFET) Microsensors

Dongmei Dong (PI), Shekhar Bhansali (Co-PI), Tinsley Benhaddouch

Develop F- Sensor as FC Diagnostics Tool

ISFET Electrochemical Sensor:

Patent No.: US 10,739,305 B1
Date of Patent: Aug.11, 2020



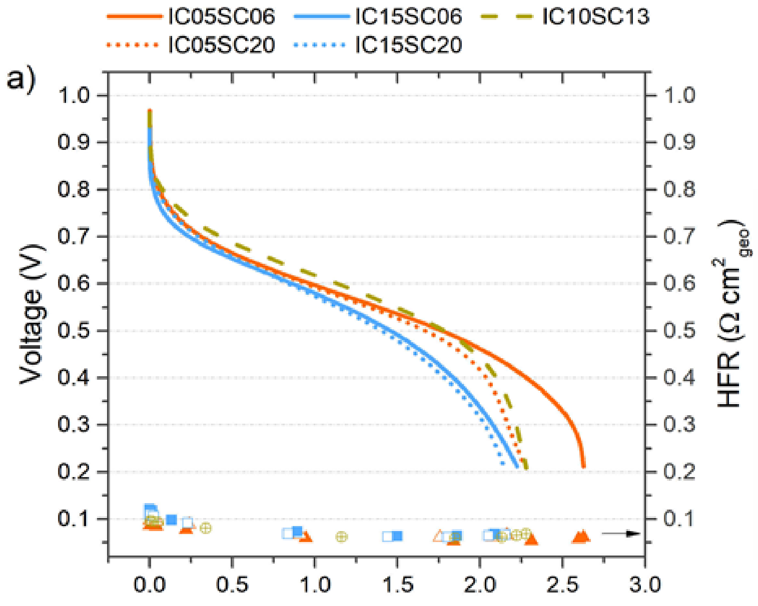
Integrate microsensors to new FC vehicles design

- Develop *Operando* complementary characterizations to existing techniques:
  - Functionalize sensing gate: Hydrogel/MOF
  - Probe membrane degradation rates with increased temporal resolution
- Employ AI/DL architectures for sensor-based predictive maintenance of fuel cells
- Bring a miniaturized/portable/user-friendly specific diagnostics tool for fuel cell degradation quantitative analysis
- Ionomer Degradation:
  - Advanced sensors and spatial/temporal probing diagnostics
  - Membrane degradation rate analysis

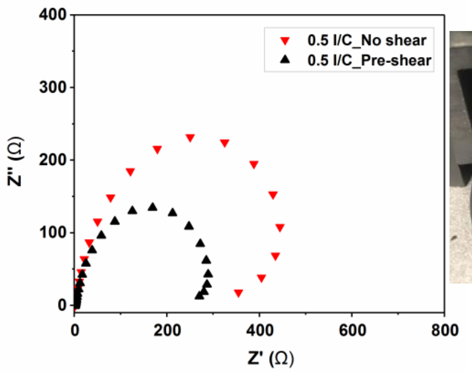
Characterization and Simulation of Interfacial Transport Phenomena in the electrode using Impedance and Imaging Tools

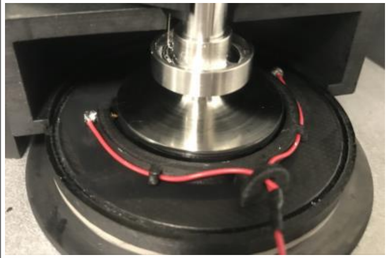
Abel Chuang (PI), Nitul Kakati, Marc Labata, Shirin Mehrazi, Donglei Yang

Rheological study of catalyst ink dispersion.



Develop an in-situ rheo-impedance characterization tool to effectively study the interaction of ionomer and the catalyst.

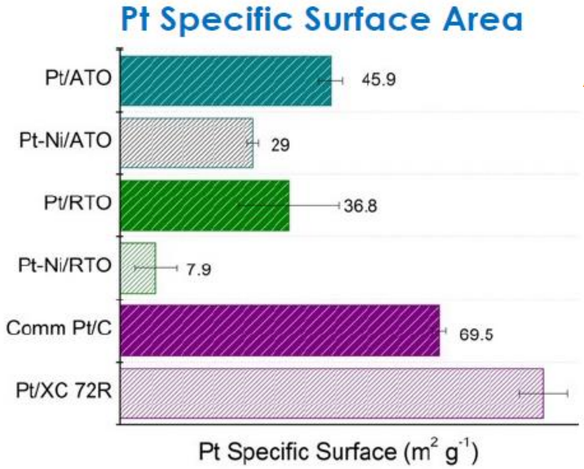




- Apply rheo-impedance to study inks with various constituents, such as solvent, ionomer content, and other additives, for developing a fundamental understanding of the effectiveness of the tool.
- Develop the capability of STEM-EDS study of the catalyst ink in its liquid state by fabricating a graphene liquid cell.
- Perform fuel cell testing to quantify ECSA and proton transport resistance of the catalyst layer and evaluate overall performance.
- Develop 1-D catalyst layer model for performance prediction.

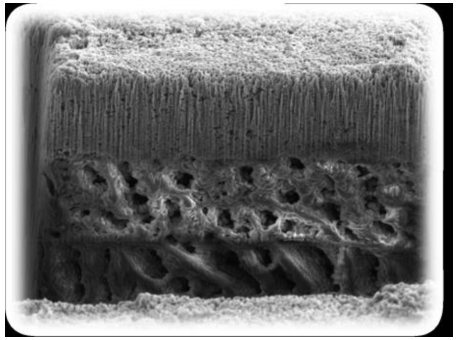
Materials and Design Solutions for PEMFC Durability

Plamen Atanassov and Iryna Zenyuk

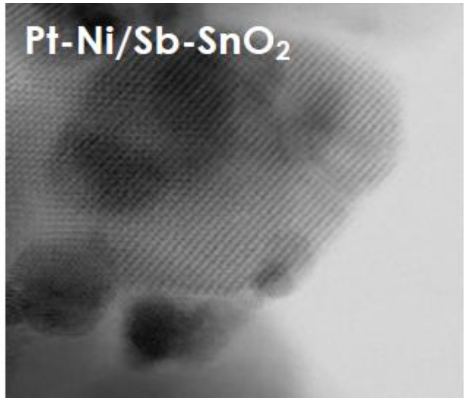


Analysis

Develop degradation analysis based on *Glow Discharge – Optical Emission Spectroscopy* (GD-OES). Establish methodology for structure-to-property relationship with in-house XPS surface analysis and M2FCT FIB-SEM for dept profiling and 3D structure elucidation.



GD-OES and micro-XCT. MEA prior, during (*operando*) and post AST will be evaluated.



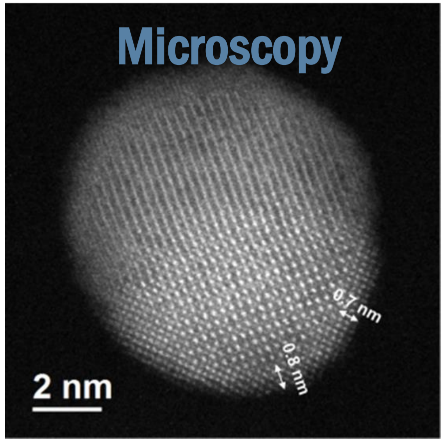
Materials

Develop non-carbon catalyst supports based on highly conductive transition metal oxides Sb-SnO_x and/or Nb-TiO_x families. Evaluate stability 95°C and corrosion resistance. Establish materials/chemical stability at 120°C.

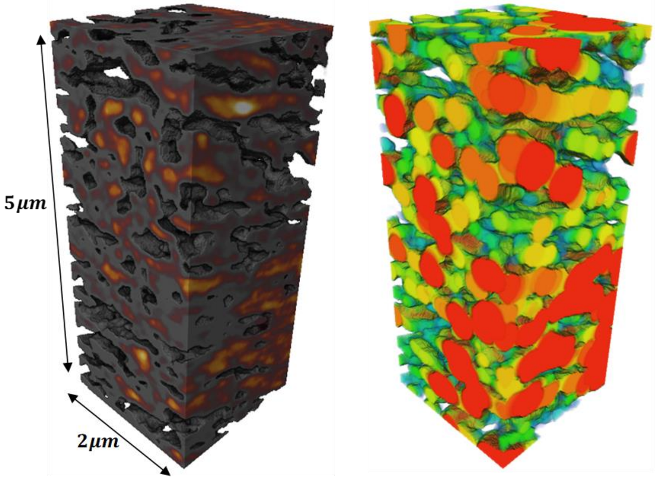
Develop composite carbo/ceramic structures based on integrating those oxide materials with specialty carbons: industrial research samples and/or in-house synthetic carbons.

M2FCT User Facilities Engaged

- Advanced Light Source (ALS)
- Advanced Photon Source (APS)
- Center for Integrated Nanotechnology (CINT)
- Center for Nanophase Materials Sciences (CNMS)
- Los Alamos Neutron Science Center (LANSCE)
- Molecular Foundry
- NIST Center for Neutron Research (NCNR)
- High Flux Isotope Reactor (HFIR)
- Spallation Neutron Source (SNS)

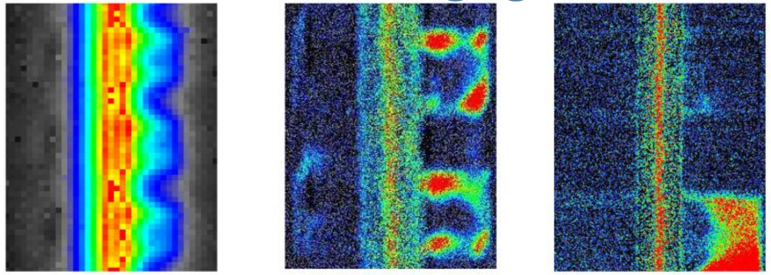


XCT



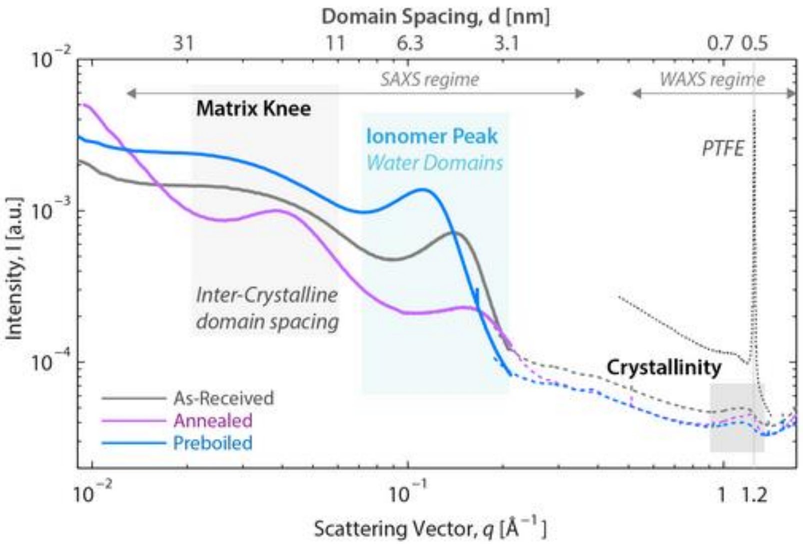
Spatial distribution of agglomerates

Neutron Imaging

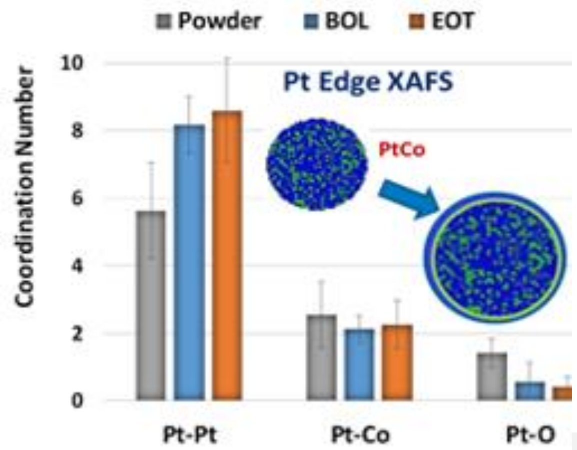


250 μm 25 μm 10 μm

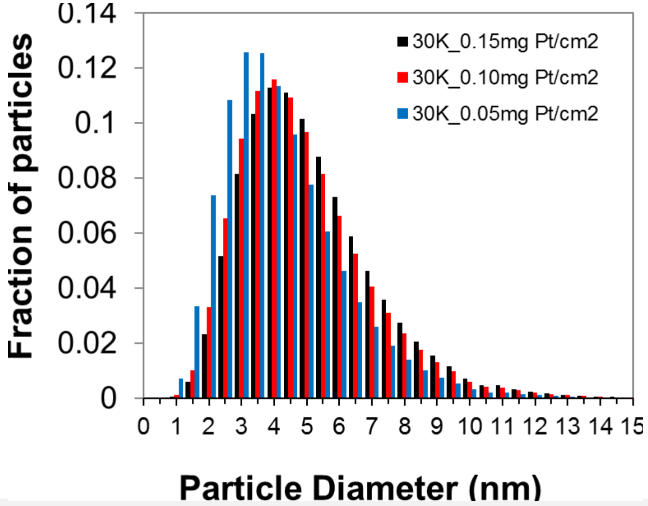
SAXS-WAXS



XAFS



SAXS

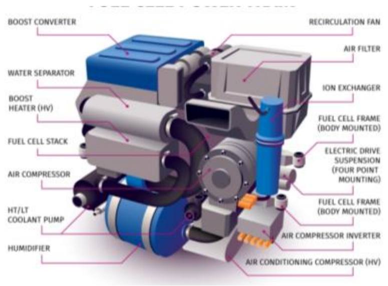


Analysis

Baselining Materials, Performance, Durability and Cost of Fuel Cell Systems for Heavy Duty Trucks

Overall Target: 2.5 kW/g_{PGM} PGM utilization at 0.7 V (1.07 A/cm^2 current density) after 25,000 hour-equivalent accelerated durability test, \$80/kW_e FCS cost

Select stack materials, air management subsystem, fuel management subsystem, heat rejection subsystem, and controls



1. Define system configuration

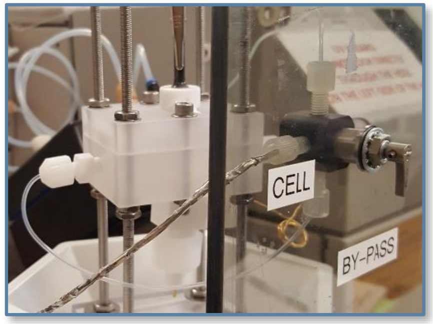
Select system and component sizes

2. Identify Class-8 truck duty cycle

Measure and model cell performance and degradation for leading SOA materials

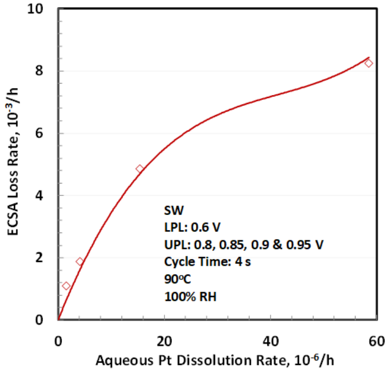
3. Baseline materials on ASTs

Measure and model cell performance and degradation for leading SOA materials



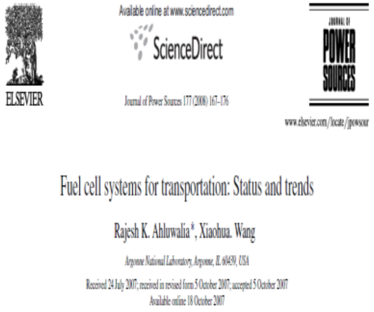
4. Characterize degradation mechanisms

Correlate H_2/air cell and aqueous data



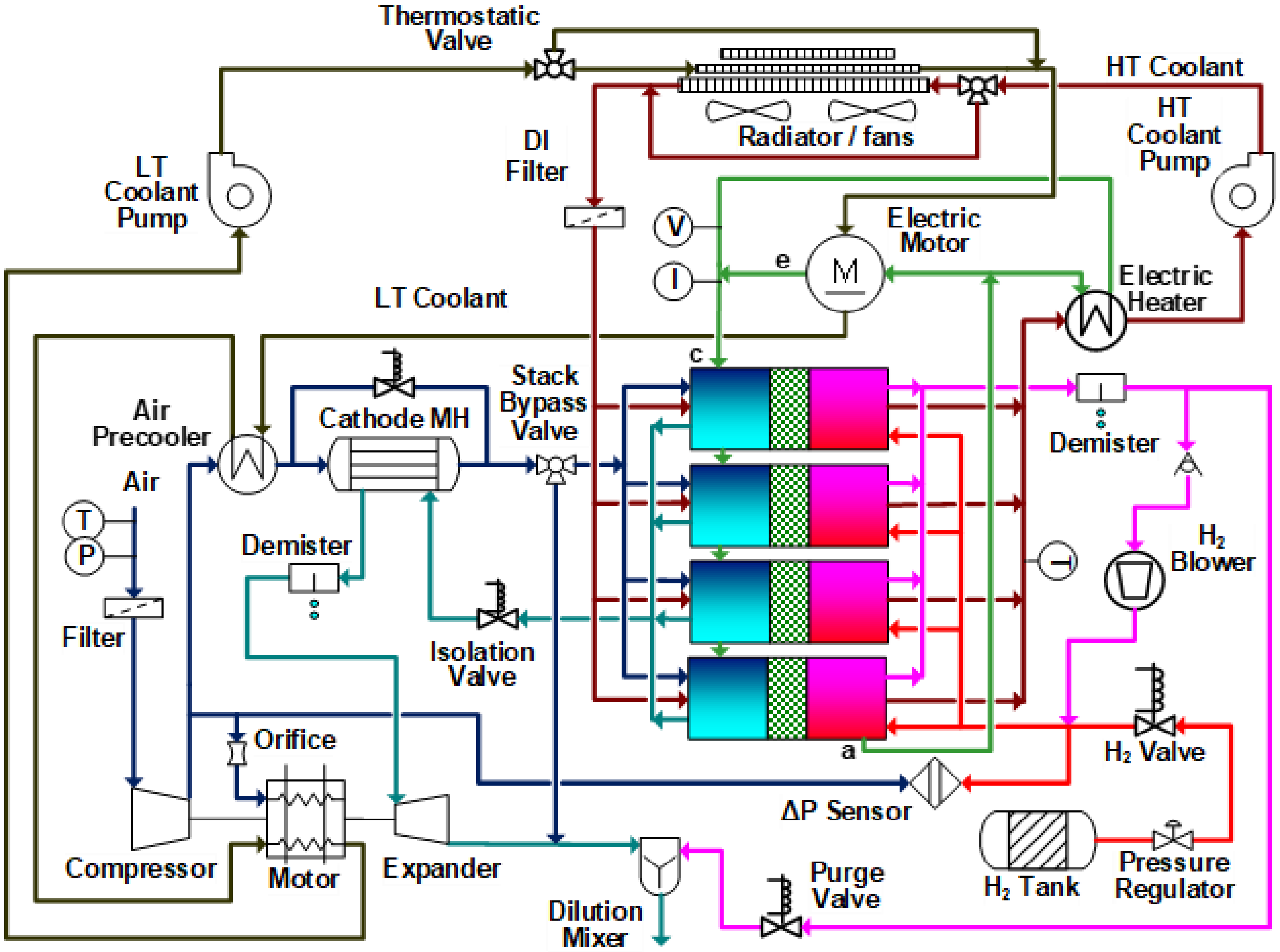
5. Measure degradation rate in H_2/air

Compare performance, durability and cost against M2FCT targets



6. Run simulations

M2FCT Reference Fuel Cell System for Heavy-Duty Trucks



Salient Features

- 275 kW net (70-kWh ESS) at EOL
- Multiple stacks: 4
- Electrodes
  Cathode: a-Pt/C, 0.25 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
- Single air system with expander
- Single anode system with recirculation blower
- Cathode humidifier: Cross-flow with high flux WVT membrane
- Rated power conditions at EOL: 2.5 atm, 90ºC, 700 mV
- Control valves for startup and shutdown, cold start and OCV

21st Century Truck Partnership Long-Haul Duty Cycle

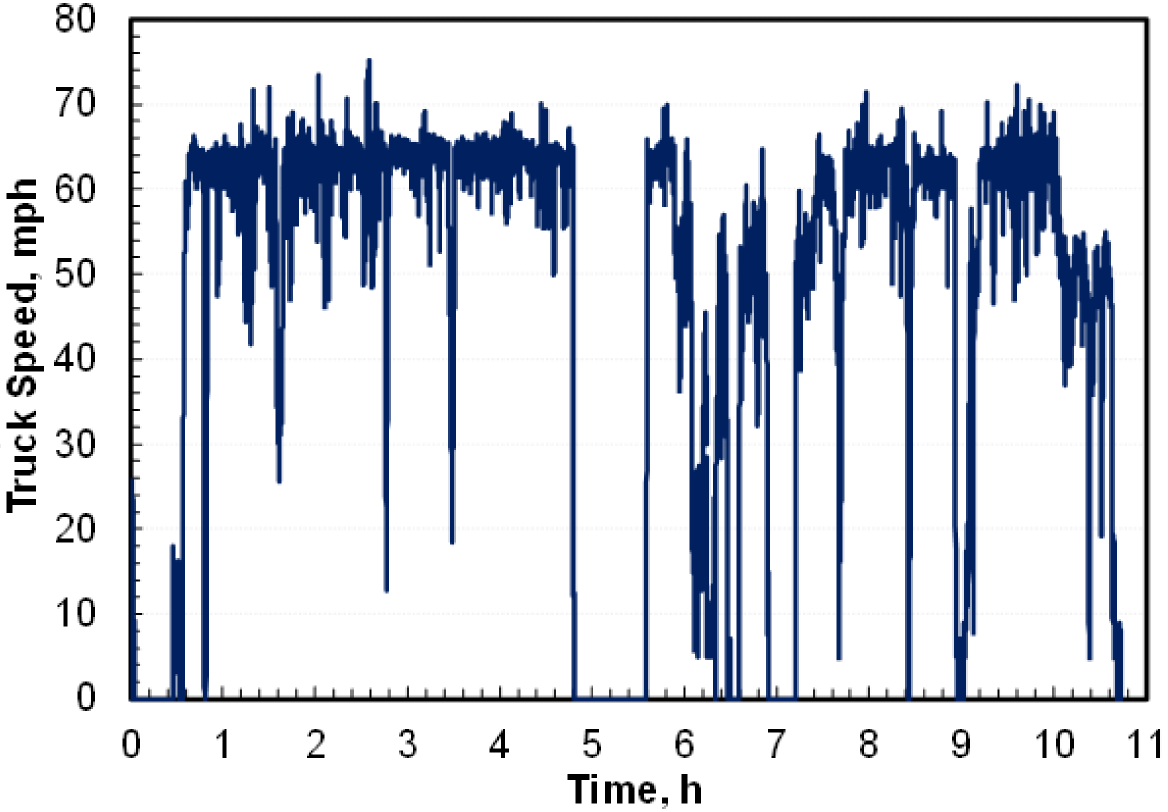
Real World Drive Cycle

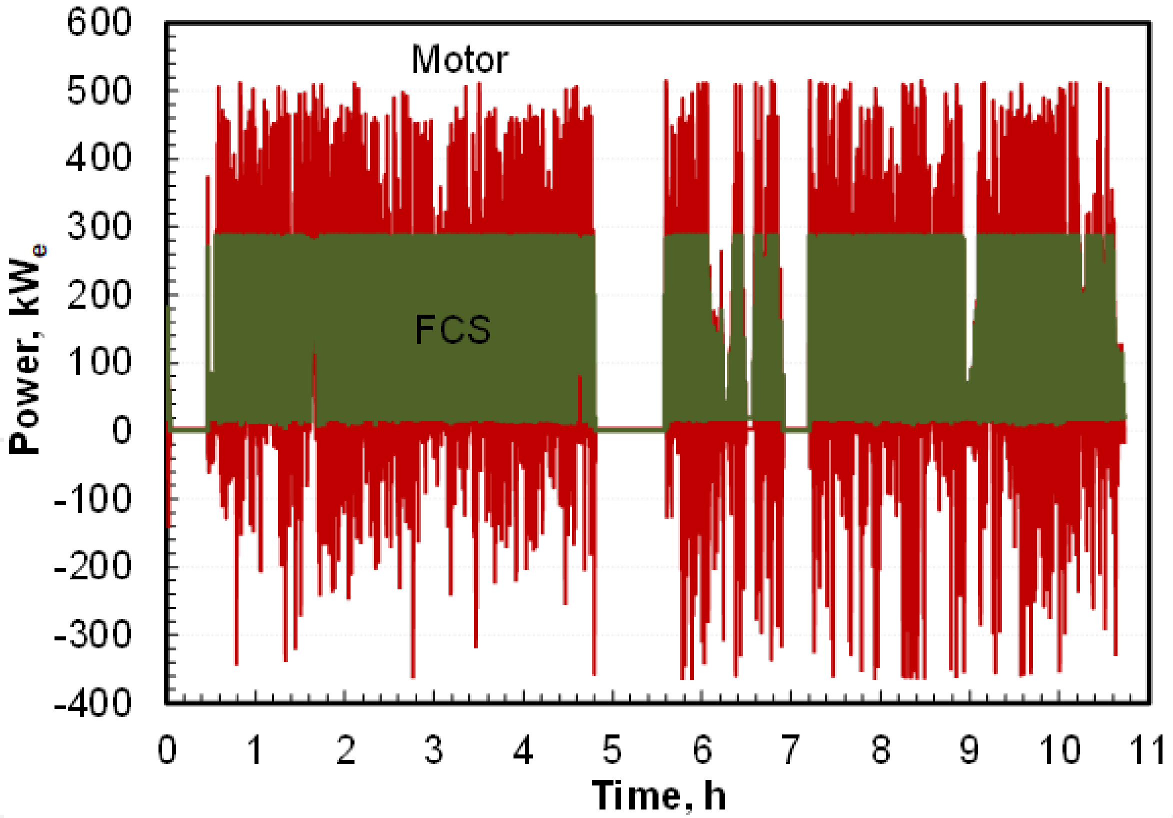
- Gross Vehicle Weight (GVW): 36.3 MT (80,000 lbs)
- 10.5-h drive segment, 3 extended idles with engine off
- Truck speed around 65 mph

Motor Power

- Maximum power on drive cycle: 500 kW_e
- Motor input power on 6% grade at 30 mph, 36.3-MT GVW: 400 kW
- Motor input power at 65-mph, 0% grade: 160 kW

- Selected 400-kW_e hybrid platform to meet power demand on 6% grade at 30 mph subject to heat rejection constraint: 275 kW_e FCS, 70 kWh ESS



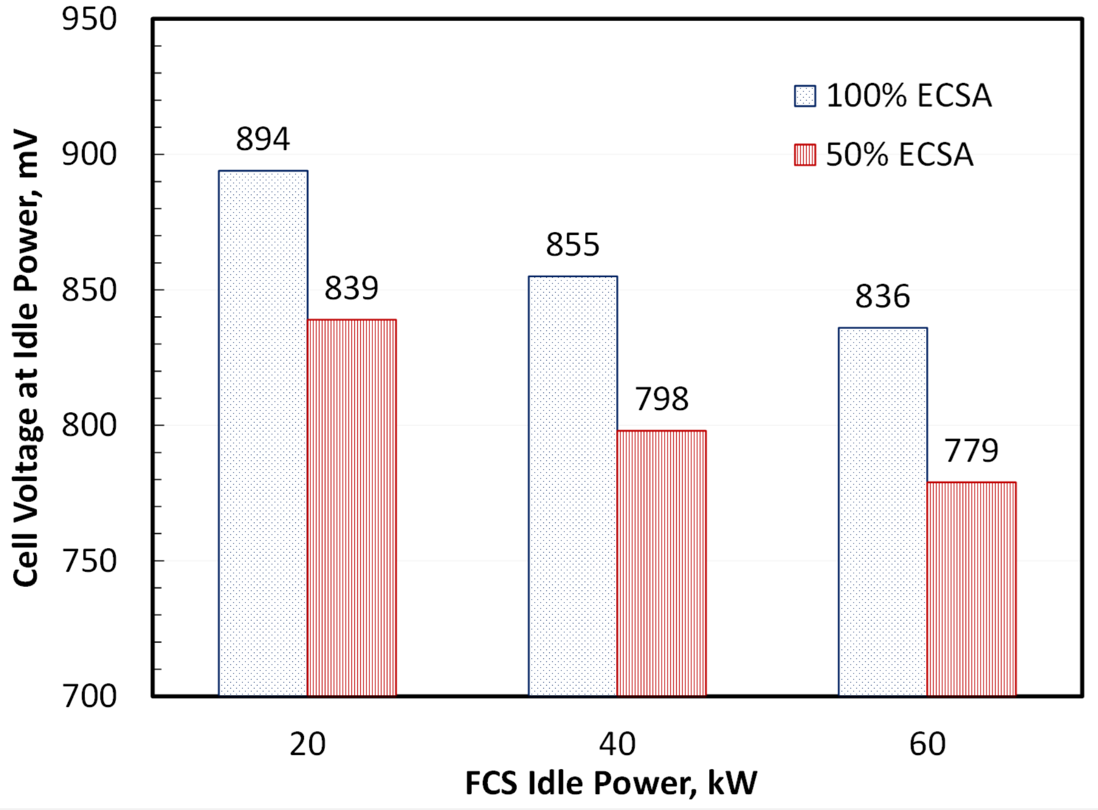


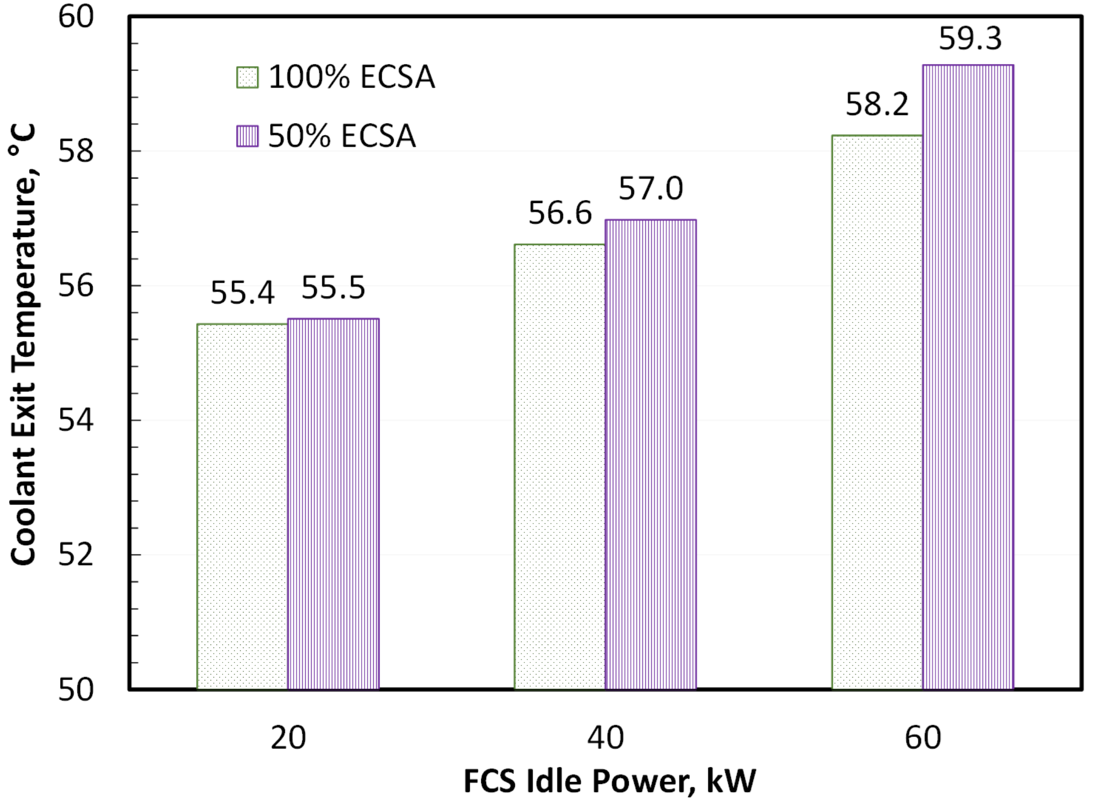
Autonomy simulation results provided by Vincent Nicolas Freyermuth, Argonne National Laboratory

Key Parameters that Control Electrode Lifetime: Idle Power and Coolant Exit Temperature

Target average ECSA loss rate parameter for 50% ECSA Loss after 25,000 h: 40×10^{-6}/h (0.04/1000 h)

- Peak cell voltage at BOL: Below 835 mV
- FCS idle power: 60 kW
- Coolant exit temperature during idling (BOL): 58°C
- Idle fan power: 750 W for 60 kW FCS idle power, 12.5 W(fan)/kW(FCS)

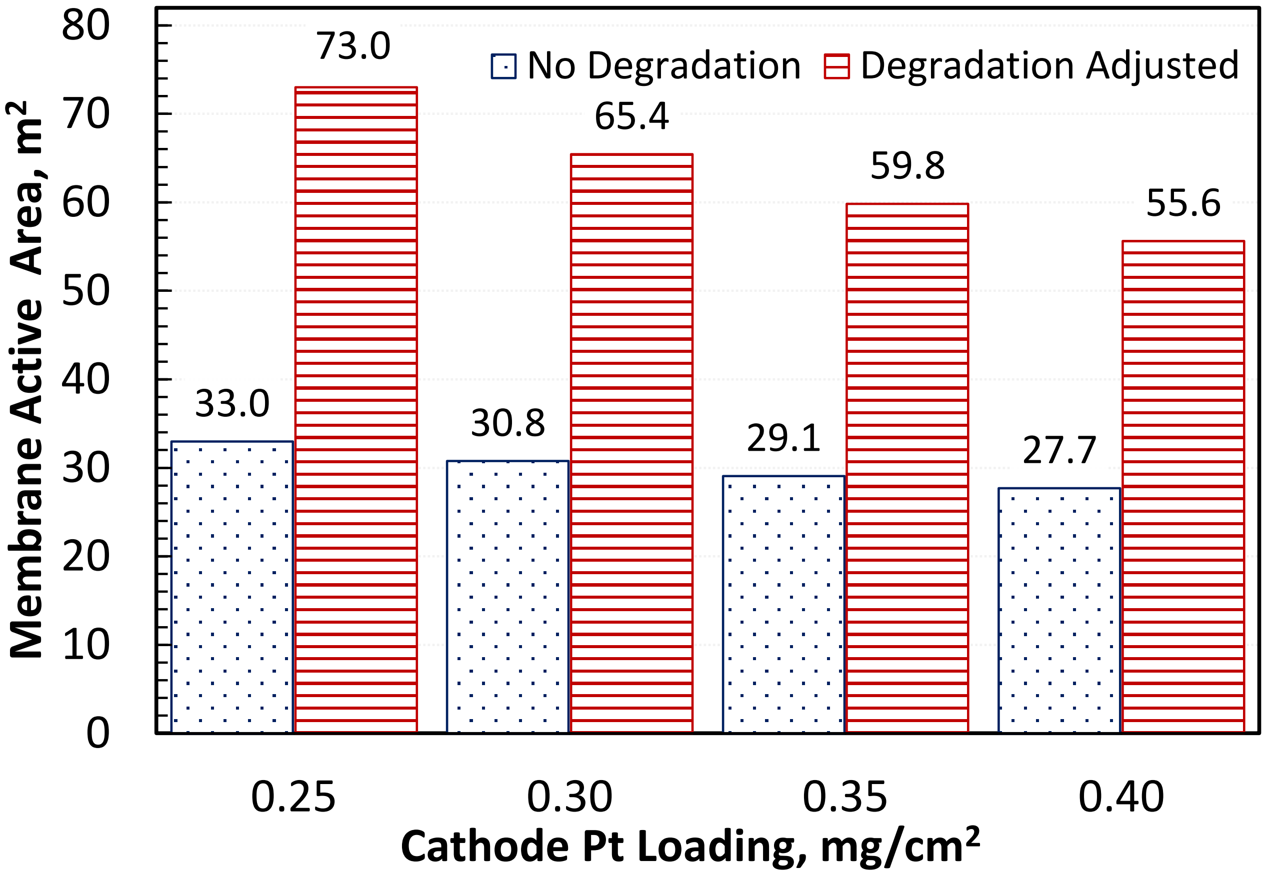




Degradation Adjusted Performance – Total Active Membrane Area

Required membrane area and allowance for degradation to reach 25,000-h electrode lifetime are smaller for higher Pt loading in cathode electrode

- **0.25 mg/cm² Pt loading: 73 m² membrane area, 120% allowance for catalyst degradation**
- **0.40 mg/cm² Pt loading: 59.4 m² membrane area, ~100% allowance for catalyst degradation**



Electrode Lifetime

- **EOL Definition: Produce 275 kW_e at 0.7 V, 2.5 atm, 90°C coolant exit temperature, 1.5 cathode stoichiometry**
- **ECSA Loss at EOL on Class-8 Drive Cycle from degradation Model: 56.5%**
- **BOL Conditions from Performance Model: Rated power at 0.780 V and heat rejection conditions listed above**

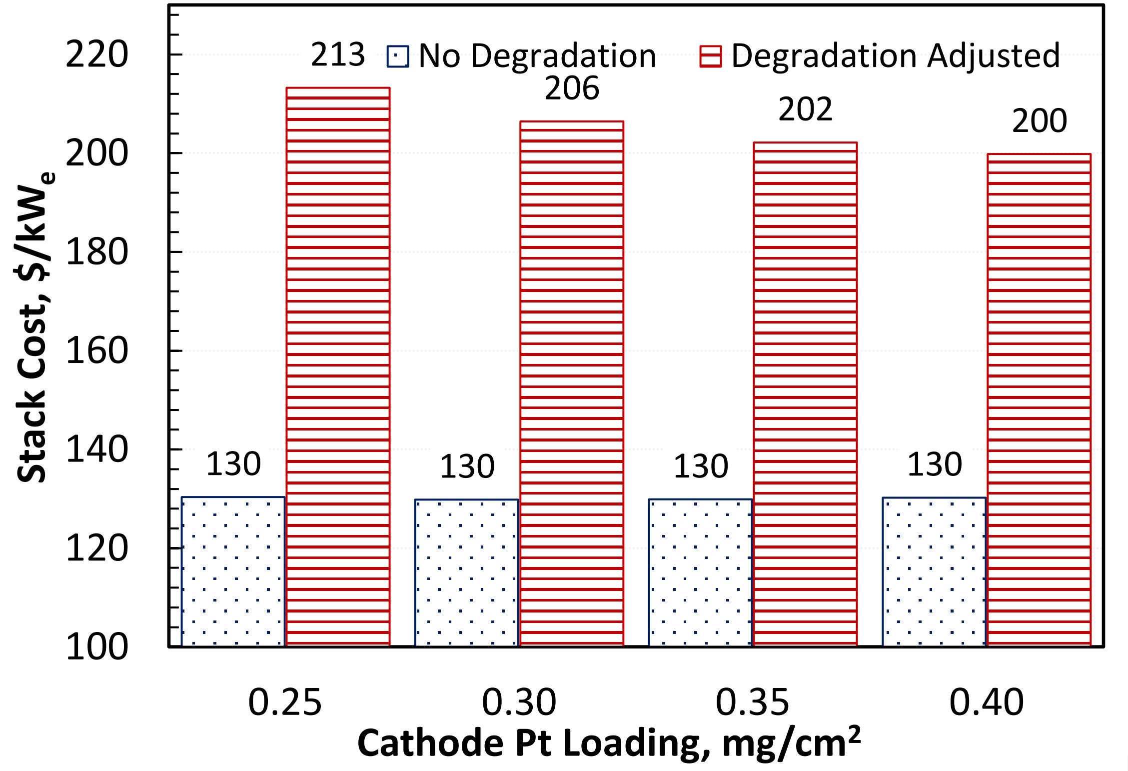
Degradation Adjusted Cost

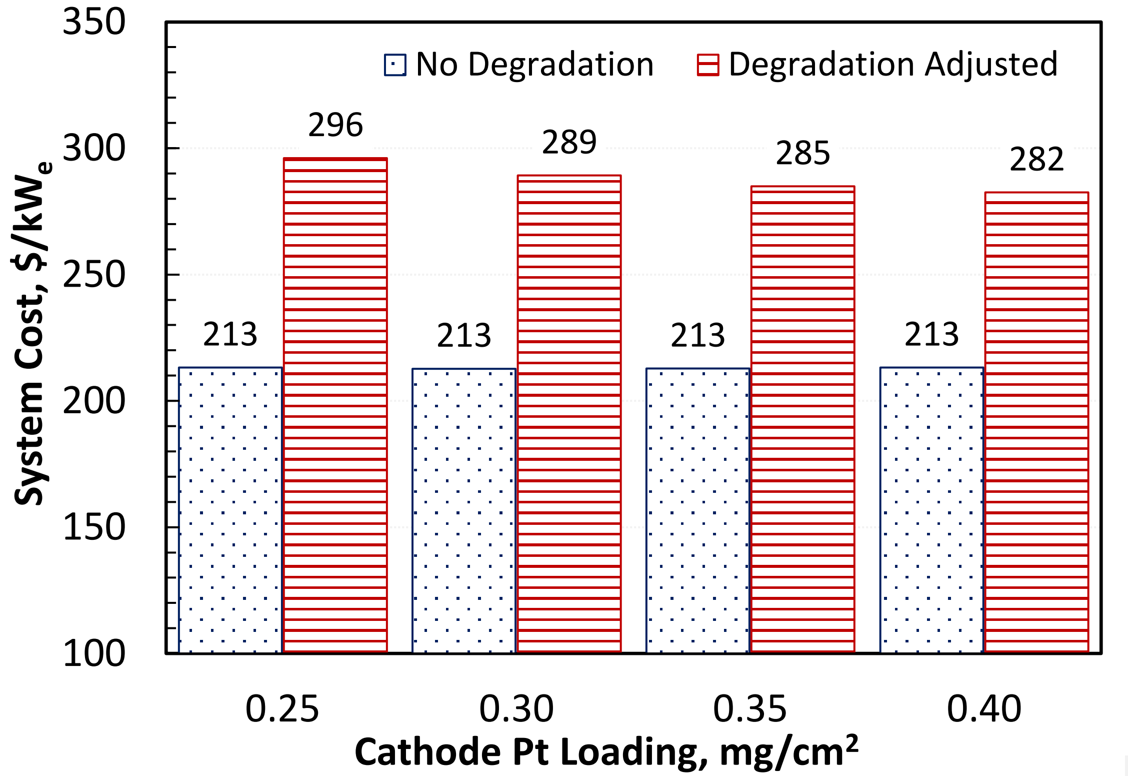
Preliminary projections of degradation adjusted system cost for 1,000 units/year manufacturing rate using cost correlations from Jennie Huya-Kouadio, Strategic Analysis

- **64% increase in stack cost and 39% increase in FCS cost for 0.30 mg/cm^2 total Pt loading**

Case for Higher Pt Loading in Cathode (0.40 mg/cm^2)

- **Pro: 24% more compact stack**
- **Pro: 6% lower cost**
- **Con: 12% penalty in Pt utilization**

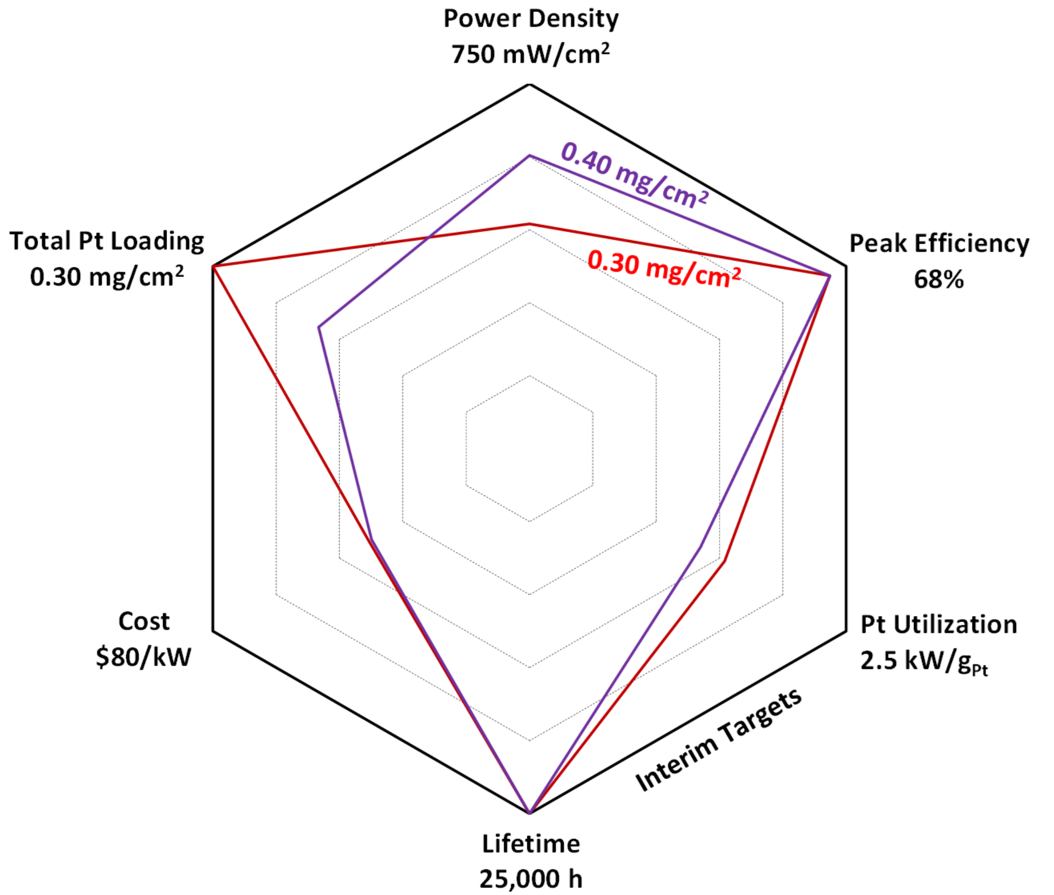




Analysis Status

Status of Fuel Cell Systems for Class-8 Heavy Duty Trucks

- Key barrier to meeting the interim targets: Active and stable catalysts capable of meeting the target of 750 mW/cm² power density at EOL with 0.3 mg/cm² Pt loading
- Need to address membrane durability



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

Future Work

- Drive cycle modified for durability
- Membrane durability
- Carbon corrosion
- Higher activity and more stable catalysts

Small Stack (175 kW$_e$ System), Large Battery Hybrid System

- Minimum battery capacity for 22-min hill climb at 6% grade: 127 kWh
- Stack coolant exit temperature: 85°C
- EOL Stack power density at 0.7 V: TBD
- Stack oversizing to accommodate performance degradation: TBD
- Total system parasitic power at EOL: 17.3%

Large Stack (425 kW$_e$ System), Small Battery Hybrid System

- Minimum battery capacity for transients on reference duty cycle and idling: 10 kWh
- Stack coolant exit temperature: 95°C
- Radiator frontal area: 50% larger than in diesel truck
- EOL Stack power density at 0.7 V: TBD
- Stack oversizing to accommodate performance degradation: TBD
- Total system parasitic power at EOL: 23.7%

Materials Development

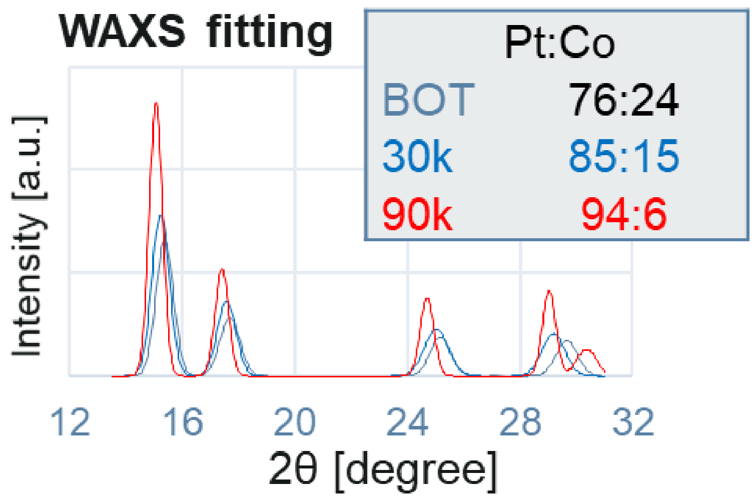
MILLION MILE
FUEL CELL TRUCK
U.S. DEPARTMENT OF ENERGY

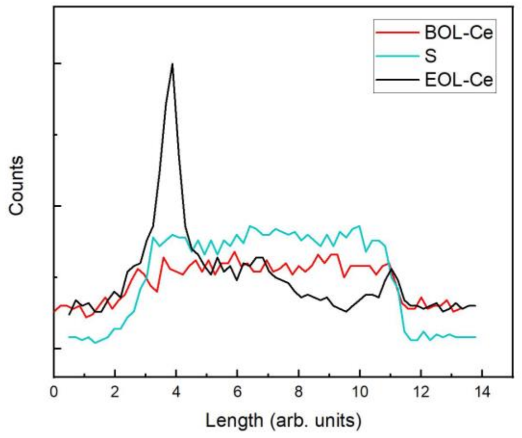
Issues with Current Catalysts and Ionomers for Heavy-Duty Applications

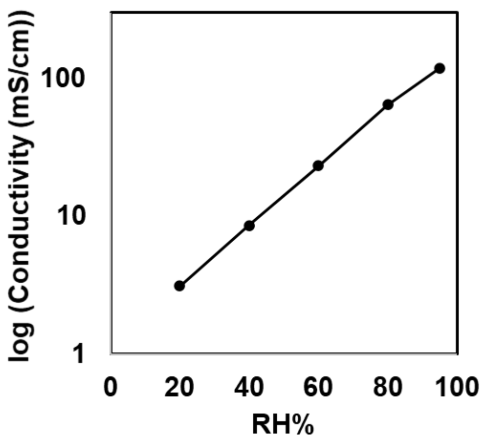
Cathode Catalysts:

- Transition metal alloys of Pt (e.g., Pt-Co, Pt-Ni) have higher oxygen reduction reaction (ORR) activity per gram Pt than Pt only
- However, transition metals leach from Pt-transition metal alloy particles until reaching ≈8 mol% transition metal
- Platinum particle size increases with cycling due to particle size dependence of Pt dissolution; Pt dissolution is significant at potentials > 0.85 V
- Example: 90K cycles between 0.6 and 0.95 V for Umicore Pt_3Co/HSC:
  - ✓ 50% ECA loss, MA 824 to 152 A/g-Pt, loss of Co from 24 to 6 mol%
- High efficiencies need for heavy-duty applications require higher performance at high cell voltages throughout 25,000 h lifetime
  - ✓ Requires higher-performing, stable cathode catalysts that resist particle growth and loss of ORR specific activity

Umicore Elyst 0690 Cathode Catalyst; AST cycling: H_2/N_2, 0.6 to 0.95 V trapezoid, 2.5 s each potential







Ionomers/membranes:

- Current perfluorinated sulfonic acid (PFSA) ionomers have performance and durability limitations, particularly at high temperatures and low RH
- Hydrogen crossover through current membranes decreases efficiency
- Sulfonate in PFSA ionomers poisons Pt catalyst; PFSA ionomers cause local O_2 transport resistance
- Main chemical degradation mechanism is radical attack – traditional radical scavengers (e.g., Ce) are mobile

M2FCT's Materials Development Sub-Tasks

- Catalyst/Support Development
  - "Pt-Co Intermetallics", Los Alamos NL, Jacob Spendelow
    - "Metal oxide-metal-carbon junction to stabilize PtM NPs catalysts", Pacific Northwest NL, Yuyan Shao
  - "Nitrogen-Doped PtMN Catalysts and Supports", Brookhaven NL, Kotaro Sasaki
  - "Tailored Pt-nanomaterials, supports, and interfaces", UC-Irvine and Argonne NL, Vojislav Stamenkovic
- Membrane/Ionomer Development
  - "High-conductivity Novel Perfluorinated Ionomers", National Renewal Energy Laboratory, Bryan Pivovar
  - "Low Molecular Weight Oligomeric Electrode and Membrane Ionomers", Los Alamos NL, Yu Seung Kim
- Supporting Efforts
  - Electron microscopy, Michael Zachman/Dave Cullen, Oak Ridge NL
  - Synchrotron X-ray characterization, Debbie Myers, Argonne NL
  - Fundamental studies of ionomers and membranes, Ahmet Kusoglu, Lawrence Berkeley NL

Materials Development QPM-Go/No-Go

Catalyst Development Go/No-Go Decision

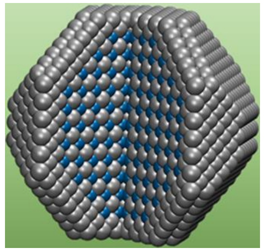
Q6 (3/31/2022) QPM: Demonstrate ≥ S.O.A. at 0.8 V on hydrogen-air at 150 kPa, 100% RH, 80°C cell temperature after 90,000 catalyst AST cycles using an MEA with ≤0.3 mg/cm^2 total PGM loading (comparison with S.O.A. at same loading). S.O.A is Umicore annealed Pt/HSC with 138 mA/cm^2 at 0.8 V after 90,000 AST cycles. (see slide #)

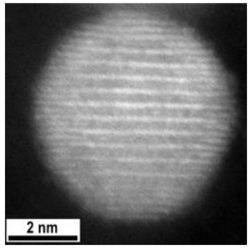
Q12 (9/30/2023)	**Go/No-Go Decision**	**Decision:** Decision point on materials to move forward into MEAs Integration and Optimization. Component (membrane, Catalyst) Performance after AST Materials passing Go/No-Go moved to scale-up and integration efforts.	**Criteria: Catalysts/supports:** >0.44* A/mg-Pt MEA ORR activity after equivalent 25,000 hr AST. **Membrane:** < 0.02 Ω-cm^2 and < 1-2 mA/cm^2 H_2 crossover after AST equivalent of 25,000 hr durability *To be revised

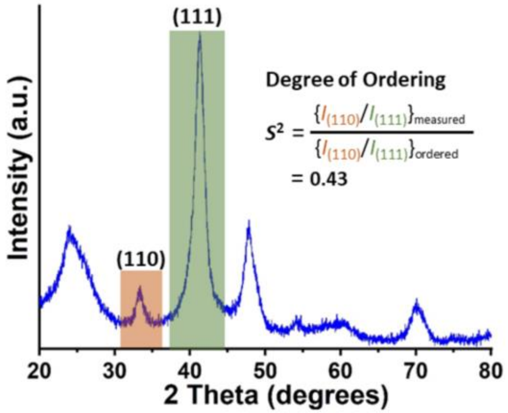
Approach: Structure Engineering of Pt Alloy Particles

Pt-based Intermetallic Particles

- Ordered structure mitigating base metal leaching
- Multi-monolayer Pt shell mitigating base metal leaching
- Strained Pt surface leading to high intrinsic activity

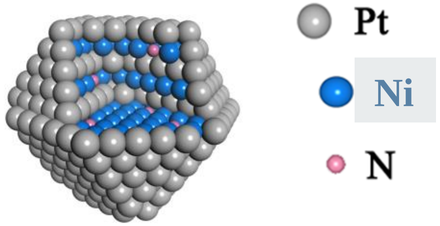




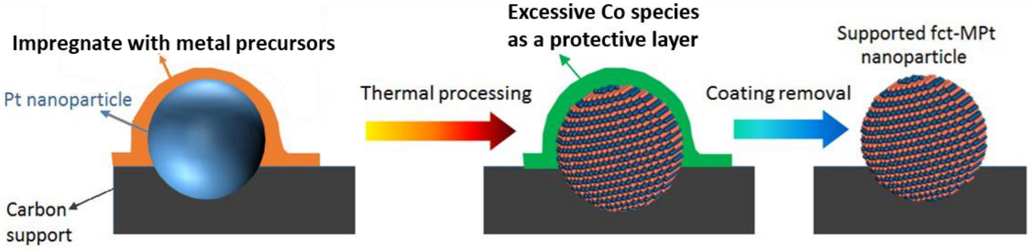


Further stabilizing base metal and straining Pt surface using nitrogen doping of Intermetallic Particles

N-doped nitride core + Intermetallic structure



Using a facile Pt intermetallic synthesis method: impregnation of Co into Pt/C

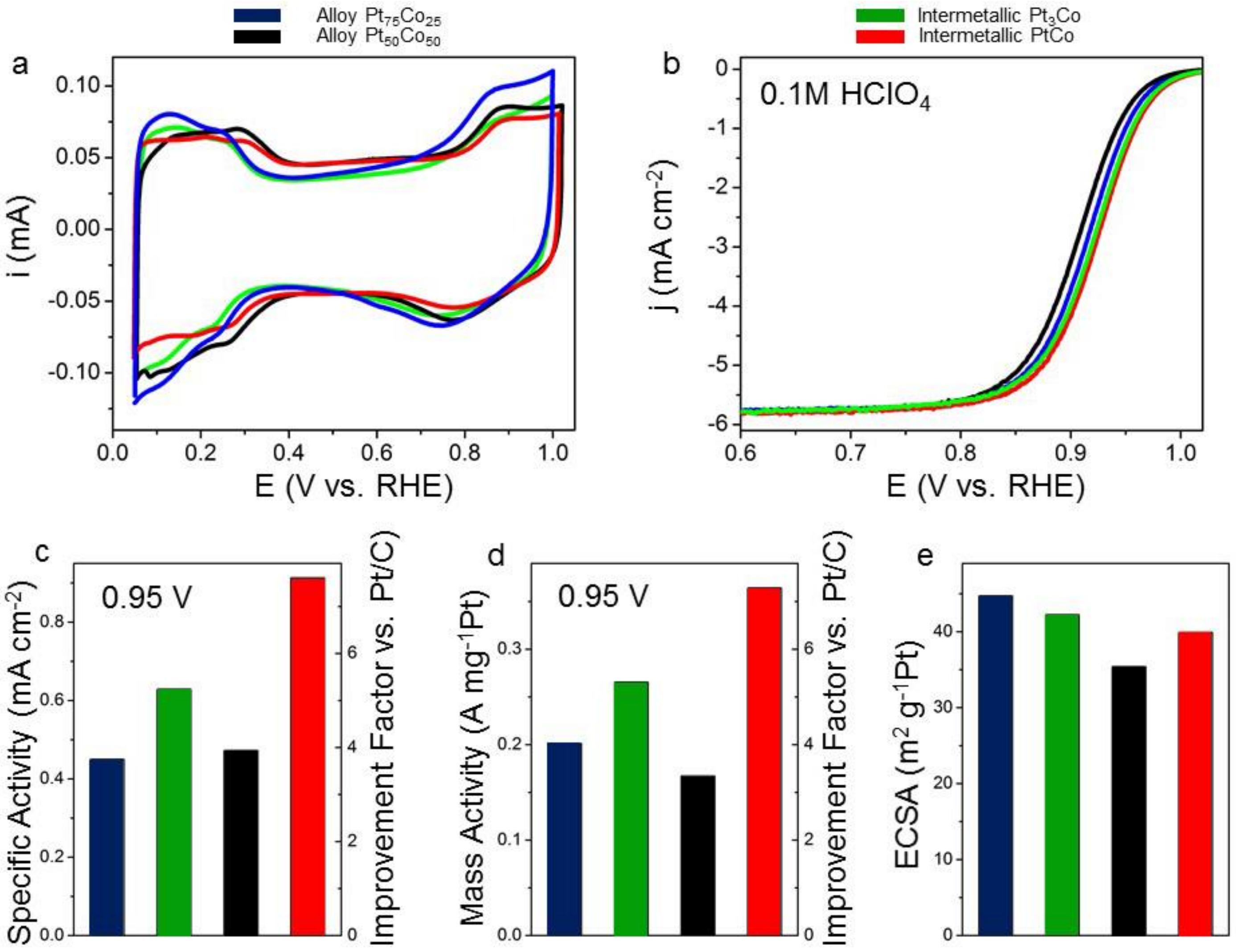


Using a facile nitriding method

- Nitriding a mixture of precursors and carbon by annealing in NH_3 gas

- Alloying Pt with Ni gives lattice contraction, while N-doping and structural ordering impart tensile strain → optimal contraction in Pt surface atoms

Accomplishment: Intermetallic Structure Increases Activity and Decreases Pt,Co Dissolution



	Pt dissolution*	Co dissolution*	Retained activity*	ECSA(CO):ECSA(H)
$Pt_{75}Co_{25}$ A	0.37 μg L^{-1}	4.0 μg L^{-1}	53 %	1.10
$Pt_{75}Co_{25}$ IM	0.25 μg L^{-1}	2.1 μg L^{-1} **	86 %	1.25
$Pt_{50}Co_{50}$ A	0.36 μg L^{-1}	14 μg L^{-1}	48 %	1.02
PtCo IM	0.24 μg L^{-1}	4.7 μg L^{-1} **	84 %	1.22

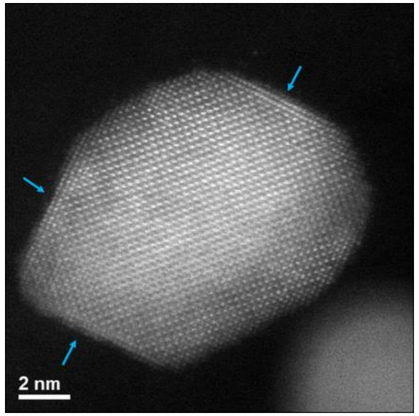
A - solid solution PtCo alloy;

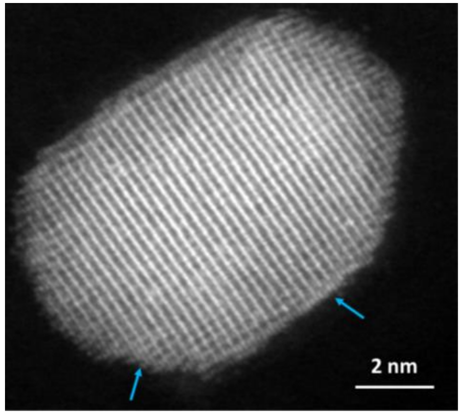
IM – intermetallic structure

* after 10,000 cycles

** the Co dissolution from preconditioned catalysts:

1.1 μg L^{-1} ($Pt_{75}Co_{25}$) and 1.2 μg L^{-1} (PtCo)

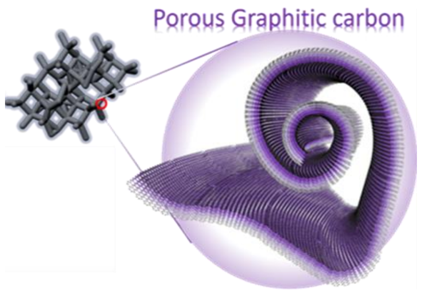




Approach: Structure Engineering of Supports

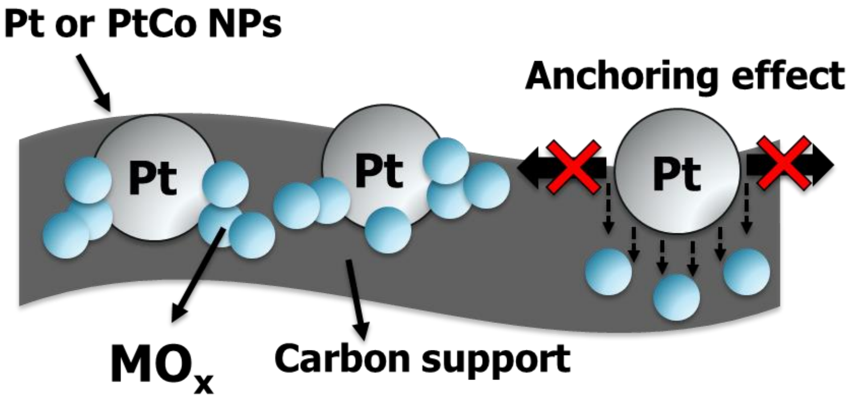
N-doped Porous Graphitic Carbon (NPGC) Support

- Intermediate porosity balances particles protection and accessibility to oxygen
- Nitrogen doping leads to an even distribution of ionomer on particles and within the catalyst layer



- Co-existence of mesopores and micropores in carbon support
- Pt particle deposition in mesopores
- Uniform doping of carbon with nitrogen
- High-pressure nitriding to increase N content

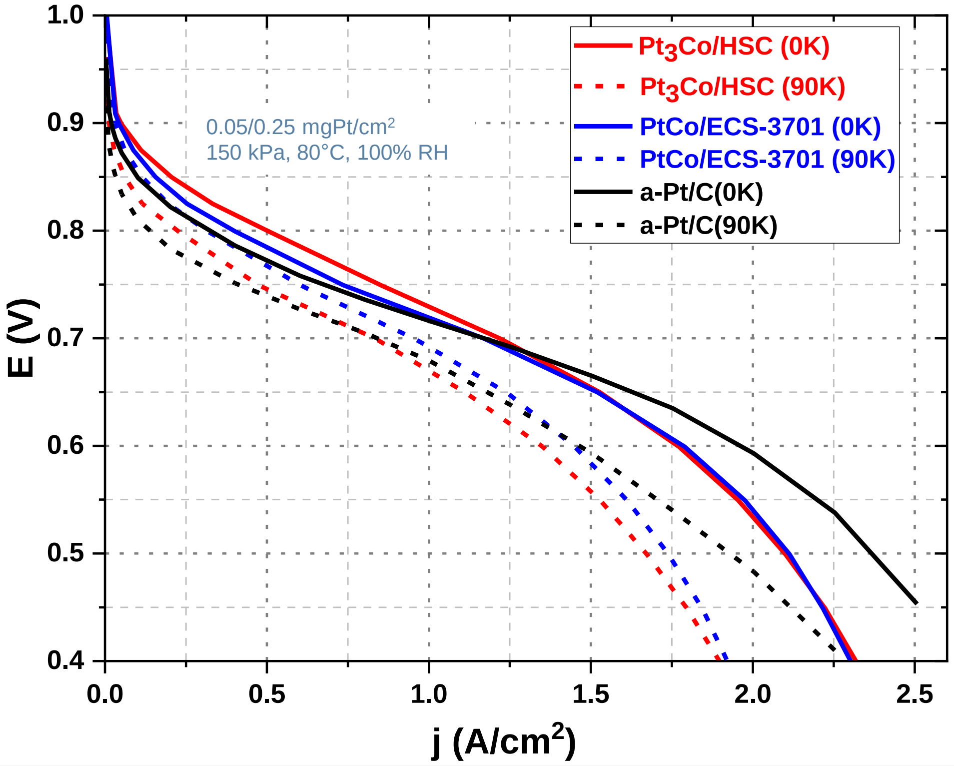
Co-Deposition of Oxide Particles and Pt Nanoparticles to Stabilize Pt

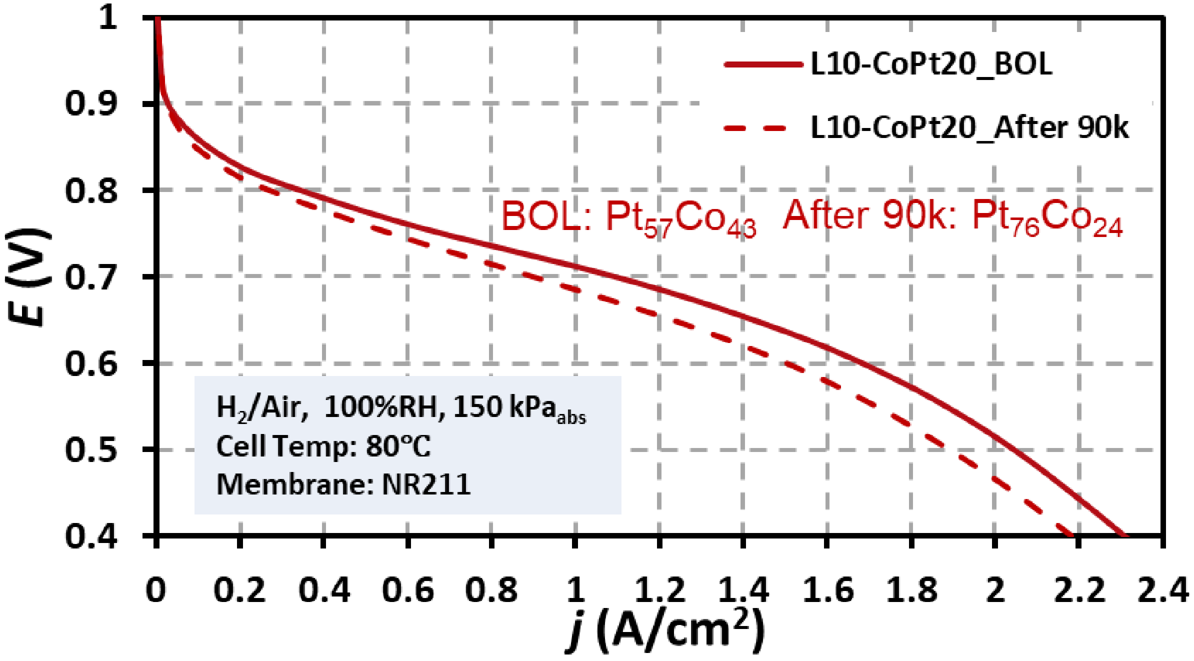


Accomplishments: PtCo Intermetallic Catalysts on Mesoporous Carbon

New PtCo intermetallic catalysts provide high performance and durability on various carbon supports

- Solid carbon (Vulcan XC-72)
- High surface area carbon (HSC)
- Mesoporous carbon (Pajarito ECS-3701)
- MOF-based carbon





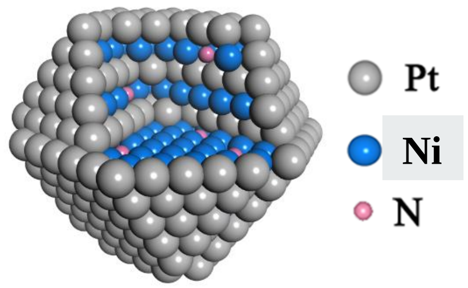
Achieved **130% higher post-90K performance at 0.8 V** vs. Umicore annealed Pt/HSC, surpassing Go/No-Go

Accomplishments: Nitrided Intermetallic PtNi/C

Intermetallic- & alloy-structured nitrogen-doped PtNiN/C

- ✓ Established facile one-step synthesis - scalable ≥ 1 g per batch
- ✓ Optimization of structure, particle size, Pt & Ni concentrations to improve ORR performance

Intermetallic PtNiN



Comparison of PtNiN/C and benchmark performance of a-Pt/C

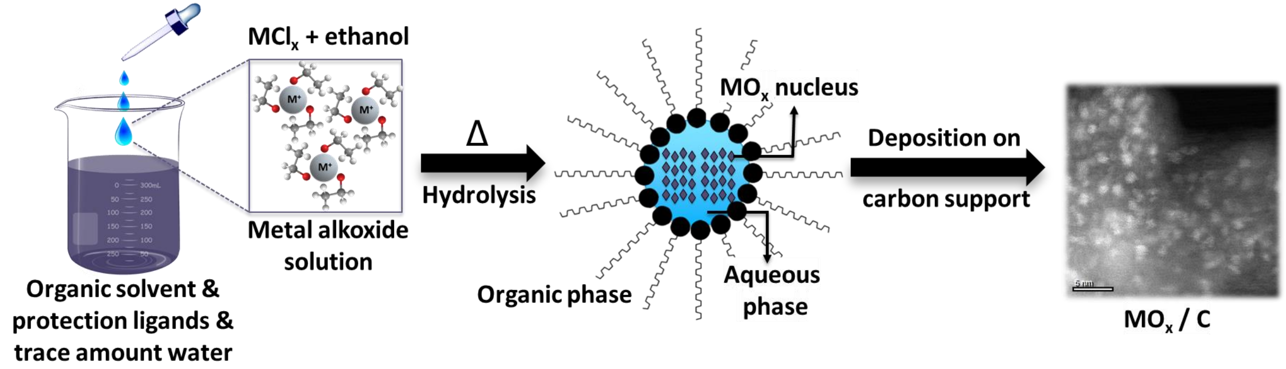
Catalysts (particle size)	Number of cycles	ECSA (m^2/g_{Pt})	MA (mA/mg_{Pt})	*j* at 0.7V (A/cm^2)	*j* at 0.8V (A/cm^2)
SA-Int-PtNiN§ BNL-C (4.5 nm)	0K	**53**	**550**	**1.22**	**0.50**
	90K	**22**	**210**	**0.96**	**0.25**
Int-PtNiNᶲ BNL-D (4.8 nm)	0K	**51**	**560**	**1.19**	**0.45**
	90K	**25**	**246**	**0.95**	**0.27**
a-Pt/C (4~5 nm)	0K	39	418	1.17	0.32
	90K	20	178	0.85	0.14

5 cm^2 differential cells, 150kPa, 100% RH, 80°C, 0.25 mg/cm^2 at cathode, 0.6-0.95V square wave
SA-Int-PtNiN/C§ is acid-washed Int-PtNiN/KBᶲ (the same batch)

- Intermetallic PtNiN/C (Int-PtNiN, 4.8 nm) and acid-washed Intermetallic PtNiN/C (SA-Int-PtNiN, 4.5 nm) showed higher durability in 90K AST testing than reference a-Pt/C
- Post-mortem analysis showed particle growth and Ni leaching, indicating there are avenues for improvement of these already promising catalysts

Accomplishments: Anchoring Catalyst Particles Using Metal Oxides

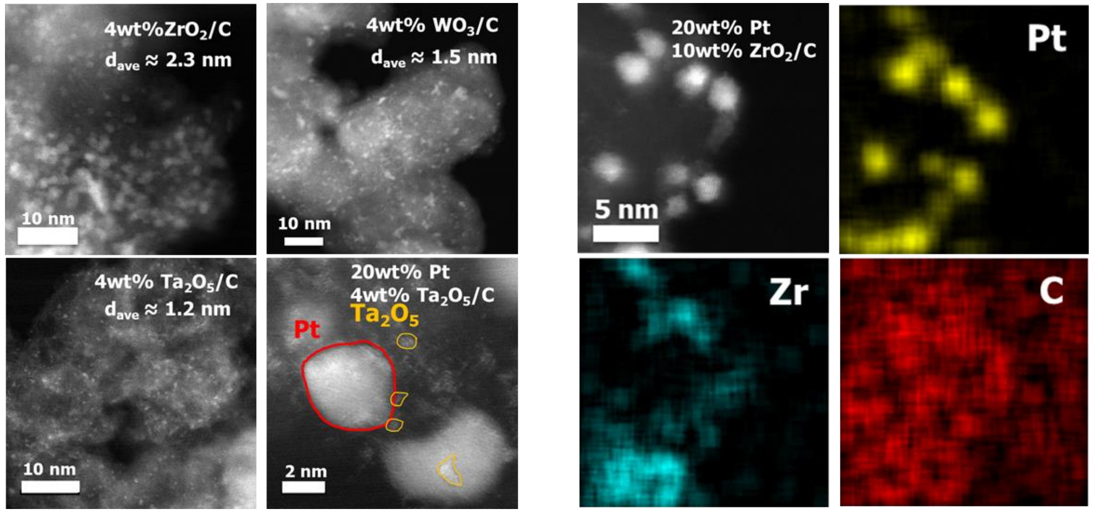
- Zr, W, Ta oxides selected as the metal oxide candidates



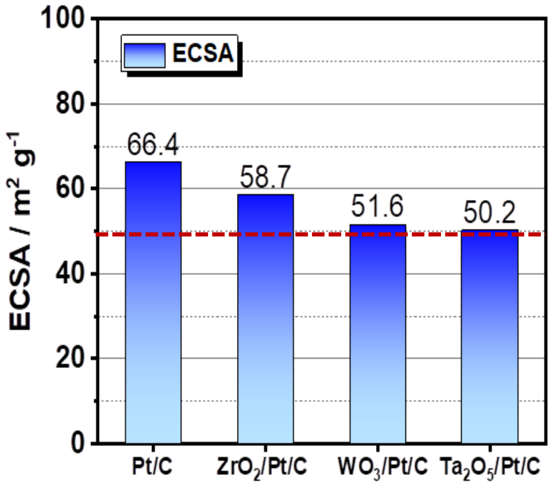
Chemical stability of metal oxide candidates screened

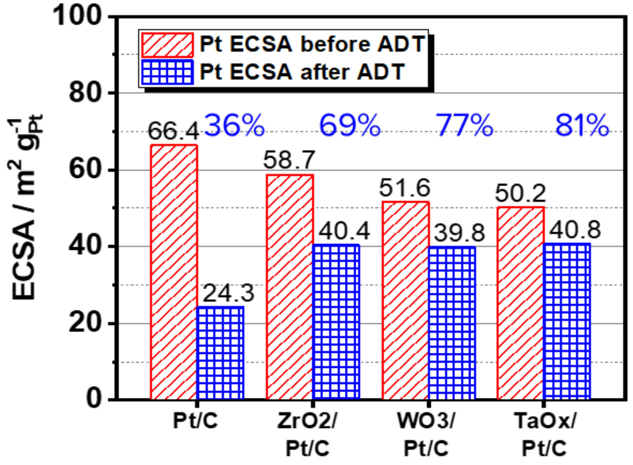
MOx	Dissolution after 10 h 90 °C 0.5 M H_2SO_4
ZrO_2	< 0.2% (total 3.8wt%)
WO_3	< 1.1% (total 4.2wt%)
TaO_x	< 0.1% (total 3.7wt%)

Uniformly dispersed <3 nm metal oxide nanoparticles by modified reverse microemulsion (RME) method



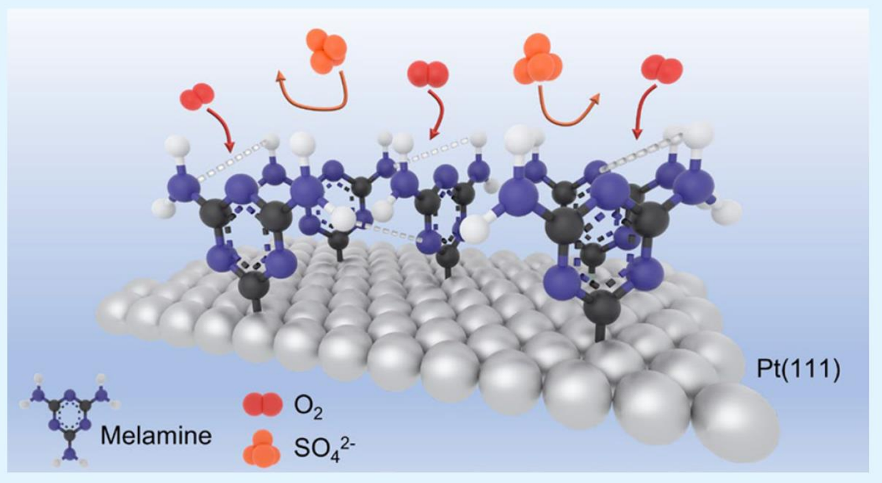
- ECSA retention enhanced to over 80% by presence of MO_x nanoparticles in RDE measurements





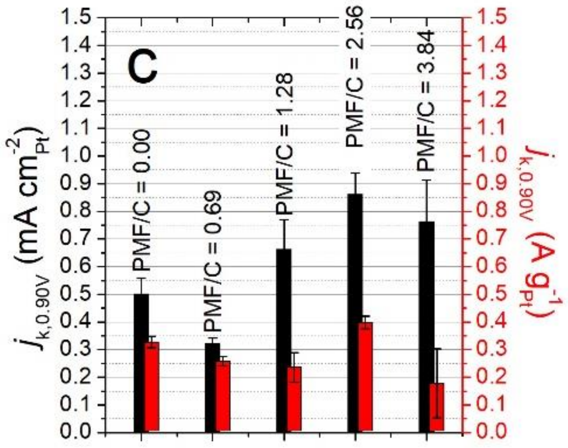
Approach: Modification of Catalyst-Ionomer Interface

- Introduce selected molecules into well-defined electrochemical interfaces with the thickness from a few *Å (2D) towards nm* scale (3D)
- Tailor properties of electrochemical double layer
- Form *the interphase* that enables control of charge transfer and adsorption of reactants and spectators (e.g., sulfonate)
- Selected molecules are spectators and do not have an active role in electrochemical processes
- Candidate modifier molecules: melamine and poly(melamine-co-formaldehyde) methylated (PMF)

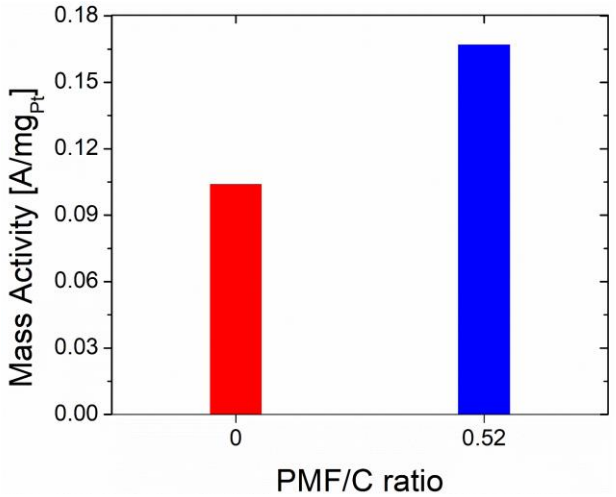


Accomplishment: Poisoning of Pt Catalyst by PFSA Decreased using poly(melamine-co-formaldehyde) methylated (PMF)

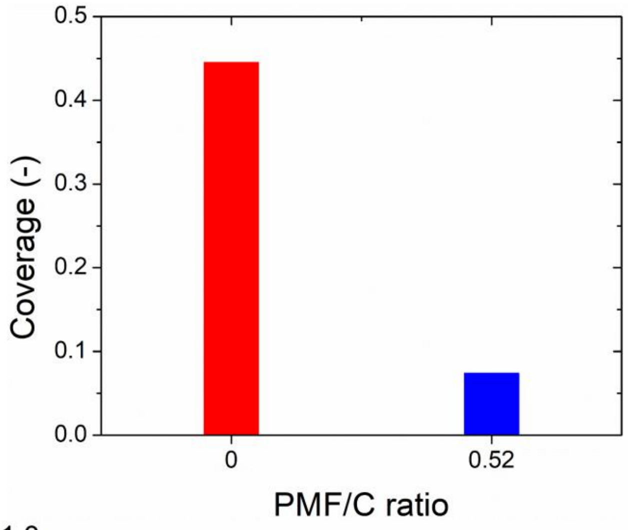
Rotating-disk electrode

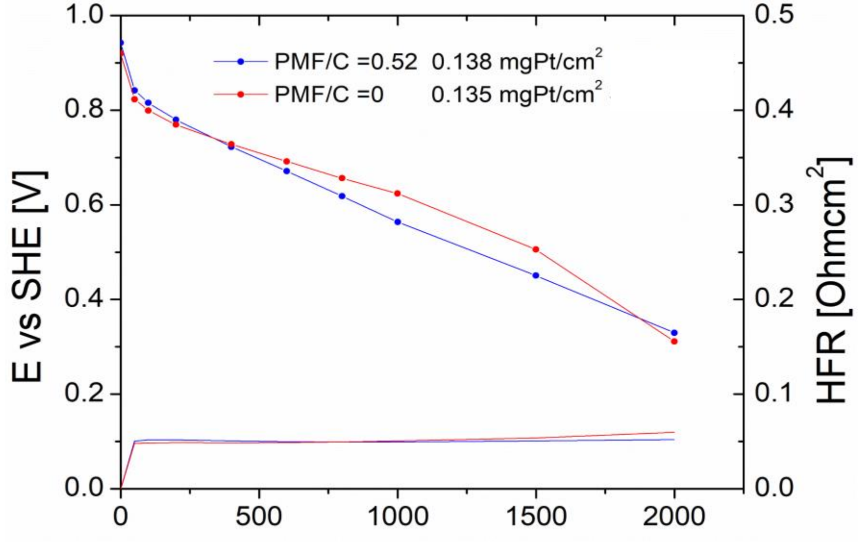


Membrane-electrode assembly



Coverage of SO_3^- on Pt



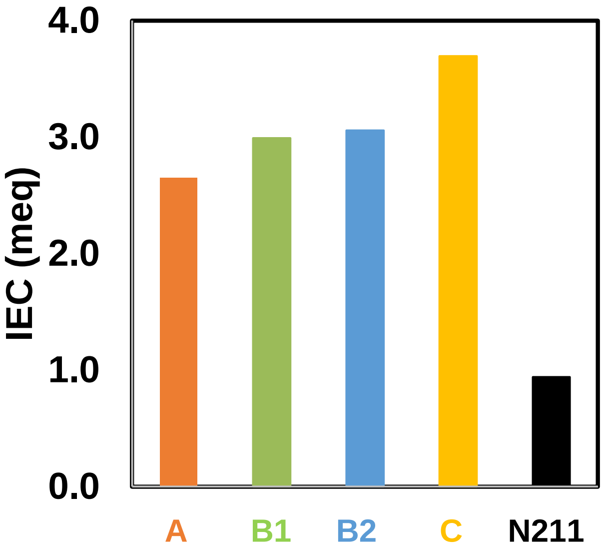


- PMF was incorporated into Pt/C to modify interface between Pt and PFSA
- PMF/C = 0.52 was selected as it was identified by RDE as optimal (Pt/PMF = 0.78)
- Hydrogen-air polarization curves shows better performance in kinetic and mass transport regions with PMF, but higher ohmic losses arising from cathode catalyst layer
- Increase in mass activity for MEA with PMF in cathode catalyst layer
- This is due to less poisoning by sulfonic acid groups
- SO_3^- coverage on Pt is reduced from 0.45 to less than 0.1

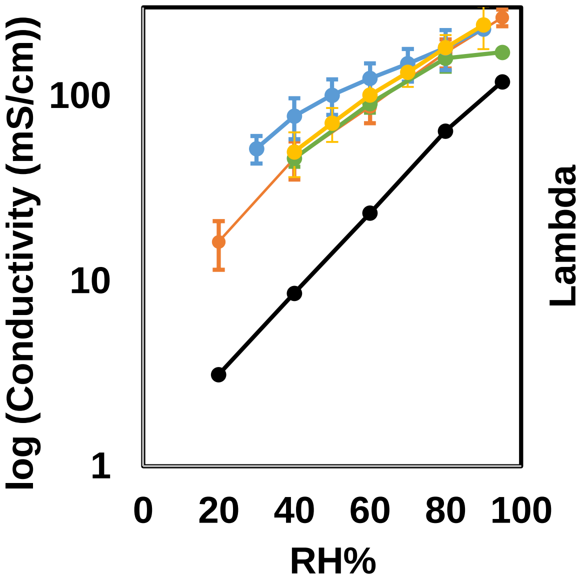
Approach: Development of Novel Perfluoro Ionomers/Membranes

- Fuel cell electrolytes have been limited to a narrow range of perfluorinated sulfonic acid (PFSA) copolymers. While current PFSAs exhibit high durability and conductivity, they still have performance and durability limitations, particularly at high T and low RH.
- Novel perfluorinated polymers offer the potential to improve lifetimes, performance and durability through advances in conductivity, durability and catalyst interactions by exploring novel chemistries with super acids at extreme local concentrations
- Ultimate goal is improved conductivity and selectivity (conductivity/hydrogen permeability) to improve high temperature/low RH performance and to improve electrocatalyst/ionomer interactions.
- Using novel ionomer chemistry, advantageous for crosslinking and grafting approaches and for:
  - decoupling mechanical/transport properties
  - restraining water uptake for decreased swell
  - increasing selectivity
  - limiting polymer mobility at catalyst
  - reaching extreme local acid concentrations

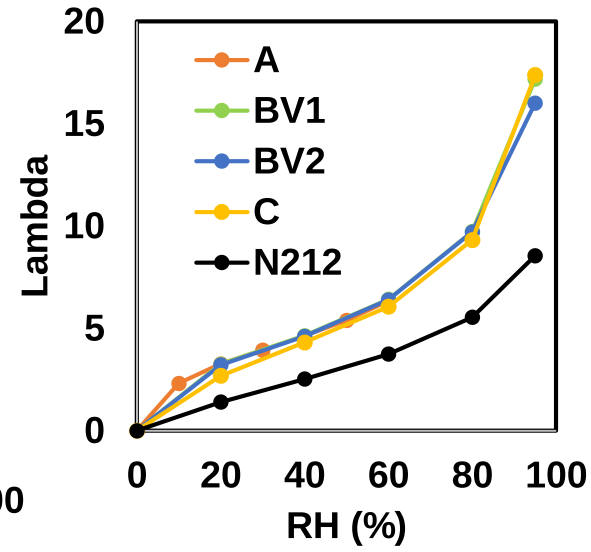
Accomplishments: High Ion-Exchange Capacity (IEC) Ionomers Synthesized and Evaluated



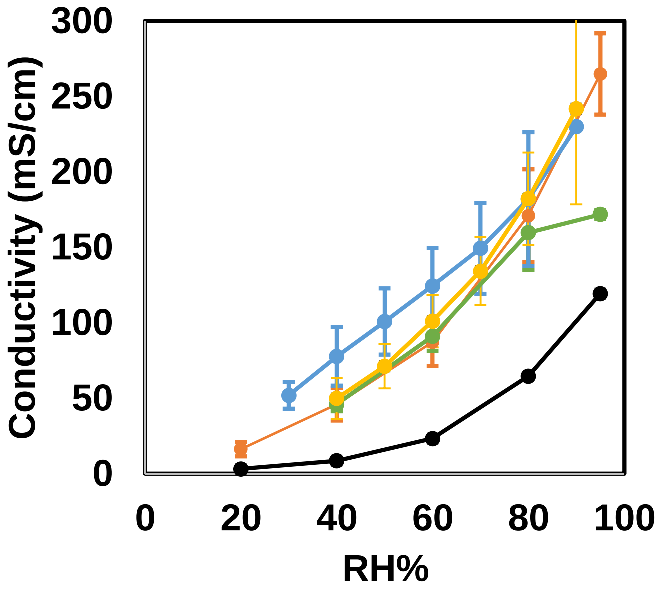
Three target chemistries of focus have been synthesized and characterized for IEC, conductivity and water uptake.



All novel ionomers have demonstrated a 5-10x conductivity improvement over Nafion 211.



Higher lambda values at lower RH suggest increased water affinity of these novel polymers compared to Nafion. Water uptake per acid site identical across chemistries explored.

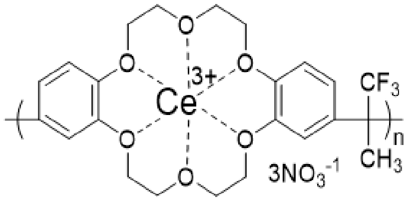


Polymers synthesized to date lack water robustness under high RH and liquid water exposure.

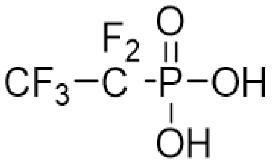
Approach: Low Molecular Weight Oligomeric Electrode and Membrane Ionomers

- Development of oligomers to be used as membrane additives or electrode binders.
- Functional structure targeting improving:
    - Radical scavenger immobilization
    - Oxygen reduction reaction kinetics
    - Mass transport of reactant gases
- Optimization of membrane and electrode processing towards:
    - Higher mechanical toughness of PEMs
    - Uniform distribution of ionomer and catalyst nanoparticles in the electrodes
    - Enhanced morphological stability
    - Formation of ideal three-phase interface

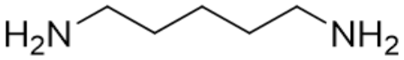
Stabilizer candidates



Crown ethers



Phosphonic acids

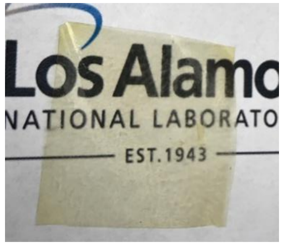


Amines

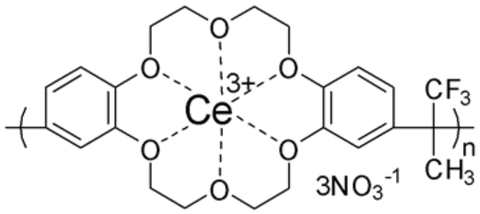
Accomplishment: Radical Scavenger Immobilization

- Successfully synthesized 18-crown-6 functionalized polymer, complexed Ce^{+3}; TGA indicates the composite polymer contained 38% $Ce(NO_3)_3$ (1:1 ratio: 42%).
- Successfully synthesized wholly perfluorinated phosphonic acids for increased compatibility with PFSAs versus commercial perfluorooctanephosphonic acid (PFOPA)
- The synthesized C8 and C10 phosphonic acids showed superior properties over crown ether or commercial PFOPA.
- Effect of oligomer additives on catalyst activity is being investigated.

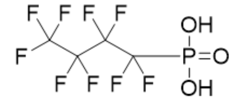
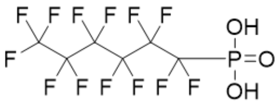
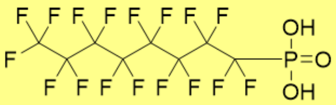
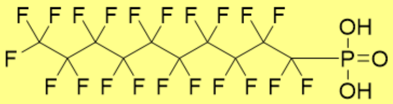
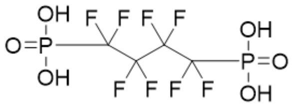
Properties	PFOPA	PDBCE	C8
Cerium Retention (%)	**89.6**	5	70.2
FER (mg_F/g_{Nafion} h)	0.26	0.42	**0.20**
Nafion conductivity (S m^{-1})	15	18	**20**
Compatibility	Hazy	**Clear**	**Clear**
Catalyst poisoning	?	?	?



$PDBCE\text{-}Ce(NO_3)_3$



Perfluorinated Phosphonic acids Synthesized

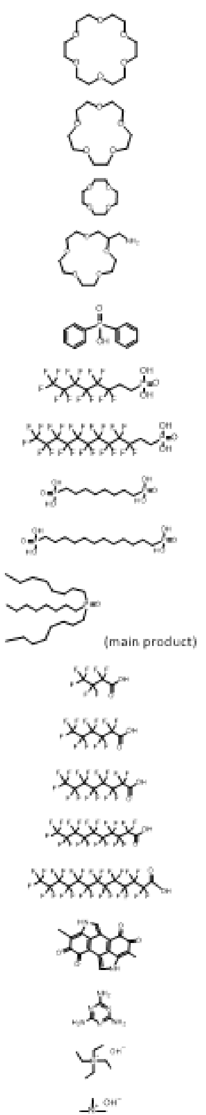
Ligand name	Chemical structure	CR(%)
C4		2.9
C6		4.6
C8		**70.2**
C10		**71.9**
C4DPA		4.4

* PA to Ce ratio = 6

CR = cerium retention

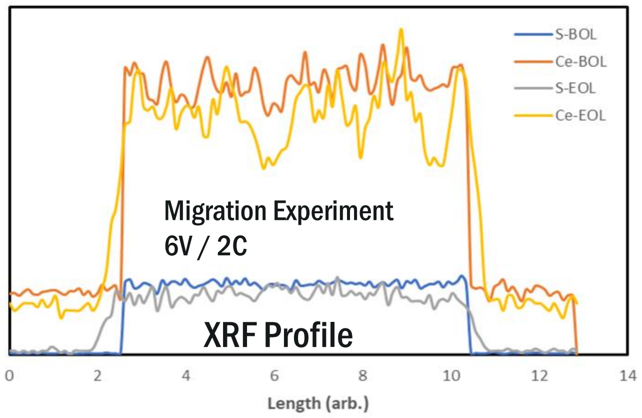
Stabilization of Cerium in Membranes

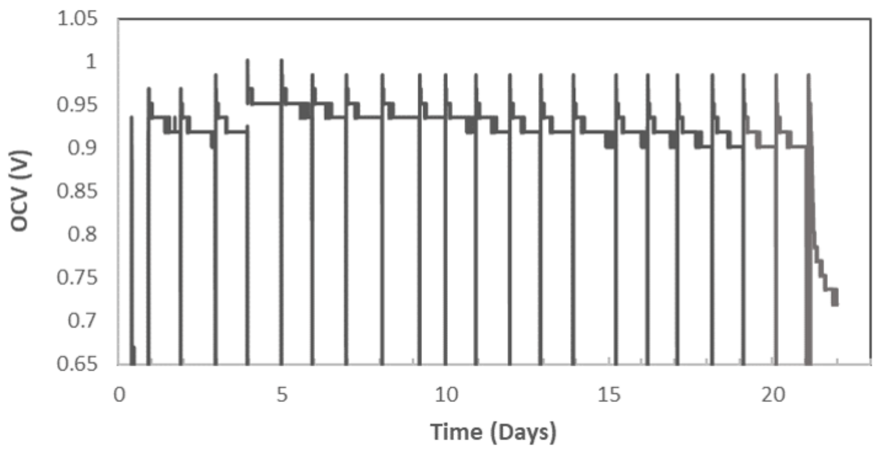
No.	Category	Name [a]	Ligand's chemical structure	CR (%)[b,c]
1	Control	Ce-Nafion	None	2.0 (3.2)
2	Crown ether	18C6		3.4 (5.3)
3	Crown ether	15C5		4.8 (17.6)
4	Crown ether	12C4		4.0 (11.8)
5	Crown ether	Amino-15C5		5.6 (12.4)
6	Phosphinic acid	DPPA		6.0
7	Phosphonic acid	PFOPA		35.7
8	Phosphonic acid	FHPA		8.9
9	Phosphonic acid	ODPA		31.4
10	Phosphonic acid	PDPA		58.6
11	Phosphine oxide	Cyanex 923	(main product)	88.3
12	Carboxylic acid	NFVA		1.3
13	Carboxylic acid	PFHA		1.1
14	Carboxylic acid	HDFNA		1.5
15	Carboxylic acid	HIFUA		3.4
16	Carboxylic acid	PFTDA		1.4
17	Amine	Melanin		0.0 (2.2)
18	Amine	Melamine		0.0 (0.9)
19	Ammonium	TEAOH		0.0 (3.6)
20	Ammonium	TMAOH		0.0 (2.3)



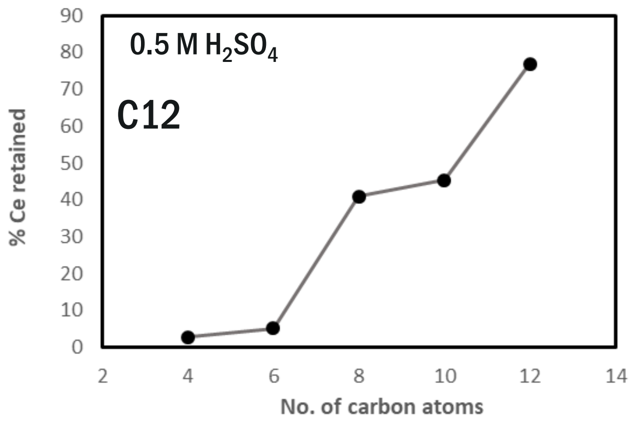
Results

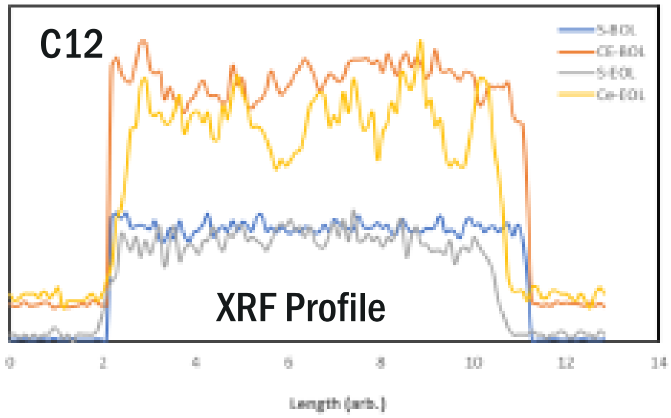
CRE Stabilization of Ce





Phosphonic Acid Stabilization of Ce



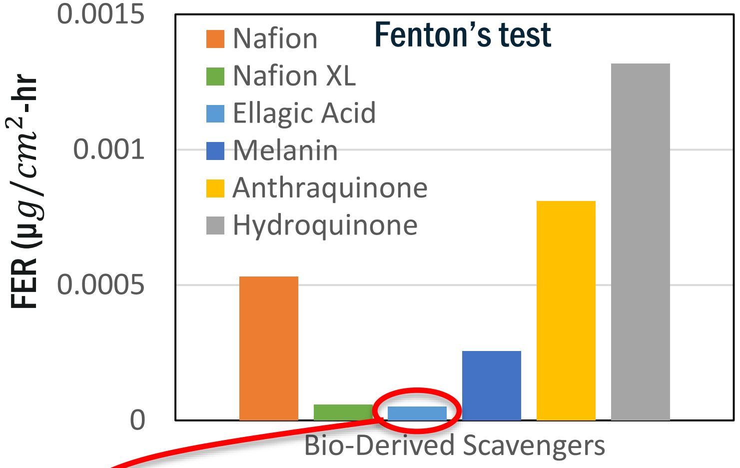


Both CREs and phosphonic acid show promise as Ce stabilizers

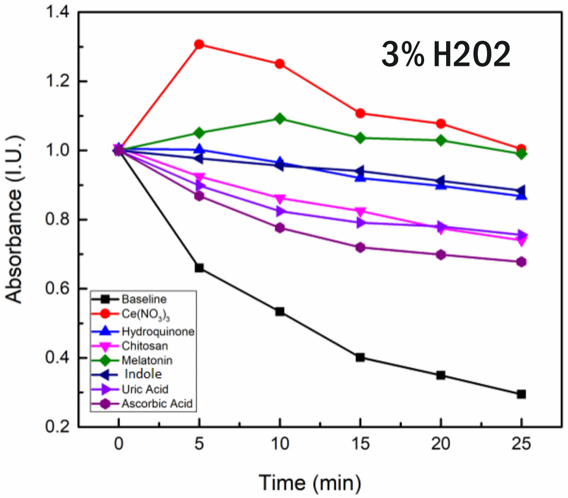
Radical Scavengers (non-Ce based)

Novel Radical Scavengers – Nature-inspired Scavengers

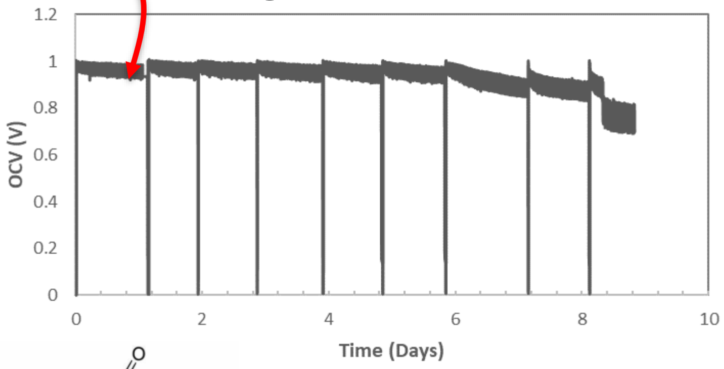
Ex-situ FER emissions



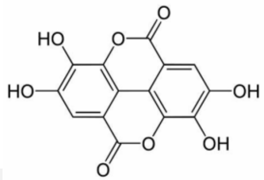
In situ Radical Monitoring



Ellagic Acid – OCV AST

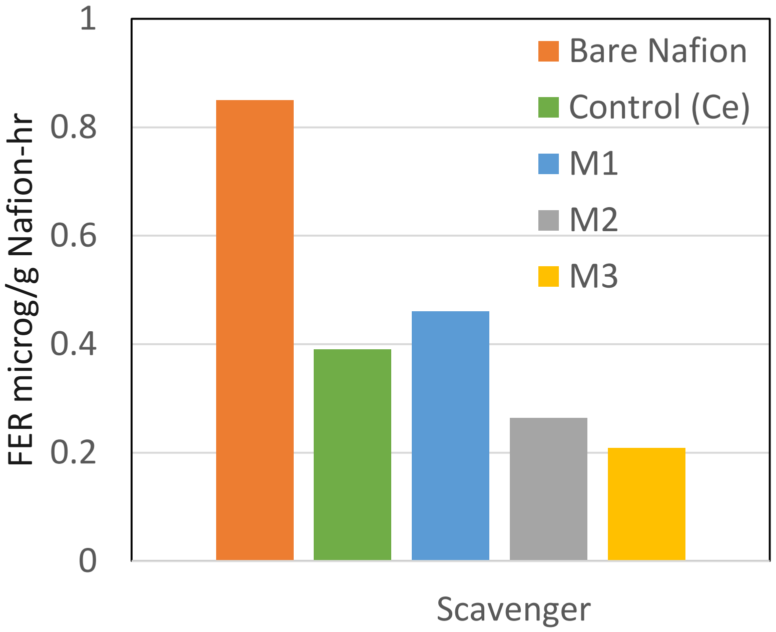


- Radicals change color of Rhodamine dye
- Best performance is no change in Absorbance



- Many bio-derived scavengers unstable in fuel cell environment
- Some bio-derived scavenger do not self-regenerate
- Ellagic acid seems the most promising – repeating OCV test

Novel Radical Scavengers – Synthesized

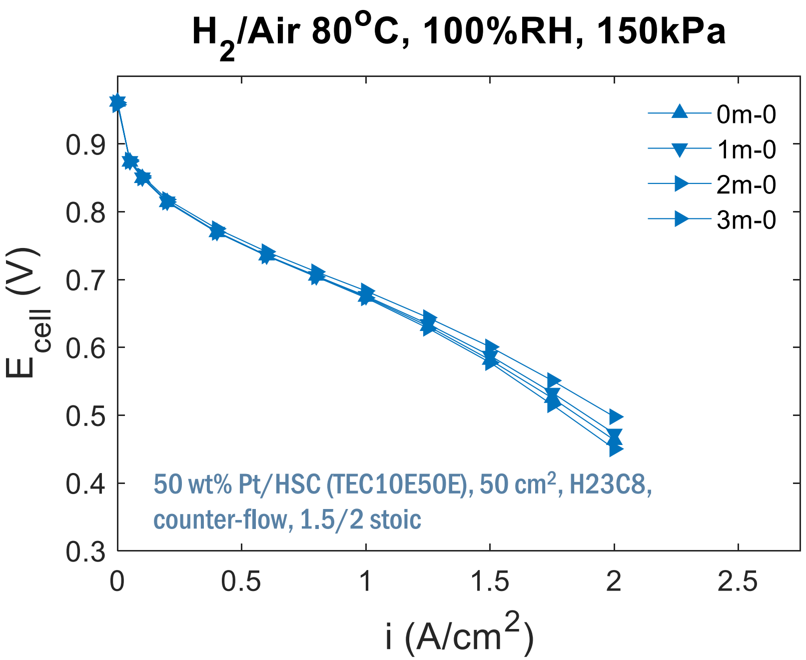


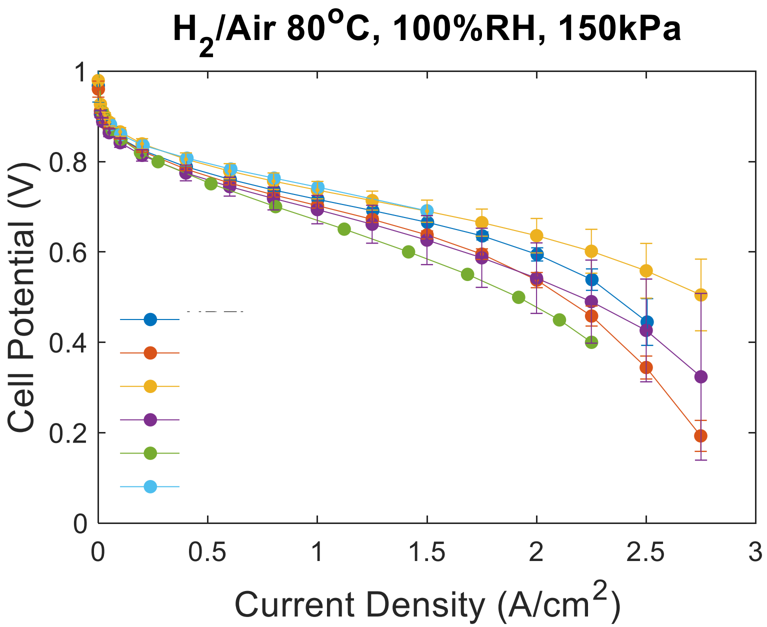
Invention disclosure filed; patent in process

Benchmarking and Baselining

Common R2R MEA Testing

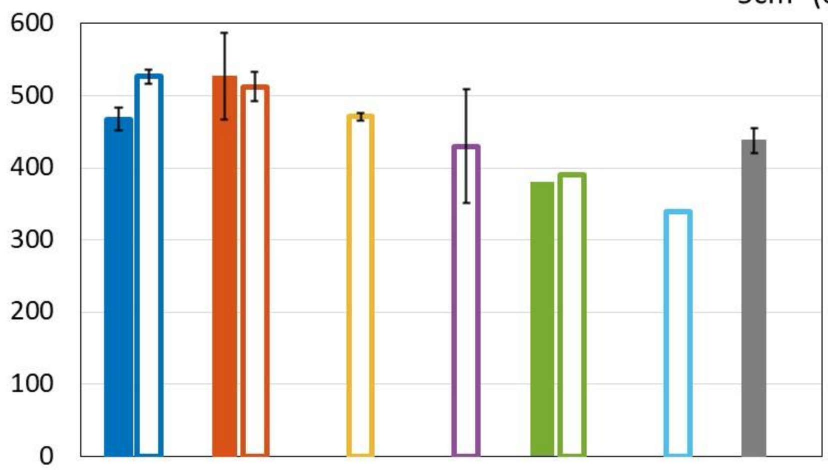
Roll-to-Roll System





Location	50 cm2	5 cm2
LBNL	2	3
GM	2	3
LANL	2	3
ANL	0	2
NIST	2	
CMU	2	2
Nikola	2	
NREL	4	3

- Very reproducible results across GDE roll (4-50 cm^2 MEAs tested at same site)
- Relatively consistent results across labs for mass activity
- More variation in polarization performance
- Outliers identified and addressed



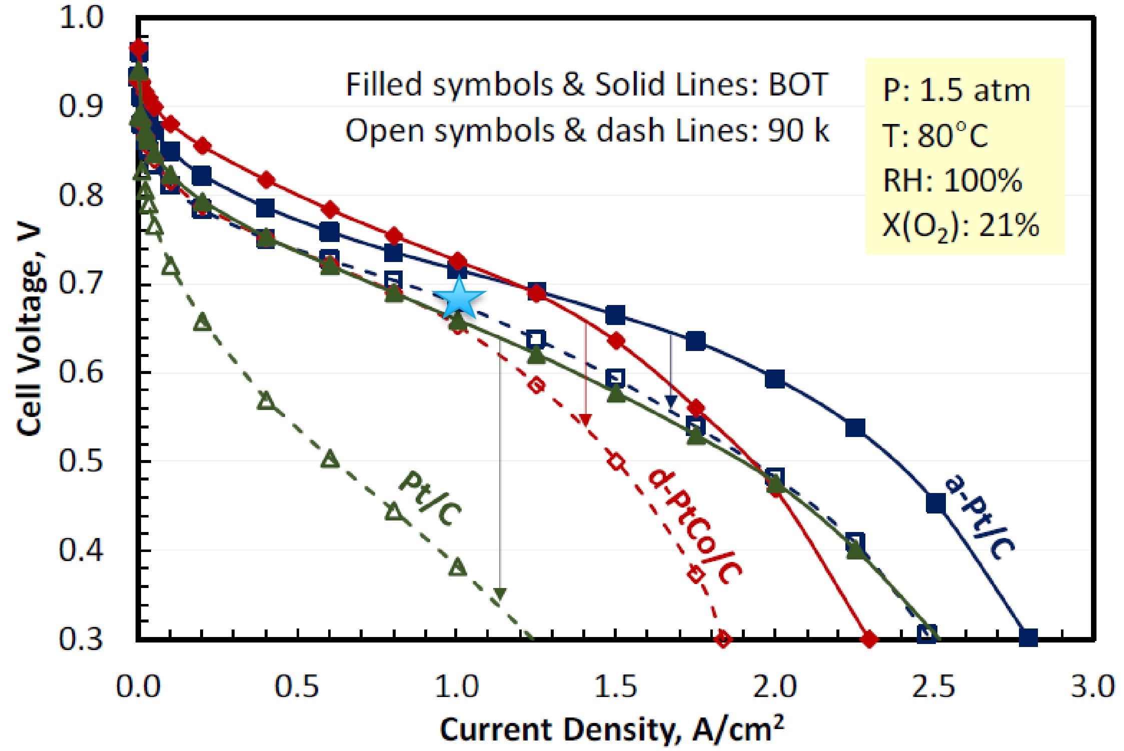
Baseline Performance for Catalyst Down Selection

- Materials chosen to contrast mean particle size and alloying
- 90k cycles from 0.6 to 0.95 V

Supplier		Umicore	Umicore	TKK
Catalyst		Elyst Pt50 0690 (PtCo)	Elyst Pt50 0550	TEC10E50E
Carbon Support		HSA	HSA	HSA
Support BET [m^2/g]		780-800	780-800	800
Pt/Co ratio [mol/mol]		2.2 – 3.0		
Mean Pt Crystallite size [nm]		[a]4.4 - 5.4	[a]5.1 - 5.7	[c]2.4 - [b]2.6
Catalyst BET [m^2/g]		240 - 280	310 – 350	300
ECSA [m^2/g_{Pt}]		40-45	45-50	72-77

Baselining MEA Performance and Durability in Differential Cells

	Number of Cycles	ECSA m^2/g_{Pt}	MA mA/mg_{Pt}	PD at 0.7 V		PD at 0.8 V	
				mW/cm^2	kW/g_{Pt}	mW/cm^2	kW/g_{Pt}
Pt/C	0k	65	651	519	1.70	144	0.47
	90k	20	73	94	0.31	20	0.07
a-Pt/C	0k	39	418	818	2.60	253	0.80
	90k	20	178	594	1.89	110	0.35
d-PtCo/C	0k	34	824	829	2.59	403	1.26
	90k	16	152	526	1.64	125	0.39



➢ Data given to analysis team

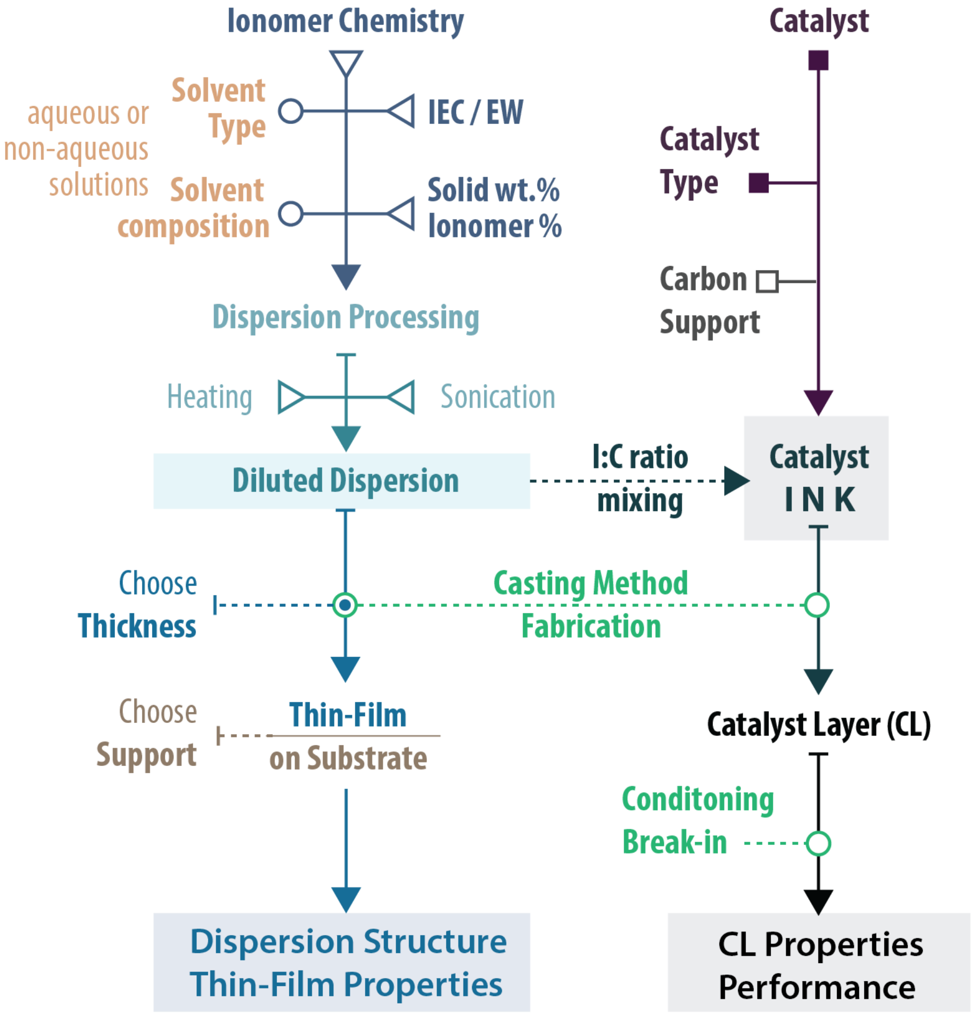
★ EOT integral cell target 749 mW/cm^2

Multicomponent Integration

Relevance to FY22 M2FCT Milestones

- **Q3 - Characterization of Ionomer adsorption**
- Measurement of ionomer adsorption and binding energy (GIXS/QCM/ITC) and characterization of the size and shape of aggregated catalyst clusters in catalyst inks (USAXS/SAXS) and correlation with catalyst-layer structure, coverages, and performance. (LANL, LBNL, ANL, NREL)
- **Q4 – MEA Fabrication**
- Demonstrate at least 10% improvement in current density at 0.7 V over baseline materials by varying MEA fabrication method, solvent ratio in catalyst-ionomer ink, ionomer type and content, etc. Total PGM loading constrained to 0.4 mg/cm2 or lower. MEA test conditions: 88°C, 2.5 atm, SR: 1.5 cathode/2 anode, 40% RH inlet, simulated integral cell. (All)
- **Overall** - Cell demonstrated 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.

Ionomers & Catalysts | Thin Films | Catalyst Layer
Structure-Property-Performance Correlations

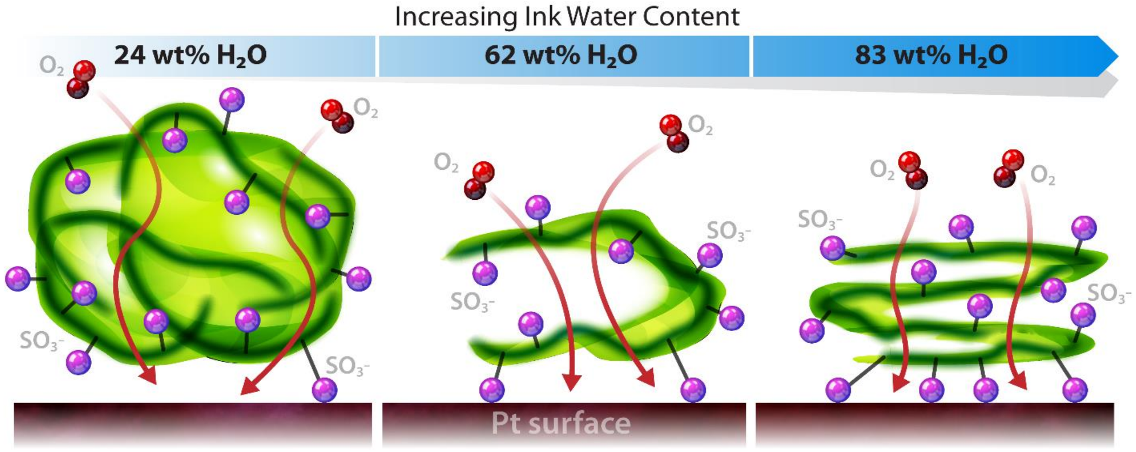


Million Mile Fuel Cell Truck (M2FCT)

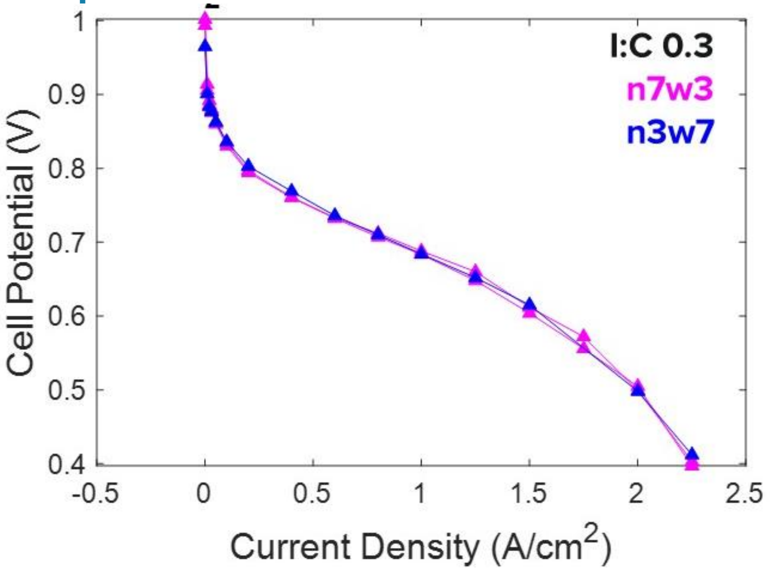
Created by A.Kusoglu

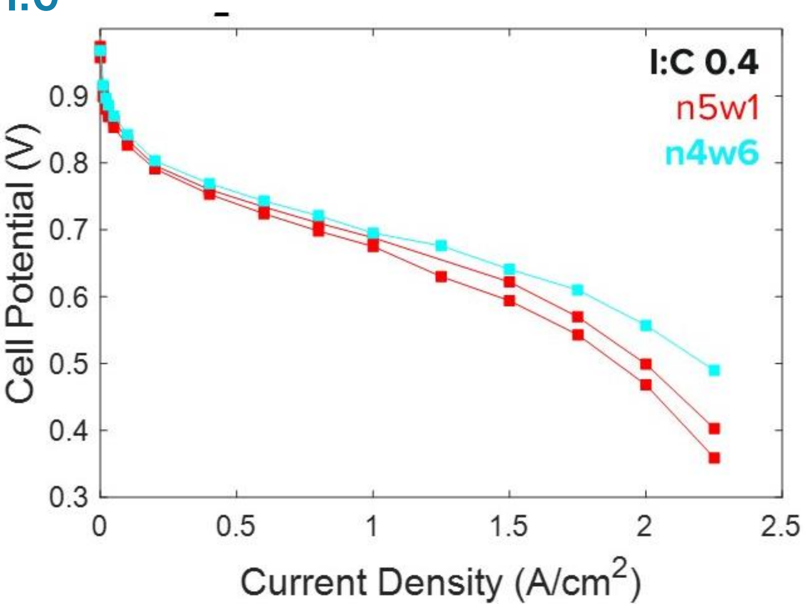
Accomplishments

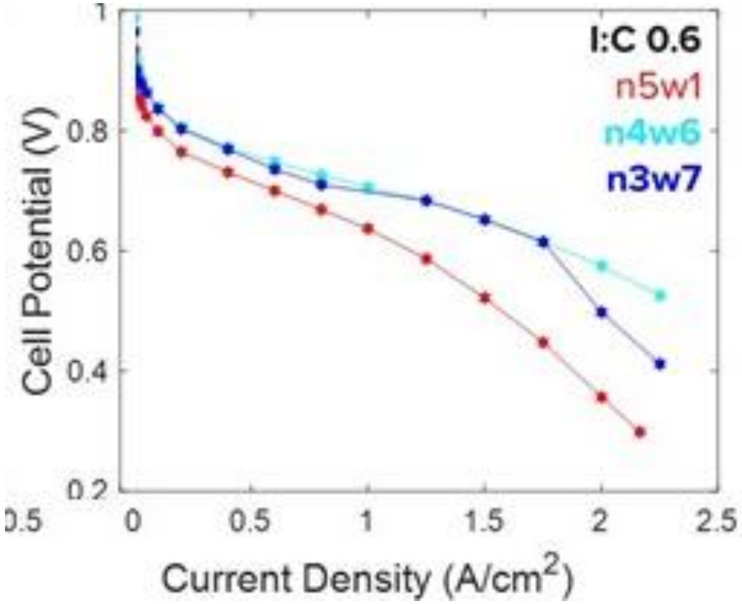
- **Motivation**
  - Extend prior work on electrocatalyst interfaces to lower I:C and chemistries



- Impact of solvents less critical at lower I:C





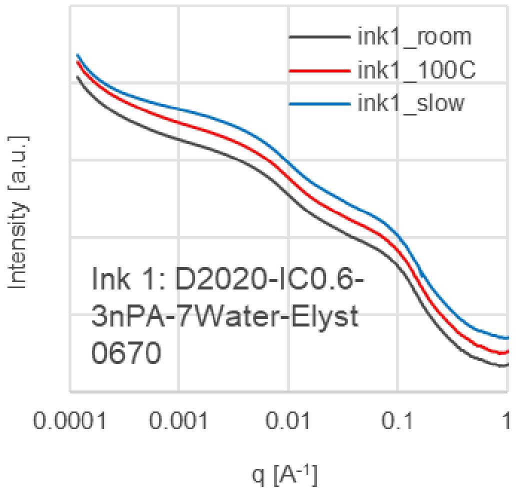


Pt/Vu D2020 - H_2/Air 90ºC 40% RH 250 kPa

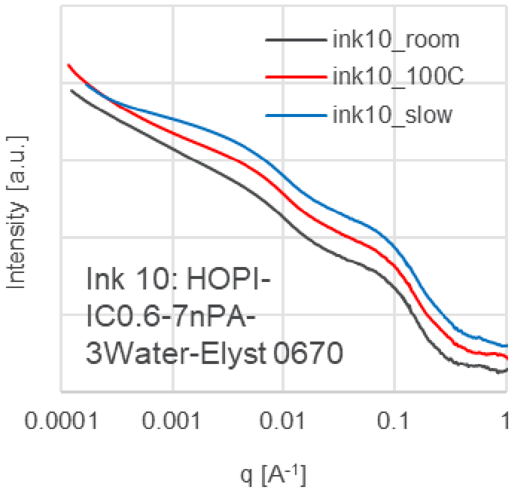
ACS Appl. Mater. Interfaces **2019**, 11, 50, 46953–46964

Impact of Ionomer, Solvent, and Solvent Removal Rate on Electrode Structure

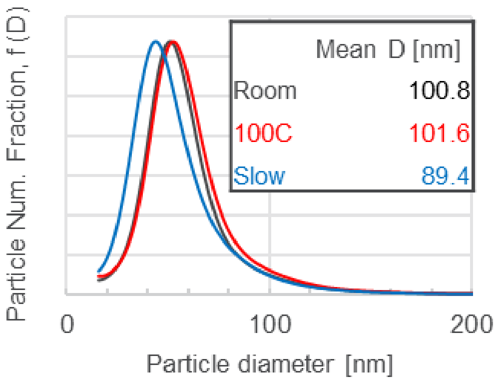
D2020



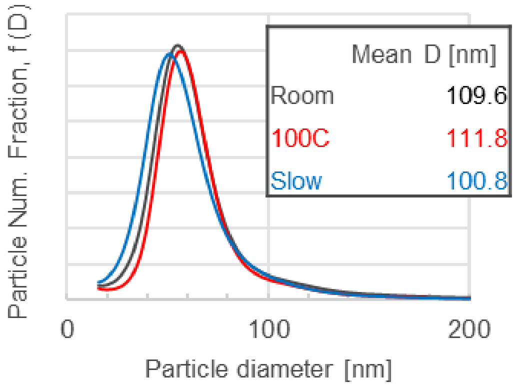
HOPI



Ink 1

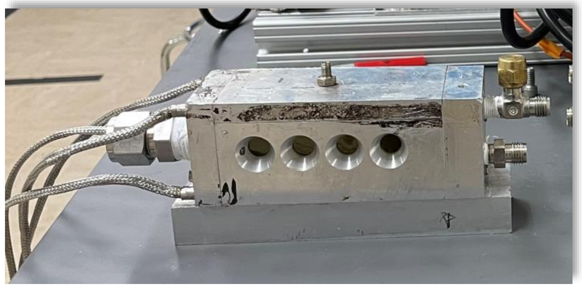


Ink 10

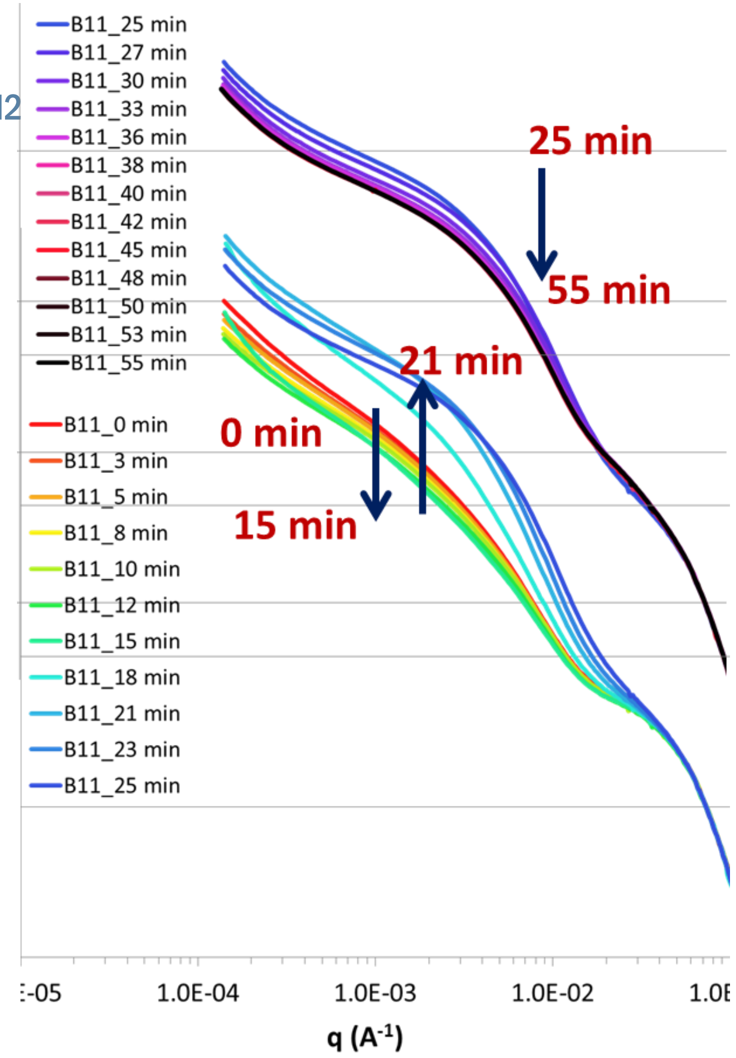


In situ drying in a humidity cell fixture

~1 mm thick ink painted onto Kapton with dry N2 flowing over the ink (30 wt% Pt/C catalyst)



- PSD changes during ink drying indicate structural evolution
- Solvent removal up to 15 mins
- Compaction during 18-27 mins
- No further change after 30 mins

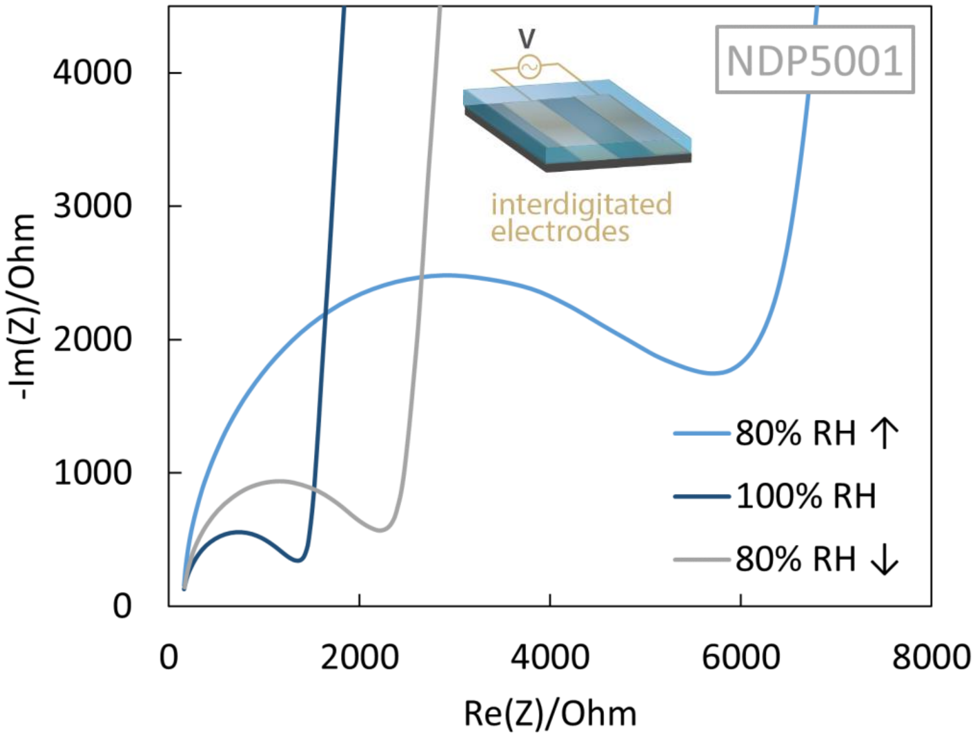


- HOPI consistently shows larger agglomerates
- Slow drying results in smaller agglomerates for both D2020 and HOPI inks
- MEA performance tests are on-going.

Ionomer Thin Film Characterization

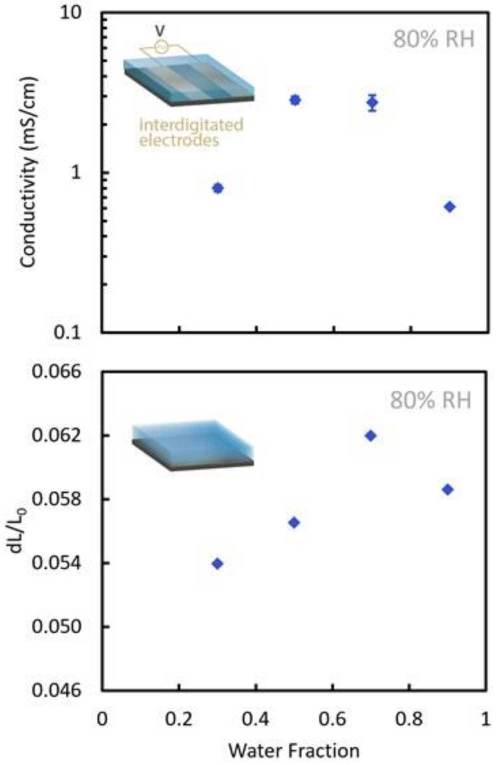
Electrochemical Impedance for Thin Films

- Ionomer films on inter-digitated substrates
- Conductivity in controlled environment

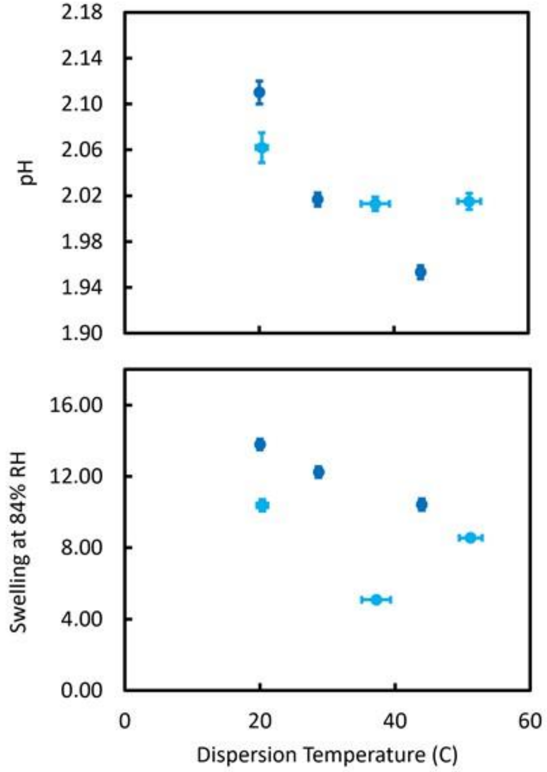


Processing Nafion Ionomers

Effect of Solvent Composition: on Swelling and Conductivity

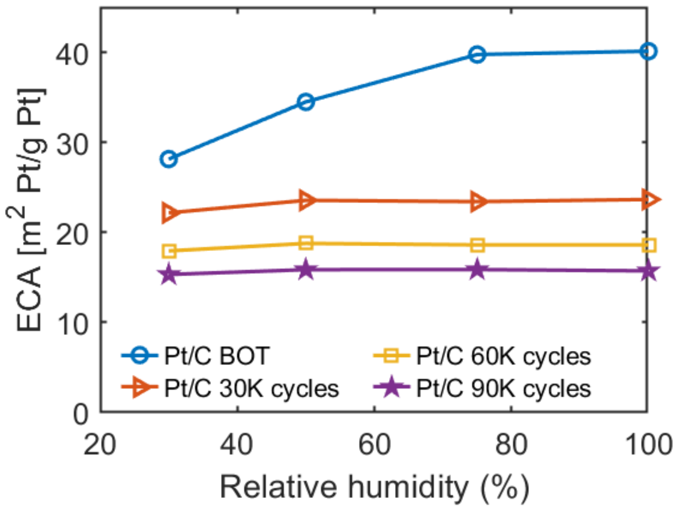


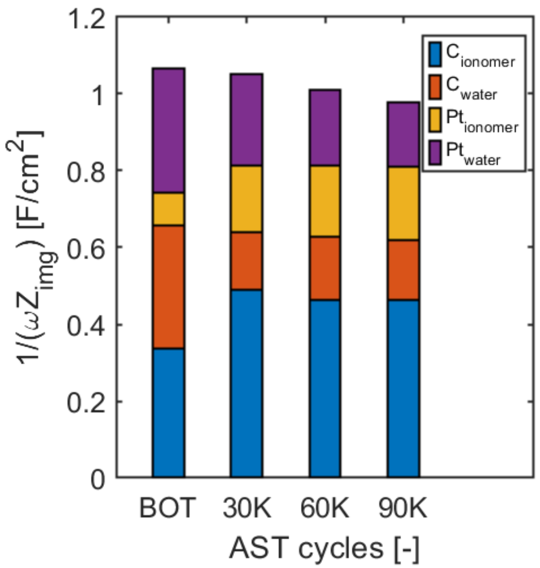
Effect of Dispersion Temperature: on pH and Swelling of cast-films

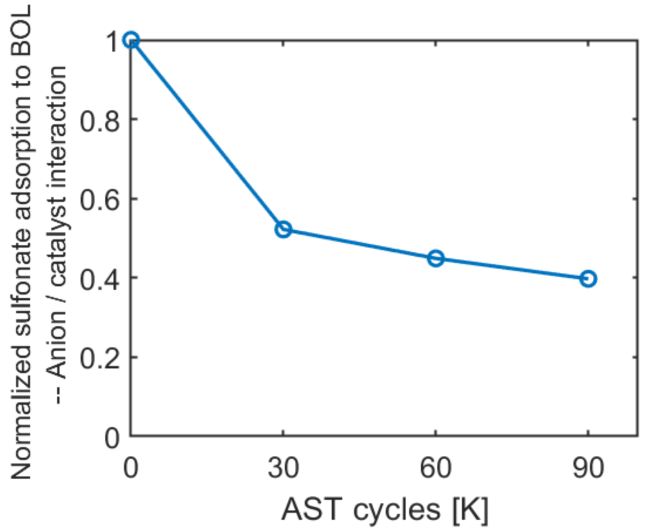


Accomplishments

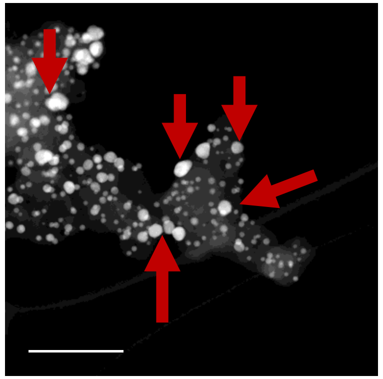
- **Objectives**
  - Assess benchmark performance for subsequent materials down selection
  - Inform systems analysis efforts
  - Examine evolving interfaces and structures

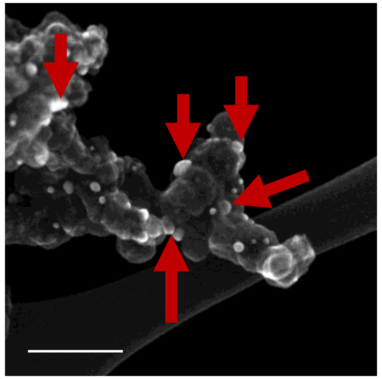


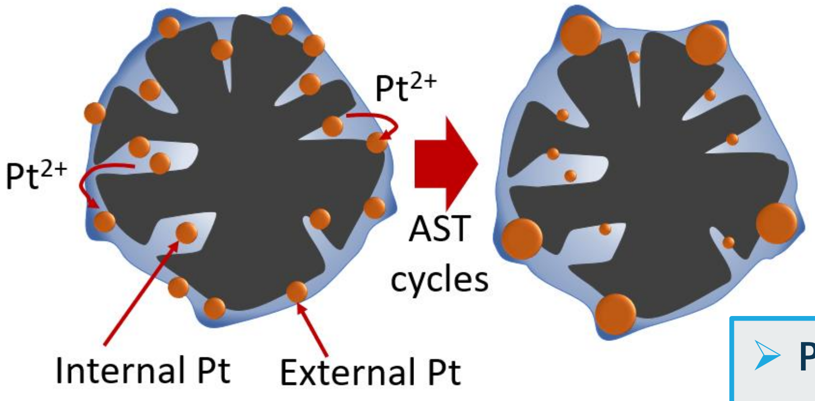




*From Cullen & Yu





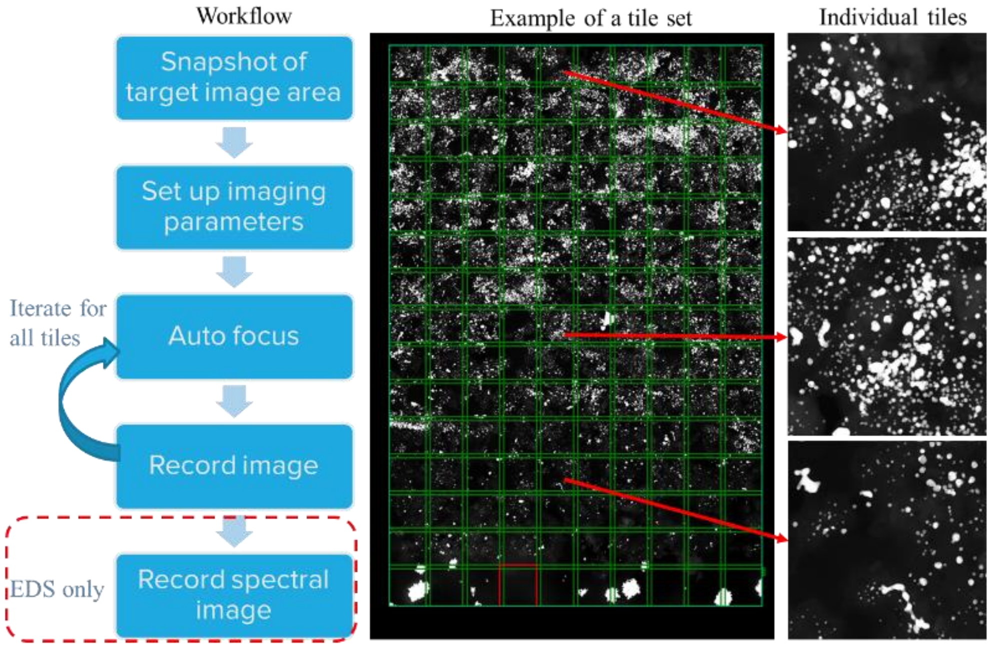


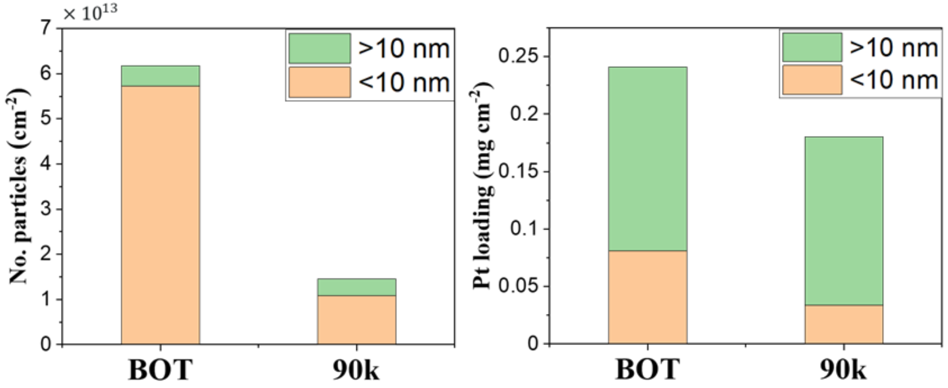
- Increased particle size
- Increased Pt accessibility
- Decrease in total capacitance
- Increase in C/I and Pt/I
- Decrease in C/W and Pt/W

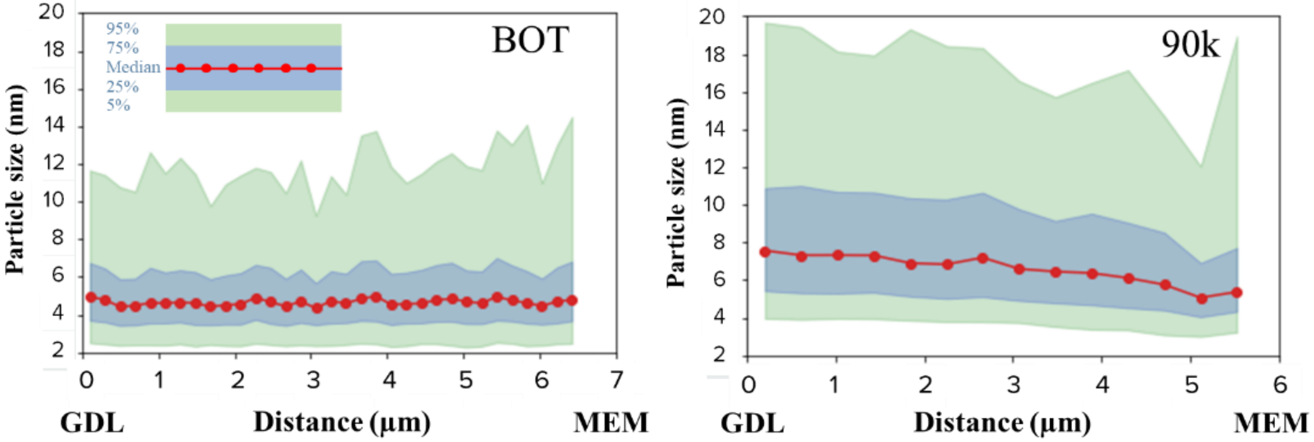
> Pt interactions with ionomer backbone become more relevant at EOL

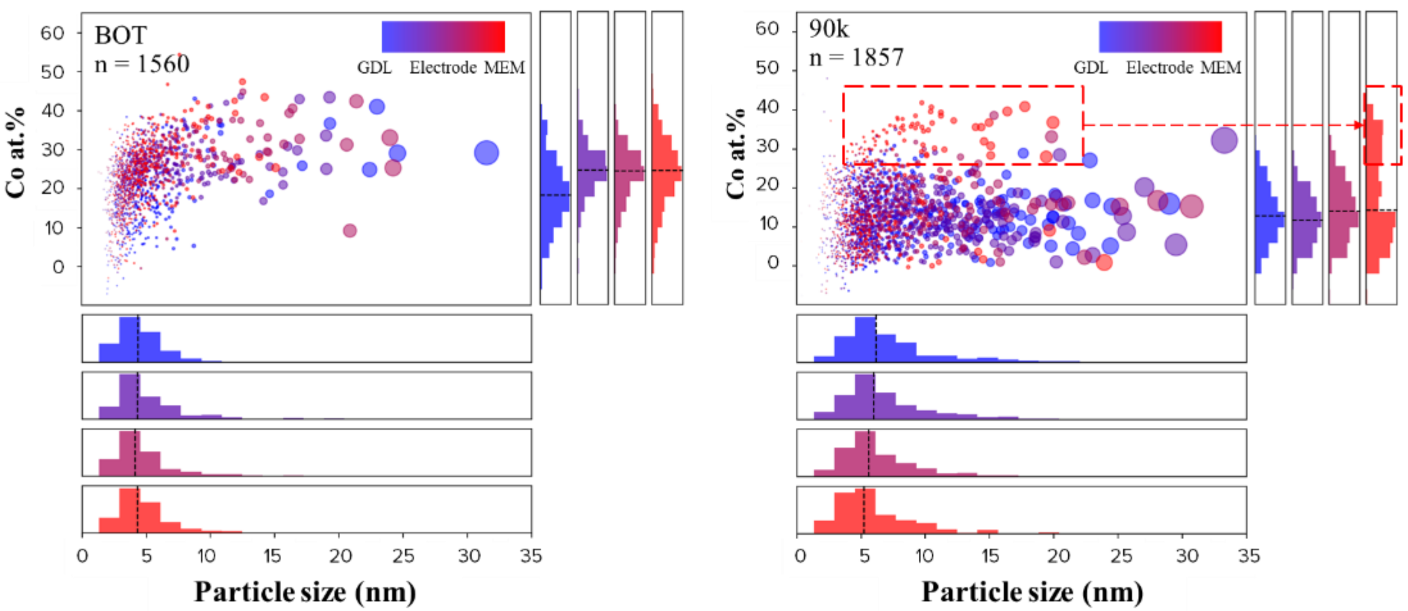
80oC, H_2/N_2, 100% RH, 150 kPa, 0.6 – 0.95 V square wave, 50 and 5 cm^2 data taken

Accomplishments | Automated Electron Microscopy



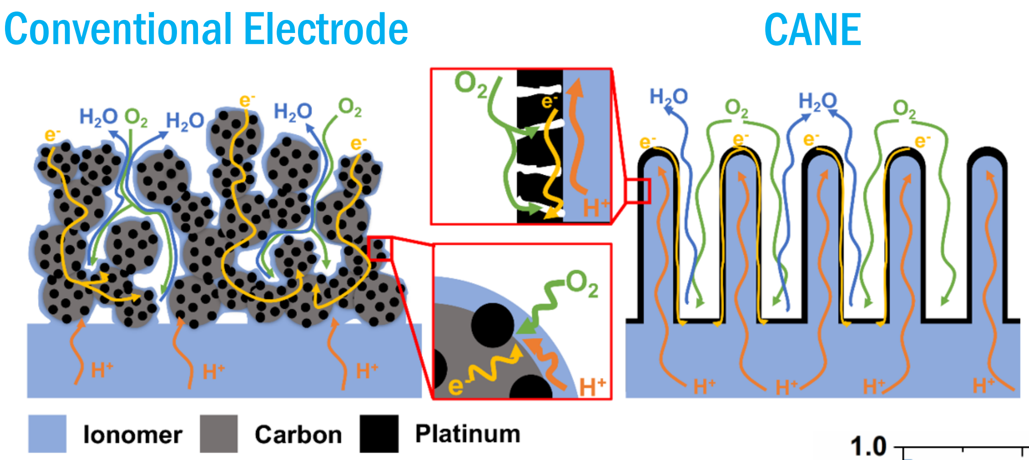


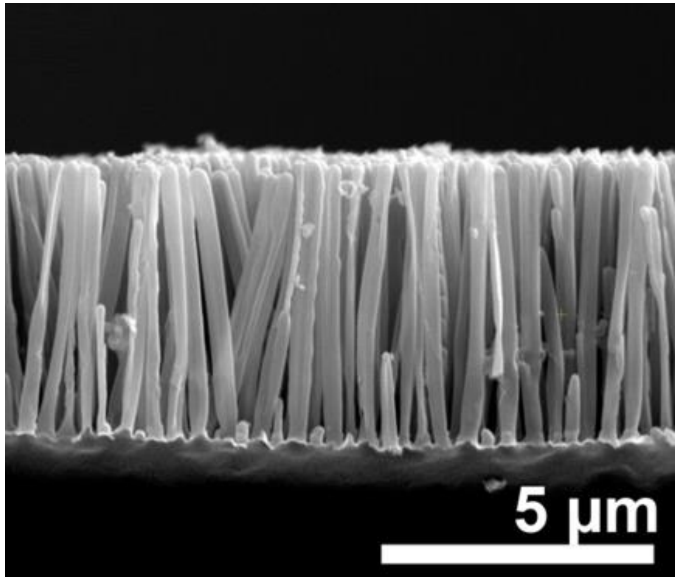




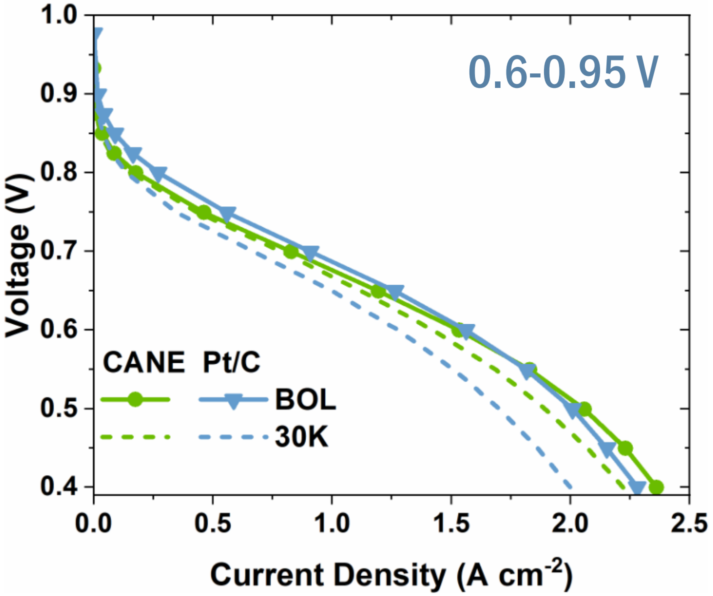
- Implemented automated electron microscopy workflow for particle size analysis
- Able to identify regions of more significant particle growth and Co loss

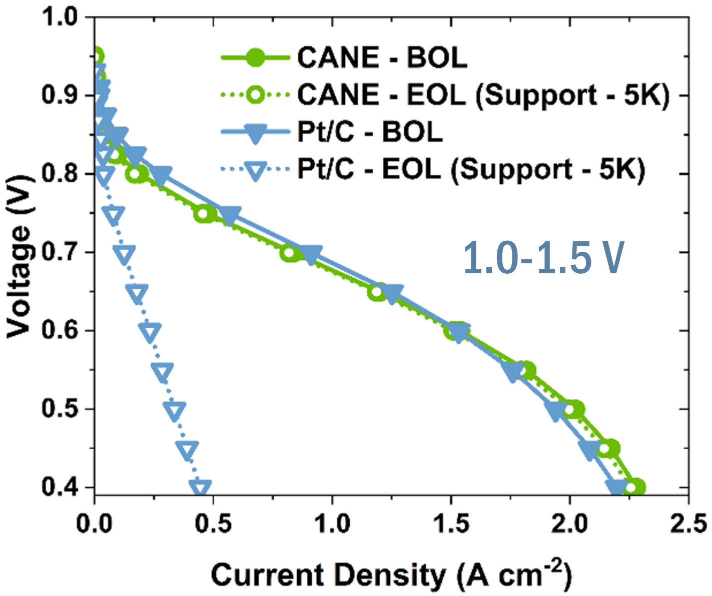
Coaxial Nanowire Electrode (CANE)





- CANE design uses coaxial nanowires with Pt shells encapsulating ionomer cores
- CANE provides BOL performance similar to Pt/Vulcan, but higher durability on catalyst and support ASTs





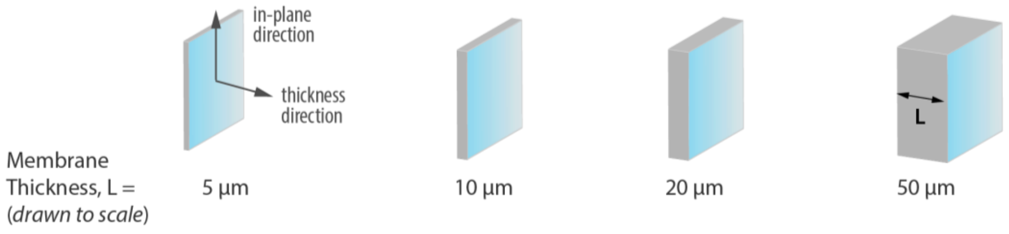
Nafion XL, 0.1/0.3 mg_{PGM}/cm^2, 80°C, H_2/air, 150 kPa, 100% RH

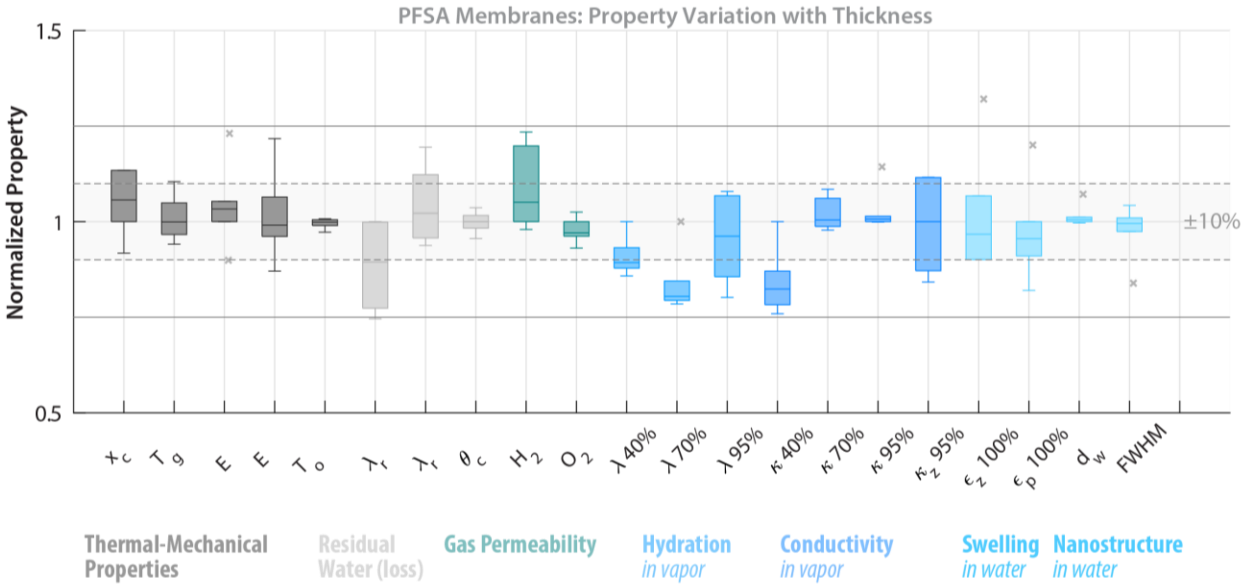
Manufacturing

Accomplishments | Membrane Studies & Database

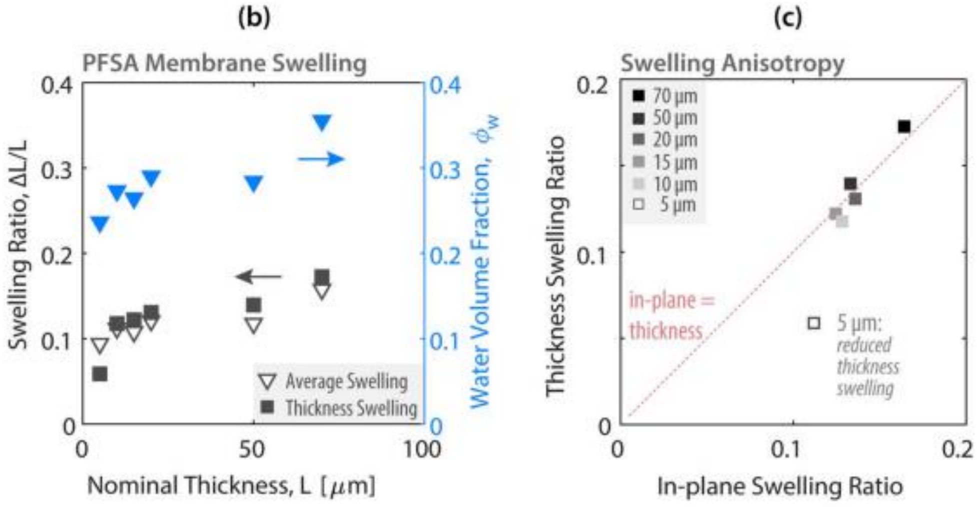
Membrane Properties: Effect of Thickness

- Thickness-dependent property map established for PFSA membranes





- ➢ Anisotropy for membranes dispersion-cast < 10 μm
- ➢ Implications for thin reinforced membranes and for durability where RH cycling is pronounced



(2021) J. Electrochem. Soc. 168 104517 https://iopscience.iop.org/article/10.1149/1945-7111/ac2973

Integration & Manufacturing-Scaling Science

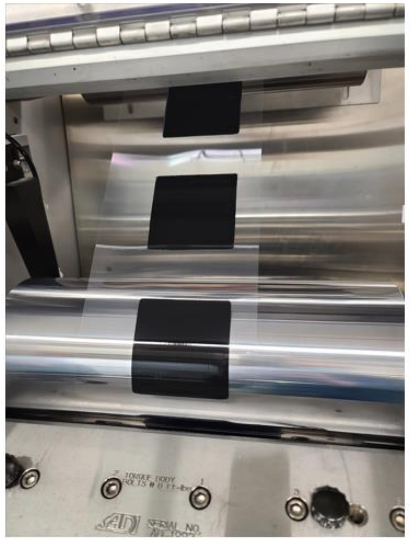
- Scaling Science for Ink and Electrode Development
  - Scaling science activities to support Clean H2 Manufacturing in the BIL related to manufacturing

Types of MEA Coating Methods

Slot-die Coater

Spray Coater

R2R Patch, Uniform and Controllable Coating



Doctor Blade

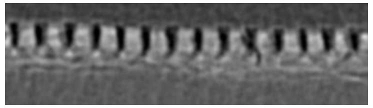
Mayer Rod

Baker bar

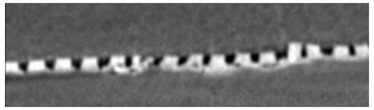
Effect of cracks on electrode durability

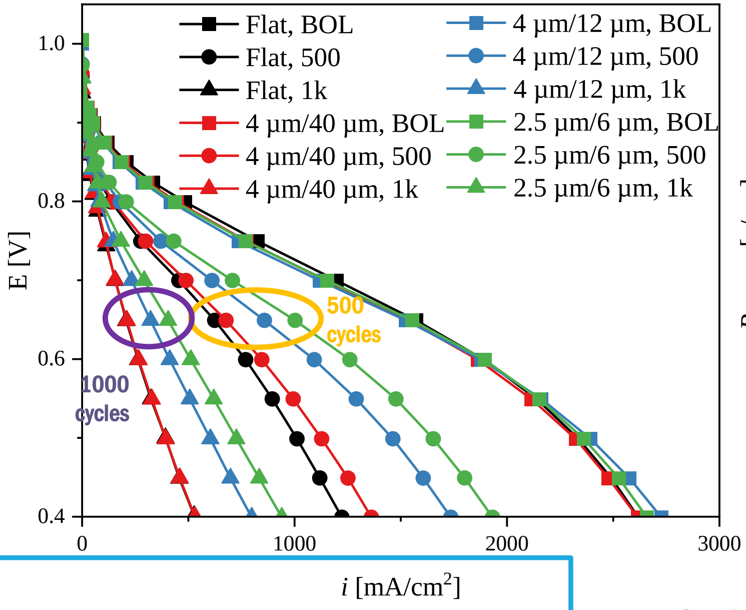
- Denser cracks help performance after carbon corrosion (1.0 – 1.5 V)
  - Cracks reduce R_{O2} at EOT

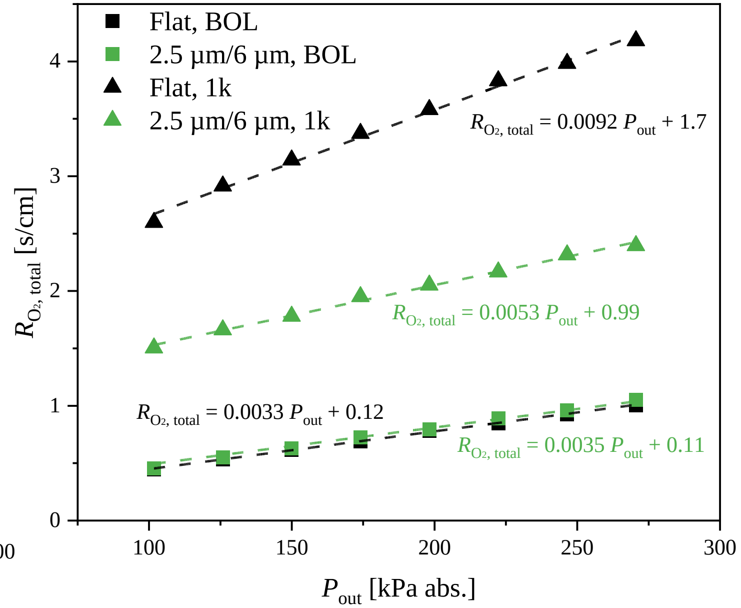
4 µm/12 µm, BOL



4 µm/12 µm, 1k

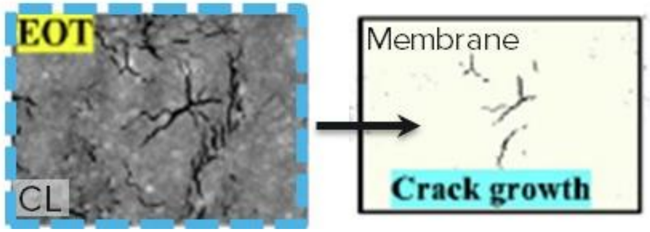


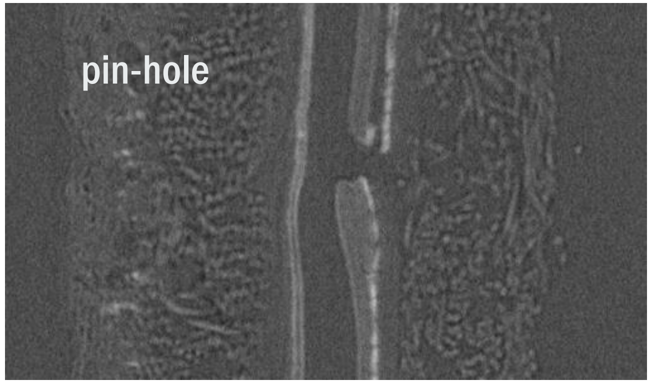




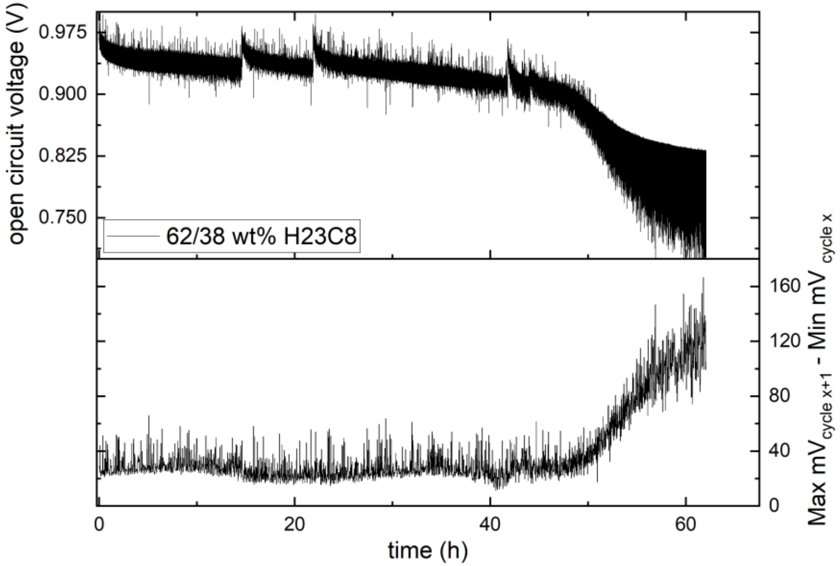
- **Objectives**
  - Elucidate correlation/causation between electrode cracking and MEA failure
  - Identify allowable feature sizes for HD applications

- Crack propagation to membrane



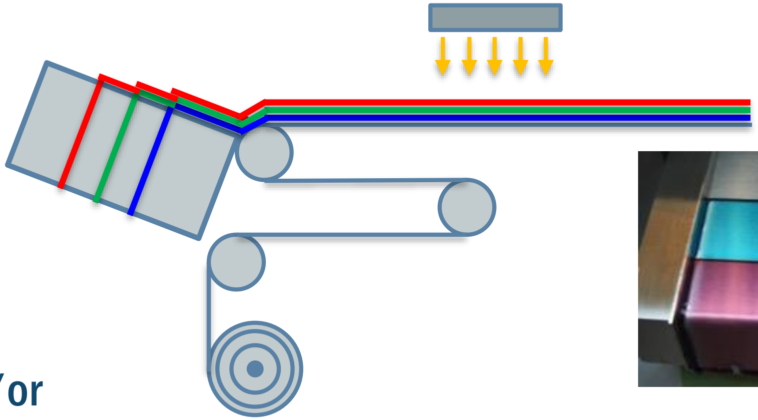


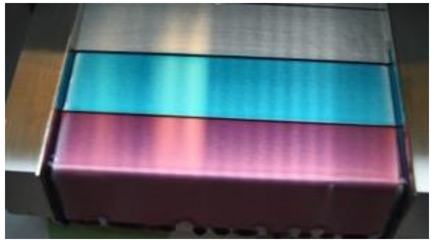
AST 101 kPa, 30/45s, 90C, 2.0/2.0 slpm, H_2/Air

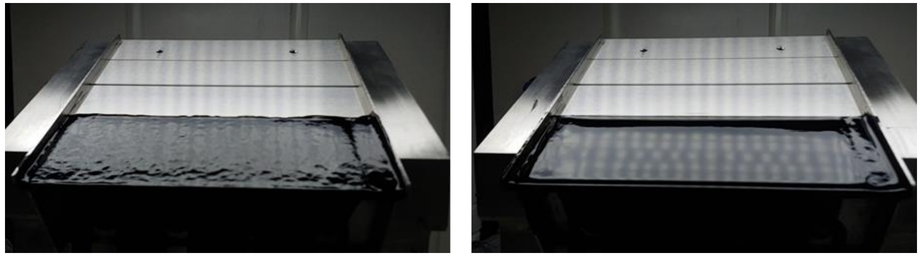


Multilayer structures for electrode optimization – Slide Die

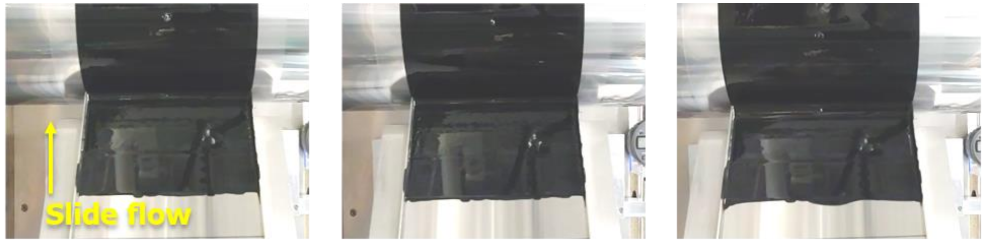
- Graded electrode structures could be beneficial to performance and durability
  - Control of properties at interfaces with membrane and GDL
  - Influence local transport and electrochemistry via different catalysts and/or ionomers
  - Functional additives to enhance durability
- Results
  - Multilayer slide die set up and validated
    - Completed modifications to R2R line
  - Established key process window parameters: impact of ink rheology, layer flowrates and thickness, die gap, line speed
    - Coordination with Sandia for flow modeling, via R2R Consortium
  - Coated 2-layer electrode as initial demonstration case
    - Pt/HSC / Pt/Vu 2-layer decals
    - Coordinated with ORNL for dual slot comparison, microscopy







Unstable → Lower catalyst solids → Stable



Loading ratio: 1:1
- Top: 3.89 ml/min
- Bottom: 6.08 ml/min

Loading ratio: 3:1
- Top: 5.84 ml/min
- Bottom: 3.04 ml/min

Loading ratio: 1:3
- Top: 1.95 ml/min
- Bottom: 9.12 ml/min

Durability

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

International Durability Working Group (i-DWG)

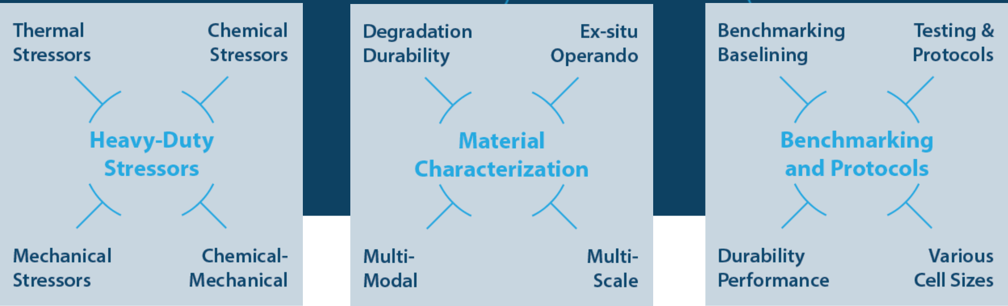
https://millionmilefuelcelltruck.org/idwg

International Durability Working Group (iDWG)

8 Countries
from America, Europe, and Asia

30 Institutions
participants representing governments, universities, industry and labs

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



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.

International Durability Working Group (i-DWG)

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.

M2FCT (funded by US DOE)

EU's IMMORTAL (by FCH 2 JU)

Japan's FC-Platform (by NEDO)

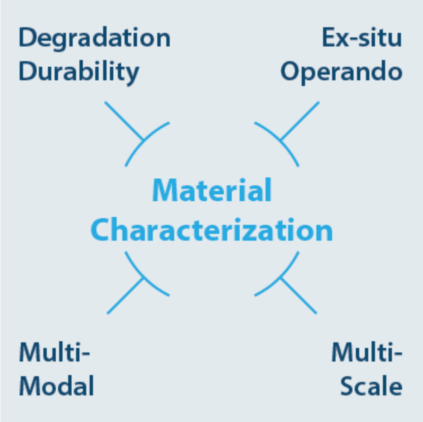
CEA-Grenoble (Embassy fellows)

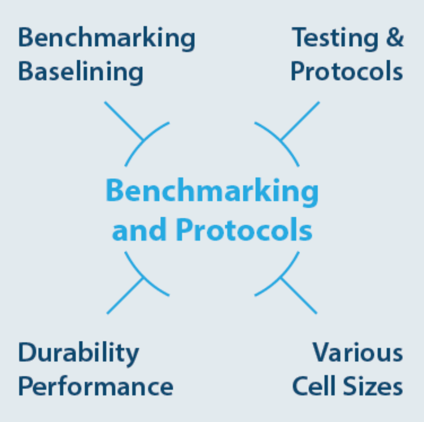
Over 80 researchers with access to resources hosted by M2FCT

ensure all relevant stressors are taken into account while developing heavy-duty ASTs

leverage the characterization tools and capabilities available to the various International groups to advance understanding of PEMFC performance and durability.

explore MEA testing at various scales to better understand the scaling of performance and durability from small differential cells to operating stacks

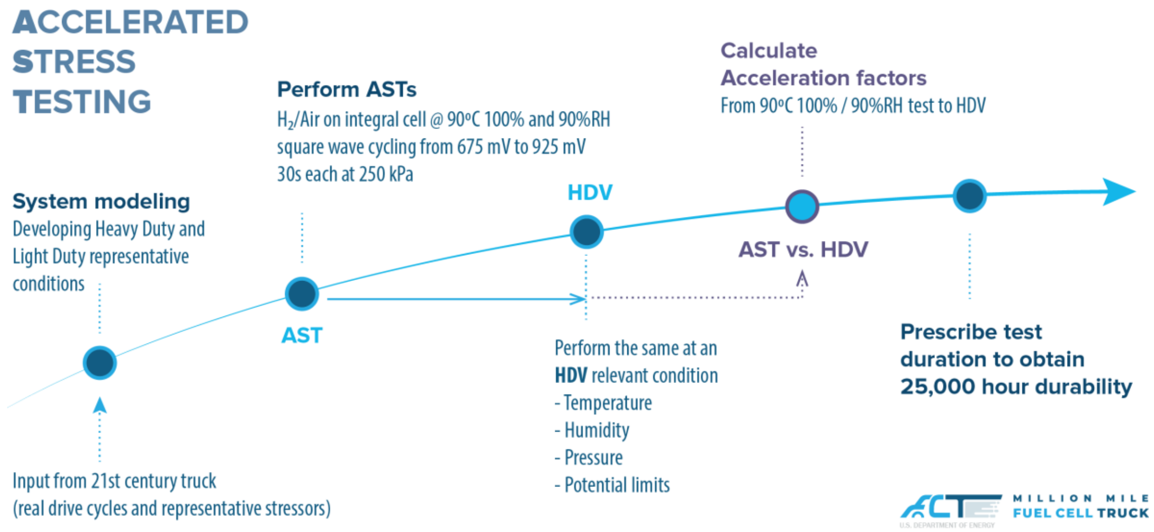




N. American AST Working Group

Define the 25,000 hour equivalent AST in the M2FCT 2025 Target

- Recommend protocols and targets related to heavy duty application of fuel cells
- ASTs for use in M2FCT for target evaluations (targets are End-of-Life)
- Accelerated Stress Tests (ASTs) to be developed
  - Catalyst
  - Catalyst support
  - Membrane chemical
  - Membrane chemical-mechanical
  - SD/SU
  - Anode H2 starvation
  - MEA drive-cycle



Participants
3M
ANL
Ballard
Carnegie Mellon
Chemours
Cummins
DOE
GM
LANL
LBNL
Nikola
NREL
ORNL
Plug Power
W.L. Gore

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

Baseline durability – Integral cells

Lab call and M2FCT Target

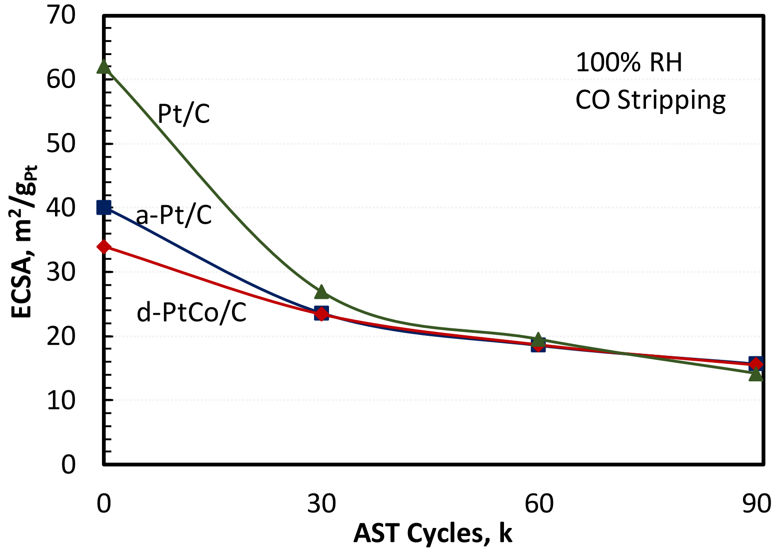
- 2.5 kW/g_{PGM} at 0.7 V (1.07 A/cm^2 current density) after 25,000-h equivalent accelerated stress testing
- 750 mW/cm^2 power density for 0.3 mg/cm^2 total Pt loading, 1.5/2 c/a stoic, 2.5 atm, 88ºC, 40% RH, integral cell conditions

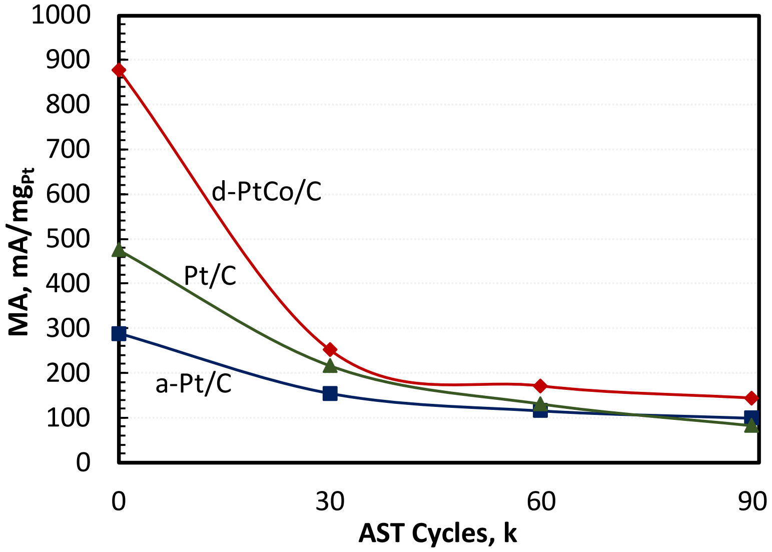
M²FCT FY2021 Q4 Milestone

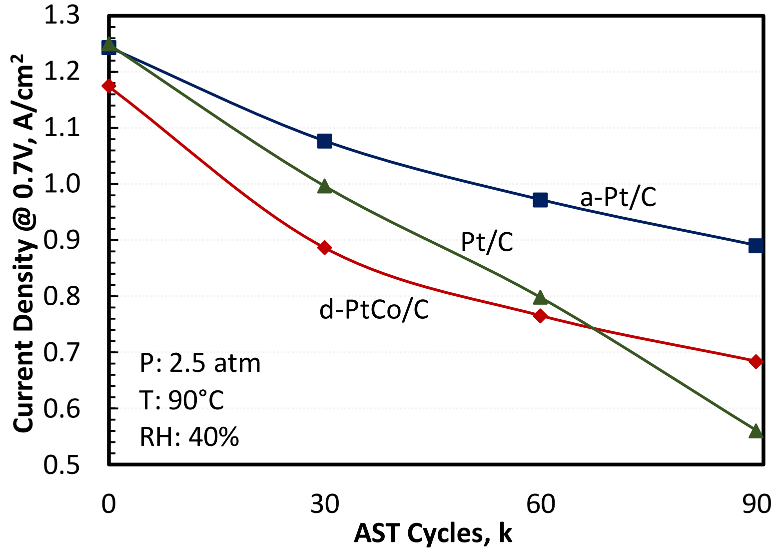
- AST defined as 0.6 V – 0.95 V square wave with 0.5 s ramp and 2.5 s hold in H_2/N_2 at 80ºC and 100% RH, 90k cycles
- Integral Cell Conditions for Pol Curves: 1.5(c)/2(a), 2.5 atm, 90ºC, 40% RH

	Number of Cycles	ECSA m^2/g_{Pt}	MA mA/mg_{Pt}	PD at 0.7 V mW/cm^2	PD at 0.7 V kW/g_{Pt}	PD at 1.07 A/cm^2 mW/cm^2	PD at 1.07 A/cm^2 kW/g_{Pt}
Pt/C	0k	62	476	874	2.9	772	2.5
	90k	14	82	393	1.3	647	2.1
a-Pt/C	0k	40	289	870	2.8	768	2.4
	90k	16	99	622	2.0	721	2.3
d-PtCo/C	0k	34	878	822	2.6	766	2.4
	90k	16	144	479	1.5	660	2.1

EOL modeling being done at 50% ECSA loss

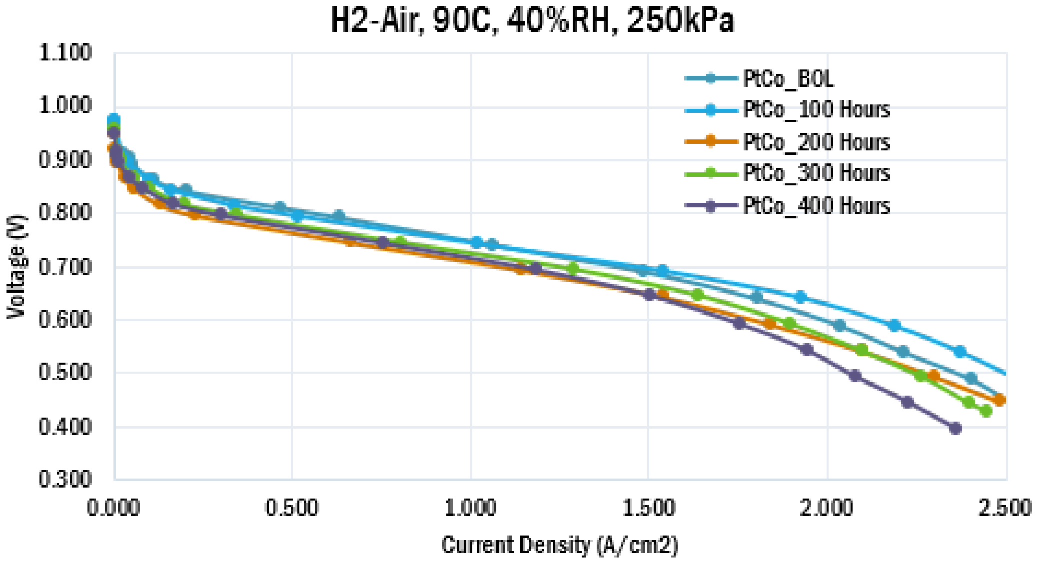


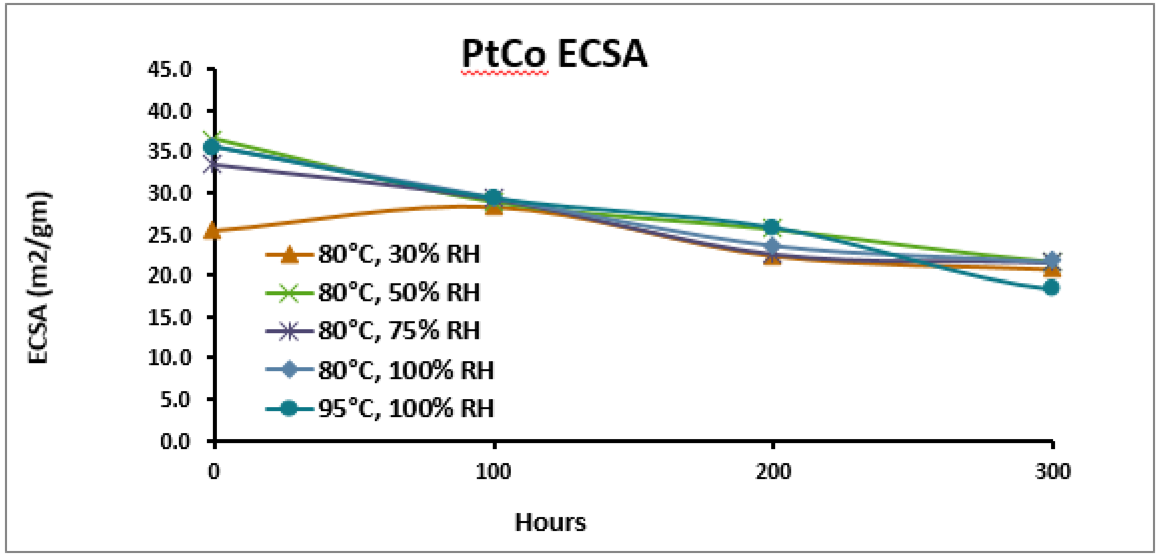


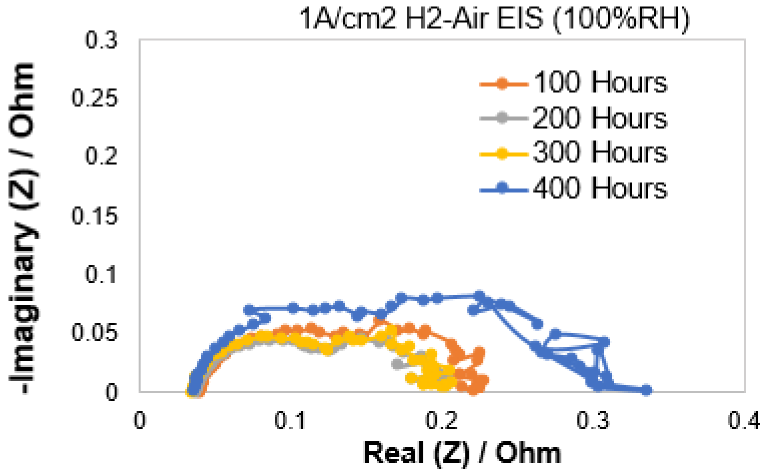


H_2/Air MEA AST

Cycle from 0.675 (30s) to 0.925V (30S)







- Similar ECSA loss for H_2/Air and H_2/N_2 tests
  - BOT = 40m^2/gm for Pt/C and 35 m^2/gm for PtCo/C
  - EOT ≈ 15 m^2/gm after 90K H_2/N_2 cycles at 80ºC or 500 hours of H_2/Air cycles (30,000 cycles) at 90 ºC
- Similar kinetic performance losses seen in H_2/N_2 and H_2/Air tests
- Mass transport losses observed in H2/Air tests

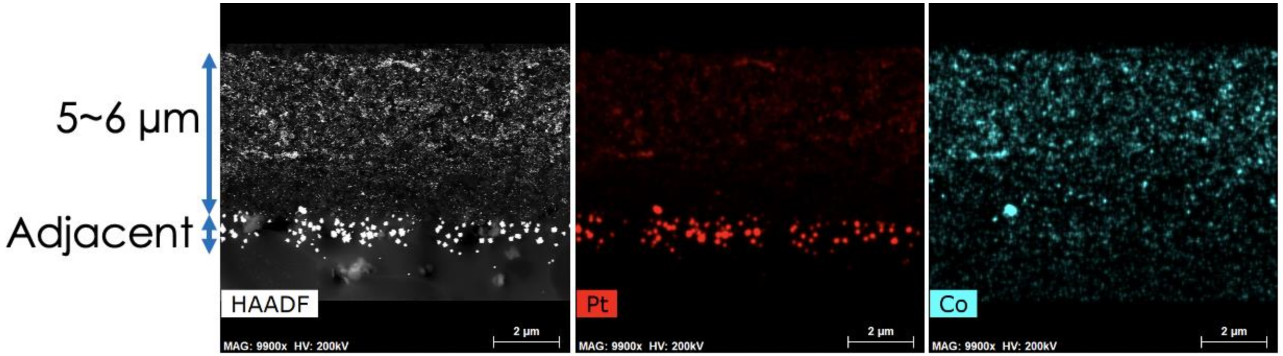
Compare H_2/Air and H_2/N_2 ASTs

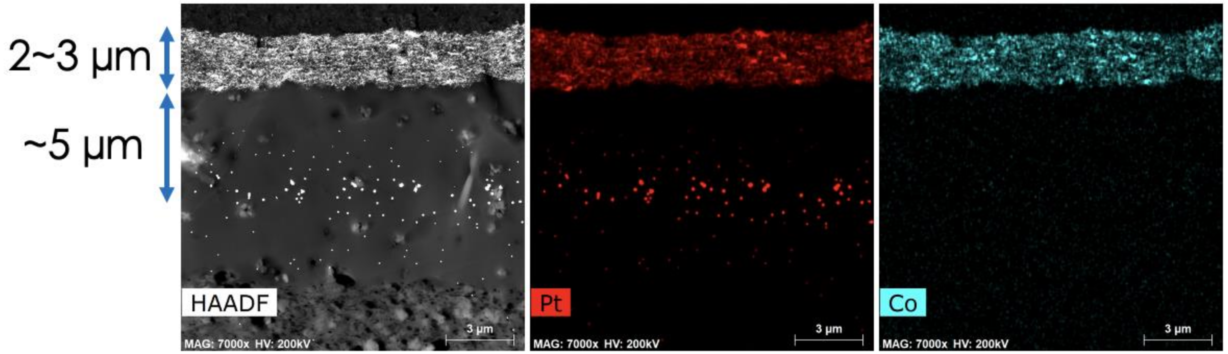
AST in N2, 80 °C, 0.6V to 0.95V, 90k cycles

	Co at.%	Pt loss%
Map1	13.0	36.7
Map2	12.6	34.2
Map3	13.9	37.9
Average	13.2±0.5	36.3±1.5

AST in Air 300 hrs. 90 °C, 90%RH, 0.675 to 0.925V

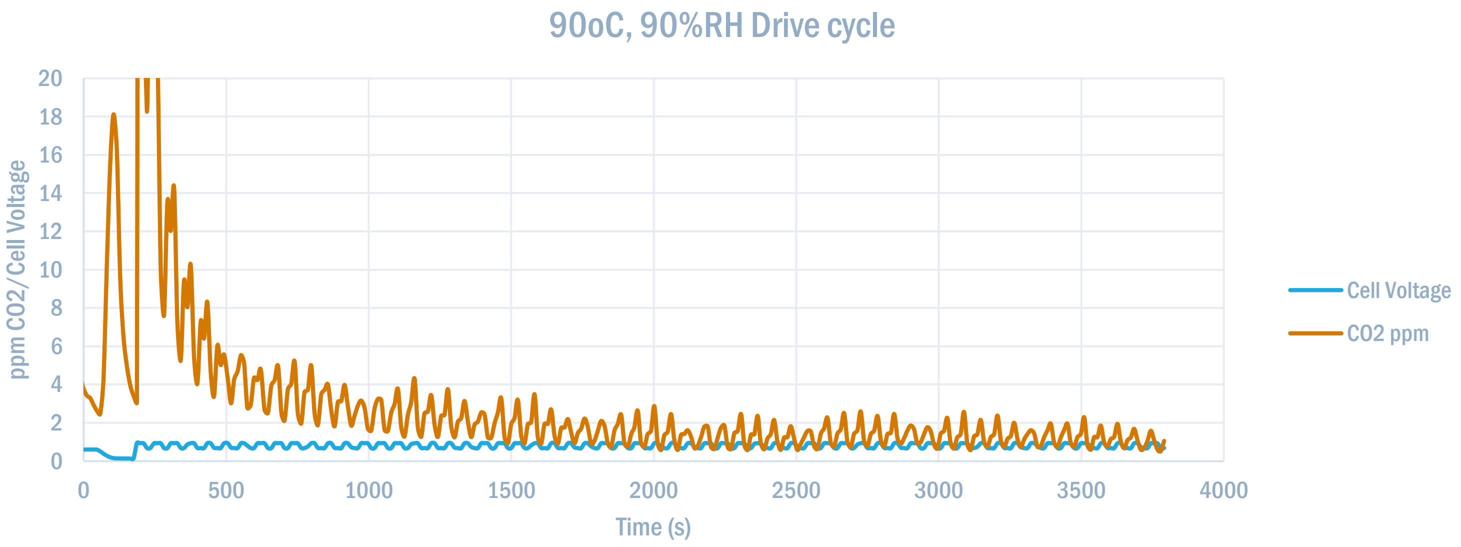
	Co at.%	Pt loss%*
Map1	21.3	12.1
Map2	21.8	15.8
Map3	21.6	10.7
Average	21.6±0.2	12.9±2.2



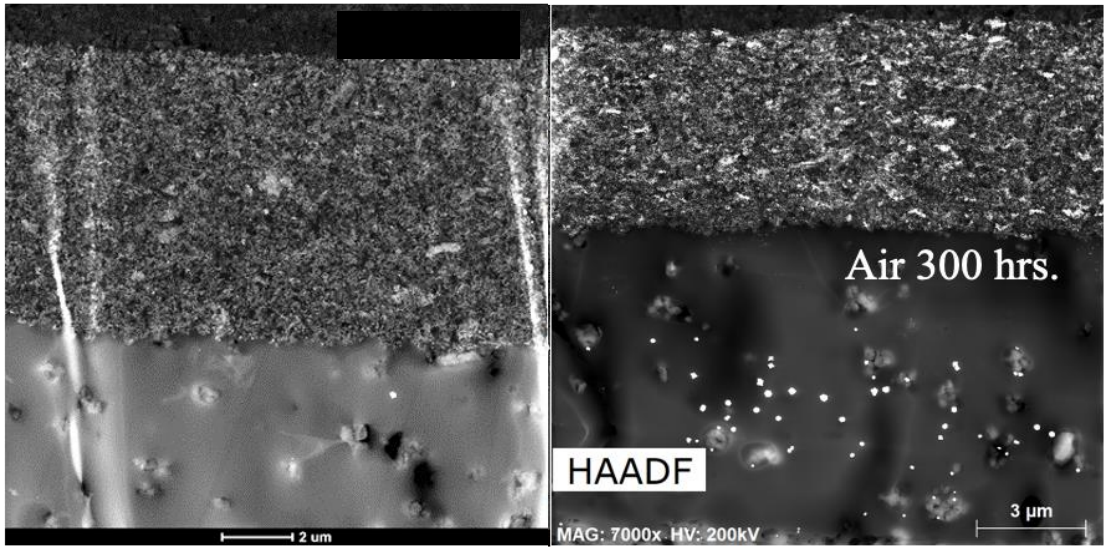


- Slower Co dissolution and lower mass activity decay in H_2/Air AST
- Pt depletion layer in H_2/N_2 tests (not observed in H_2/Air tests)
- Pt band further in the membrane for H_2/Air AST

Carbon Corrosion and Membrane Degradation During H_2/Air AST



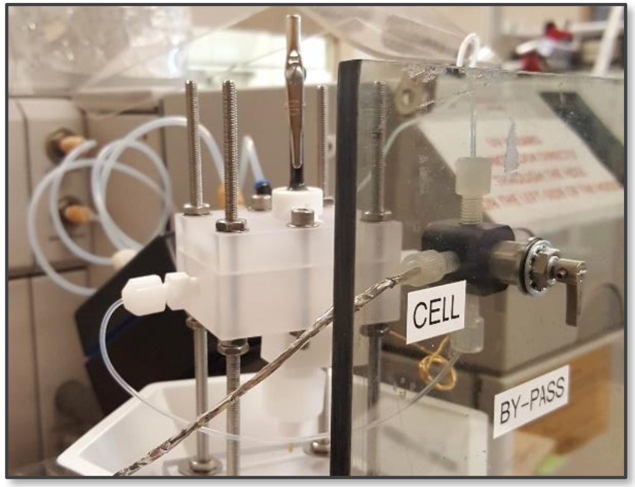
- Significant carbon corrosion observed during H_2/Air AST testing
- Drops dramatically over time (not all carbon the same, easily accessible carbon corrodes first)
- Parametric study under way



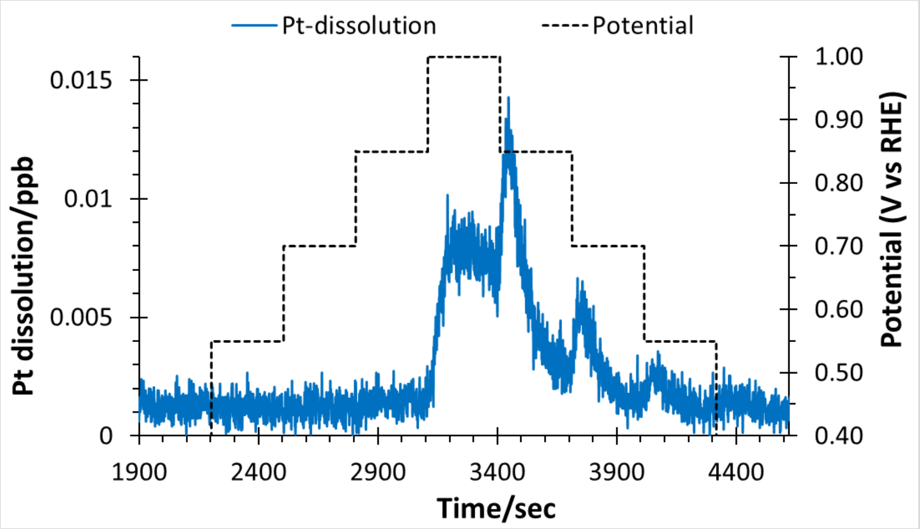
- Fluoride emission rates during H_2/Air AST at 90°C ≈ 3.5 $\mu g/cm^2$.hr for N211
- For Nafion HP membranes the rate increase from ≈ 0.2 to 2 $\mu g/cm^2$.hr over the 500 hours of testing
- Need to evaluate advanced membranes (NC700)
- Also performing parametric study with GM supplied MEAs

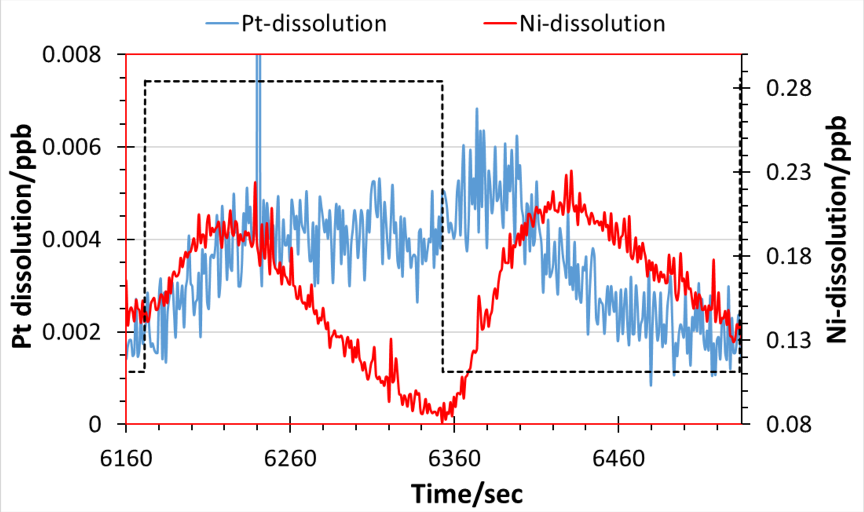
Understanding Catalyst Degradation using Online ICP-MS

Pt and Ni dissolution from 30 wt% PtNi/HSC

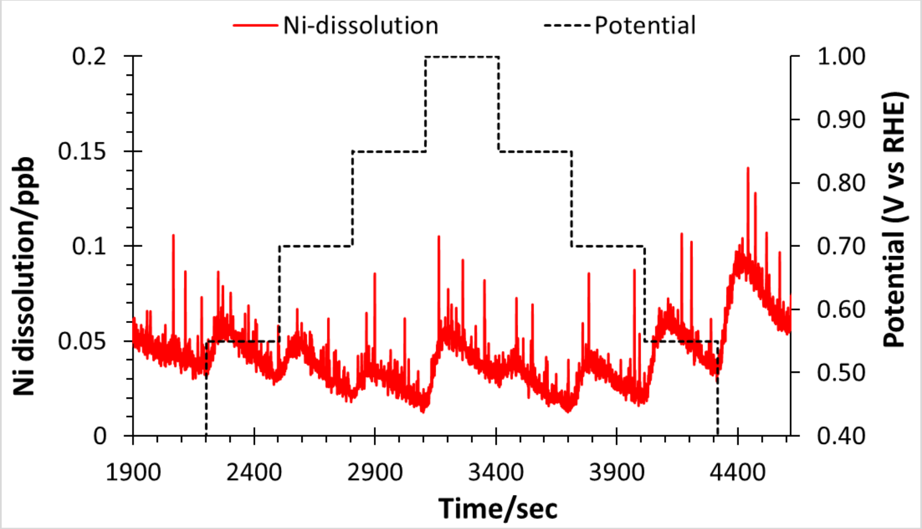


Electrode loading: 4 μg-Pt/cm^2
Electrolyte: Ambient 0.1 M $HClO_4$
Square wave: 0.4 V and 0.95 V, 3 min each



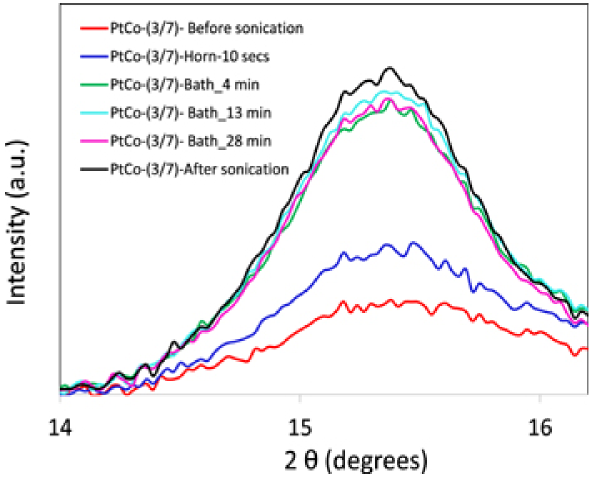


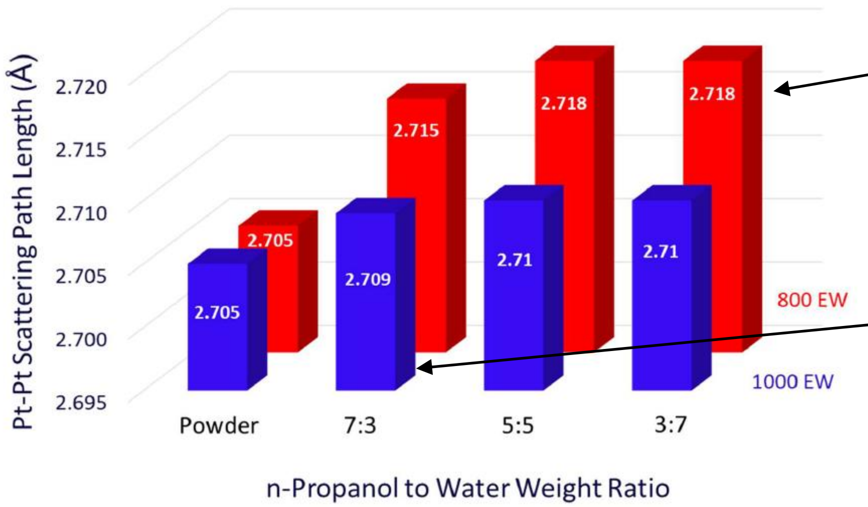
- Ni dissolution rate 40x Pt dissolution rate
- Pt and Ni dissolution increase after potential transitions
- Pt dissolution stabilizes while Ni dissolution passivates
- Initial studies of BNL nitride PtNi show comparable results



Degradation of Platinum-Cobalt Alloy Cathode Catalysts

Umicore Elyst Pt30 0670





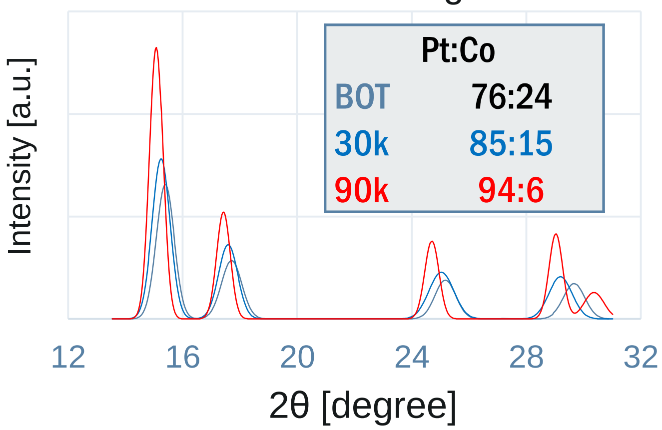
Co^{2+}/Sulfonate in CCL = **0.26** (extent of Co^{2+} loss from particles calculated from change in WAXS peak position)

Co^{2+}/Sulfonate in CCL = **0.13**

Umicore Elyst Pt50 0690

AST Cycling for 30k and 90k

WAXS fitting



	Pt:Co
BOT	76:24
30k	85:15
90k	94:6

78% of initial Co lost after 90k:

100% exchange of SO_3^- groups in CCL

FY'21 Q3 Milestone: Define acceptable transition metal loss from alloy catalysts (% of sulfonic acid sites in ionomer layer) with respect to electrode layer losses.

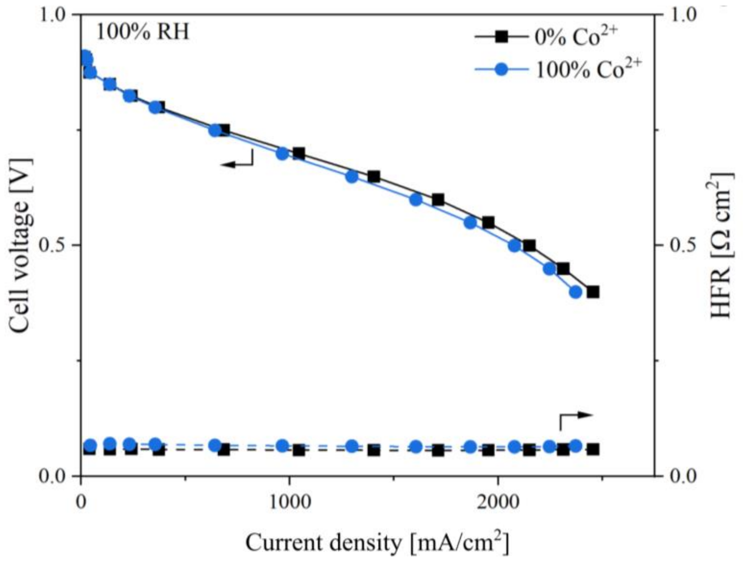
- Umicore Elyst Pt50 0550 0.25mg_{Pt}/cm^2, 2cm x 2cm
- Ink: D2020, 0.06 g catalyst, 16.6 g H_2O:NPA (4/3 v/v), I/C=0.9
- Soak Pt/C decals in Co^{2+}/HNO_3 solutions (0.15 M HNO_3)
- Measured Pt and Co content using XRF
- Prepare CCMs from these decals and measure H_2/Air performance as a function of RH, diagnostics

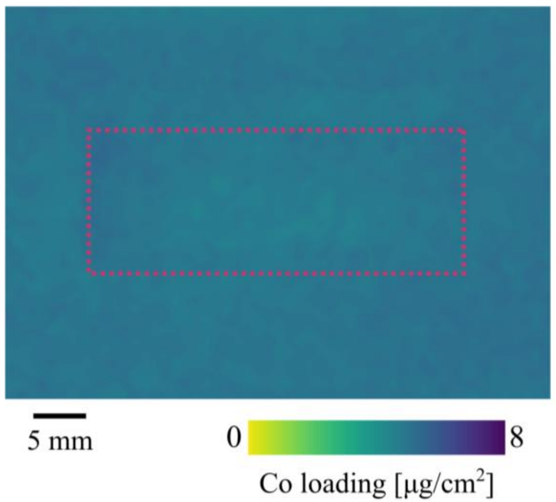
D. Myers et al., J. Electrochem. Soc., 2021, 168, 044510.

Effects of Co^{2+} Concentration in Electrode on Performance

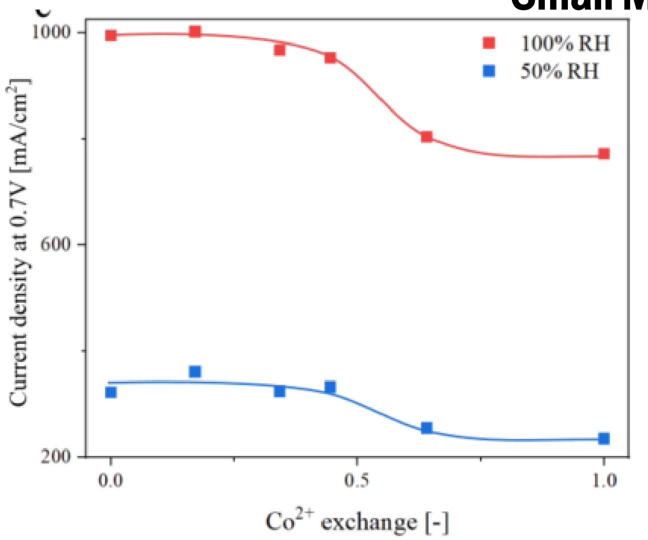
- Co^{2+} was doped into a decal electrode.
- Reduction in inactive membrane area led to stronger Co^{2+} effects
  - Inactive membrane area acts as Co^{2+} sink
- Co^{2+} exchange fraction and performance showed a sigmoidal behavior.
- Co^{2+} was also doped in membranes with the same total Co^{2+} amount in the MEA.
  - The performance was the same as the electrode-doped MEA.
  - Co^{2+} content in the active area governs the performance loss.
- A thicker membrane (N212) suppressed effects of Co^{2+} doping.

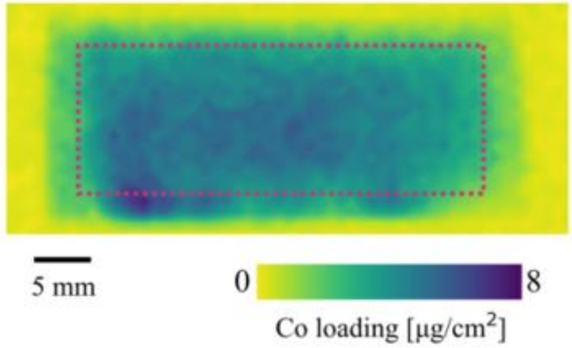
Large Membrane





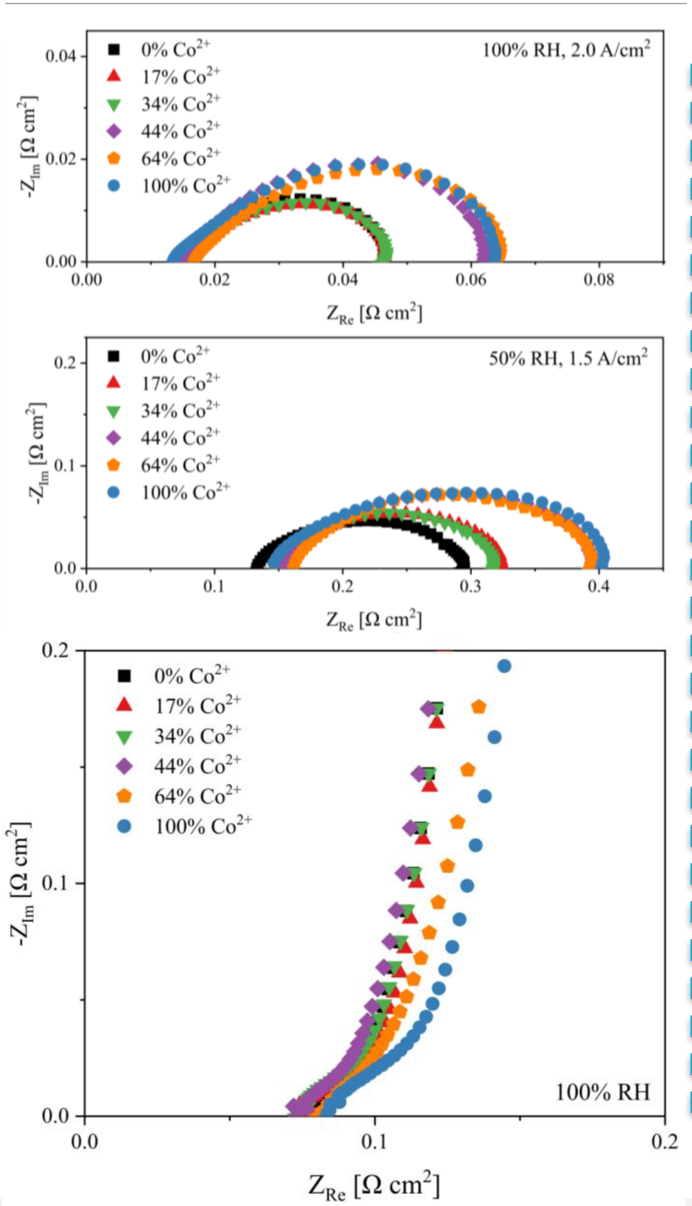
Small Membrane

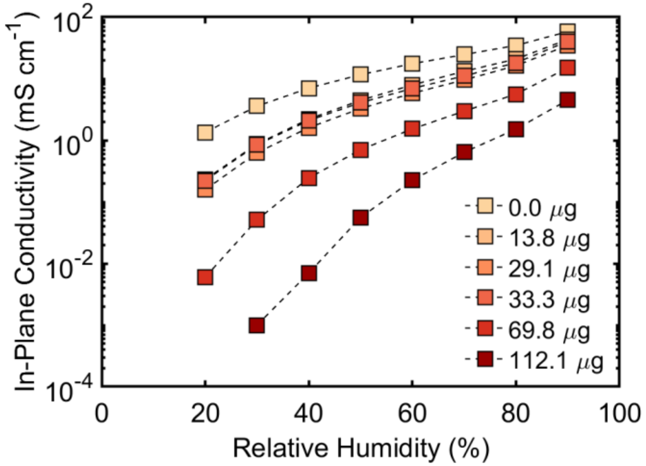


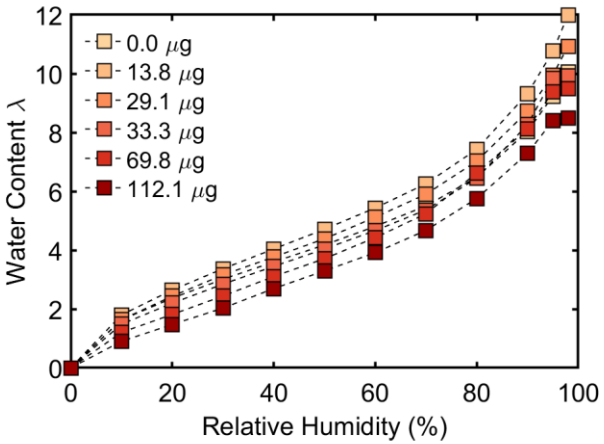


Co^{2+} Effects on Proton and Oxygen Transport

- R_{MT} increased with increasing Co^{2+} doping.
  - The volume of hydrophilic domain in the ionomer reduces, resulting in lower O_2 permeability.
- Sheet resistance increased with increasing Co^{2+} doping.
  - Co^{2+} ion-exchanging with sulfonic acid sites lead to poor proton transport.



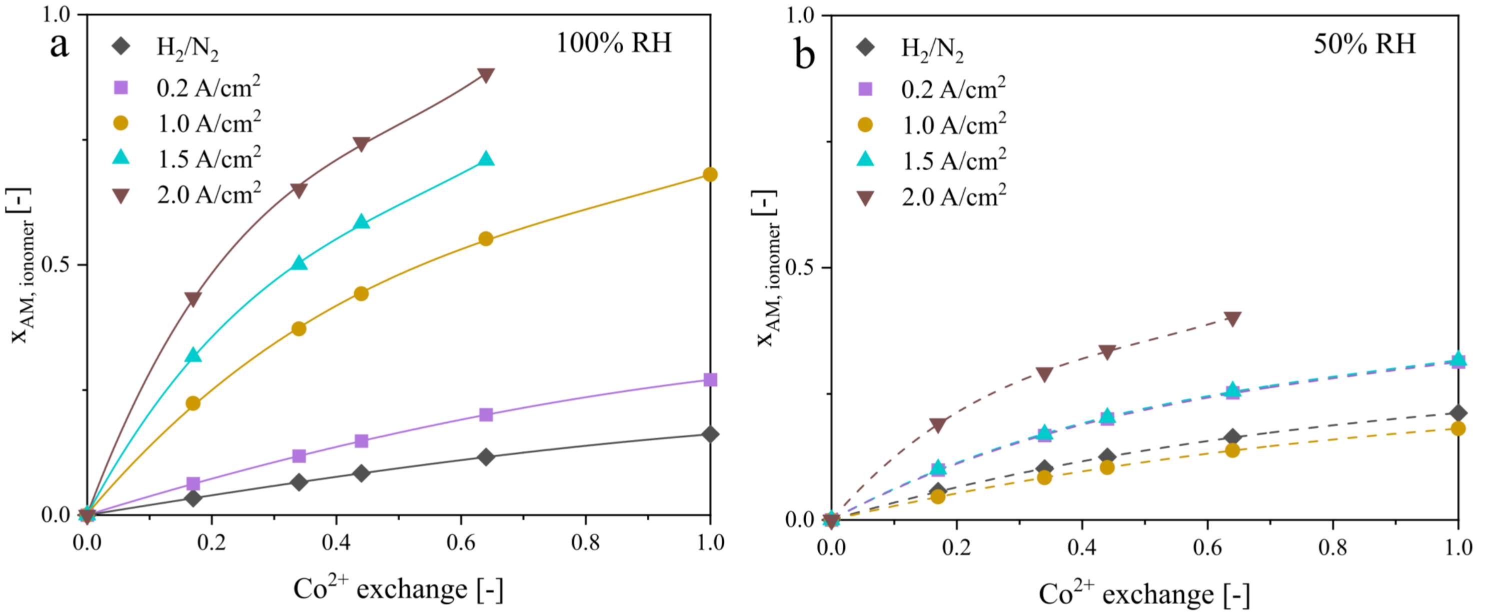




- LBNL performed ex situ measurements on membranes provided by LANL to study Co^{2+} effects on λ and conductivity.
- Conductivity decreased with initial Co^{2+} doping, but plateaued between 13 and 33 μg/cm^2. Then, conductivity decreased with increased Co^{2+}.
- Water uptake decreased monotonically with Co^{2+}.

Partitioning of Co^{2+} Between Membrane and Electrode

- Combining LANL's impedance results and LBNL's λ and conductivity measurements, ANL modelled [Co^{2+}] across the MEA
  - Different x_{AM} trends at 50%RH, due to *i* affecting ionomer λ, subsequently affecting Co^{2+} mobility and flux from membrane.
- Co^{2+} effect on kinetic resistance was negligible.
- At 100%RH, R_{MT} increases when 1.0 → 1.5 A/cm^2 due to flooding
- At 50%RH, R_{MT} decreases with increasing *i* due to higher ionomer λ.



Conclusions

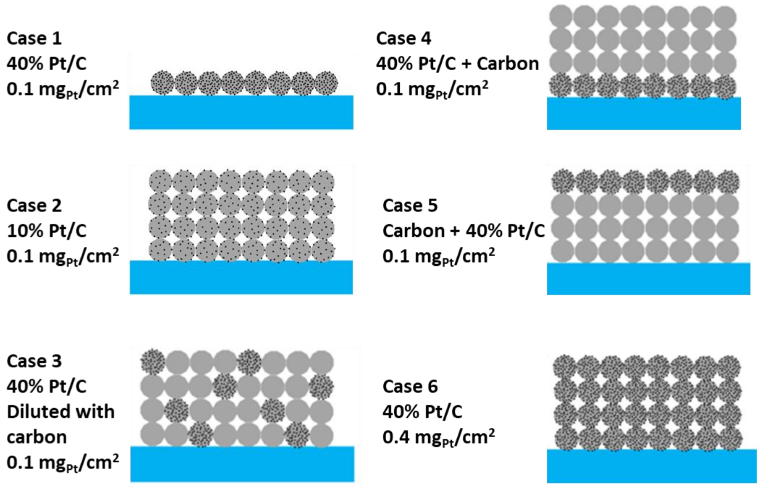
- ~ 45% of Co^{2+} exchange of the ionomer can be tolerated in the electrode
- Loss in performance is predominantly induced by mass transport losses and proton transport losses,
- Co^{2+} preferentially resides in the electrode under wet operating conditions

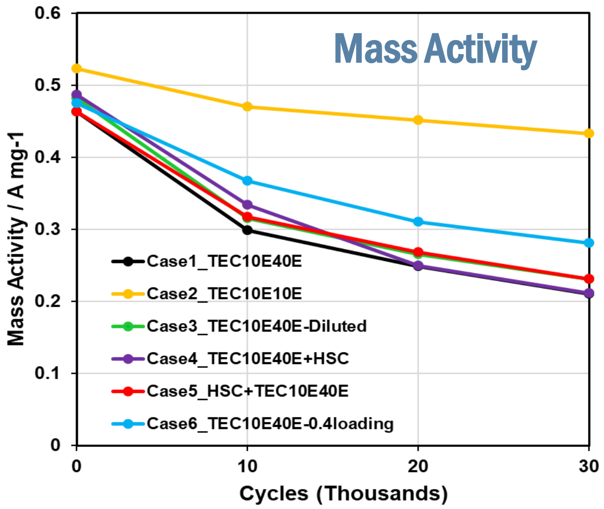
This defines the catalyst alloying element loss allowed in catalyst development projects - not related to changing kinetics

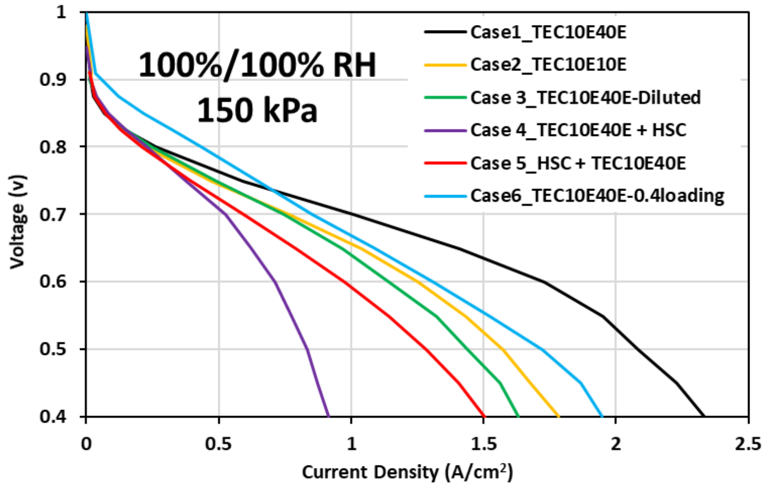
Effect of Electrode Thickness and Pt Distribution (HSC)

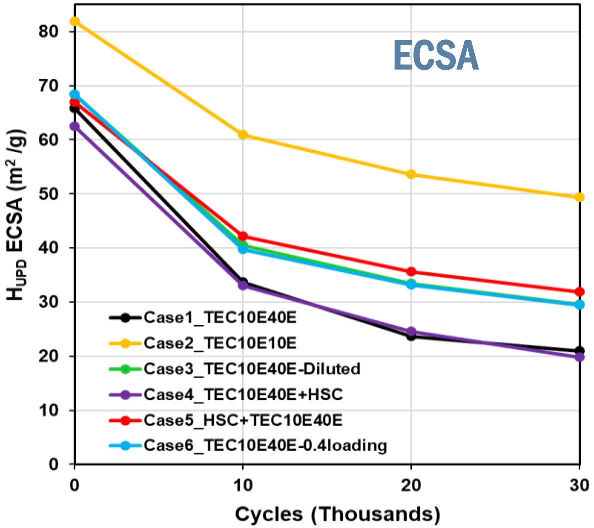
- Model electrode case studies guide our design of HDV MEAs with increased efficiency and durability, including studies on effect of electrode thickness, local Pt distribution, ionomer, and carbon support properties
- For Pt/HSC
  - Increasing electrode thickness dramatically decreased cell performance
  - Local concentration of Pt affects durability
  - Selection of Pt wt% is a balance between durability (lower wt% better) and transport (higher wt% better)

Schematic illustration for 6 case studies







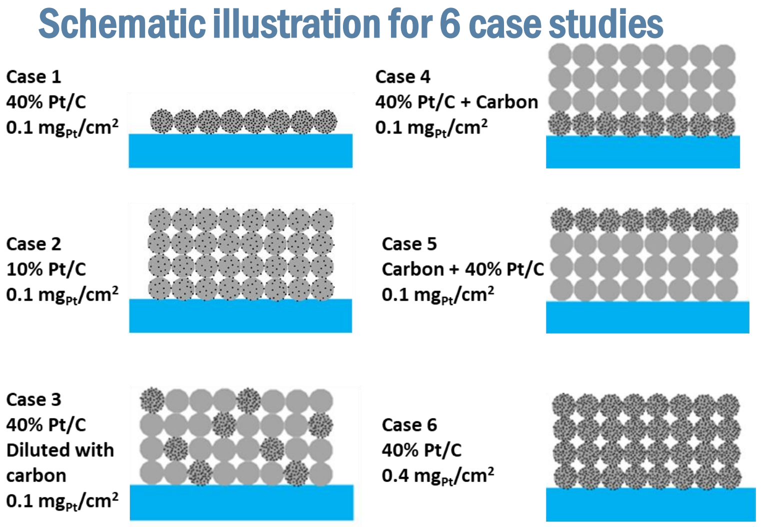


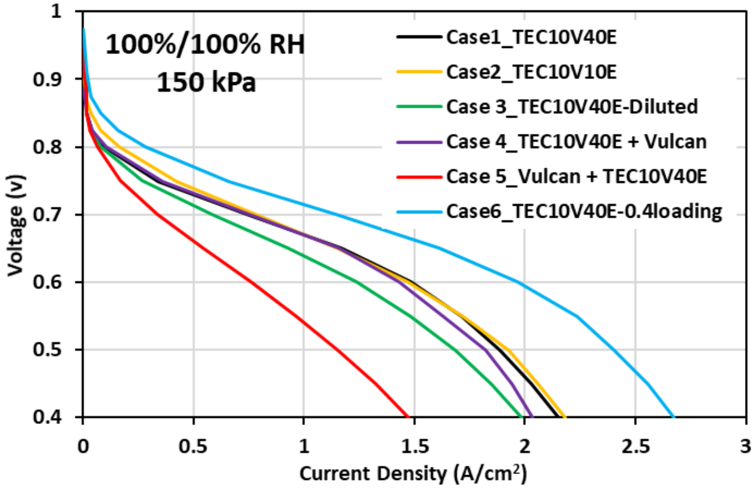
Cell performance is limited by higher mass transport resistance in thicker electrodes

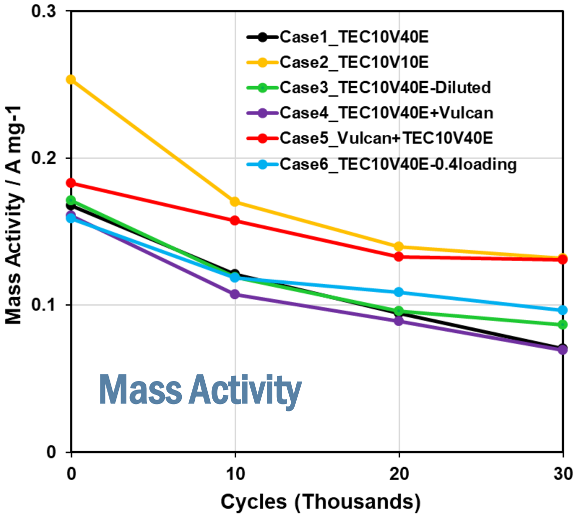
Pt/HSC with lower Pt wt% shows better durability

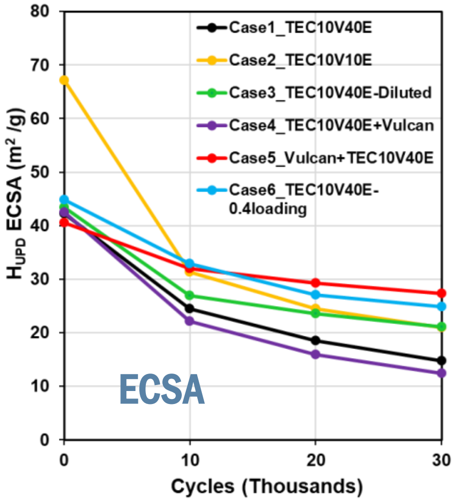
Effect of Electrode Thickness and Pt Distribution (Vulcan)

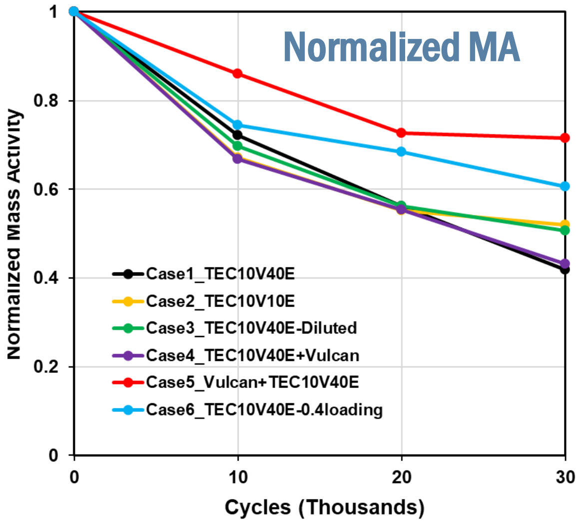
- For Pt/Vulcan
  - Durability of Pt/Vulcan shows no obvious trend with Pt local concentration
  - Selection of Pt wt% is more flexible, since it is less sensitive to flooding
- Improved understanding of how catalyst and electrode properties control electrode behavior enables design of improved HDV MEAs

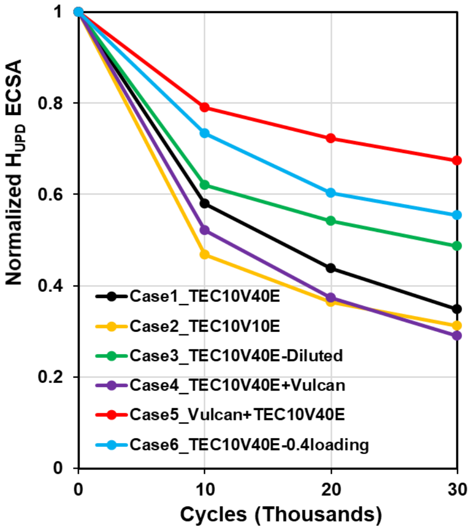






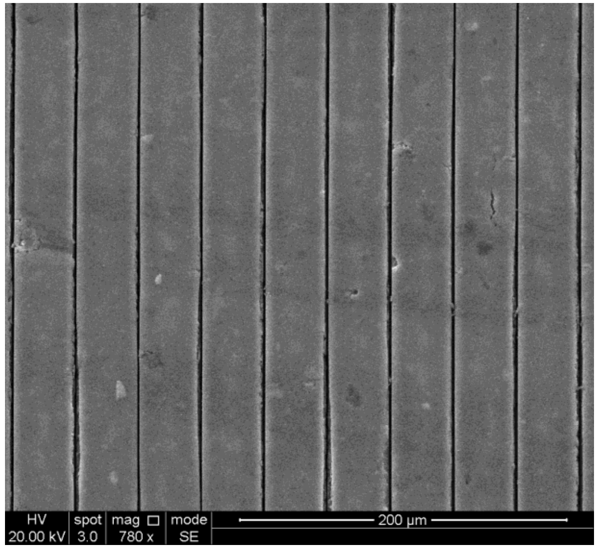


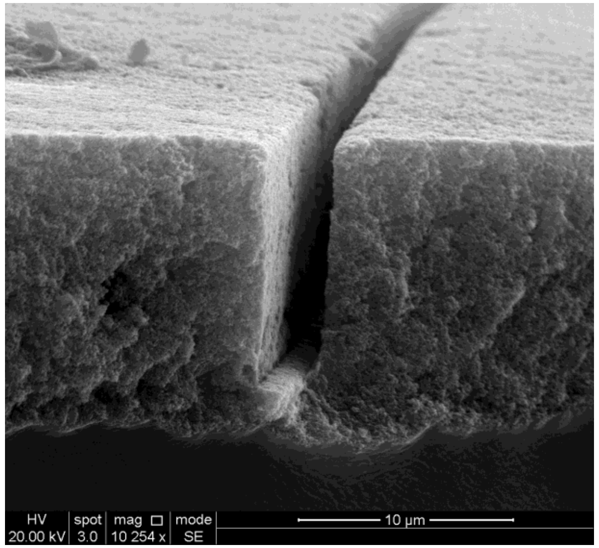


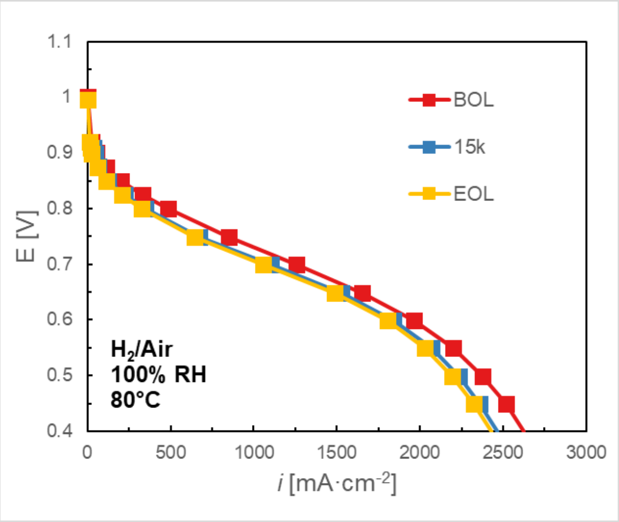


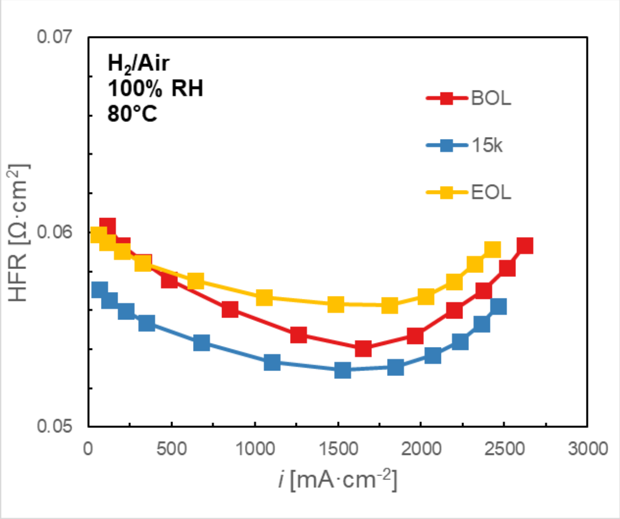
Catalyst located far away from membrane shows better durability

Effect of Cracks/Grooves on Electrode Durability

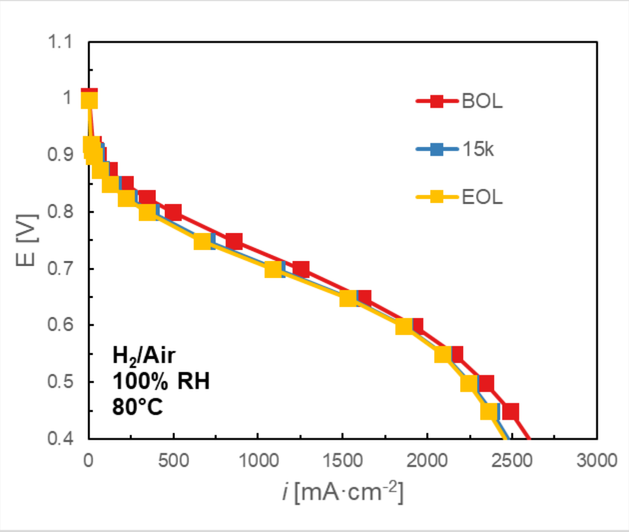


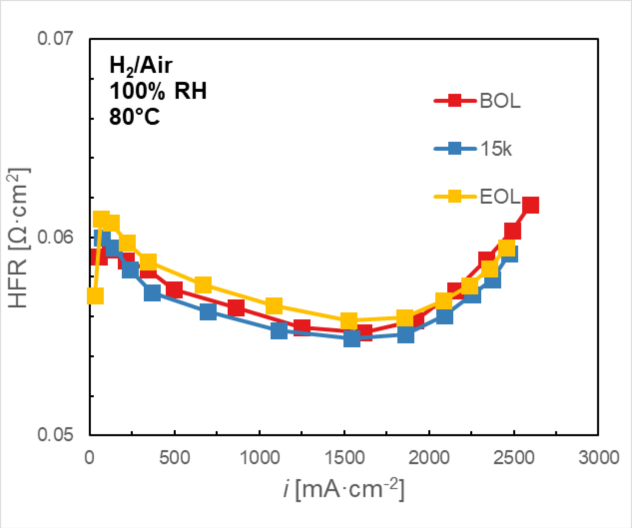






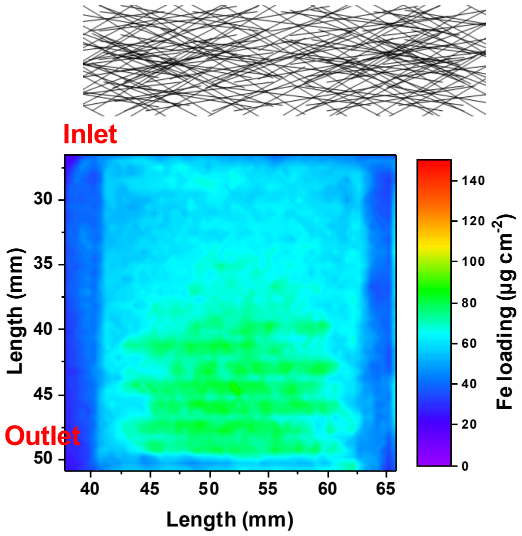
- Cracks have negligible effect on catalyst durability
- ECSA, mass activity, and transport losses are similar with and without cracks
- Dense grooves improve performance of un optimized electrodes
  - Thick electrodes
  - High I/C ratio electrode
  - Electrodes subjected to carbon corrosion



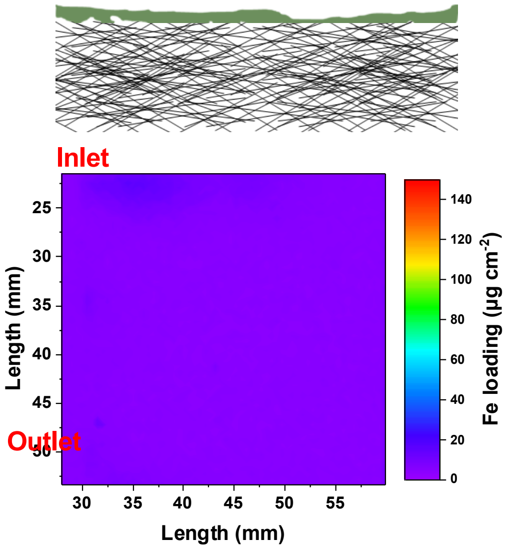


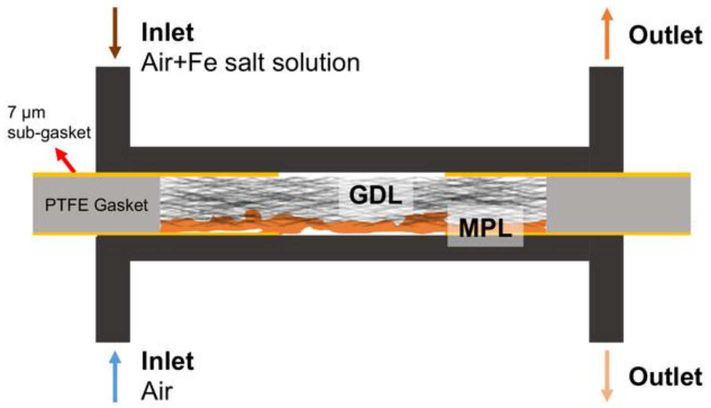
Fe Transport Rates Across GDL/MPL

29 BA

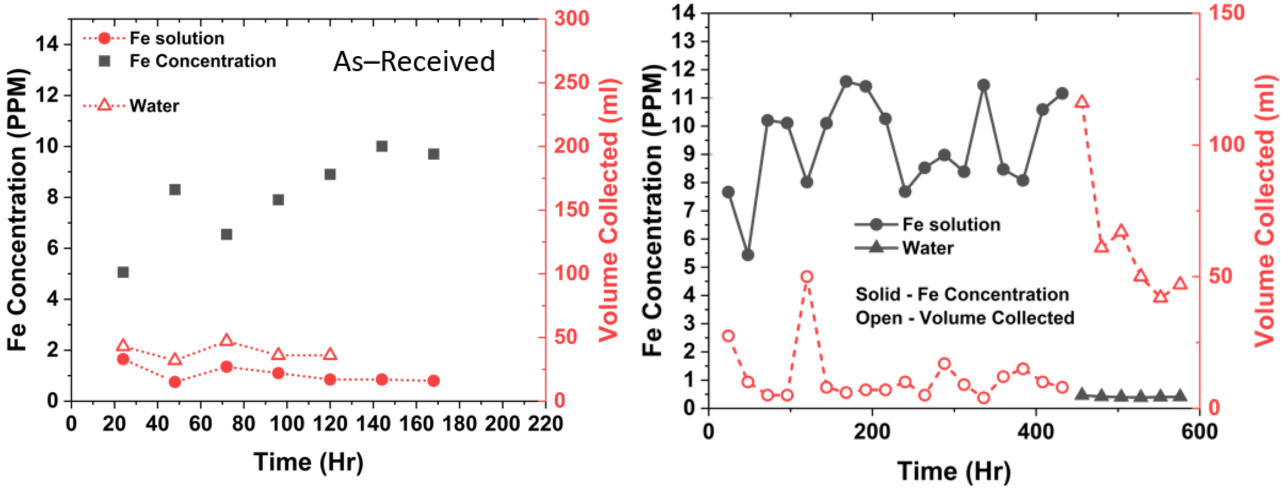


MCT





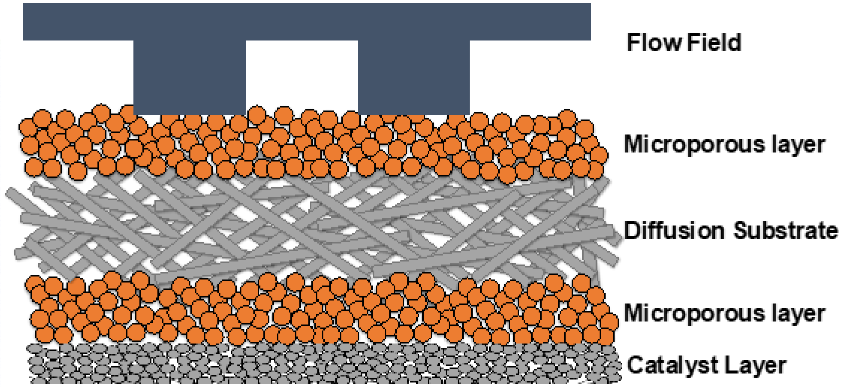
29 BC



- H23C8 experiment where no water was transported through
- No crack in the MPL has a higher water break through pressure

Cation Suppression Layer

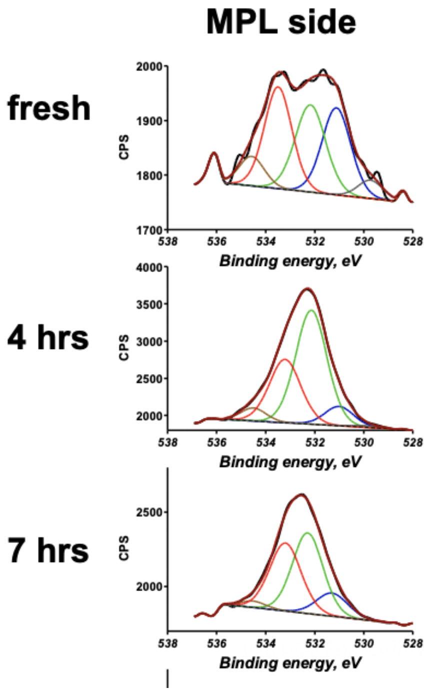
Dual-Layer Cation Transport layer

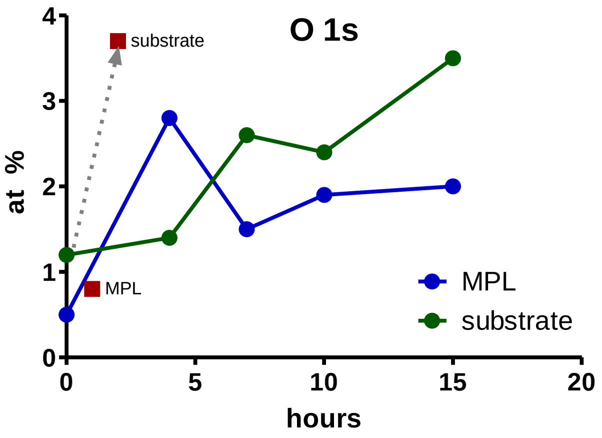


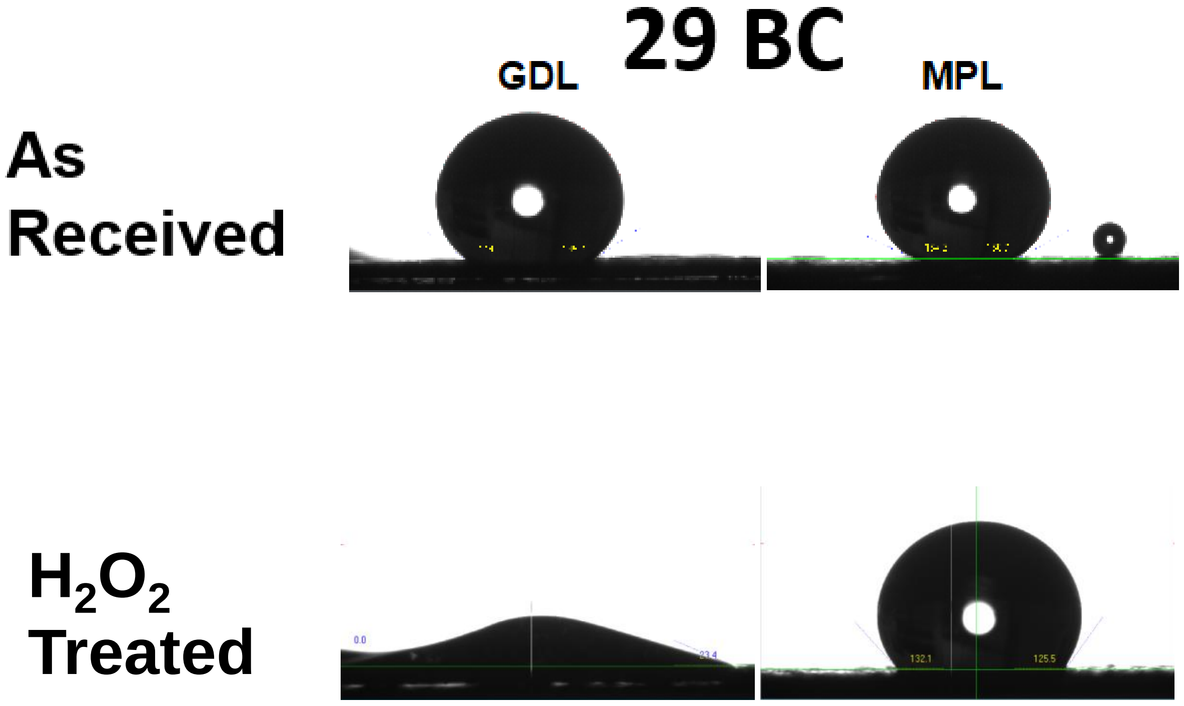
Development of GDL AST

GDLs

1) **Chemical Ageing – peroxide:** 30% H_2O_2
2) **Chemical Ageing – peroxide:** 3% H_2O_2
   Sometimes refluxed at 80C
3) **Submersion in H_2O** (with N_2, H_2 or Air)
4) **Potential Cycling (1 to 1.5), 80 °C, 200%RH**







- Aged GDLs exhibit higher oxygen content
- Aged GDLs are more hydrophilic
- Need to match ex-situ ageing with in-situ ageing studies

Collaborations: Non-FOA activities

Entity	Scope of collaboration
AvCarb	R2R gas diffusion electrode fabrication
3M	Ionomer materials and discussions for ionomer studies
N.E. Chemcat	Development of Pt core-shell catalysts
Toyota North America	Development of catalysts for light-duty vehicles
IUPUI	Development of PBI-modified carbon
Umicore	Provide tailored MEAs
U Delaware	Membrane durability with radical stabilization
Robert Bosch	Voltage loss analysis and modeling discussions
Celadyne	Evaluation of advanced membrane
CEA - Grenoble	X-ray and neutron scattering experiments

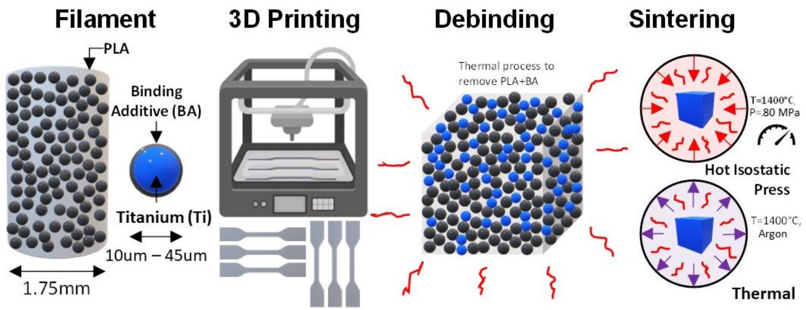
Entity	Scope of collaboration
SUNY Buffalo	Catalyst carbon supports
Advent	Membrane development
U. South Carolina	Catalyst development
U. Louisville	Electrode structures
Georgia Tech	Lattice Boltzmann modeling
Virginia Tech	Membrane characterization
RPI	Membrane development
Texas A&M University	Sulfonated ionic liquid block co-polymer
Toyota Research Institute	Machine learning for membrane design
Chemours	Membrane durability

Inclusion, Diversity, Equity, Accountability

- **Outreach and Workforce Development**
  - Our labs hosted UGS, GRA interns and DOE SCGSR Fellows visiting national labs to gain hand-on experience working with fuel cell systems and materials and learn about hydrogen technologies
  - Two new MSI students with M2FCT starting summer 2022
  - Internship programs (K12 and SULI) for summer 2022
- **M2FCT added Discretionary projects which includes three MSIs:**
  - Univ Cal Irvine (UCI) – AANAPISI and HSI
  - Univ Cal Merced (UCM) – HSI
  - Florida International University (FIU) – HSI

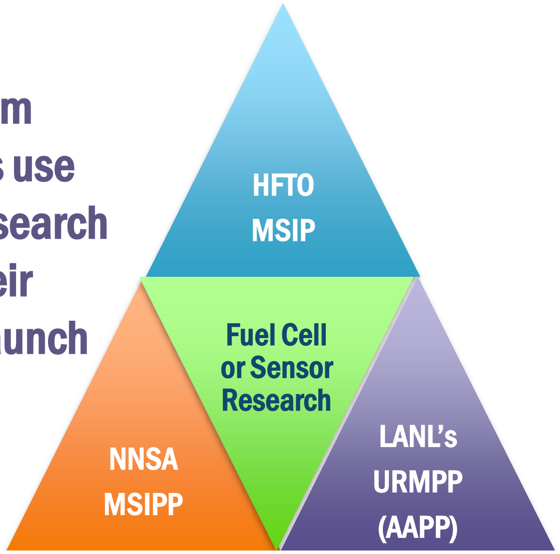
Dr. David Alexander
Ph.D Thesis: April 2022
HBCU - Undergrad

Bound Metal Deposition
for fuel cell bipolar plates



M2FCT working with Minority Serving Institution Partnership Program

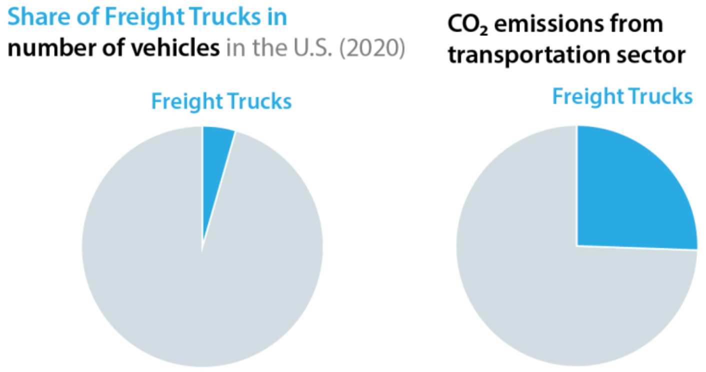
Students from HBCU/HSI's use Fuel Cell Research to obtain their PhDs and Launch Careers

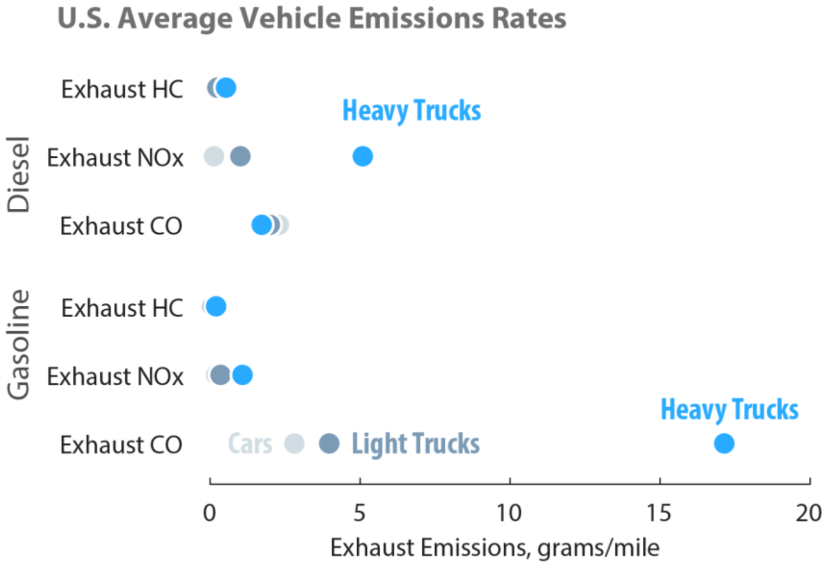


Energy Justice and Equity

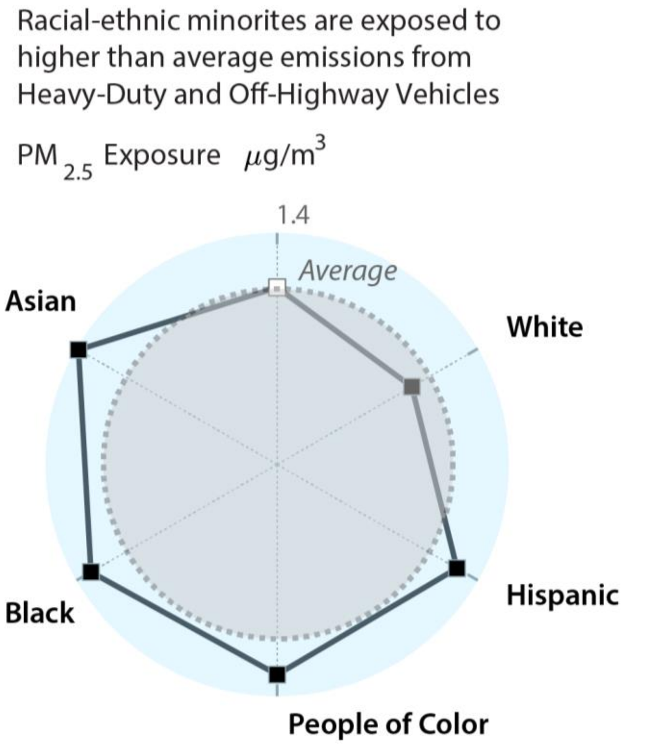
Decarbonizing freight transportation and long-haul trucking

- **Freight transportation is one of the major contributors to the emissions**
- In particular, significantly higher NOx and CO emissions from trucks (vs. LDVs, cars) are a major environmental and health concern
- Underserved communities (e.g., near highways, ports and freight centers) are more vulnerable to exposure and experience adverse health effects





- Disadvantaged neighborhoods will be favorably impacted by decarbonization of freight transport and improvements to long-haul trucking corridors



Source: EIA, BTS, EPA; Tessum et al. *Science Advances*, 2021 Liu et al., *Environ. Sci. Technol.* 2015, 49, 19, 11569–11576

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
  - Targets are end-of-life performance
  - Greater efficiency (68-72%)
  - High durability (1,000,000 miles; 25,000-30,000 hrs)
  - Material down-selects ~ year 3
    - Catalyst areas Go/No-Go at Q6
- **Analysis**
  - Refine models, characterization, and diagnostics for heavy-duty operating conditions
  - Define operating conditions efficiency and durability trade-offs
  - Coordinate and harmonize truck platforms and duty cycles
  - Compare systems with different ratios of fuel cell power and battery energy storage
  - Sensitivity of performance, durability and cost to cell voltage target at EOL
  - Incorporate membrane durability in system analysis
- **Machine learning / Data analysis**
  - Correlations of metadata for material and integration studies
- **High-Performance Computing**
  - Unsteady FCS simulations on truck drive cycles
  - Electrode and agglomerate structure
  - Upscaling physics-based micro- and nano-scale models to cell models and optimization
- **Durability**
  - Develop refined ASTs for life-time prediction with heavy-duty materials and operating conditions
    - Extended catalyst AST from 30K cycles to 90K cycles
    - Developing new H_2/Air MEA AST protocols specific for HDV
  - Propose new MEA AST protocol in collaboration with ASTWG by end of FY22
  - Electrode stability – Co dissolution rates from advanced catalysts
  - Membrane and ionomer durability with additives
  - Prediction of membrane lifetime modeling FER data
  - Modeling of durability adjusted cost using real world drive cycles and degradation models
  - Understand long-term durability effects on other components (GDL, and bipolar plate AST development)

Future Work

- **Integration**
  - Baseline SOA
    - Establish benchmark performance and cost of state-of-art MEA
  - Incorporate advance catalyst ink understanding into R2R manufacturing
  - Integrate newly developed materials into optimized MEA structures
    - Membranes
    - Ionomers
    - Catalysts
    - Catalyst supports
    - GDLs
  - Catalyst layer studies
    - Understand cation migration effects on catalyst layer performance
    - Catalyst layer porosity
    - Catalyst ink to structure formation models
  - Transport Properties (Gas phase, water, cations)
    - Catalyst Layer
    - GDL

- **Material Development**
  - Assess and develop scalable PtCo intermetallic catalyst synthesis methods; study effects of Pt shell thickness, particle size, degree of ordering, core composition, and carbon support
  - Optimize synthesis and post-synthesis treatment of PtNiN catalysts, increase N content of metal particles and supports through high-pressure nitriding, synthesize and evaluate high entropy alloys
  - Evaluate durability of PtCo intermetallics and Pt/C modified with melamine and melamine-based polymers
  - Evaluate durability of Pt/C-MO_x in MEAs
  - Explore grafting, cross-linking, block co-polymer and cathode catalyst layer fabrication strategies to address water solubility of high IEC ionomers
  - Evaluate perfluorinated phosphonic acid radical scavenger retention additives in MEA cathode and membrane

Summary - Technical Accomplishments

- Analysis of operating conditions, performance & efficiency
  - Evaluated Key barrier to meeting the interim targets
    - Active and stable catalysts capable of meeting the targets
  - Compared systems:
    - Small Stack (175 kW_e System), Large Battery Hybrid System
      - Stack coolant exit temperature: 85ºC
    - Large Stack (425 kW_e System), Small Battery Hybrid System
      - Radiator frontal area: 50% larger than in diesel truck (Stack coolant exit temperature: 95ºC)
  - Stack power/size and idle power

- Durability
  - ASTWG and iDWG meeting regularly to advance heavy duty AST development
  - Quantified effect of Co cations on CL performance
  - Catalyst layer thickness effect, support interactions and location of catalyst on durability elucidated
  - Evaluated the role of catalyst layer cracks on catalyst durability
  - Improvements to membrane durability
    - Stabilized radical Scavengers; Preventing Fe migration with Crack free MPL.
  - H_2/Air MEA AST under development for 25,000+ lifetime. Test at 90 ºC for accelerating degradation

Summary - Technical Accomplishments

- Integration and Science of Manufacturing
  - Established baseline performance for 90k AST cycles for 3 catalysts (informed systems analysis and go/no-go down selection)
  - Elucidated evolving electrocatalyst structure as a function of durability to identify design needs
  - Identified promising ionomer materials to reduce EOT transport losses without sacrificing kinetic performance
  - Identified potential electrode structures to enable optimum transport characteristics
  - Mitigated crack formation without forsaking performance
  - Showcased anisotropy in membrane coatings below 10 mm and established trends for membrane properties as a function of thickness
  - Demonstrated reduced electrode agglomeration with slower ink drying time
  - Established two viable approaches for multilayer electrode coatings to facilitate improved performance/durability
- Material Developments
  - Two PtNiN BNL catalysts and three LANL PtCo ordered intermetallic catalysts have exceeded the Q6 Go/No-Go criteria for performance and durability in an MEA
  - Pt/C modified with co-polymer of melamine and formaldehyde showed reduced SO3- poisoning, improved ORR mass activity in MEA
  - Synthesis method for oxide nano-particle additive to carbon supports was developed; improved Pt surface area retention demonstrated in aqueous electrolyte tests for Zr, Ta, and W oxide particle-modified Pt/C
  - Multiple batches of novel high IEC perfluoro polymers were synthesized, showing higher proton conductivity at all RHs versus Nafion control
  - Crown ether-functionalized oligomers and perfluorinated phosphonic acid additives synthesized; demonstrated to complex/retain Ce radical scavenger, and to mitigate PFSA degradation

Who is M2FCT? National Lab Contributors

Argonne National Laboratory

Rajesh Ahluwalia
Firat Cetinbas
Nancy Kariuki
John Kopasz
Debbie Myers
Jaehyung Park
Voja Stamenkovic (UCI)
Xiaohua Wang
Andrew Star

NIST

Dan Hussey
David Jacobson
Jake LaManna

Berkeley Lab

Grace Anderson
Claire Arthurs
Vince Battaglia
Sarah Berlinger
Ashley Bird
Hailey Boyer
Anamika Chowdhury
Arthur Dizon
Kenny Higa
Samay Garg
Pryamvada Goyal
Hans Johansen
Doug Kushner
Ahmet Kusoglu
Lalit Pant
John Petrovick
Clay Radke
Mayank Sabharwal
Harsh Srivastav
Adam Weber

Los Alamos National Laboratory

Tanya Agarwal
Rod Borup
Alex Gupta
Yu-Seung Kim
Siddharth Komini-Babu
Chung Hyuk Lee
Rangachary Mukundan
Sarah Park
Fahim Rahman
Jacob Spendelow
Chenyu Wang
Xiaojing Wang

NREL – National Renewable Energy Laboratory

Carlos Baez-cotto
Leiming Hu
Sanghun Lee
Scott Mauger
KC Neyerlin
Zbyslaw Owczarczyk
Bryan Pivovar
Peter Rupnowski
Colby Smith
Audrey Taylor
Mike Ulsh
Tim Van Cleve
Gokul Venugopalan
Erica Young
James Young

Oak Ridge National Laboratory

David Cullen
Xiang Lyu
Shawn Reeves
Alexey Serov
Haoran Yu
Michael Zachman

Pacific Northwest National Laboratory

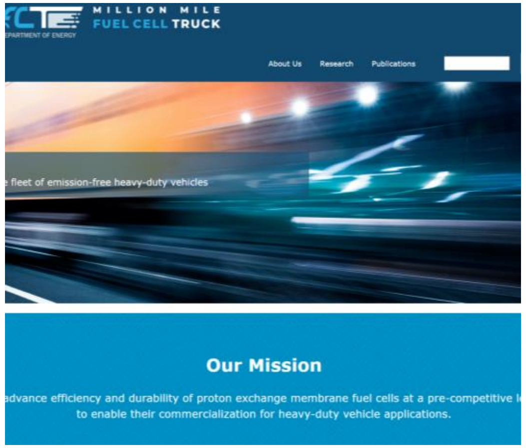
Yang Qiu
Yuyan Shao

Brookhaven National Laboratory

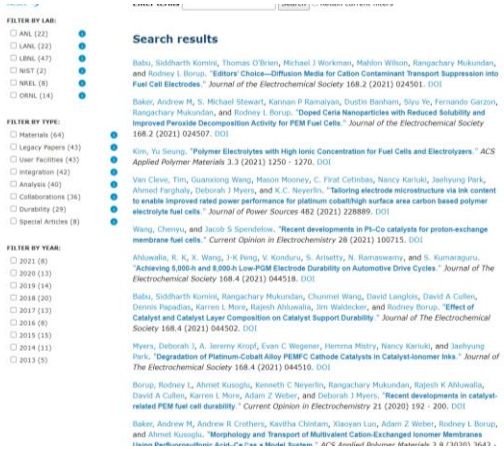
Kotaro Sasaki

Visit our website: millionmilefuelcelltruck.org

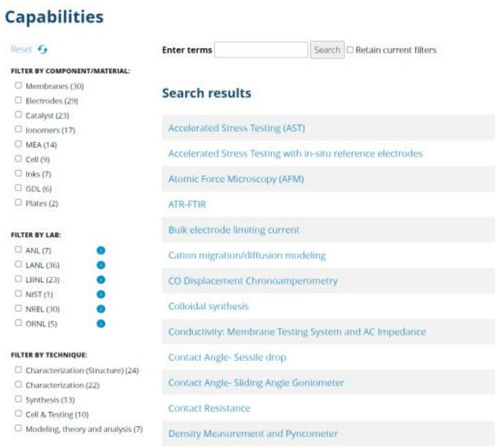
Main Page



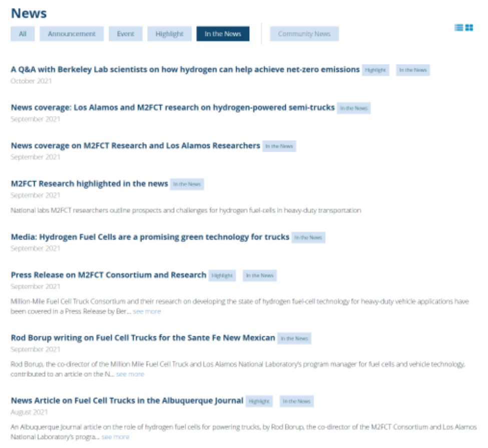
Publications



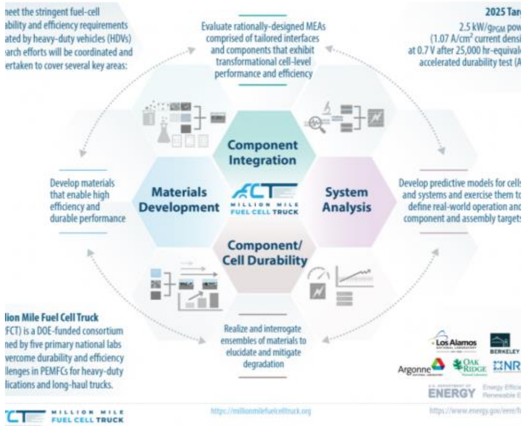
Capabilities



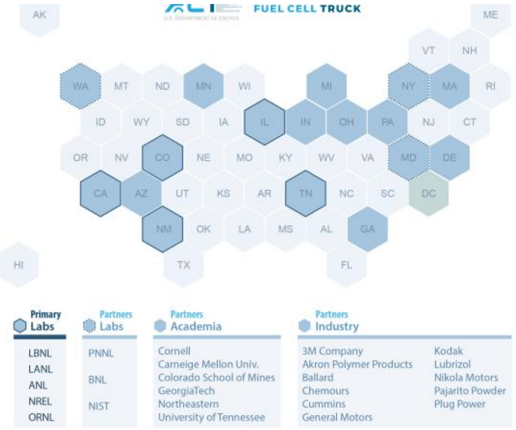
News



Research

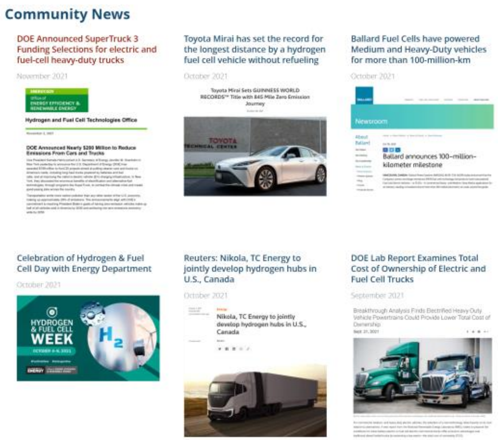


People and Partners



Outreach: Working Groups

Community News



Acknowledgements

DOE EERE Hydrogen and Fuel Cell Technologies Office

Technology Managers:

Greg Kleen, Dimitrios Papageorgopoulos

MILLION MILE FUEL CELL TRUCK

U.S. DEPARTMENT OF ENERGY

http://millionmilefuelcelltruck.org

Los Alamos National Laboratory

Berkeley Lab

NREL National Renewable Energy Laboratory

Argonne National Laboratory

Oak Ridge National Laboratory

Brookhaven National Laboratory

Pacific Northwest National Laboratory

NIST

User Facilities

DOE Office of Science: SLAC, LBNL-Advanced Light Source, LBNL-Molecular Foundry, ANL-Advanced Photon Source, LBNL-Molecular Foundry, ORNL-Center for Nanophase Materials Sciences, ANL-Center for Nanostructured Materials, NIST: BT-2

Back-up Slides

Publications, Awards

Publications

- Kim, Y.S.."Polymer Electrolytes with High Ionic Concentration for Fuel Cells and Electrolyzers." *ACS Applied Polymer Materials* 3.3 (**2021**) 1250 - 1270.
- Berlinger, SA., Garg, S, Weber, AZ.."Multicomponent, multiphase interactions in fuel-cell inks." *Current Opinion in Electrochem.* 29 (**2021**) 100744
- Petrovick, J.G., Anderson, G.C., Kushner, D.I., Danilovic, N., Weber, A.Z.."Method—Using Microelectrodes to Explore Solid Polymer Electrolytes." *J Electrochemical Society* 168.5 (**2021**) 056517.
- Berlinger, S.A., McCloskey, B.D., Weber, AZ.."Probing Ionomer Interactions with Electrocatalyst Particles in Solution" *ACS Energy Letters* 6 (**2021**) 2275 - 2282. DOI
- Garg, S., Fornaciari, J., Weber, A., Danilovic, N.."fuelcell: A Python package and graphical user interface for electrochemical data analysis." *J of Open Source Software* 6.59 (2021) 2940. DOI
- Gittleman, Craig S., Jia, Hongfei, De Castro, Emory S., Chisholm, Calum R.I., and Kim, Yu Seung."Proton conductors for heavy-duty vehicle fuel cells." *Joule* (**2021**). DOI
- Lei, C, Yang, F, Macauley, N, Spinetta, M, Purdy, G, Jankovic, J, Cullen, D A., More, KL., Kim, Y.S, Xu, H."Impact of Catalyst Ink Dispersing Solvent on PEM Fuel Cell Performance and Durability." *J Electrochemical Society* 168.4 (**2021**) 044517. DOI
- Pant, L.M., Stewart, S, Craig, N, Weber, A Z.."Critical Parameter Identification of Fuel-Cell Models Using Sensitivity Analysis." *Journal of the Electrochem. Soc.* 168.7 (**2021**) 074501. DOI
- Chowdhury, A., Bird, A., Liu, J., Zenyuk, I.V., Kusoglu, A., Radke, C.J., Weber, A."Linking Perfluorosulfonic Acid Ionomer Chemistry and High-Current Density Performance in Fuel-Cell Electrode." *ACS App. Mat. & Interfaces* 13.36 (**2021**) 42579 -42589
- Luo, X., Lau, G., Tesfaye, M., Arthurs, C.R., Cordova, I., Wang,C., Yandrasits, M.I, Kusoglu, A. "Thickness Dependence of PEM Properties," *J Electrochem. Soc.* 168 (**2021**) 104517
- Yu H., Zachman M.J., Li C., Hu L., Kariuki N.N., Mukundan R., Xie J., Neyerlin K.C., Myers D.J., Cullen D.A. "Recreating Fuel Cell Catalyst Degradation in Aqueous Environments for Identical-Location Scanning Transmission Electron Microscopy Studies," (**2022**)
- Petrovick J.G., Radke C.J., Weber A.Z. "Gas Mass-Transport Coefficients in Ionomer Membranes Using a Microelectrode" (**2022**)
- Chen M., Li C., Zhang B., Zeng Y., Karakalos S., Hwang S., Xie J., Wu G. ."High-Platinum-Content Catalysts on Atomically Dispersed and Nitrogen Coordinated Single Manganese Site Carbons for Heavy-Duty Fuel Cells." *Current Opinion in Electrochemistry* 169.3 (**2022**)

Awards

- Chung Hyuk Lee was awarded the Natural Sciences and Engineering Research Council of Canada Postdoctoral Fellowship
- Sidd Rajupet, for winning the DoD National Defense Science and Engineering Graduate (NDSEG) fellowship
- Rajesh Ahluwalia – University of Chicago Board of Governors Distinguished Performance Award
- Rajesh and Xiaohua (ANL) received the 2021 AMR Award
- Siddharth Komini-Babu – ECS Toyota Young Investigator award

Publications

Publications

- Kim, Y.S.."Polymer Electrolytes with High Ionic Concentration for Fuel Cells and Electrolyzers." *ACS Applied Polymer Materials* 3.3 (2021) 1250 - 1270. DOI
- Berlinger, Sarah A., Garg, Samay, and Weber, Adam Z.."Multicomponent, multiphase interactions in fuel-cell inks." *Current Opinion in Electrochem.* 29 (2021) 100744. DOI
- Petrovick, J.G., Anderson, G.C., Kushner, D.I., Danilovic, N., Weber, A.Z.."Method—Using Microelectrodes to Explore Solid Polymer Electrolytes." *Journal of The Electrochemical Society* 168.5 (2021) 056517. DOI
- Berlinger, S.A., McCloskey, B.D., Weber, AZ.."Probing Ionomer Interactions with Electrocatalyst Particles in Solution" *ACS Energy Letters* 6 (2021) 2275 - 2282. DOI
- Garg, S., Fornaciari, J., Weber, A., Danilovic, N.."fuelcell: A Python package and graphical user interface for electrochemical data analysis." *J of Open Source Software* 6.59 (2021) 2940. DOI
- Gittleman, Craig S., Jia, Hongfei, De Castro, Emory S., Chisholm, Calum R.I., and Kim, Yu Seung."Proton conductors for heavy-duty vehicle fuel cells." *Joule* (2021). DOI
- Lei, C, Yang, F, Macauley, N, Spinetta, M, Purdy, G, Jankovic, J, Cullen, D A., More, KL., Kim, Y.S, Xu, H."Impact of Catalyst Ink Dispersing Solvent on PEM Fuel Cell Performance and Durability." *J Electrochemical Society* 168.4 (2021) 044517. DOI
- Pant, L.M., Stewart, S, Craig, N, Weber, A Z.."Critical Parameter Identification of Fuel-Cell Models Using Sensitivity Analysis.“ *Journal of the Electrochem. Soc.* 168.7 (2021) 074501. DOI
- Chowdhury, A., Bird, A., Liu, J., Zenyuk, I.V., Kusoglu, A., Radke, C.J., Weber, A."Linking Perfluorosulfonic Acid Ionomer Chemistry and High-Current Density Performance in Fuel-Cell Electrode." *ACS App. Materials & Interfaces* 13.36 (2021) 42579 -42589. DOI
- Luo, X., Lau, G., Tesfaye, Meron, Arthurs, Claire R., Cordova, Isvar, Wang, Cheng, Yandrasits, Michael, and Kusoglu, Ahmet."Thickness Dependence of Proton-Exchange-Membrane Properties." *Journal of The Electrochemical Society* 168.10 (2021) 104517. DOI
- Yu H., Zachman M.J., Li C., Hu L., Kariuki N.N., Mukundan R., Xie J., Neyerlin K.C., Myers D.J., Cullen D.A.."Recreating Fuel Cell Catalyst Degradation in Aqueous Environments for Identical-Location Scanning Transmission Electron Microscopy Studies." (2022). DOI
- Petrovick J.G., Radke C.J., Weber A.Z.."Gas Mass-Transport Coefficients in Ionomer Membranes Using a Microelectrode." (2022). DOI
- Chen M., Li C., Zhang B., Zeng Y., Karakalos S., Hwang S., Xie J., Wu G. ."High-Platinum-Content Catalysts on Atomically Dispersed and Nitrogen Coordinated Single Manganese Site Carbons for Heavy-Duty Fuel Cells.“ *Current Opinion in Electrochemistry* 169.3 (2022). DOI

2021 Reviewer Comments

Project strengths:

- Really, everything is a strength. The team is great. The institutional structure is very good. The project is tackling the correct problems.
- The team and collaborations, baseline testing and standardizations, and incorporation of academia and industry through FOAs are all strengths.
- The project has a very strong team, excellent analytical capabilities, and the latest approaches, e.g., machine learning. There is excellent collaboration.
- The "team-of-teams" approach grouping well-recognized experts is a real strength of this project.
- The goal of the project, understanding how to increase the durability of fuel cell MEAs, is important.

↳ Reviewer Strengths greatly out-weighted weaknesses in the 2021 review

2021 Reviewer Comments

Project weaknesses:

- Even if considering all stack components in the scope of the project, it appears to be very focused (perhaps too much so) on MEA developments for HDVs. Connections with ongoing DOE-funded projects on MEAs, membranes, and stacks seem to be put in place, and it will be important to ensure regular effective exchanges.
  - ↳ M2FCT had it's first in-person meeting March 2,3 2022. This was delayed because of COVID. Two in-person meetings per year are intended. Direct communication with projects will be designed as each project needs to meet those project needs. A coordination officer exists specifically to coordinate exchanges.
- The project may be too large and all-encompassing for effective management.
  - ↳ Management was designed with co-directors, deputy directors to address the size of the consortium
- The project is not tailored to guide controls designs, and there are few new ideas in the project/presentation.
  - ↳ This consortium is working to develop material and integration solutions; past guidance has been that controls designs are topics that industry feels are proprietary and DOE does not typically fund.

2021 Reviewer Comments

Recommendations for additions/deletions to project scope:

- The DOE researchers need to do a comprehensive literature review of much of the proposed work and determine whether the proposed experimental work needs to be completed, or whether controls parameters can be recommended on existing publications and patents. The team must include one or two people from outside the DOE fuel cell community .... The researchers should not be in administrative roles.....
  - This is primarily a comment for DOE. DOE has taken the approach of developing materials solutions. M2FCT has an External Advisory Board which is mostly people from industrial positions. The M2FCT consortium was formed addressing the requirements in the solicitation, and all staff scientists are engaged in R&D.
- The project is so large and includes so many different activities that it feels like a shotgun of work. It may be better to separate out some of the work for separate reviews, rather than review all the work together. There could still be a high-level review of the consortium/approach in one normal-length review session. The team might consider studies to support freeze-start operation, such as material and MEA properties under sub-zero conditions.
  - In future years we intend to have separate posters on a number of the PI led topics.
  - Freeze-start operation can be considered; however, to-date this has been de-emphasized due to comments from industrial input.
- The scope of the project is quite large and ambitious. The challenge will be to cover all the foreseen items and to ensure effective coordination.
  - The organizational structure is set to be successful; the targets are ambitious as are many of the Hydrogen Shot targets.

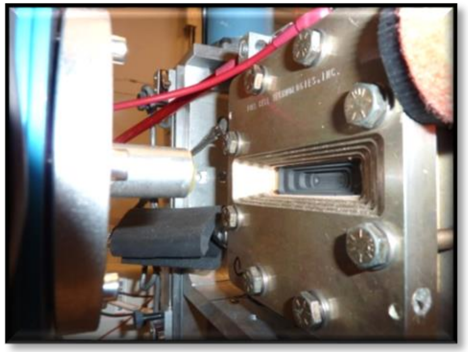
LANL and BNL Hydrogen-Air MEA Performance and Durability Summary

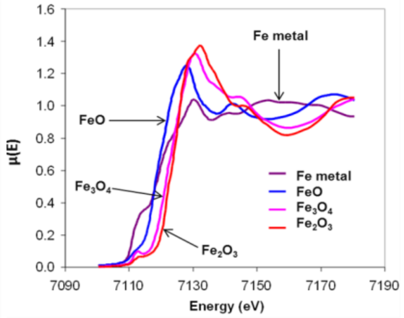
Catalyst (Particle size)	Number of Cycles	ECSA (m^2/g_{Pt})	MA (mA/mg_{Pt})	*j* at 0.7V (A/cm^2)	*j* at 0.8V (A/cm^2)
BNL 35%-DA_IntPtNiN/HSC BNL-C (4.5 nm)	0K	53	550	1.22	0.50
	90K	22	210	0.96	0.25
BNL 40%-IntPtNiN/HSC BNL-D (4.8 nm)	0K	51	560	1.19	0.45
	90K	25	246	0.95	0.27
a-Pt/C (4~5 nm)	0K	39	418	1.17	0.32
	90K	20	178	0.85	0.14
Pt/C (2~3 nm)	0K	65	651	0.71	0.18
	90K	20	73	0.13	0.03
d-PtCo/C (4~5 nm)	0K	34	824	1.18	0.50
	90K	16	152	0.75	0.16
LANL 35%-$L1_2$-PtCo/HSC (5.7 nm)	0K	36	575	1.29	0.41
	90K	13	120	0.78	0.17
LANL 20%-$L1_0$-PtCo/HSC (6.3 nm)	0K	49	719	1.09	0.35
	90K	26	362	0.90	0.27
UB/LANL 20%-$L1_2$-PtCo/MnCN	0K	67	350	1.07	0.36
	90K	22	265	0.88	0.25

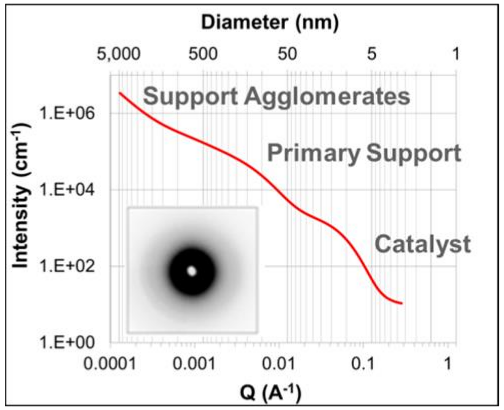
Benchmark
Umicore Pt50 0550

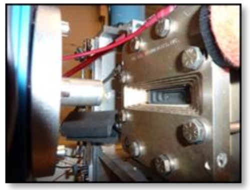
X-ray Characterization at the Advanced Photon Source

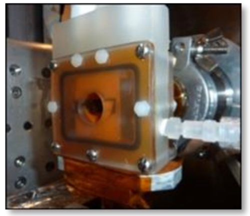
- **X-ray absorption (10-BM, 10-ID, 12-BM):**
  - oxidation state, electronic structure (density of states), local coordination of absorbing atom, identity of neighboring atoms, bond distances
  - Can be performed on powders, precursors, catalyst-ionomer inks, and electrodes in aqueous or fuel cell environments
- **X-ray scattering (9-ID, 12-BM):**
  - particle/aggregate shape, size, size distribution
  - Can be performed on powders, precursors, catalyst-ionomer inks, and electrodes in aqueous or fuel cell environments
- **X-ray imaging and tomography (32-ID):**
  - Nano- and micro-structure of materials and materials and pores in cell layers
  - Primarily ex situ, but can be performed under controlled T, RH, atmosphere, and potential

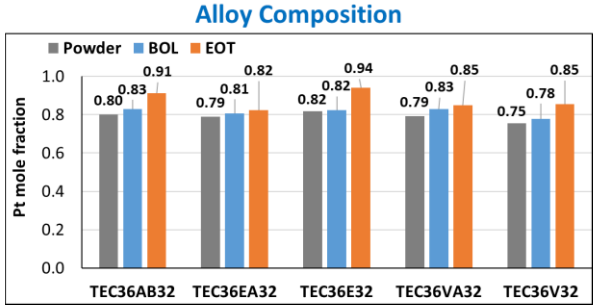


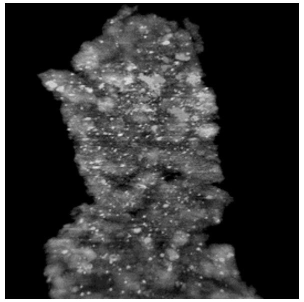


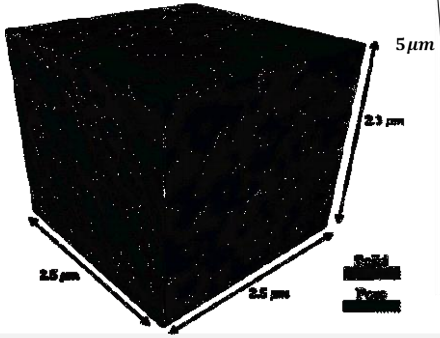


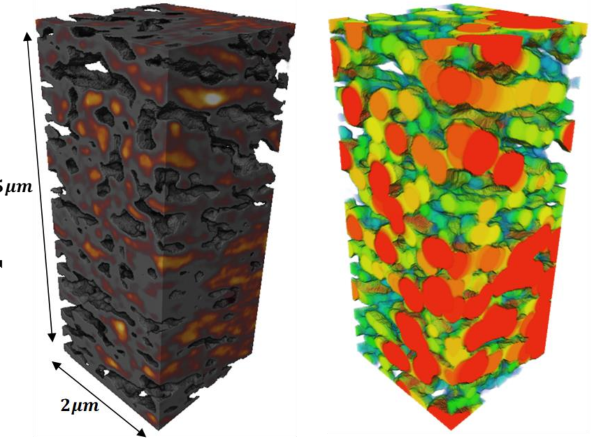












Spatial distribution of agglomerates

Baselining MEA Performance and Durability in Differential and Integral Cells

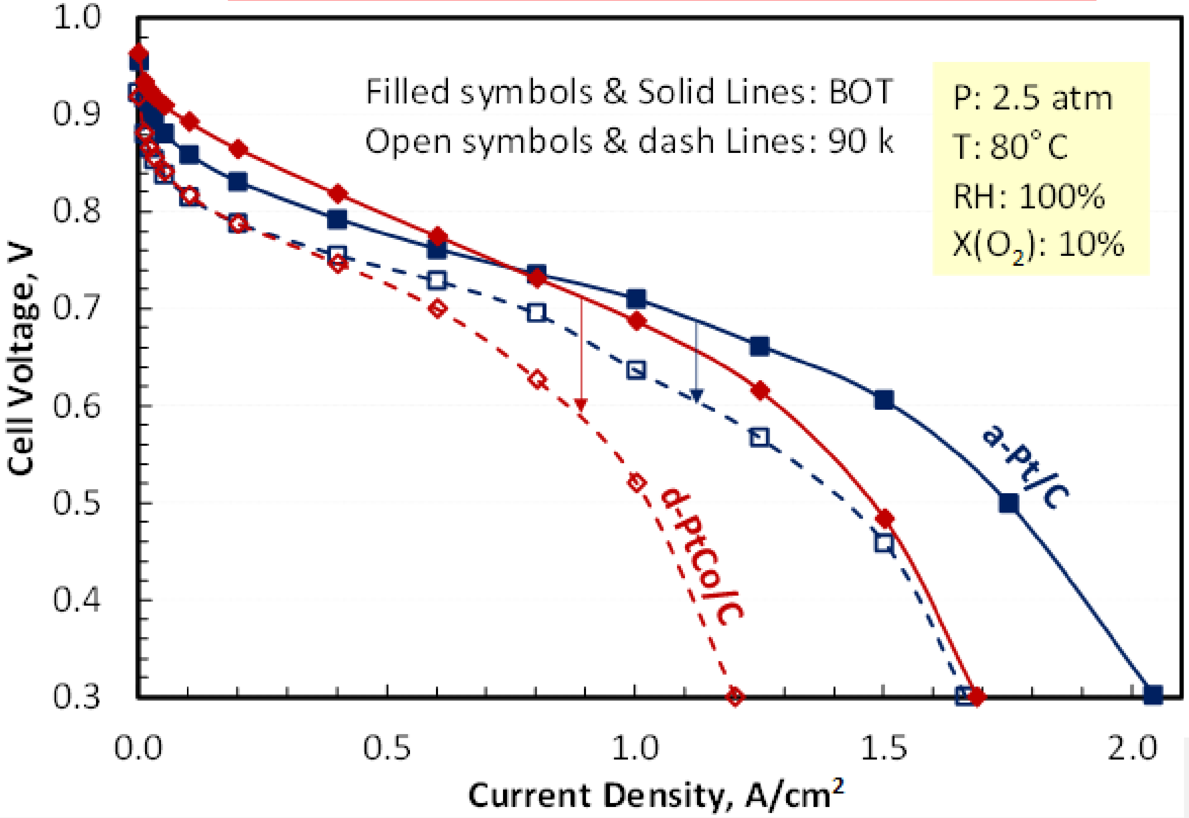
M2FCT FY2021 Q4 Milestone (ANL, LANL, NREL)

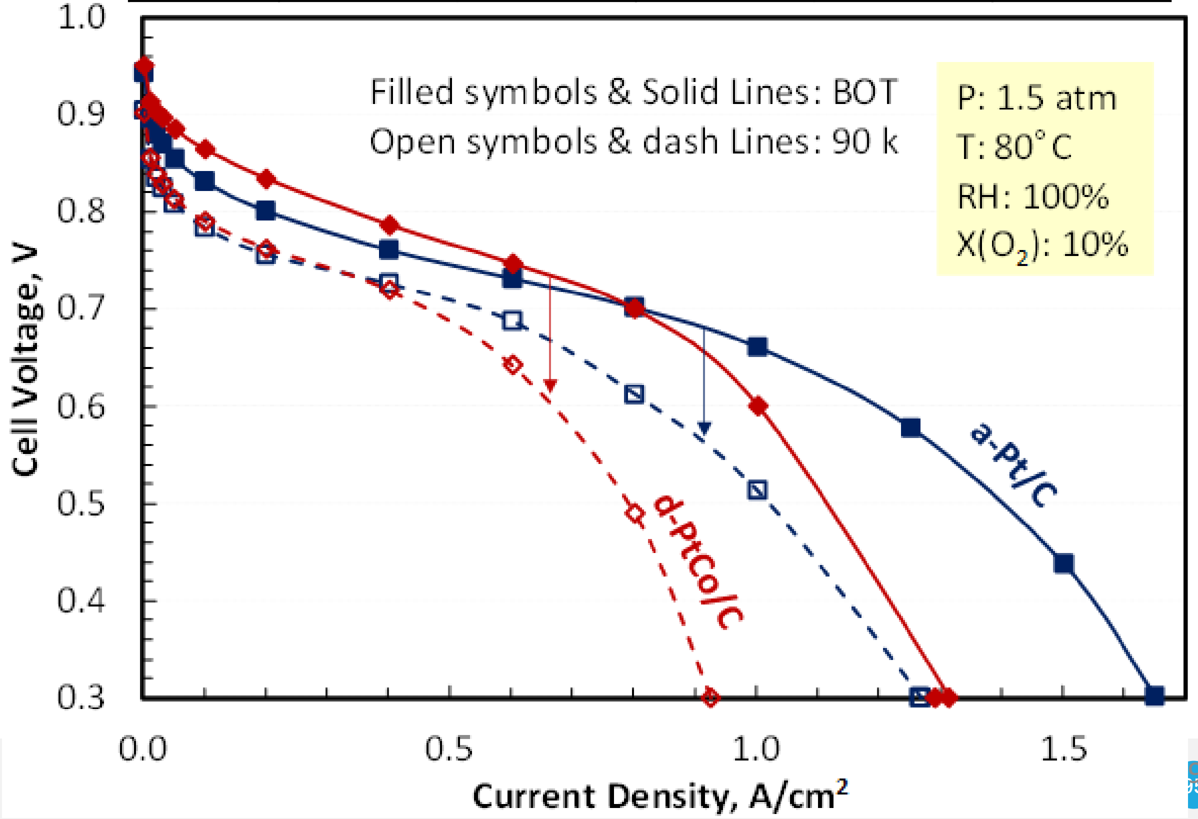
- **AST in H_2/N_2: 0.6 V – 0.95 V square wave with 0.5 s ramp and 2.5 s hold in H_2/N_2 at 80°C and 100% RH, 90k cycles**
- **AST in H_2/Air: 0.675 V – 0.925 V square wave with 30 s hold at 2.5 atm, 90°C and 40% RH, 90k cycles**

Similar electrode stability in differential and integral cells

		Integral Cell Data		Differential Cell Data	
	Number of Cycles	ECSA m^2/g_{Pt}	MA mA/mg_{Pt}	ECSA m^2/g_{Pt}	MA mA/mg_{Pt}
Pt/C	0k	62	476	65	651
	90k	14	82	20	73
a-Pt/C	0k	40	289	39	418
	90k	16	99	20	178
d-PtCo/C	0k	34	878	34	824
	90k	16	144	16	152

M2FCT Data from Leiming Hu and K C Neyerlin (NREL)





Unvalidated ASTs: GDLs an BPPs

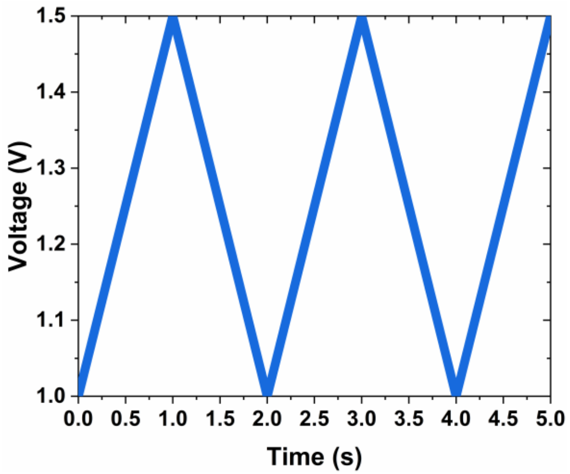
GDLs

1) **Chemical Ageing – peroxide:** 30% H_2O_2
2) **Chemical Ageing – peroxide:** 3% H_2O_2

Sometimes refluxed at 80C

3) **Submersion in H_2O** (with N_2, H_2 or Air)

4) **Potential Cycling**

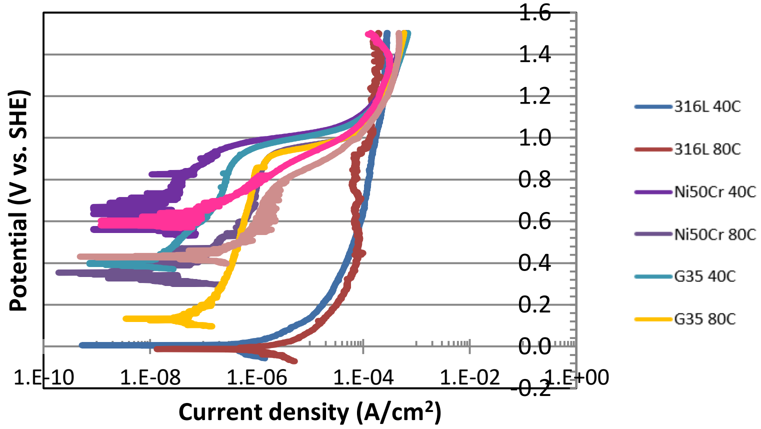


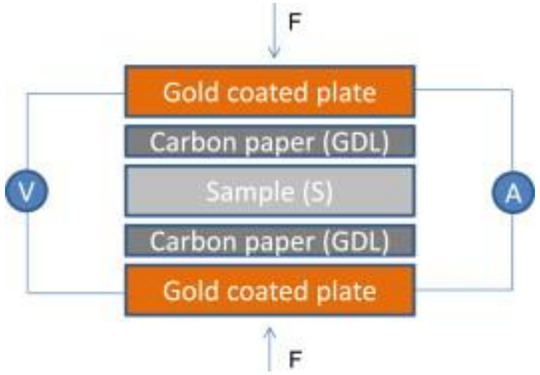
Potential Cycling

- H_2/N_2
- 80 ^oC, **200% RH**
- **7500 cycles**

BPPs

Table 3.4.8 Technical Targets: Bipolar Plates for Transportation Applications			
Characteristic	Units	2015 Status	2020 Targets
Corrosion, anode[g]	µA / cm^2	No active peak[h]	<1 and no active peak
Corrosion, cathode[i]	µA / cm^2	<0.1[c]	<1
Electrical conductivity	S / cm	>100[j]	>100
Areal specific resistance[k]	ohm cm^2	0.006[h]	<0.01





M2FCT discussing AST validation for HDV and 25,000 hrs