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

# M2FCT: Million Mile Fuel Cell Truck Consortium

DOE Hydrogen Fuel Cell Technologies Office
2023 Annual Merit Review and Peer Evaluation Meeting
June 6, 2023

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

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

**DOE AOP project award: WBS 1.5.0.402**

# M2FCT Consortium - Overview

## Timeline

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

## Budget

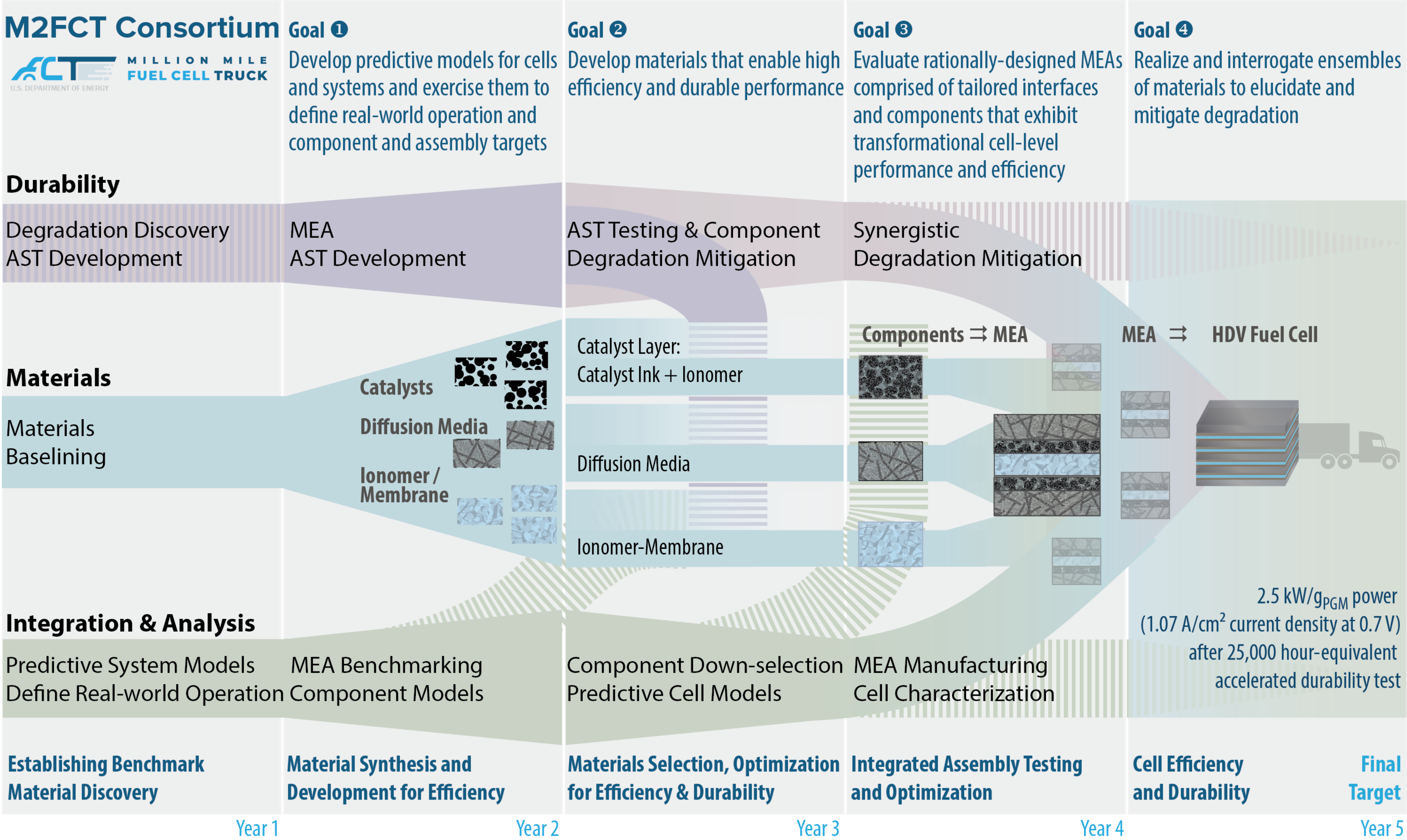
- FY23 project funding: $10M
  - $1.5M Effort to Support FOAs
  - Expansion of FOA Support for upcoming DOE Awards

## Partners/Collaborations

- DOE DE-FOA-0002044:
  - GM, Nikola, Carnegie Mellon (several ending FY23)
- DOE DE-FOA-EE0009244:
  - 3M, Lubrizol, Nikola, UT Knoxville (several ending FY23)
  - Cummins, Plug Power
- DE-FOA-0002446:
  - Raytheon, NeoGraf, GM, TreadStone, Plug Power
  - Caterpillar, Eaton, R&D Dynamics, Mahle
- No-cost collaborations

## Barriers and Targets

- Cell durability
  - 25,000 hours (2025), 30,000 hours (2030)
- Peak efficiency
  - 68% (2025), 72% (2030)
- Fuel-cell system cost
  - $80/kW (2025), $60/kW (2030)
- Overall Target: 2.5 kW/$g_{PGM}$ power - 750 mW/cm² (1.07 A/cm² current density at 0.7 V) - after 25,000 hour-equivalent accelerated durability test


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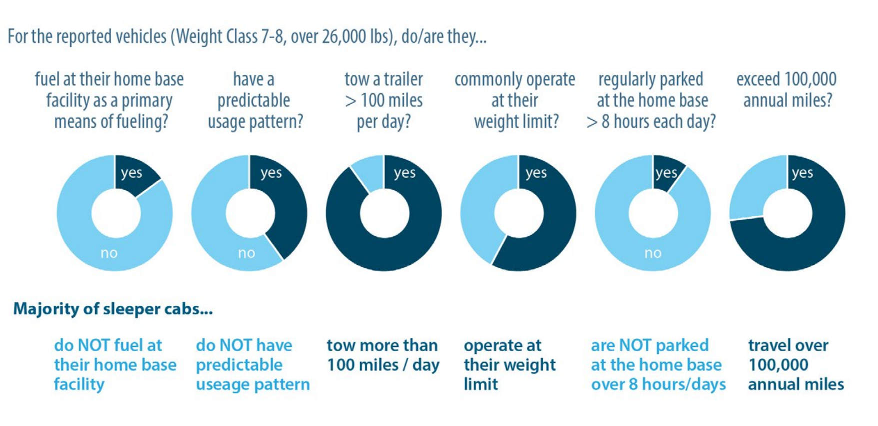
Goal ❶
Develop predictive models for cells and systems and exercise them to define real-world operation and component and assembly targets
Goal ❷
Develop materials that enable high efficiency and durable performance
Goal ❸
Evaluate rationally-designed MEAs comprised of tailored interfaces and components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

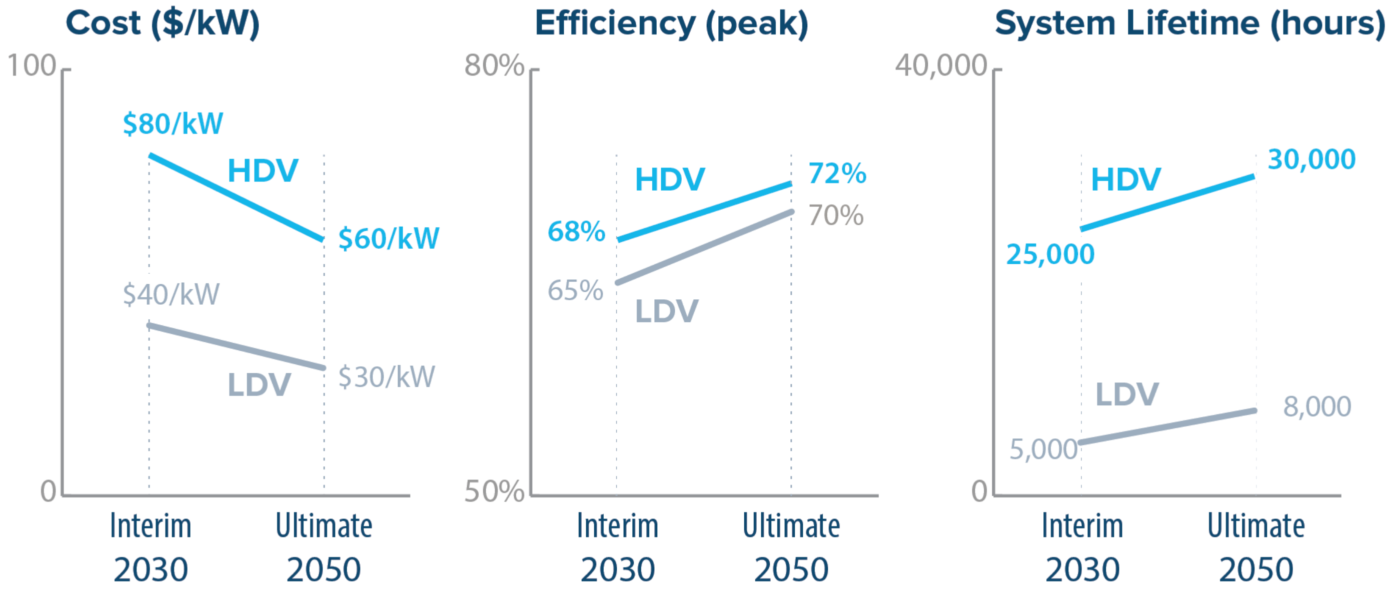
# M2FCT focuses on fuel-cell trucks that demand a greater emphasis on system efficiency and lifetimes

Heavy-duty vehicles (HDV) exacerbate durability and efficiency challenges for fuel cells




## DOE Targets for Fuel-Cell Vehicles

Light Duty Vehicles (LDV) vs. Heavy Duty Vehicles (HDV)

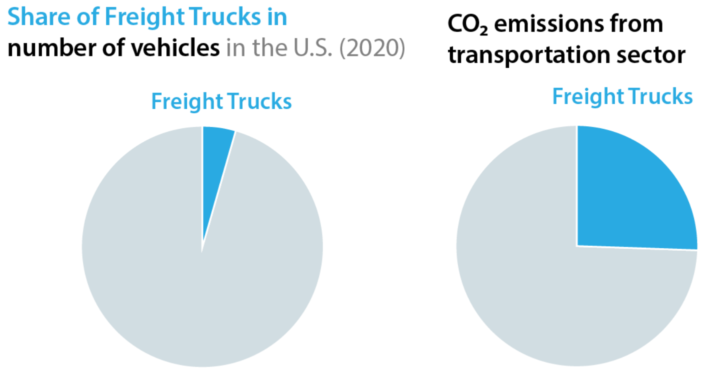


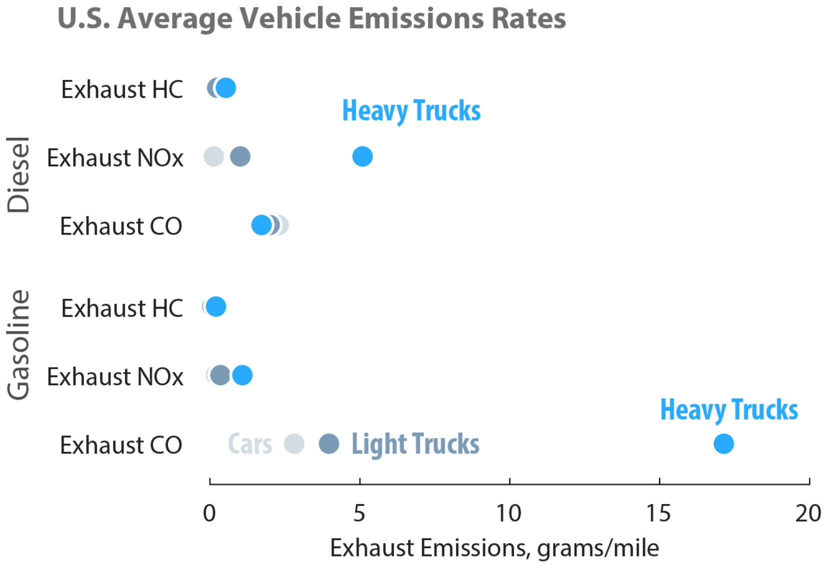

## Fuel-Cell Vehicles Durability Targets

# Energy Justice and Equity

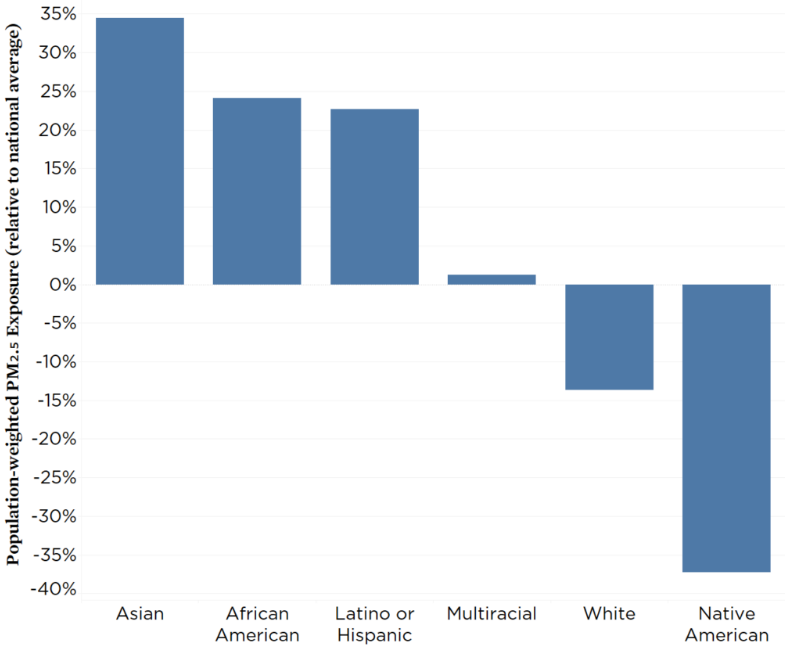
## Decarbonizing freight transportation and long-haul trucking

- **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



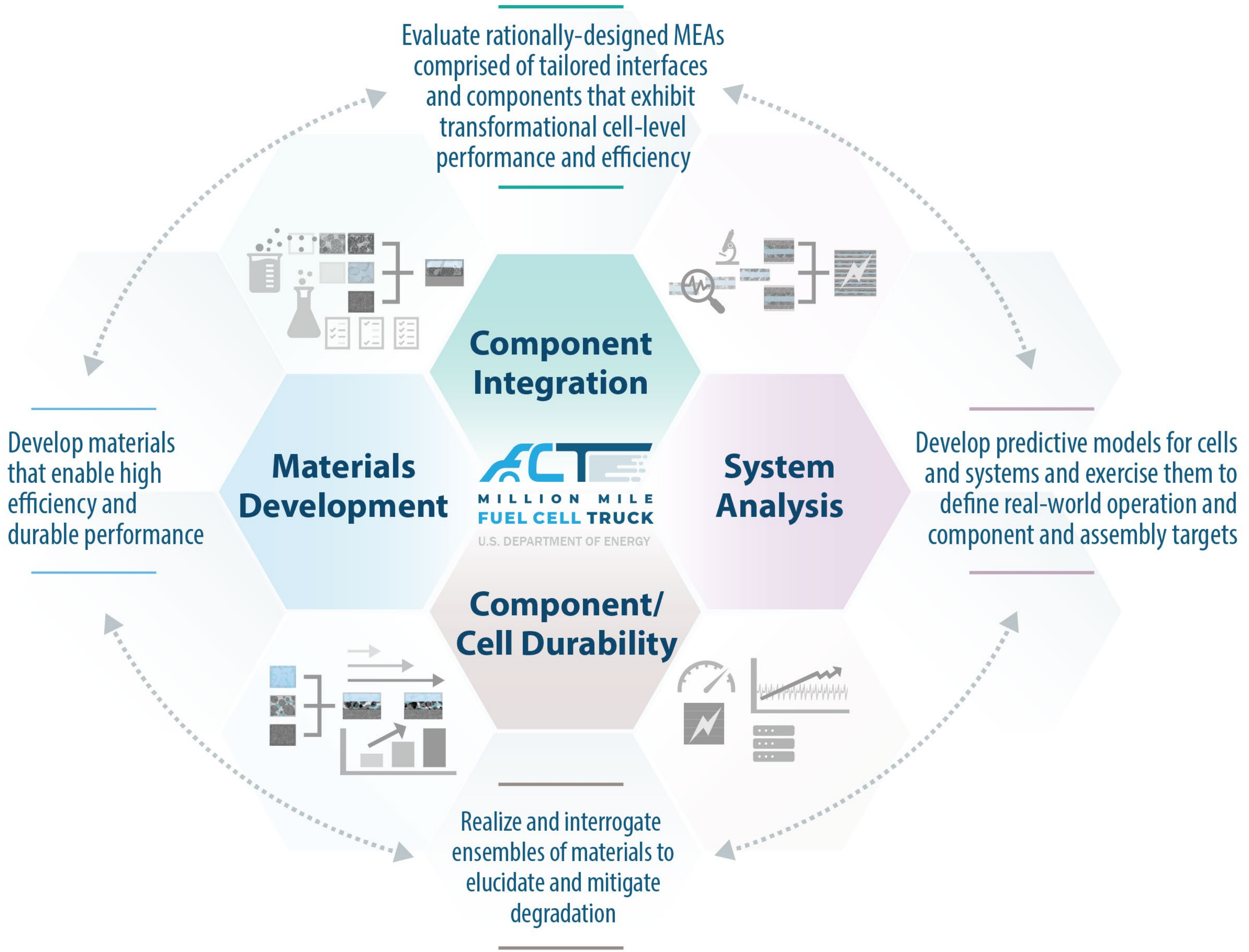



Source: EIA, BTS, EPA; Tessum et al. *Science Advances*, 2021 Liu et al., *Environ. Sci. Technol.* 2015, 49, 19, 11569–11576

# 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 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
University of Tennessee, 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

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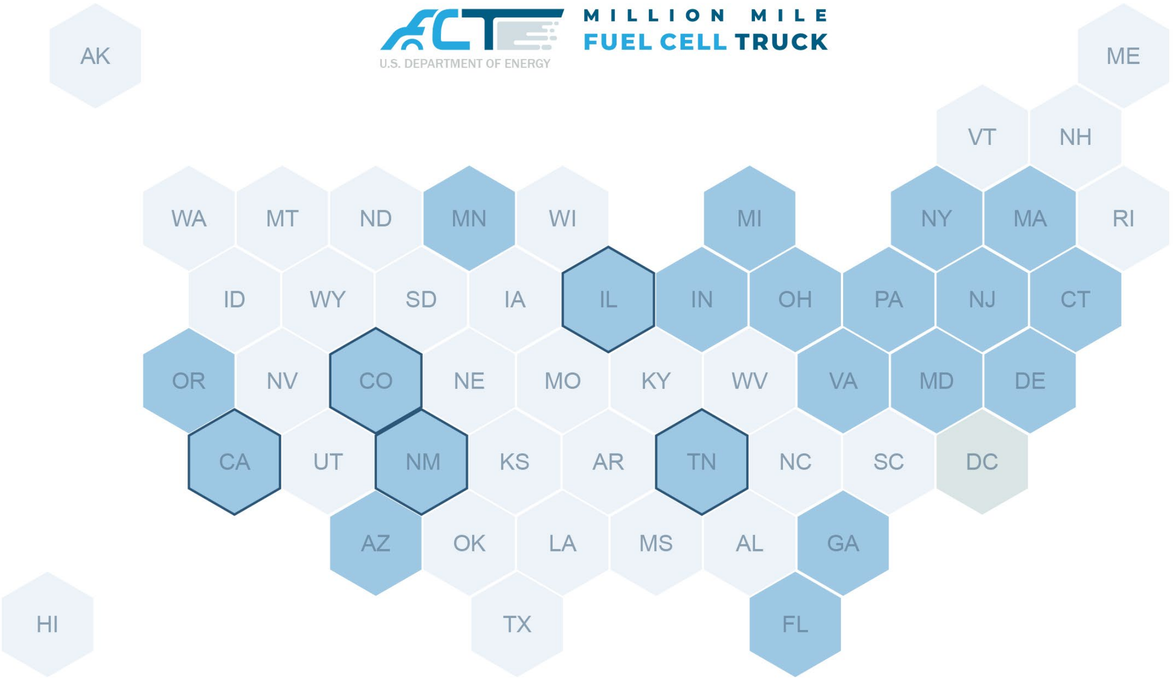


### LABS

**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, University at Buffalo, FIU Florida International University, UCI

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

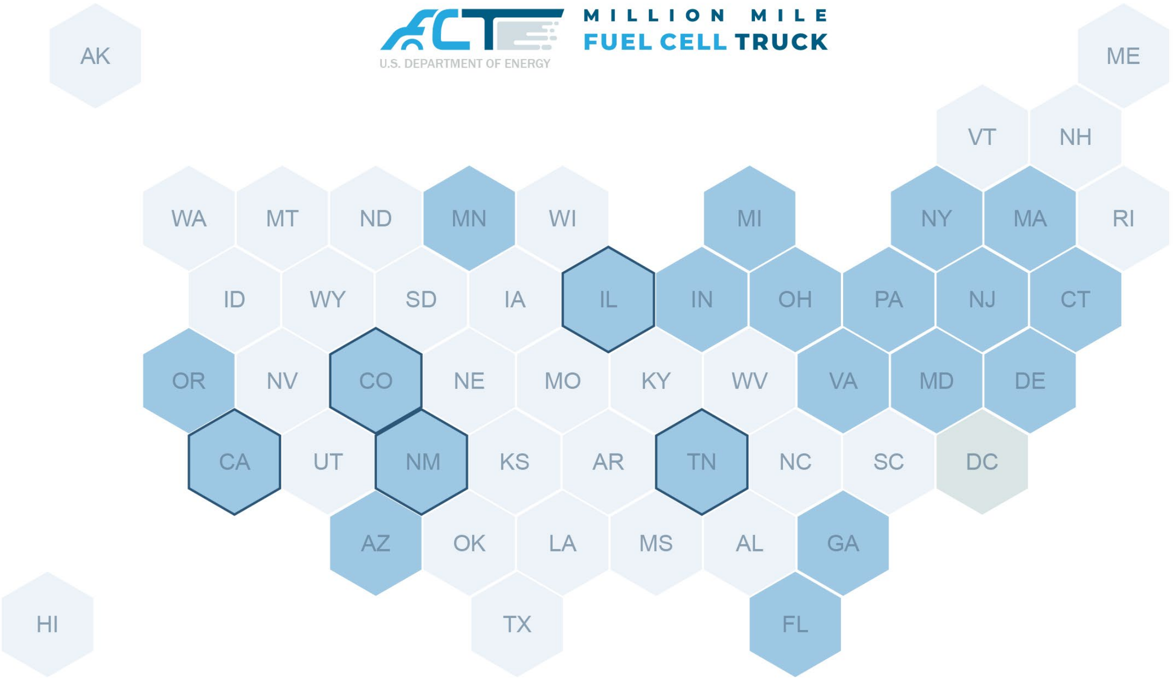
## MEA Projects
FC323 FC326 FC327

## Membrane Projects
FC335 FC336 FC333

## Stack Projects
FC337 FC338

## Bipolar Plate Projects
FC344 FC347 FC346 FC345 FC348

## Management Projects
FC352 FC350 FC349 FC351




**In this presentation**

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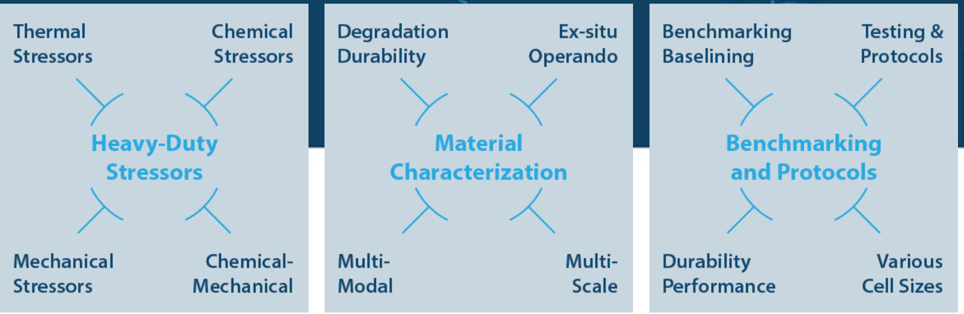
# International Durability Working Group (i-DWG)


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.




https://millionmilefuelcelltruck.org/idwg

# FY23 Milestones

| Milestone Name / Description and Criteria | Quarter | Status |
|---|---|---|
| Baseline analysis of performance, durability and cost of 175-kW fuel cell systems for heavy duty trucks relative to the previous results for a 275-kW FCS: 603 $mW/cm^2$ EOL power density, 64.5% peak efficiency, 1.35 $kW/g_{Pt}$ Pt utilization and $197/kW cost at 50,000 units/year manufacturing volume | Q1 | Met |
| Definition of bipolar plate durability testing (corrosion, cation, contact resistance, coating adhesion (if applicable)); utilize input from industrial bipolar plate projects from FOA | Q1 | Delayed to Q3 due to scheduling |
| Complete baseline total cost of ownership (TCO) analysis comparing Class-8 long haul FC trucks with reference diesel truck metrics of 50,000-lb payload, $180,000 CAPEX and $1.60/mile TCO for $3.25/gal fuel cost | Q2 | Met |
| Compare durability of SOA hydrocarbon membrane BOL performance and durability to PFSA based membranes | Q2 | In progress |
| Comparative analysis of performance and durability of catalyst layer ionomer: PFSA and HOPI. Measurements of ionomer degradation and diagnostics. | Q3 | In progress |
| Data Structure, Data Base and correlation/mining related to ionomer-catalyst interactions | Q3 | On track |
| Down-Selection of materials (catalyst, ionomer, membrane, GDL) to proceed into year 4 for manufacturing/integration and optimization | Q4 | On track |
| Determine improvement in performance, durability and cost of fuel cell system with a top-performing M2FCT catalyst relative to the baseline a-Pt/C and electrode consortium targets including meeting the 2023 HDV FCS target of $170/kW and progress towards the 2025 M2FCT target of 2.5 kW/gPGM at 0.7 V after 25,000 h with <0.3 mg/cm2 total PGM loading. | Q4 | On track |

# Materials Development QPM-Go/No-Go

Catalyst Development Go/No-Go Decision

| Milestone Name / Description and Criteria | Quarter |
|---|---|
| Name: Catalyst material Go/No-Go (PNNL, ANL, LANL, NREL, BNL)<br><br>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* |

| | | | |
|---|---|---|---|
| Q12 (9/30/2023) | Go/No-Go | Decision: Decision point on components (membrane, catalyst) materials to move forward into MEA integration and optimization pending passing performance metric after AST | Criteria<br>Catalyst/support:<br>>0.44 $A/mg_{Pt}$ MEA ORR activity after equivalent 25,000 hr AST<br><br>Membrane:<br>< 0.02 $\Omega$-$cm^2$ and < 2 $mA/cm^2$ $H_2$ crossover after AST |

# Systems Analysis

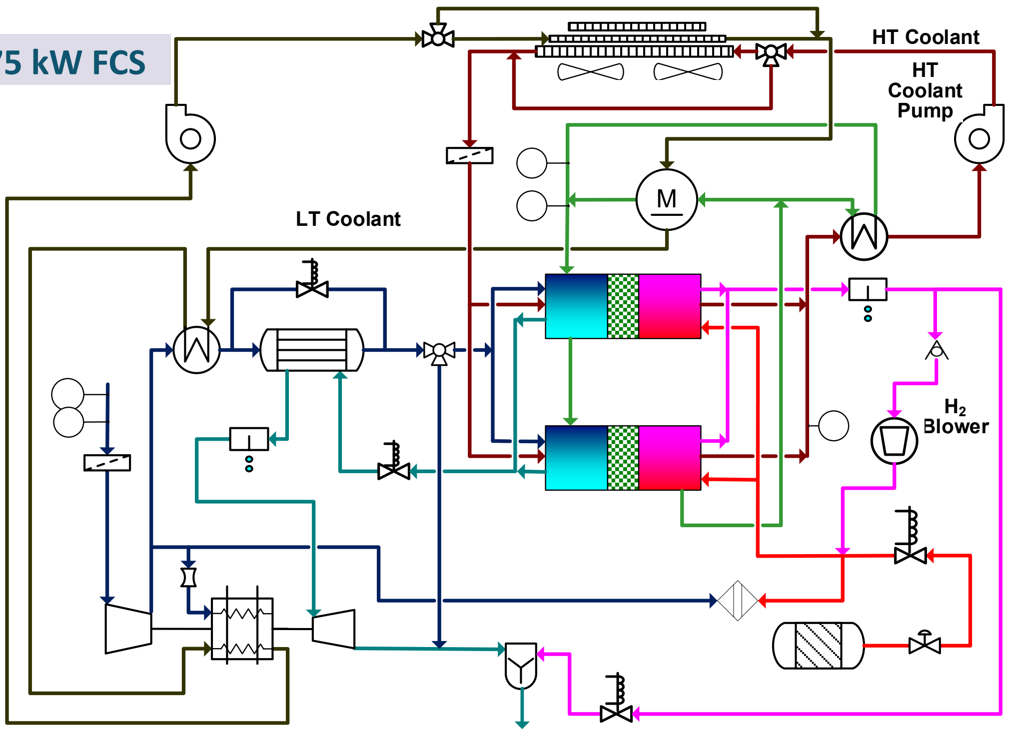
# M2FCT Reference Fuel Cell Systems for Class-8 Heavy-Duty Trucks

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

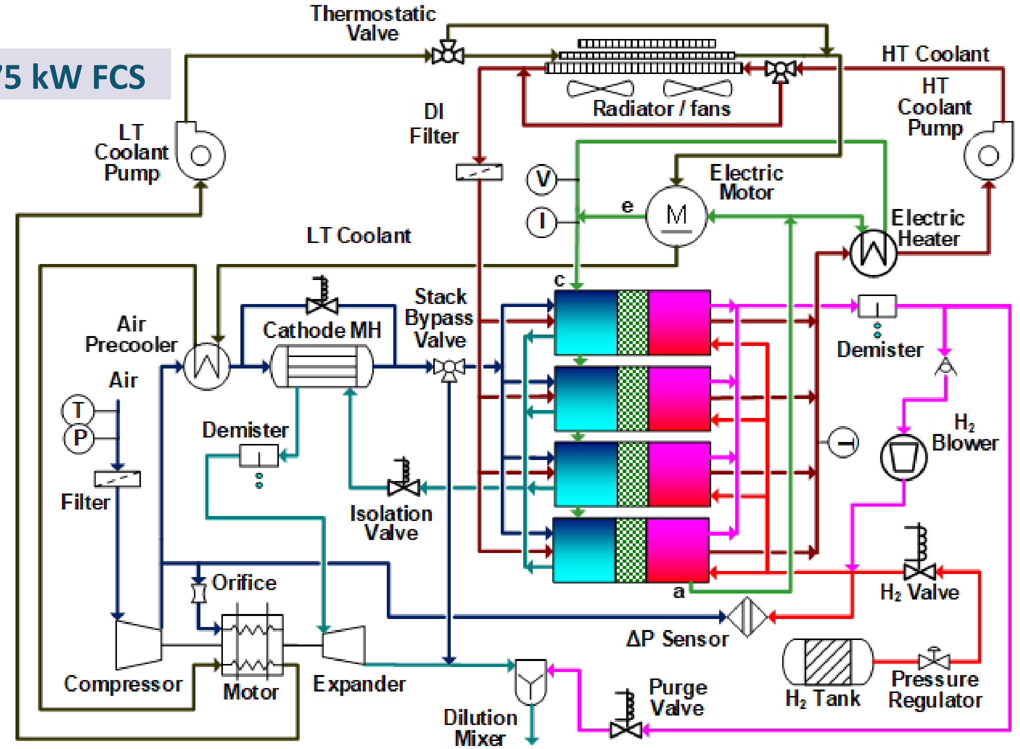
FCH-275
- 275 kW net, 360 kW stack at EOL
- Four stacks
- 106-kWh ESS

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

175 kW FCS



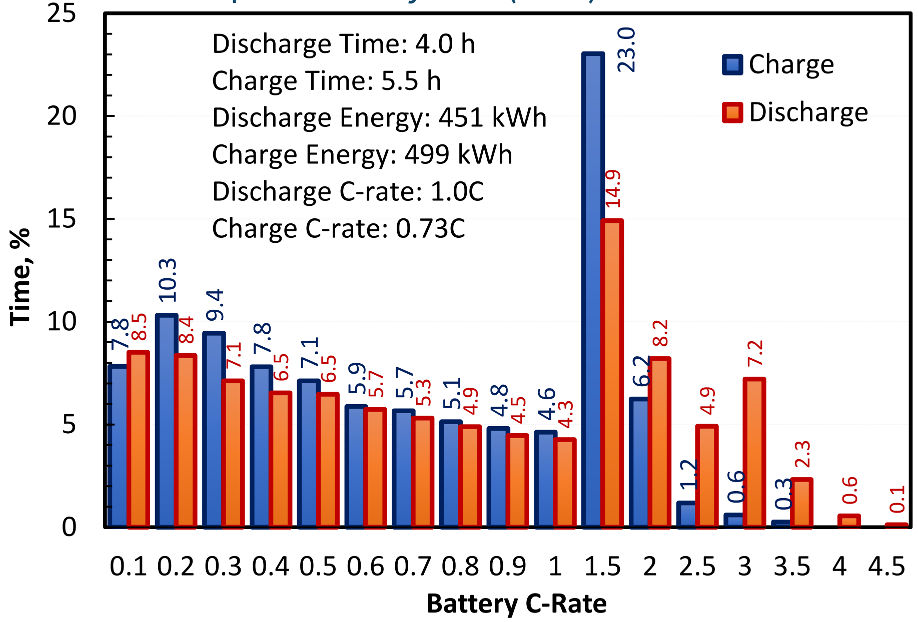

275 kW FCS



# HDV Load Sharing: ESS Duty Cycle

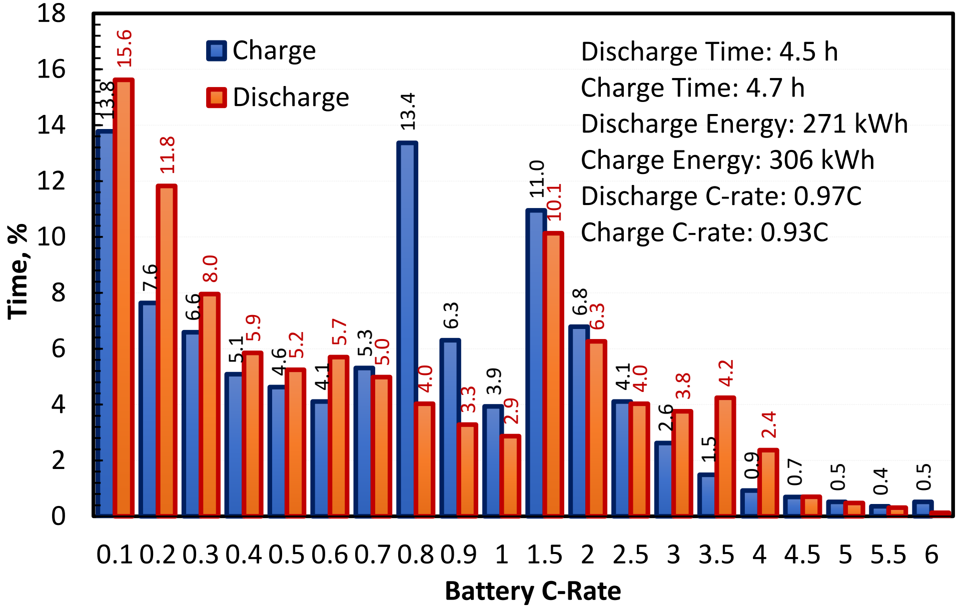
## FCH-175

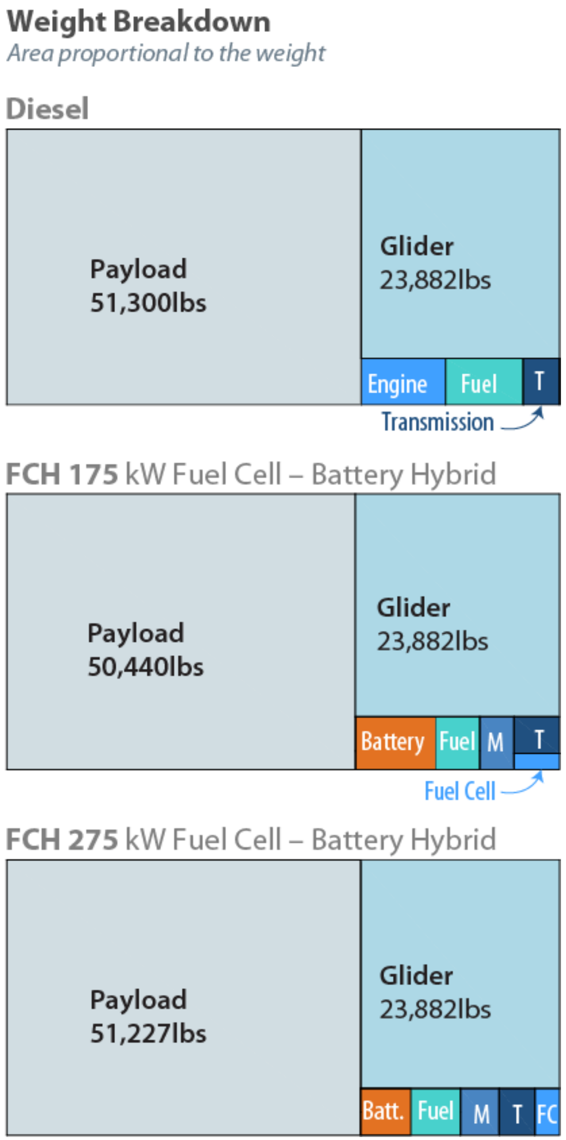
- Battery Duty Cycle
  - Battery power generally varies between ±300 kW.
  - 451 kWh discharge energy during 1 HDV duty cycle.
  - Results: 183 kWh battery capacity and 16% DOD at BOL; 35% capacity fade after 6230 full equivalent cycles (FEC)




## FCH-275

- Battery Duty Cycle
  - Battery power generally varies between ±300 kW.
  - 271 kWh discharge energy during 1 HDV duty cycle.
  - Results: 106 kWh battery capacity and 18% DOD at BOL; 38% capacity fade after 6867 full equivalent cycles (FEC)



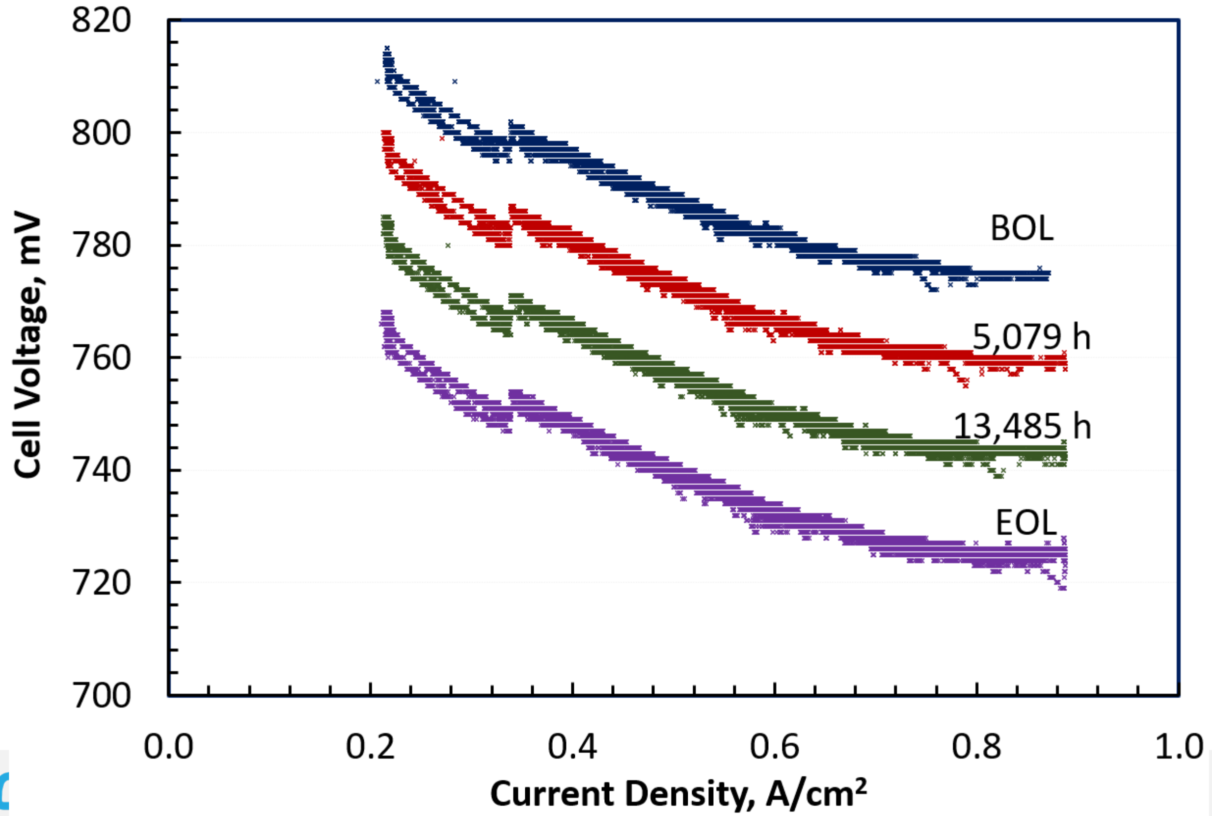
# Dynamic Polarization Curves (FCH-175)

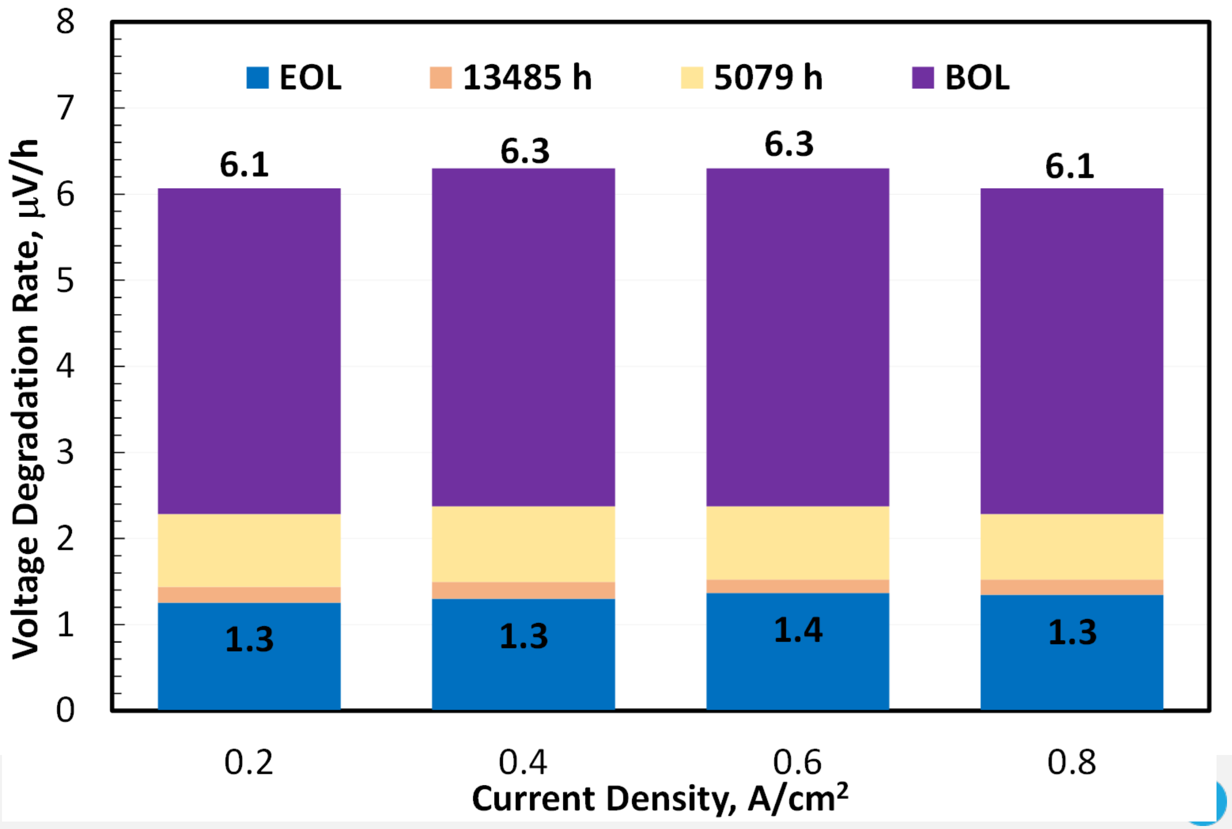
**Dynamic Polarization Curves**

- ~45 mV voltage drop (ΔV) from BOL to EOL
- ΔV is nearly independent of current density implying that it is mainly due to slowdown in ORR kinetics

**Voltage Degradation Rate**

- Mean degradation rate: 1.8 μV/h
- Degradation rate at constant idle power slows with time because of decrease in maximum cell voltage



# FCS Lifetime and Cost



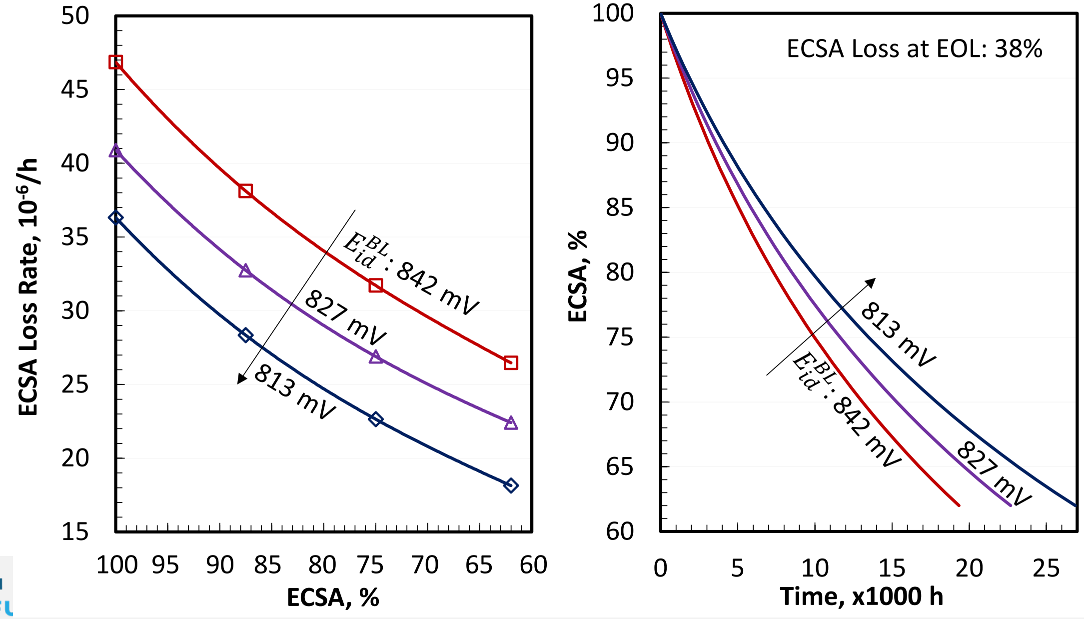
**Approach to reaching 25,000-h lifetime on Class-8 long haul truck duty cycle**

- **Voltage Clipping: 813 mV in FCH-175, 820 mV in FCH-275**
- **Idle Power Limit: 50 kW in FCH-175, 70 kW in FCH-275; Reduce SR(c) to 1.1 during idle**
- **Catalyst loading: 0.45 mg/cm$^2$ total Pt loading**
- **Active Membrane Area Oversizing: 44% in FCH-175, 67% in FCH-275**
  - Acceptable ECSA Loss: 38% for 750 mW/cm$^2$ EOL PD in FCH-175, 50% for 642 mW/cm$^2$ EOL PD at EOL in FCH-275
  - At constant idle power, electrode degradation rate slows with aging as the max. cell voltage decreases by 43 mV

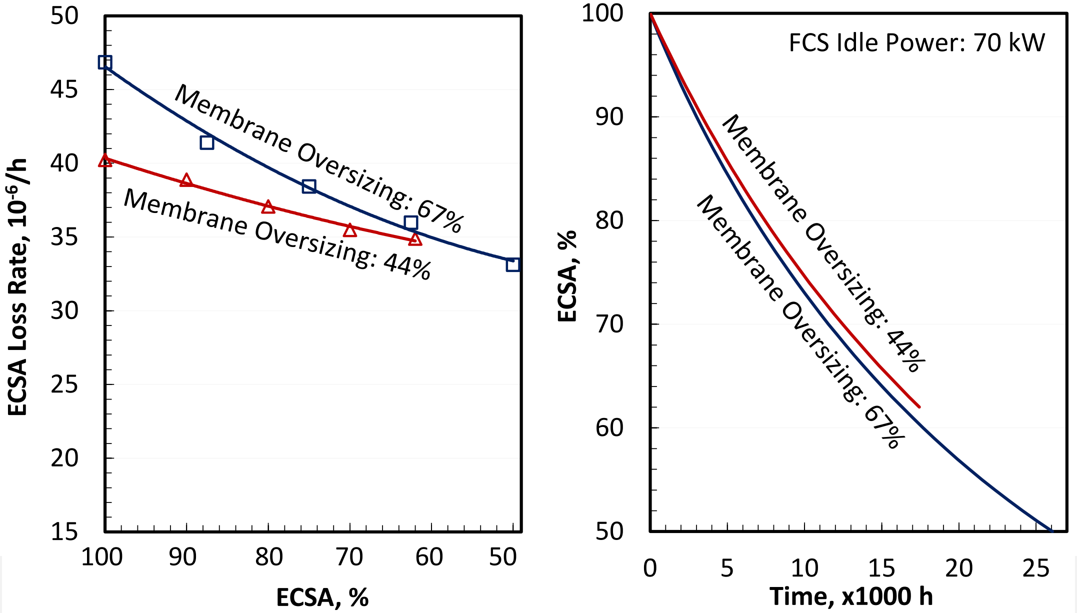
**Projected Costs**

- FCH-175: $194/kWe FCS, $154/kWe FCH
- FCH-275: $198/kWe FCS, $197/kWe FCH

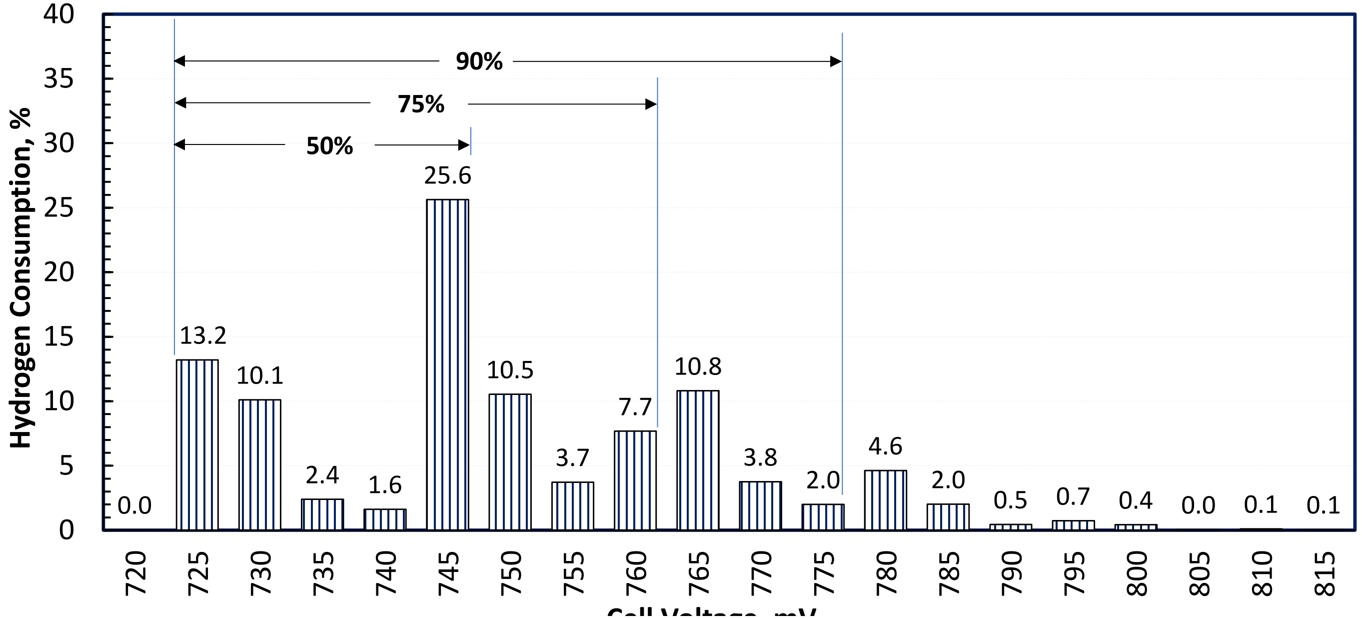
## FCH-175

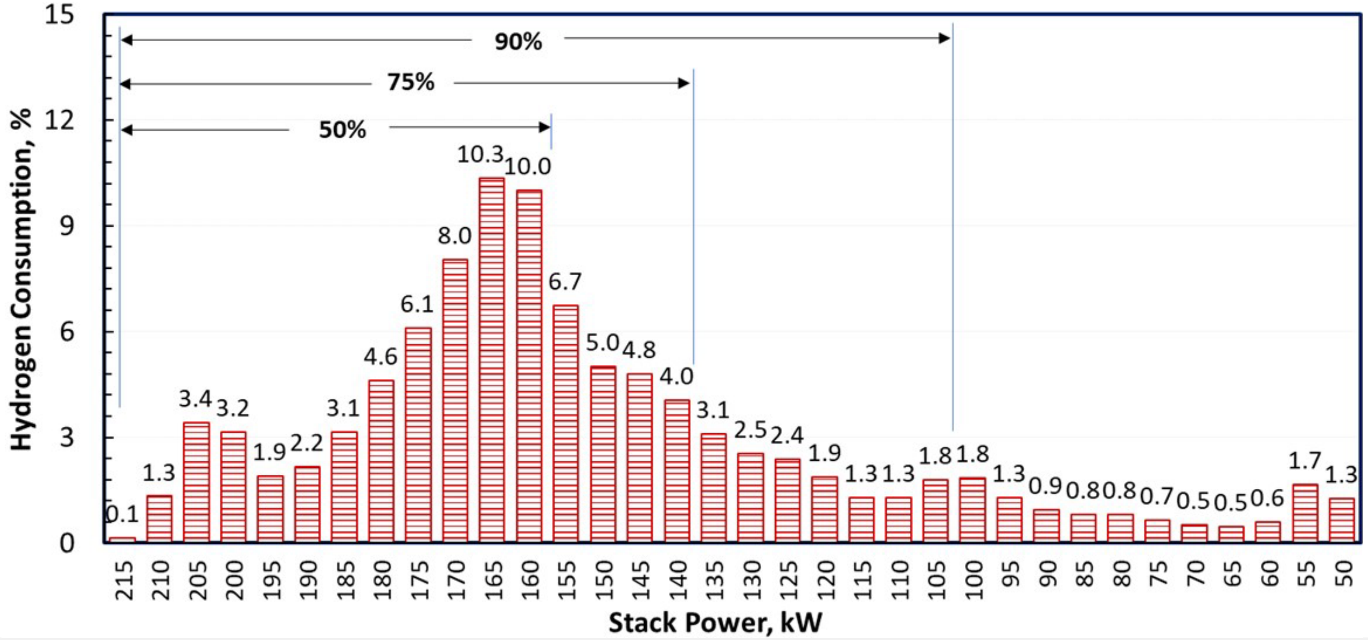



## FCH-275



# Hydrogen Consumption in FCH-175




**Lifetime $H_2$ Consumption on Class-8 Long Haul Truck Duty Cycle**

**Voltage clipping has minimal effect on fuel economy**

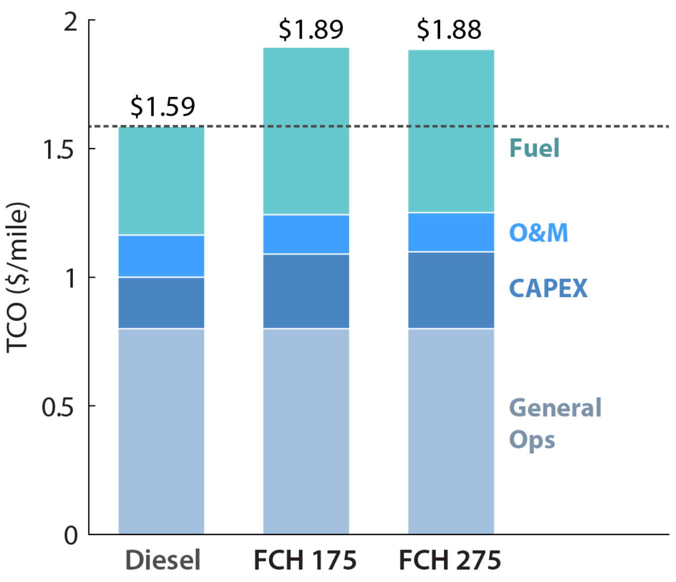
- Less than 10% $H_2$ consumed at cell voltage above 775 mV

**$H_2$ consumption during idling**

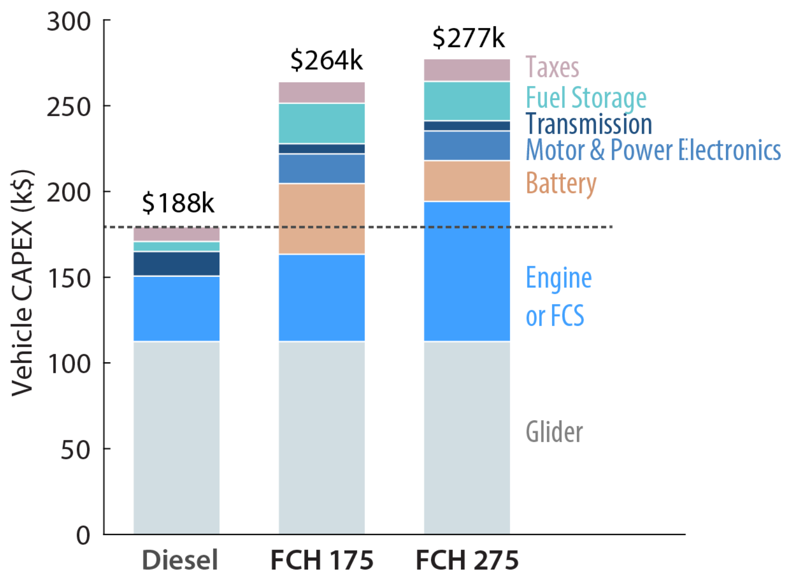
- Less than 5% $H_2$ consumed at stack power below 75 kW, i.e., below 43% of rated power

# Total Cost of Ownership

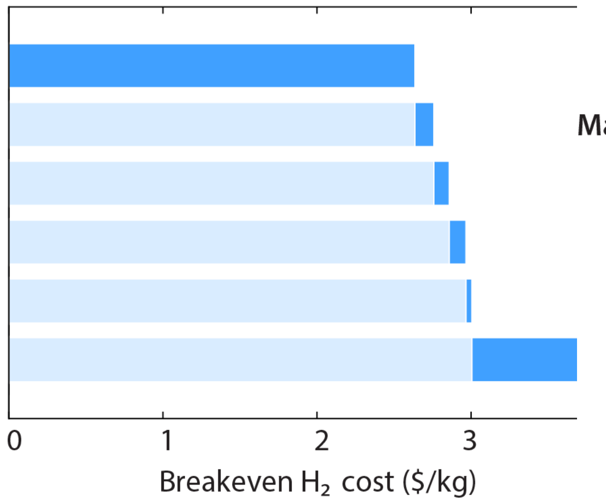
TCO Breakdown



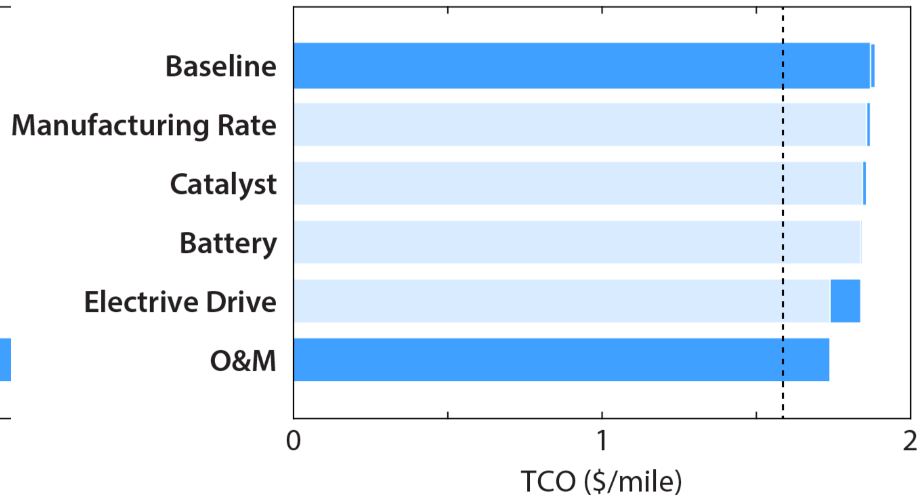

CAPEX Breakdown




TCO Breakdown: FC-275

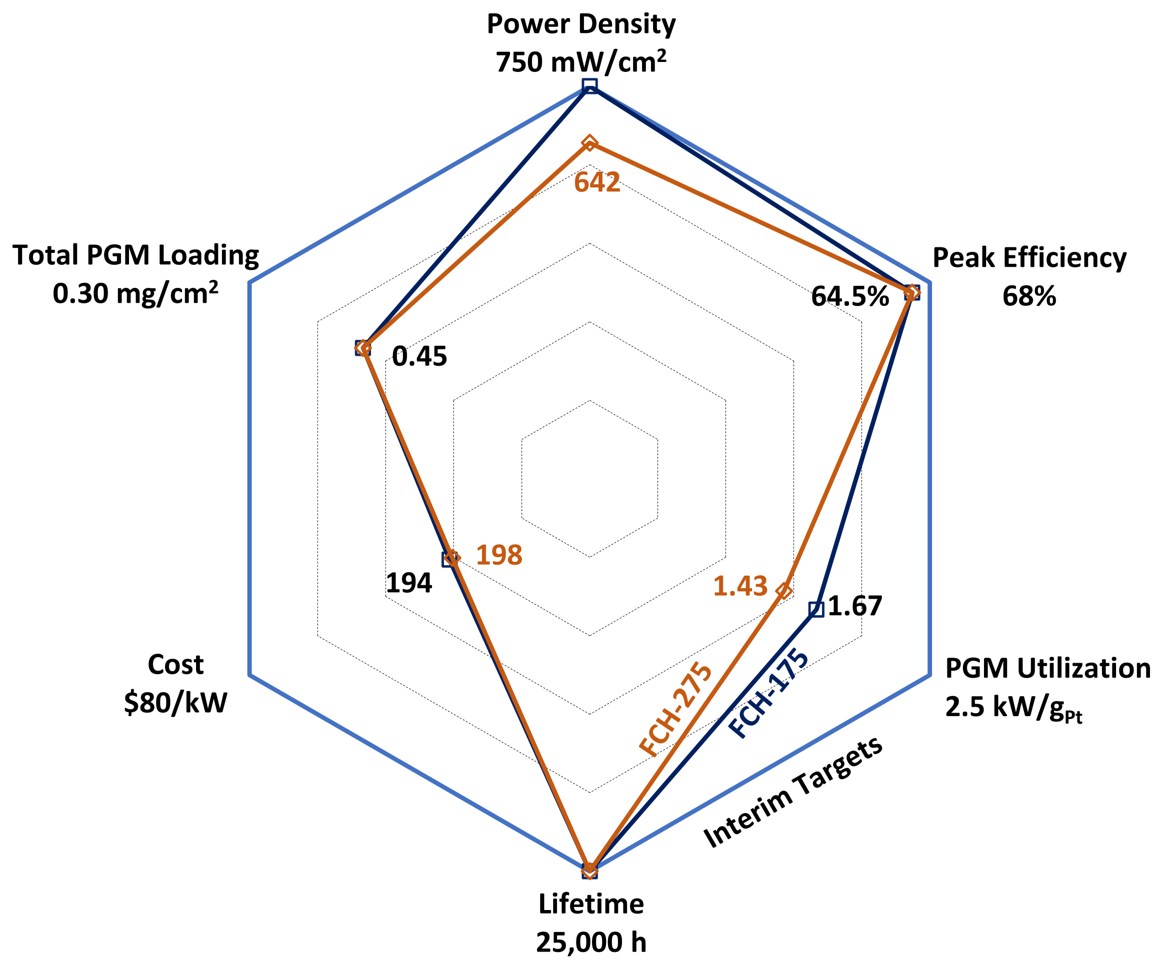



CAPEX Breakdown: FC-275




- **FCS Manufacturing Rate: 10,000 units/year**
- **No loss in payload capacity even without factoring in exemption for alternative fuels and engines**
- **FC vehicle CAPEX 50-60 % higher than conventional diesel vehicle CAPEX Breakdown FCS > Battery > Fuel Storage > Transmission > (Motor + Power Electronics)**
- **No dwell time cost as $LH_2$ can be charged within 10 mins**
- **TCO of FC vehicle ~$0.30/mile higher than conventional diesel vehicle due to higher fuel cost and CAPEX**
- **Breakeven fuel cost relative to $3.25/gal diesel: $2.60/kg-$H_2$ for FCH-175 & $2.64/kg-$H_2$ for FCH-275**

# Fuel Cells for Heavy Duty Trucks –Status




Projected cost at 10,000 units/year manufacturing rate using cost correlations from Jennie Huya-Kouadio, Strategic Analysis

**2023 Status of Fuel Cell Systems for Class-8 Trucks**

- **Key barrier to meeting the interim targets: Active and stable catalysts capable of meeting the target of 750 mW/cm$^2$ power density at EOL with 0.3 mg/cm$^2$ Pt loading**
- **Need to address membrane durability**
- **Demonstrate 25,000-h electrode lifetime with voltage clipping, catalyst overloading and membrane oversizing**

**M2FCT Gen-2 175-kW Fuel Cell – Battery Hybrid**

**Thermal Management**

- Ambient temperature: 40°C
- Radiator size: Same frontal area as in diesel truck
- Battery Thermal Management system (BTMS)

**Power Management**

- Load sharing between FCS and energy storage system (ESS): Optimized stack and battery durability
- FCS end of life (EOL) criteria: 750 mW/cm$^2$ power density after 25,000 vehicle operating hours
- ESS EOL criteria: Provide supplemental power and energy for 6% hill climb after 25,000 vehicle operating hours

# Multicomponent Integration

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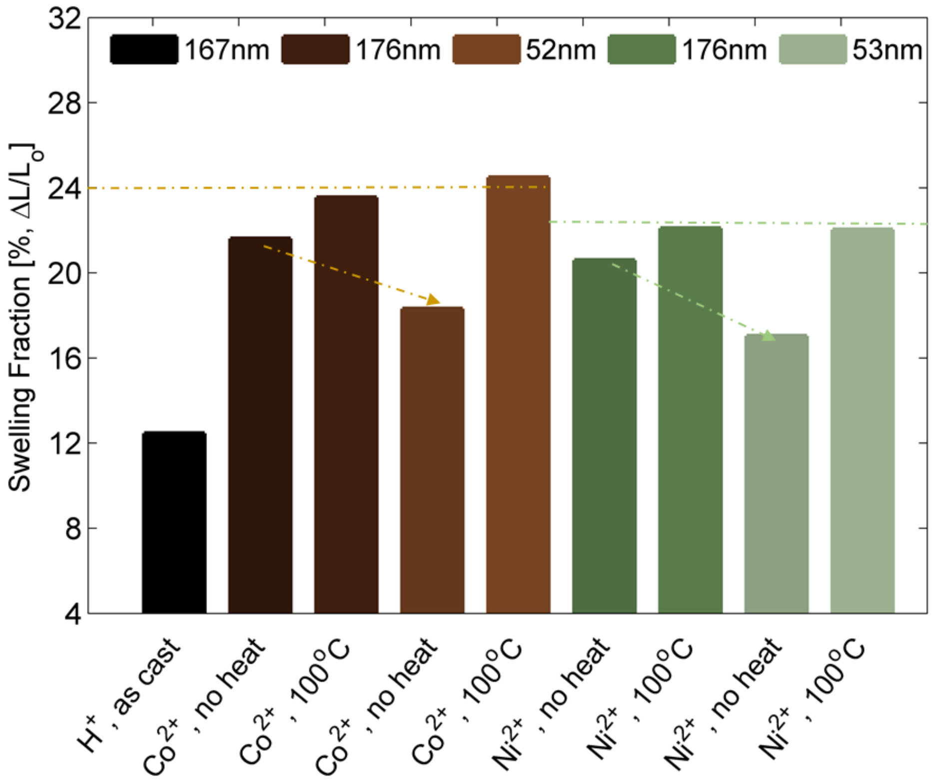
# Not All Dispersions are the Same

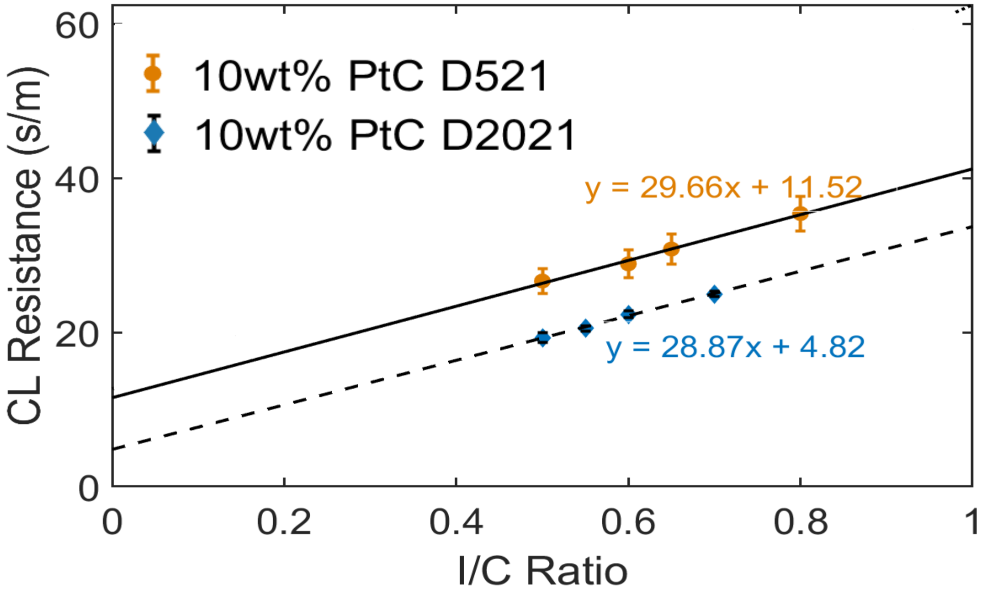
## Dispersion temperature

- Heat treatment and casting temperature or contaminant ions impact thin-film properties




## Starting stock solution concentration

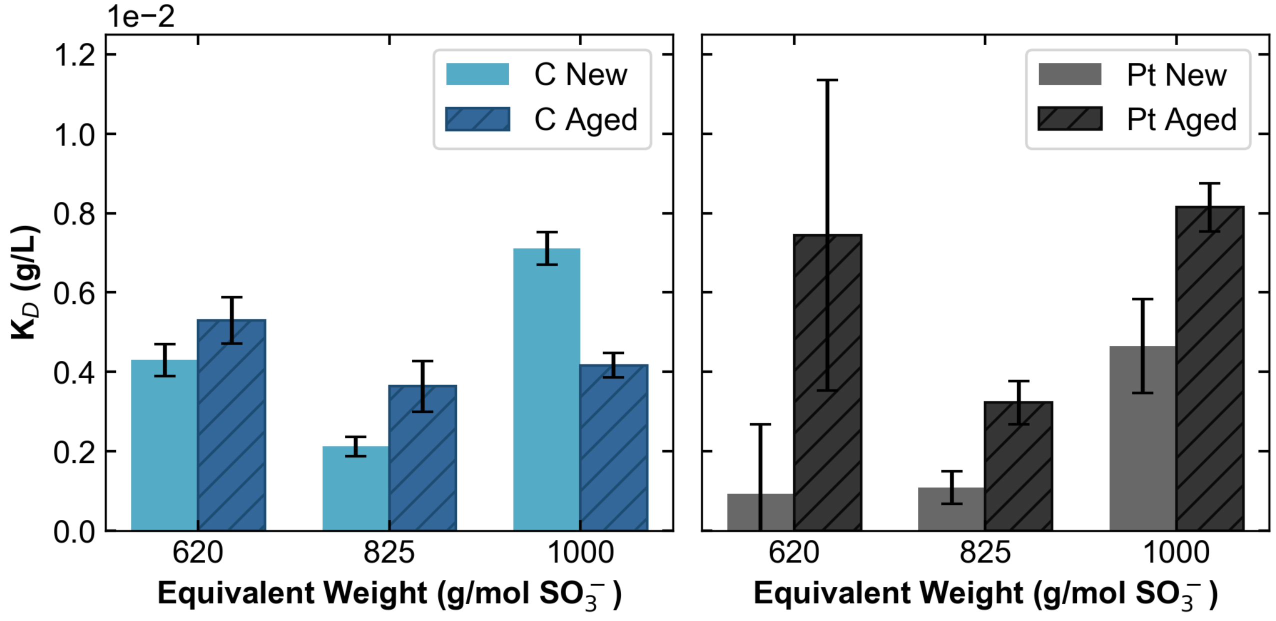
- Stock solution and dilution amount impacts performance, in particular the ionomer interaction with the electrocatalyst
  - Interfacial resistance



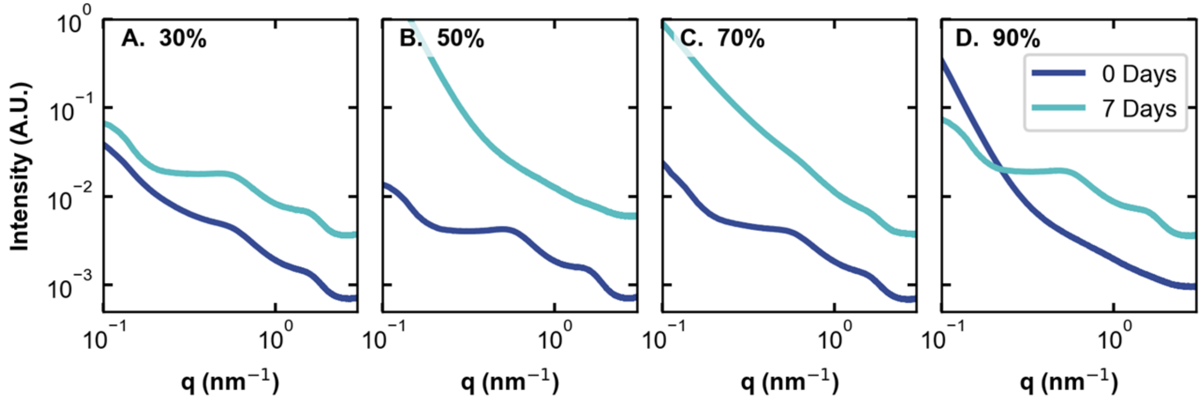
# Not All Dispersions are the Same

## Effect of time (ageing)

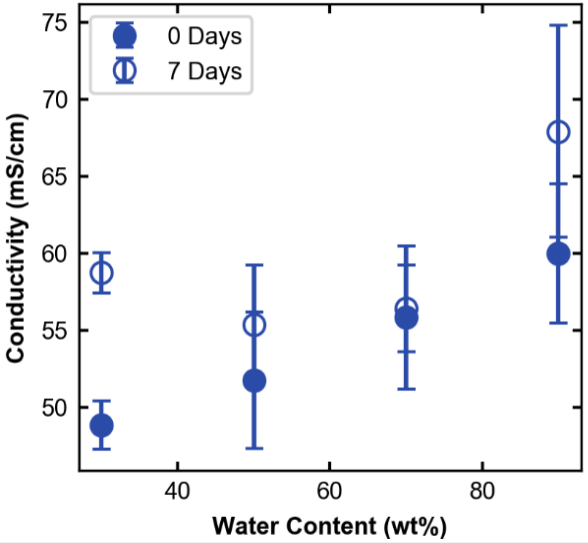
- Casted membrane morphology and properties are different
  - Change depends on water content (i.e., how different from stock solution)
- Changing in binding constant and interaction with the electrocatalyst




Ionomer/catalyst binding via ITC for new and 7 day-old dispersions



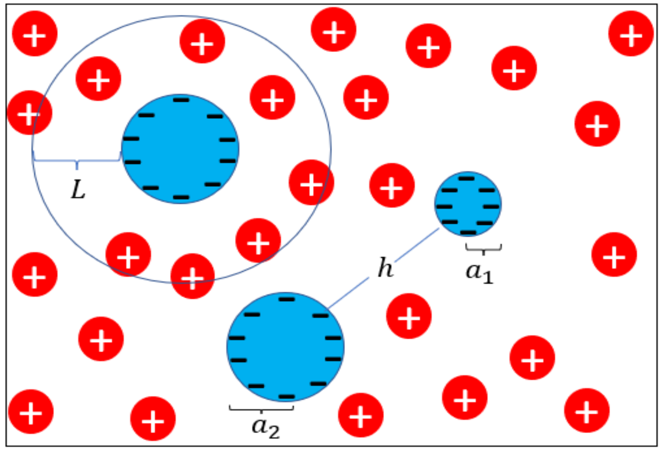

Membrane SAXS




Membrane conductivity

# Modeling Ink Colloidal Particles

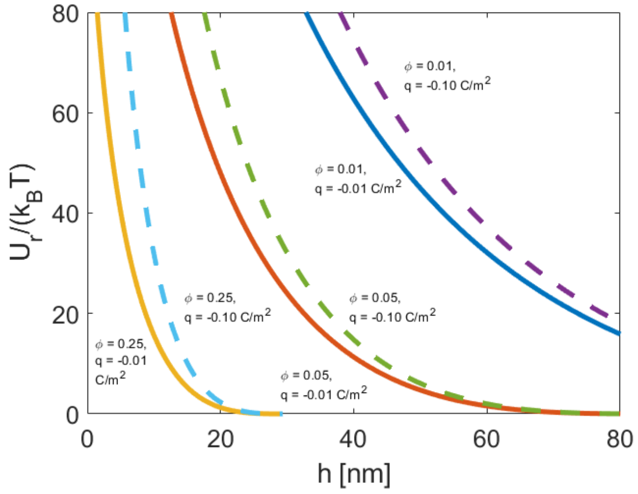
- Electrostatic model created to find the interaction energy between colloidal (e.g., ionomer) particles
- Model shows volume fraction between particles strongly changes forces
  - Increased sizes lead to exponentially greater repulsion, revealing an equilibrium aggregation size



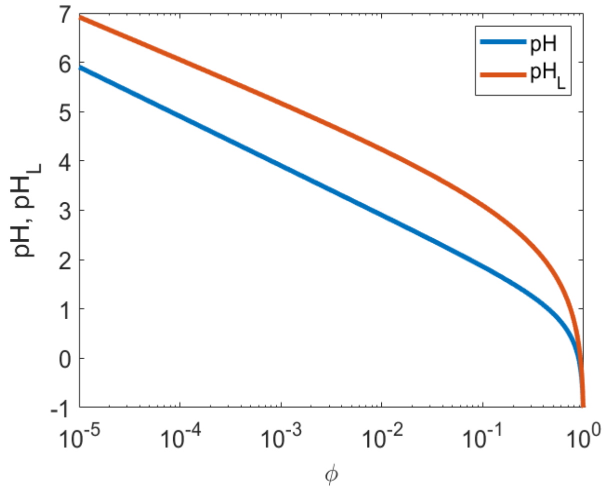

$$U_r(h) = \frac{2\pi a_1 a_2}{a_1 + a_2} RT \int_h^{2L} (y-h)(C_m(y) - C_L)\,dy$$

$$\frac{L}{a} = \frac{1}{\varphi^{1/3}} - 1$$

Calculated by solving P-B with the given surface charge numerically




**Volume fraction dominates charge density in repulsion**



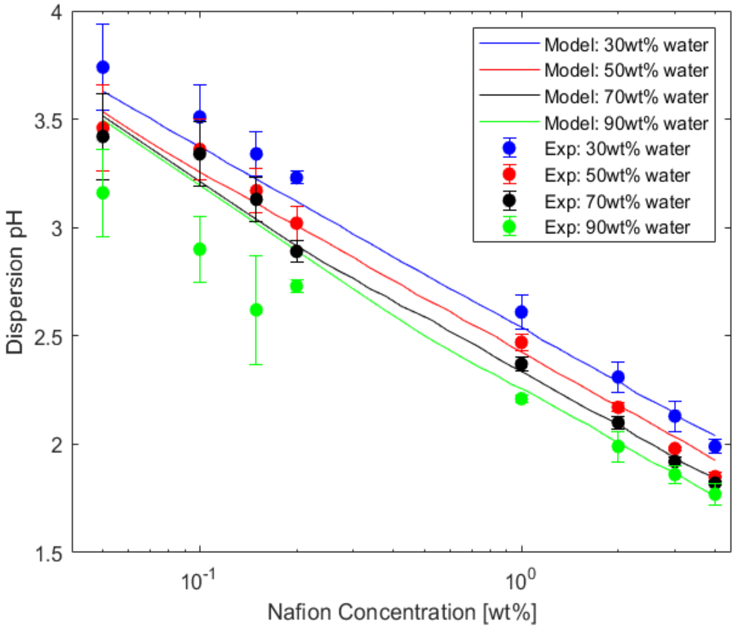

**Predict pH for different volume fractions due to coalescence and concentration**



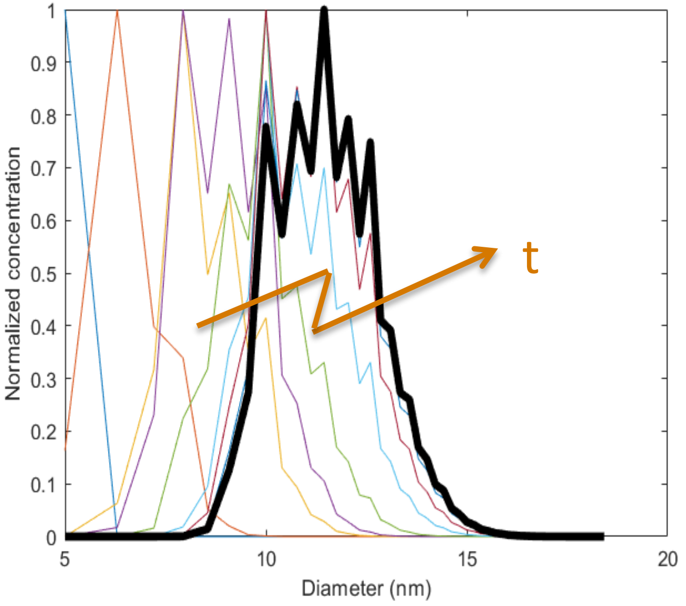
Harsh Srivastav, Adam Z. Weber, and Clayton J. Radke, *J. Phys. Chem. Letters* (submitted)

# Modeling Ink Particles Aggregation

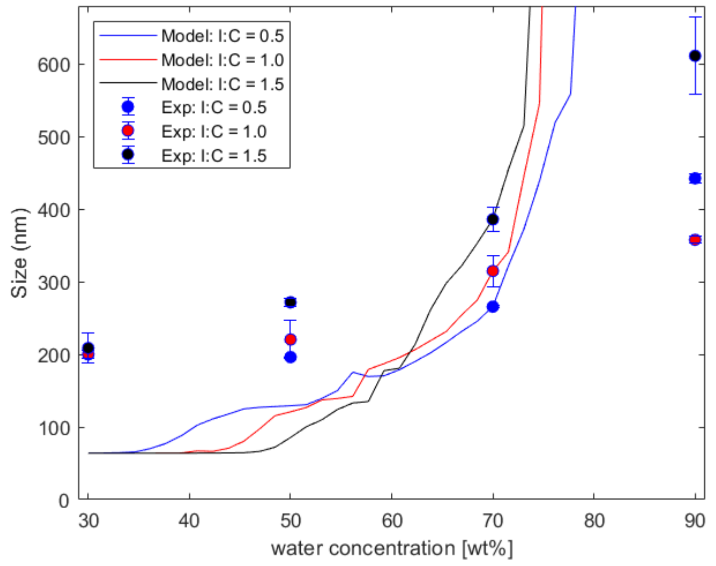
- Utilize colloidal and kinetics model to predict aggregation
  - Relate pH of inks to sizes of particles
- Model shows increasing aggregation as either Nafion or Carbon are concentrated
  - Particle size distributions and porosity can be obtained from the formed catalyst layers




Nafion only pH model results vs. data




Size evolution of Nafion until equil. size




Carbon + Nafion model results vs. DLS

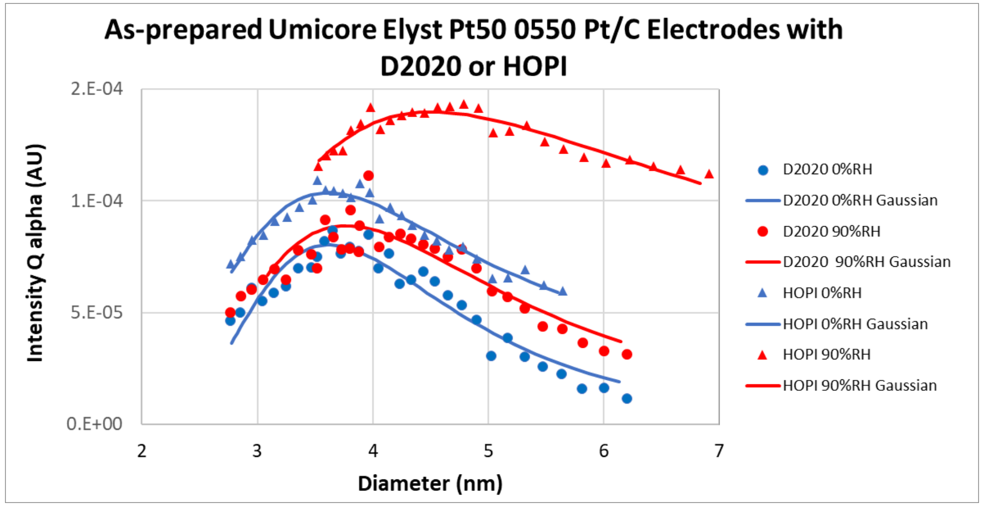


Harsh Srivastav, Adam Z. Weber, and Clayton J. Radke, (in preparation)

# Evaluation of Ink Constituents on Catalyst-Layer Structure

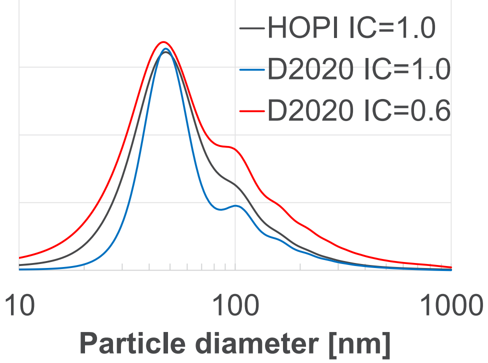
- Developed capability to resolve size distribution of ionomer in electrodes using RH-controlled SAXS
  - Size distribution of dry D2020 and HOPI in annealed Pt/C electrodes similar
  - High RH atmosphere increases size of HOPI more than that of D2020

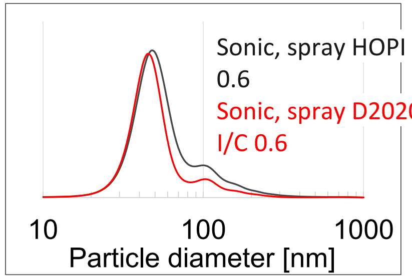
Collaboration with Arnaud Morin, CEA, Grenoble

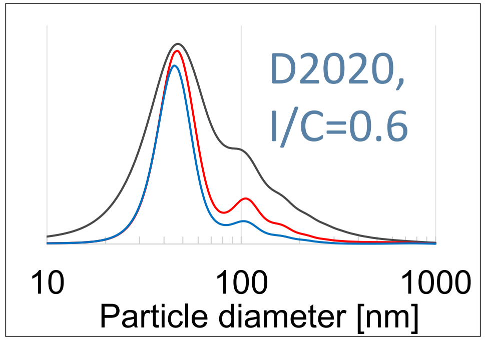


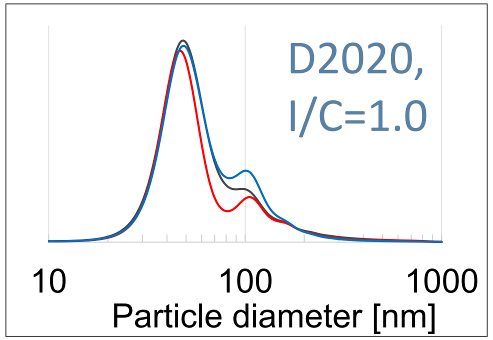

Ball-mill, rod; ball-mill, spray; ultra sonic, spray

7 nPA:3 water



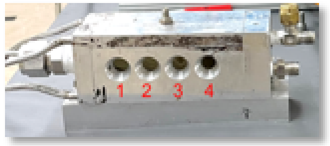

- Determined effects of HOPI vs D2020, I/C, and mixing /coating method on aggregate size distribution
  - Pt/C catalyst agglomerate/aggregate size larger with HOPI vs D2020
  - Higher D2020:carbon ratio yields smaller Pt/C agglomerates/aggregates and narrower distribution
  - Spray-coating results in smaller agglomerates/ aggregates vs ball-milling

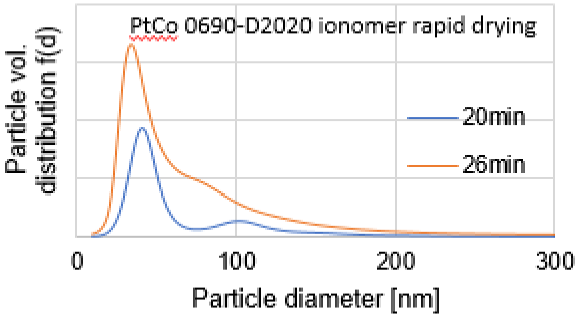
# Impact of Solvent Removal Rate on Catalyst-Layer Structure

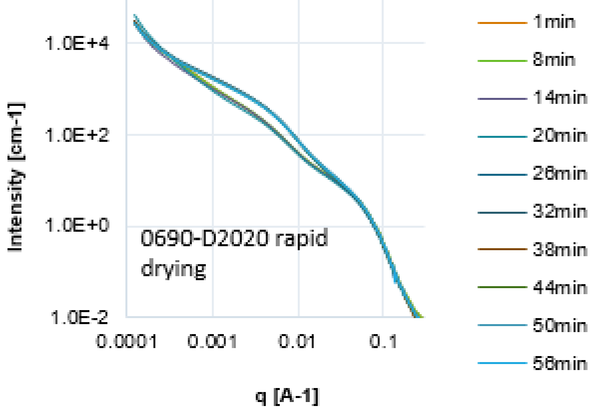
- $H_2$/Air performance has been correlated with agglomerate / aggregate size in electrodes in past work with Pt/C and PtCo/C catalysts*
- Smaller sizes result in lower $R_{NF}$ and higher current densities at <0.9 V
- Many ink composition, ink processing, and casting variables can be used to control agglomerate dispersion
- Decreasing solvent removal rate from catalyst-ionomer inks with either HOPI or Nafion ionomer results in smaller agglomerates in electrode layer

- Tracking evolution of ink structure during solvent removal and resulting electrode structure using in situ ultra-small angle X-ray scattering (USAXS)
- Studying effect of catalyst type (Pt/C, PtCo/C), ionomer type, solvent composition, and solvent removal rate

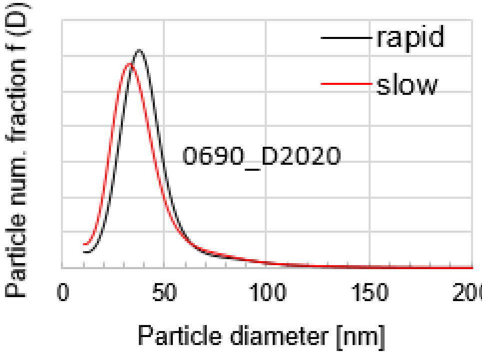
Environmental chamber for USAXS during solvent removal from catalyst-ionomer inks

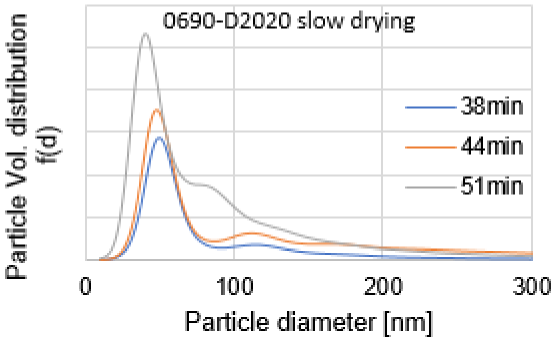


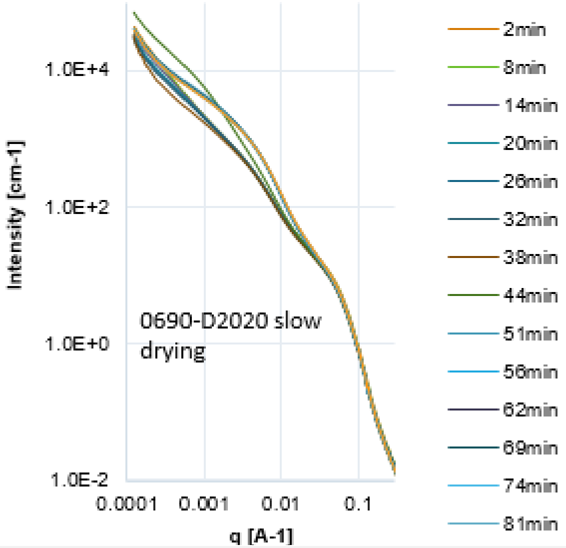




After complete removal of solvent



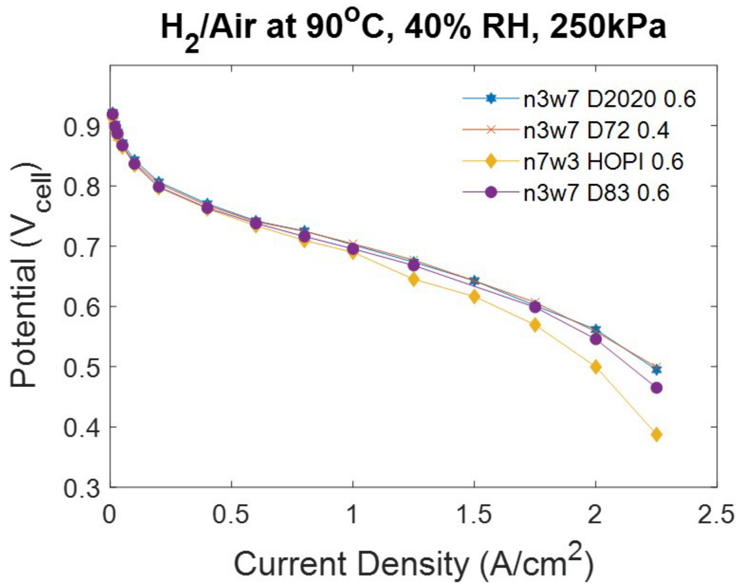



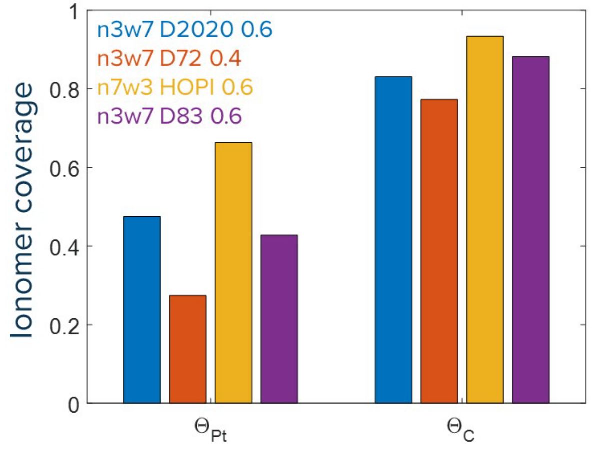



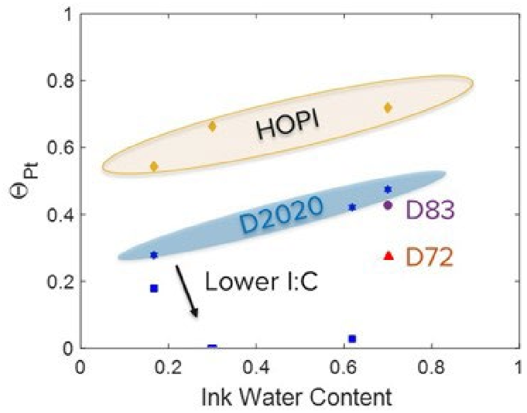


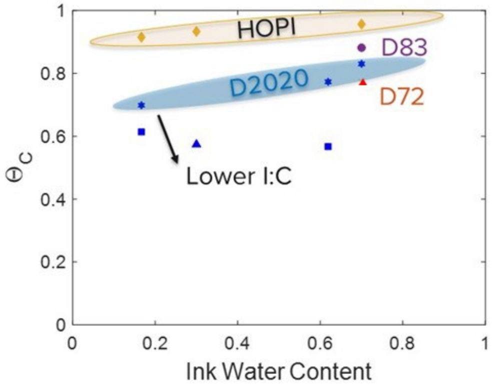

Van Cleve et al., J. Power Sources, 482 (2021) 228889

# High Oxygen Permeable Ionomer (HOPI)



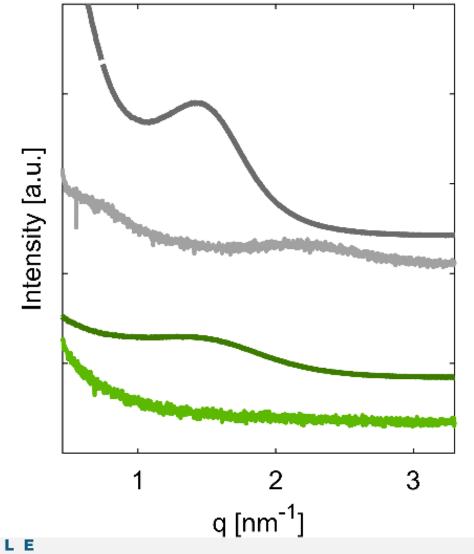



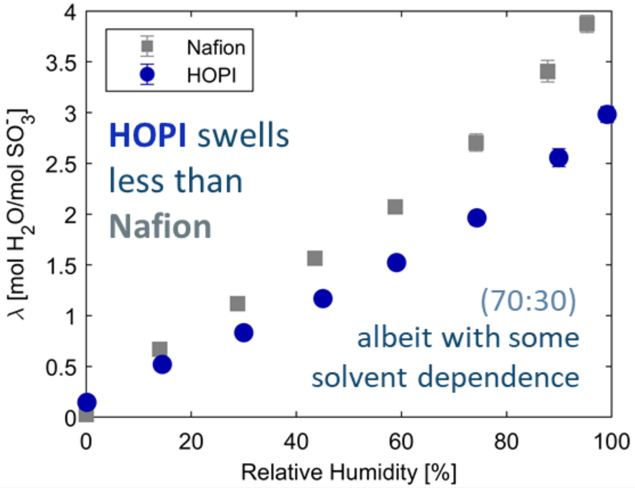



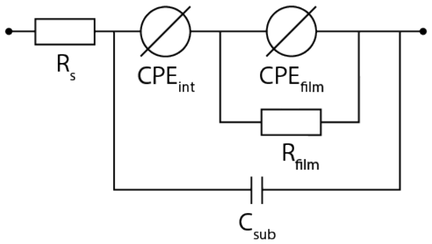




- **Compared to Nafion, HOPI demonstrates**
  - Weaker adsorption to Pt
  - Less water uptake (reduced swelling)
  - Less phase separation (more disordered)
  - Greater conductivity at same water volume fraction

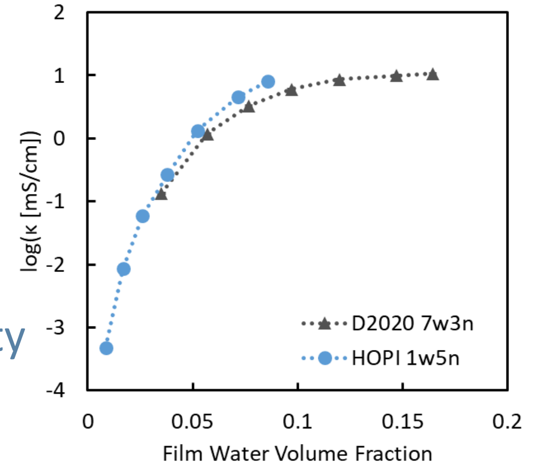
**(GI)SAXS data:**
HOPI ionomer thin film shows weaker nano-s phase separation owing to its bulkier groups



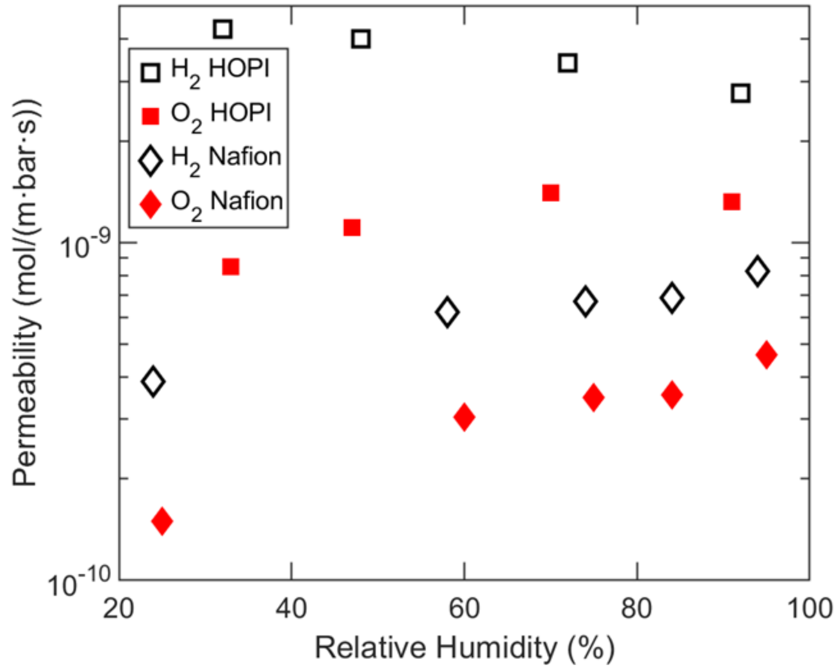



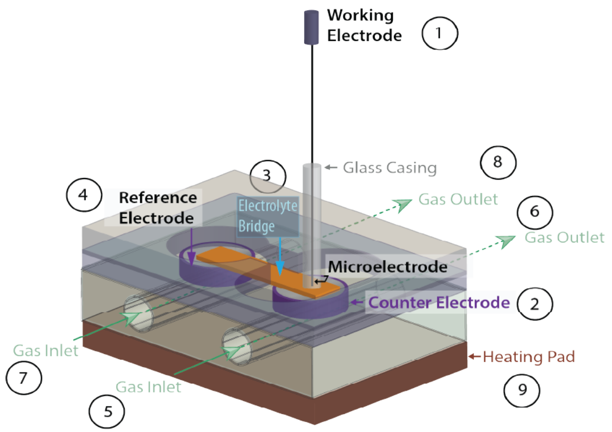

Thin Film Conductivity

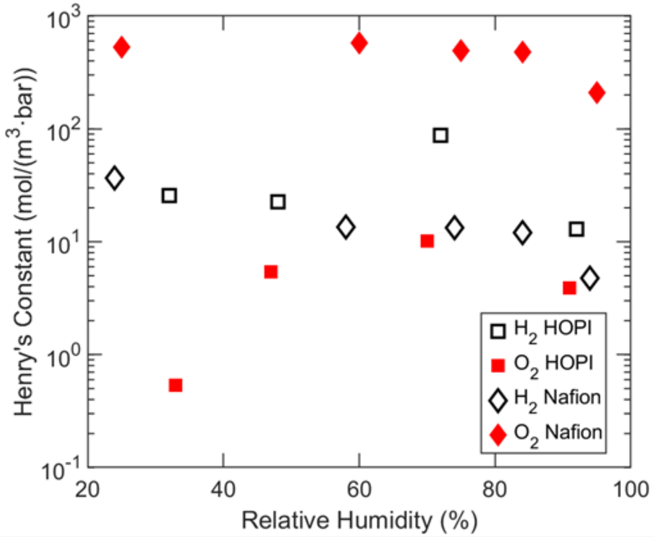


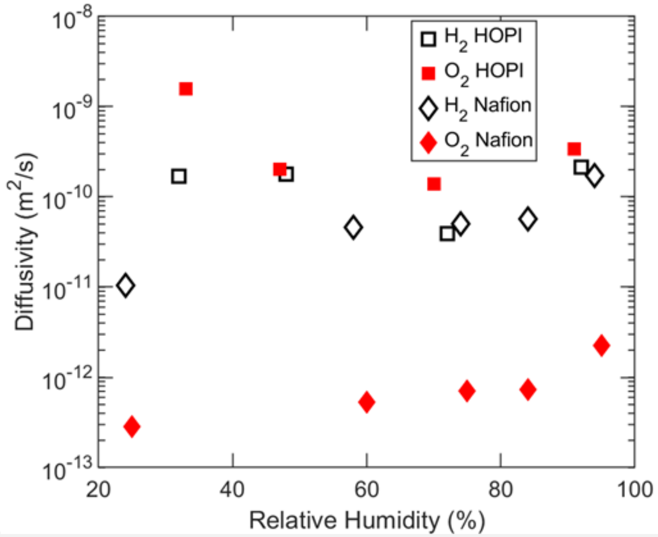
# HOPI Membrane Gas Properties

- HOPI membrane demonstrates greater diffusivity and permeability of both gases compared to Nafion
  - Hydrogen solubility is greater than Nafion 211 while oxygen solubility is lower
  - All values weakly depend on water content, perhaps due to more amorphous structure of HOPI compared to Nafion 211



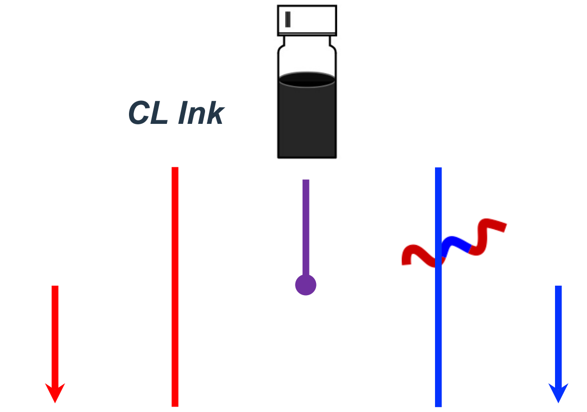



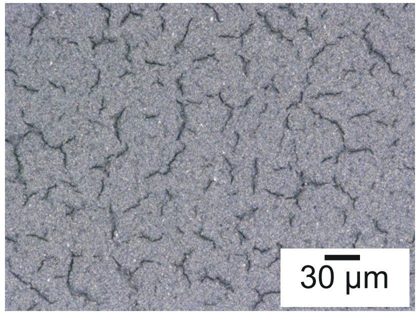



# Mitigation of Catalyst Layer Cracks with Additives

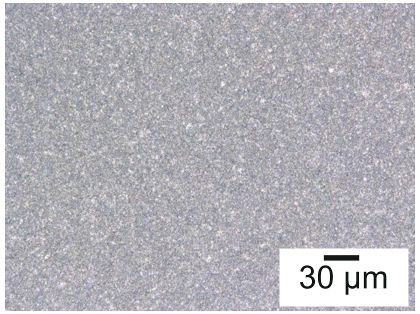
| Catalyst | Ionomer | Pt Loading |
|---|---|---|
| Pt/HSC | Nafion | ~0.25 mg/cm$^2$ |



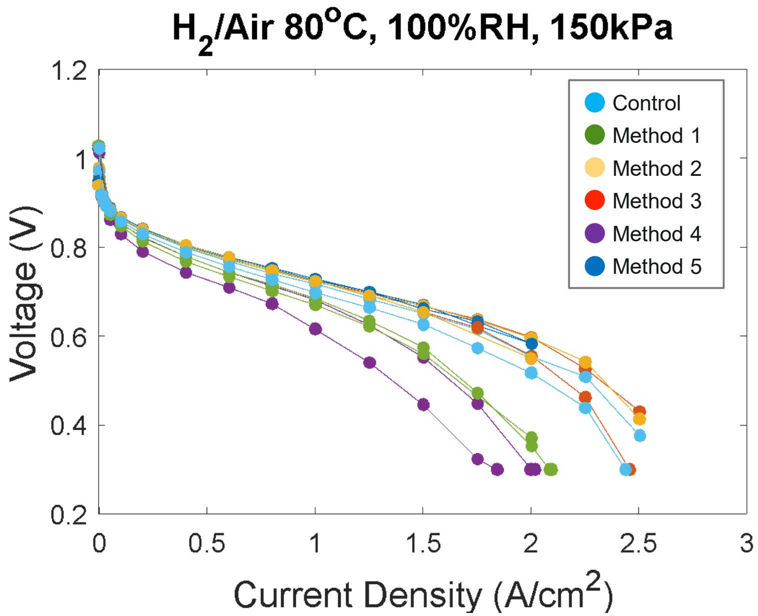

Control

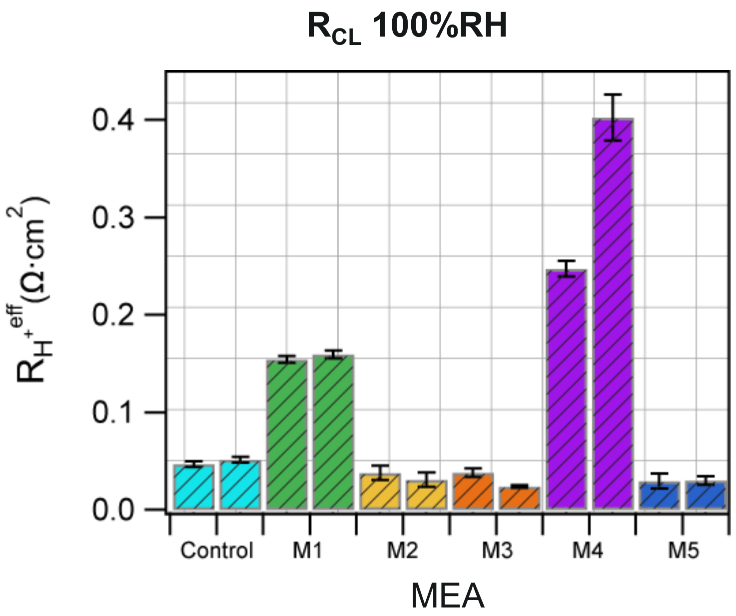



Additive-loaded



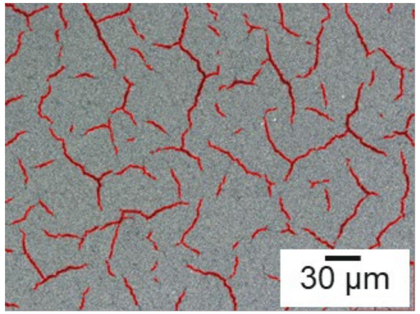

**Problem: Fabrication of thick CLs may form cracks, which could be detrimental to fuel cell durability**



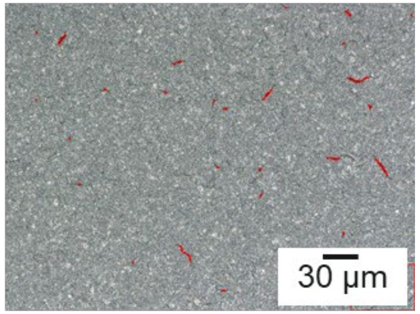




**Method of addition may improve initial electrochemical performance.**

Control (HOPI)



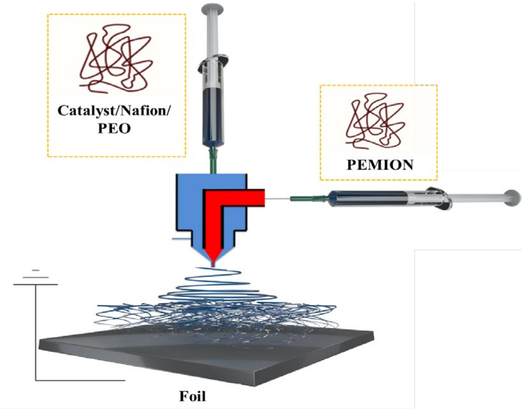

Additive-loaded



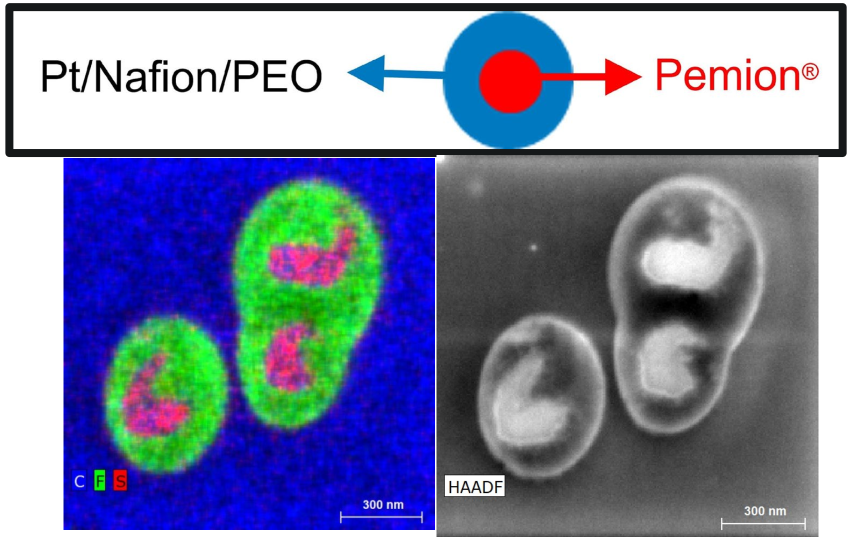

**Formulation optimization technology can be transferred to other CL systems (i.e., High Oxygen Permeability Ionomer)**


Baez-Cotto, Carlos; Mauger, Scott; Ulsh, Michael. Record Of Invention filed.

# Coaxial Electrospun Electrode Development

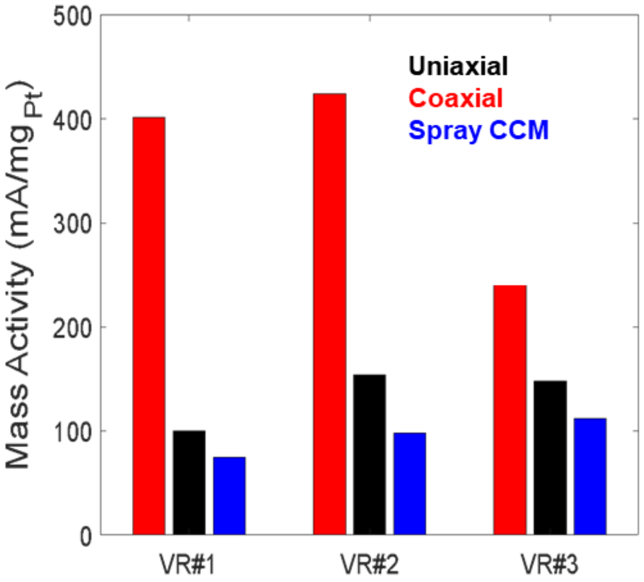


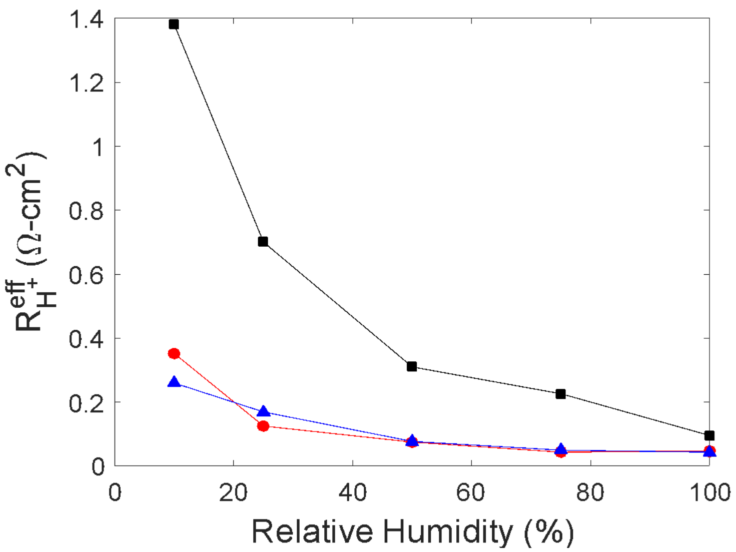

Co-axial Electrospinning

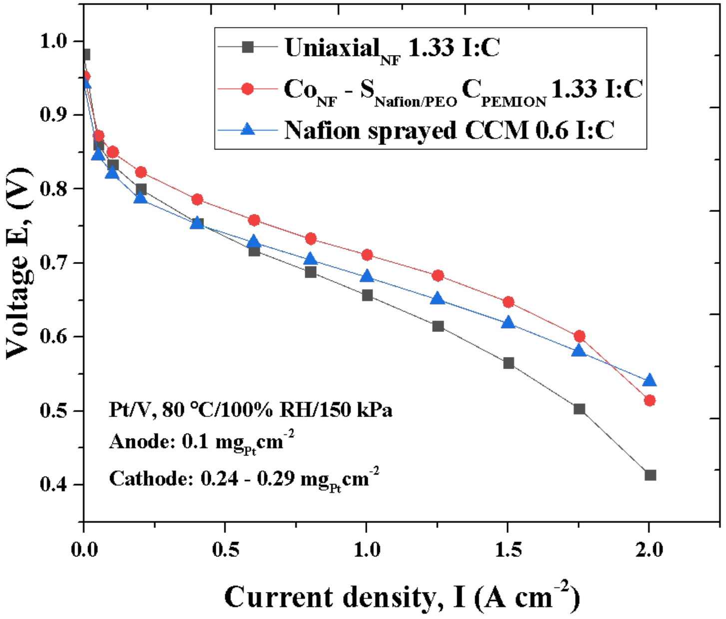



Coaxial Fiber Cross-section

- EDS maps of co-axial fibers illustrate core-shell fibers
- Co-axial nanofiber electrodes exhibit higher mass activity
- Cathode electrode ionic resistance is decreased with co-axial approach relative to uniaxial
- Net benefit to performance



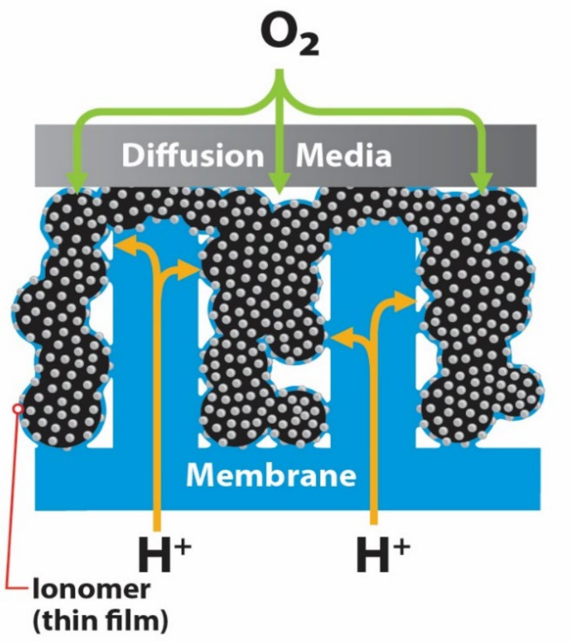



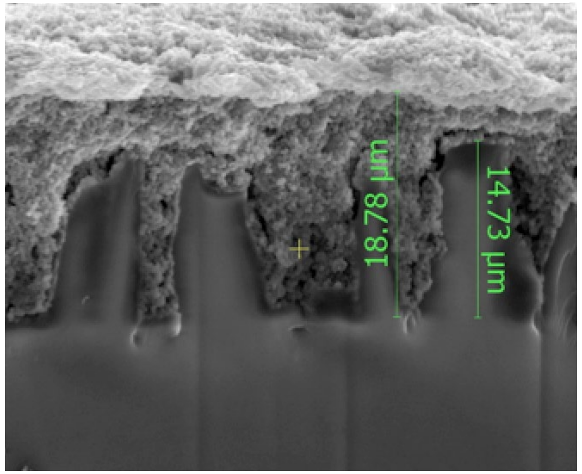





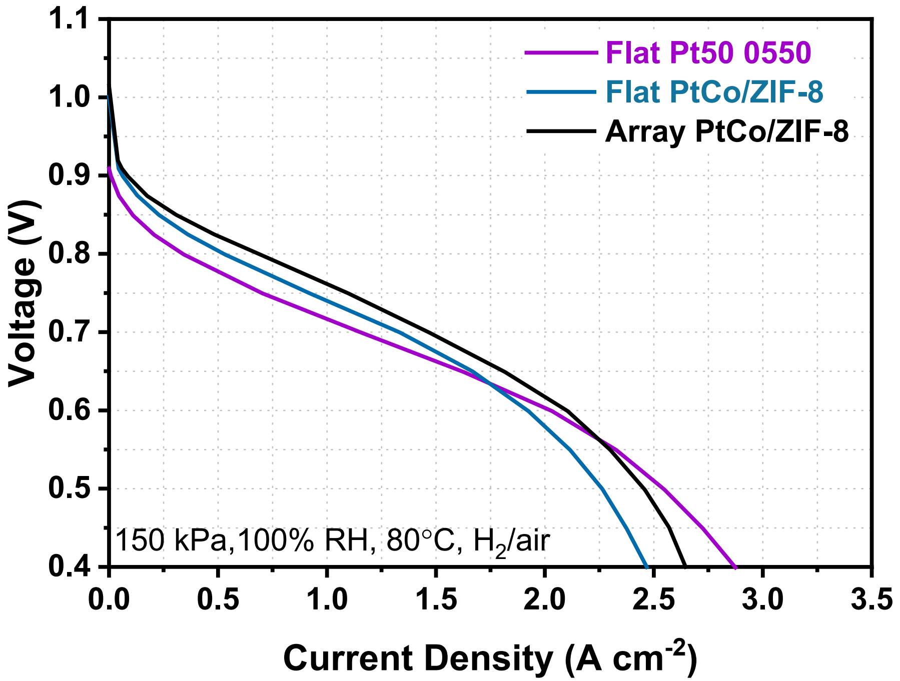

Lou, K.; Venugopalan, G.; Neyerlin, K.C. (In preparation)

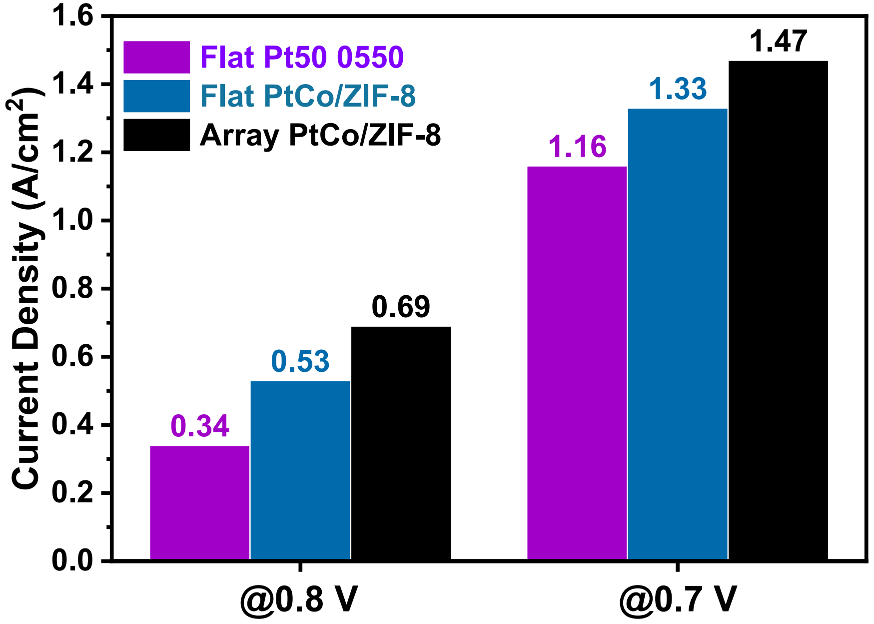
# Array Electrode: Advanced Catalyst Incorporation




Array Electrode

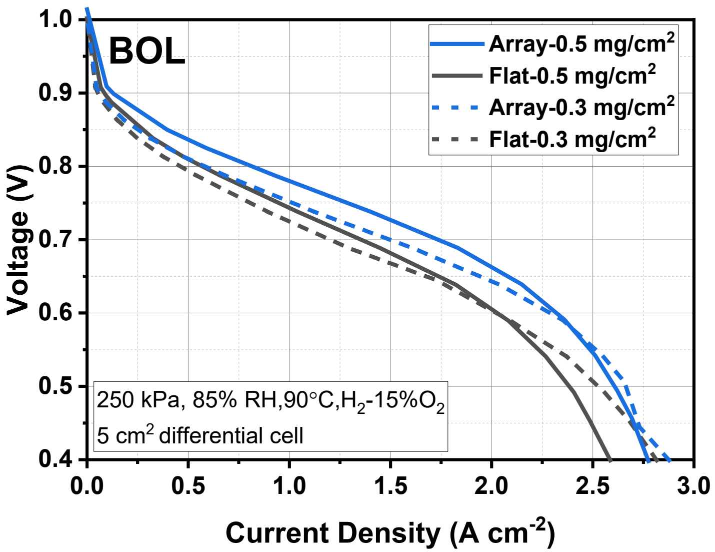


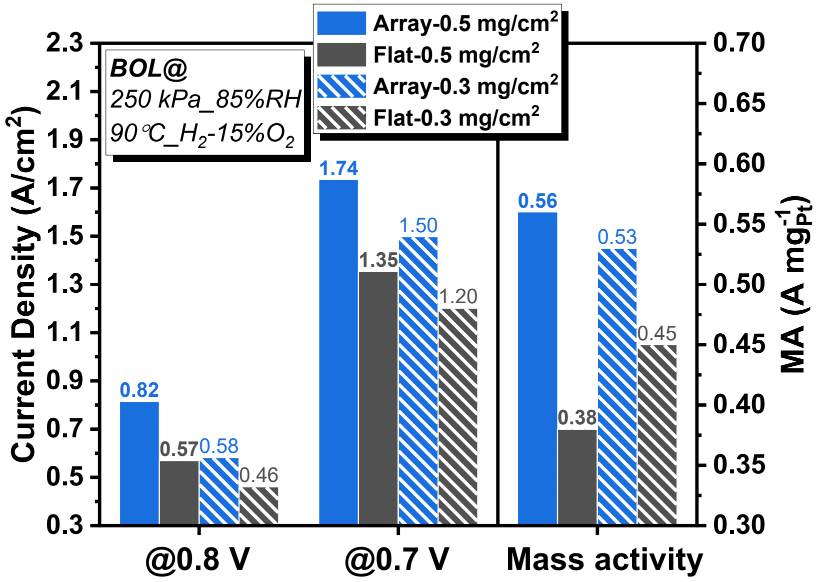



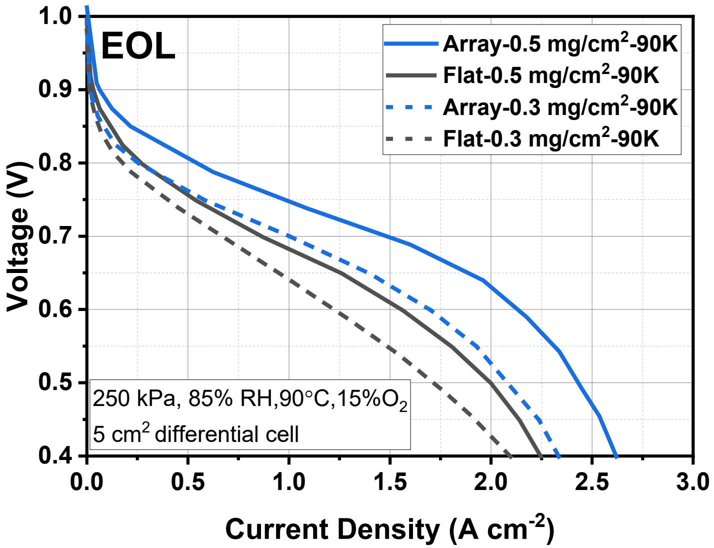

- Advanced catalysts can be integrated in array electrodes for synergistic enhancements
- Combining $L1_0$-PtCo/ZIF-8 catalyst and array electrode provides 2X performance at 0.8 V compared to baseline

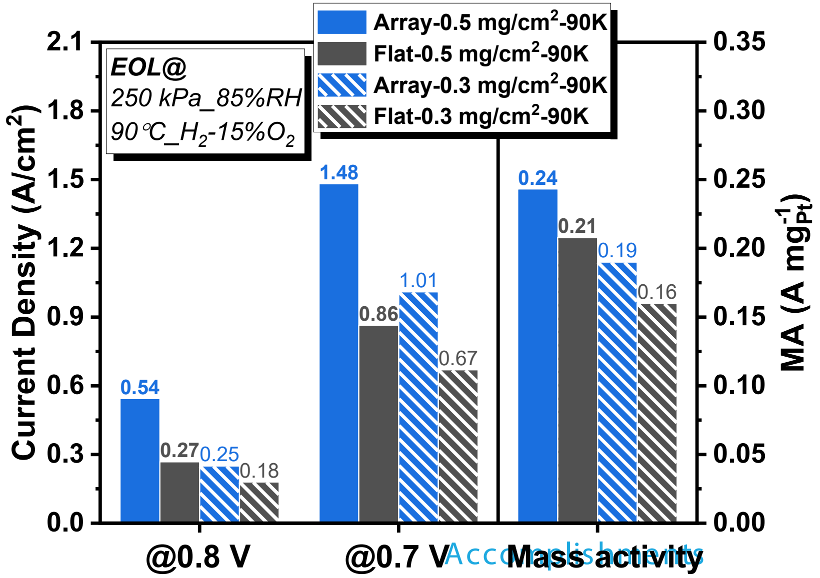
# Array Electrode: Enable Higher Loading

- Array electrodes provide fast transport even in thick electrodes, enabling higher loading and higher durability
- Array electrode performance enhancement at 0.8 V: 26% at 0.3 mg/cm², and 44% at 0.5 mg/cm²
- After 90K catalyst AST, array electrode with 0.5 mgPt/cm² loading provides 1.48 A/cm² at 0.7 V with only 15% degradation, vs. 36% for a flat electrode



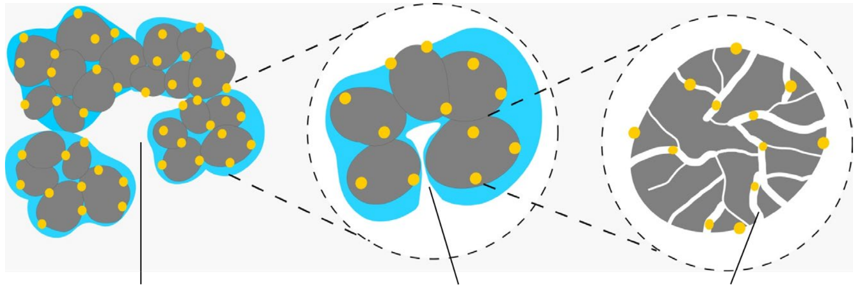

Accomplishments

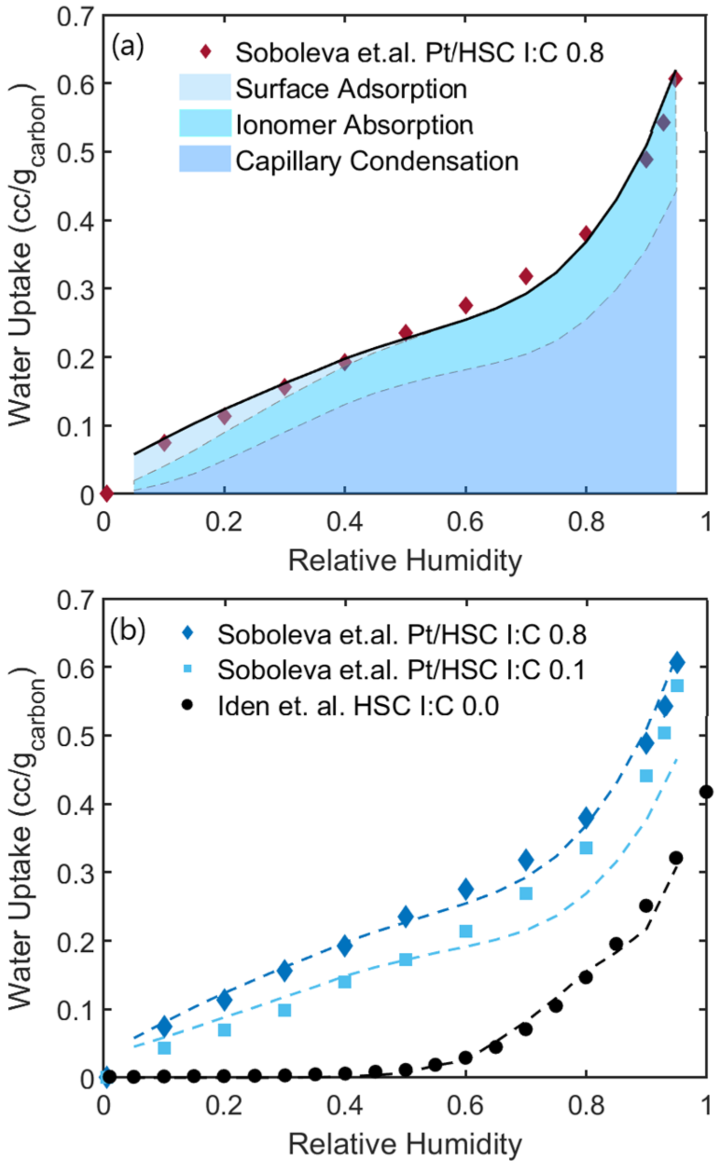

K. Li, X. Wang, S. Komini Babu, J. S. Spendelow, in preparation

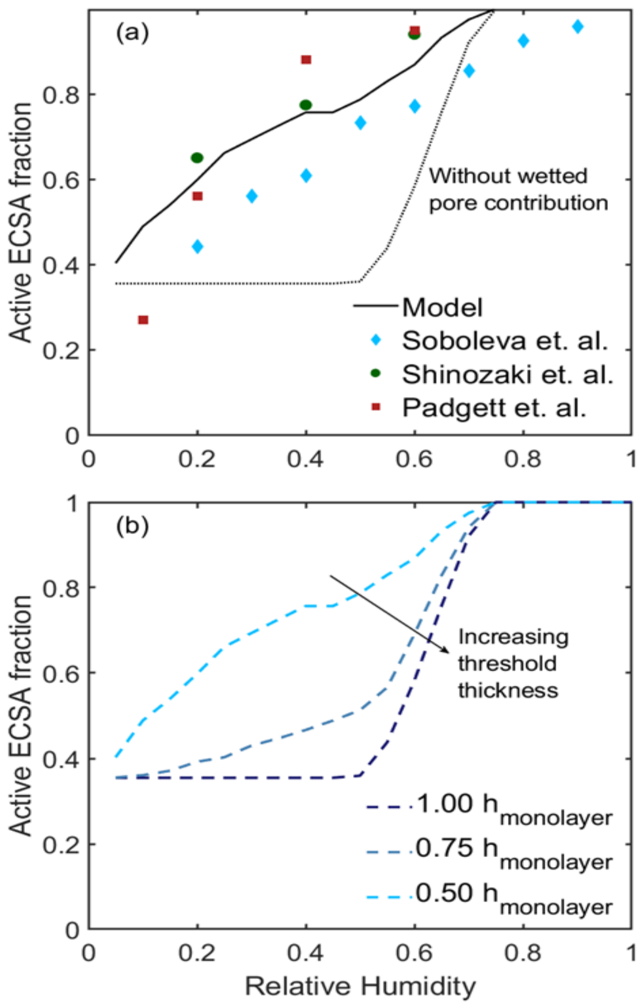
# Modeling HSC Pores

## Water uptake

- Model water condensation via multilayer Langmuir adsorption with capillary condensation
  - ↳ Enables prediction of HSC connectivity and operation at various humidity
- Water uptake dominated by capillary condensation and pore hydrophobicity but surface transport in wetted conditions significantly impacts ECSA




**Fraction of Pt particles electrochemically active versus RH**

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

# Durability

# M2FCT MEA AST

**Heavy Duty MEA Protocol and Metrics**

| | | |
|---|---|---|
| **Cycle** | Square wave between 0.675 V (30s) and 0.925 V (30s); Single-cell 50 $cm^2$ [a] | |
| **Number** | 500 hours or 30,000 cycles | |
| **Cycle time** | 1 minute | |
| **Temperature** | 90°C | |
| **Relative Humidity** | Anode/Cathode: 50%/50% | |
| **Fuel/Oxidant** | Hydrogen/Air ($H_2$ at 1000 sccm and Air at 2500 sccm for a 50 $cm^2$ cell | |
| **Pressure** | 250 kPa | |
| | | |
| **Metric** | **Frequency** | **Target** |
| **Catalytic Mass Activity**[b] | At BOT[c], after 100h, 200h, 300h, 400h, and 500h | ≤ 60% loss of initial catalytic activity |
| **ECSA/Cyclic Voltammetry**[d] | At BOT, after 100h, 200h, 300h, 400h, and 500h | ≤ 40% loss of initial area |
| **Hydrogen Crossover**[e] | At BOT, after 100h, 200h, 300h, 400h, and 500h | < 2mA/$cm^2$ |
| **Polarization curve**[f] | At BOT, after 100h, 200h, 300h, 400h, and 500h | 2.5 kW/$g_{PGM}$ EOT PGM utilization at 0.7 V[g] |

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

b. Mass activity in A/mg @ 150 kPa abs backpressure at 900 mV iR-corrected on H2/O2, 100% RH, 80°C, anode stoichiometry 2; cathode stoichiometry 9.5 (as per Gasteiger et al. Applied Catalysis B: Environmental, 56 (2005) 9-35). Measured ORR current should be corrected for H2 crossover and shorting.

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

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

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

f. $H_2$/Air, 250 kPa abs backpressure, 90°C, 40% RH, cathode stoichiometry 1.5, anode stoichiometry 2; Recommend taking pol curves from high to low current densities at 0.01, 0.02, 0.03, 0.05, 0.1, 0.2, 0.4, 0.6, 0.8, 1, 1.25, 1.5, 1.5 and 2 A/cm2, 240s hold time at each data point

g. $H_2$/Air, 250 kPa abs backpressure, 90°C, 40% RH, cathode stoichiometry 1.5 (300 sccm minimum flow), anode stoichiometry 2 (100 sccm minimum flow); 0.25(c)/0.05(a) mg/$cm^2$ maximum PGM loading

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

# Cell Design for ASTs

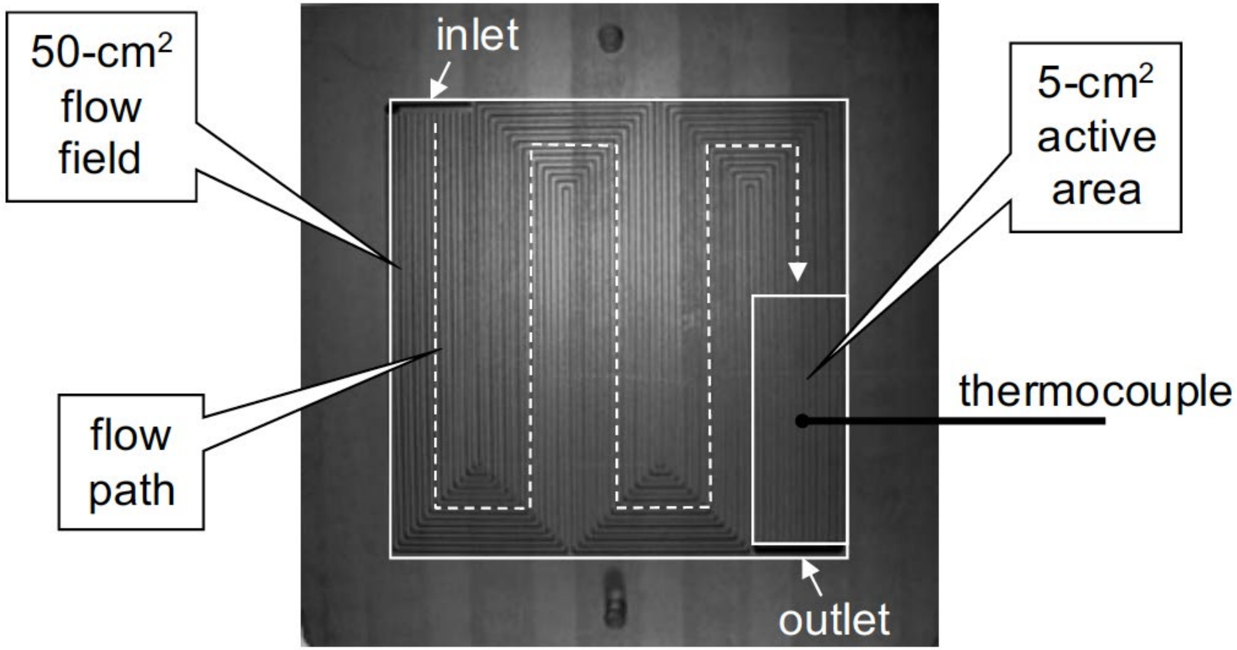


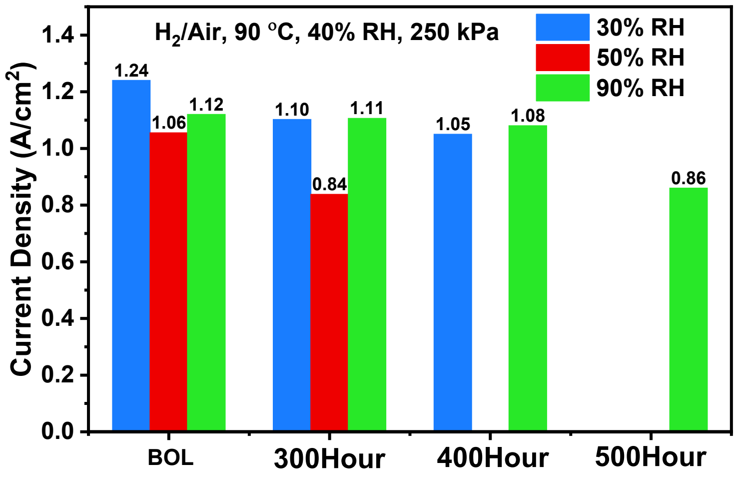

**Figure 1.** The serpentine flow field (50 cm$^2$) used for limiting current experiments, masked down with gaskets to a 5 cm$^2$ active area near the outlet.

- Differential Cell testing:
  - Use masked off 5 cm$^2$ MEAs in 14-channel serpentine 50 cm$^2$ hardware
  - Straight channels and high flows to create differential conditions
  - Pol curves obtained under different conditions to simulate integral cell performance (90°C, 85% RH, 15% $O_2$)
- Integral Cell testing:
  - Use the entire 50 cm$^2$ hardware for integral cell testing
  - Counter-flow conditions for more realistic water balance
  - Stoichiometric flows for more realistic pol curves under various temperature and RH conditions

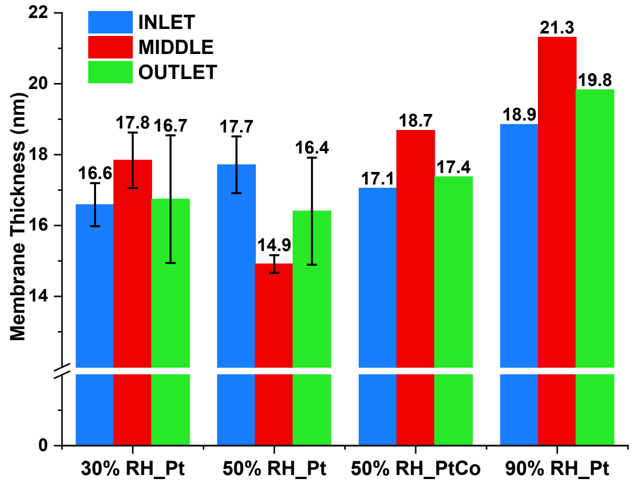

J. Electrochem. Soc., 156 (9) B991-B1003 (2009)

# M2FCT MEA AST

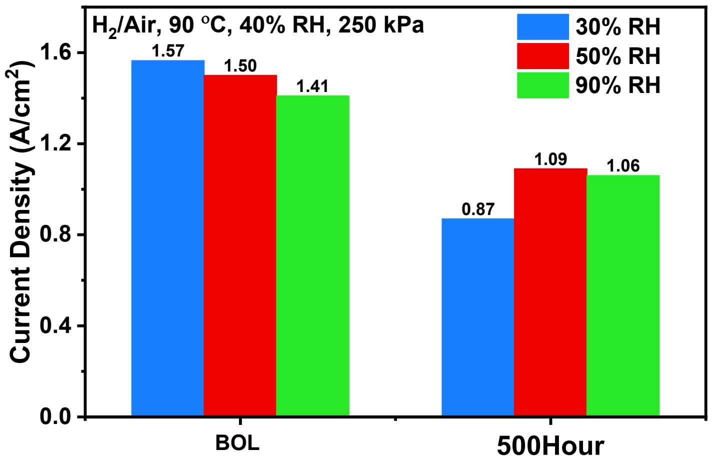
## Baseline Pt50 0550 catalyst fails to meet targets (1.07 A/cm2 @ 0.7V) with MEA protocol



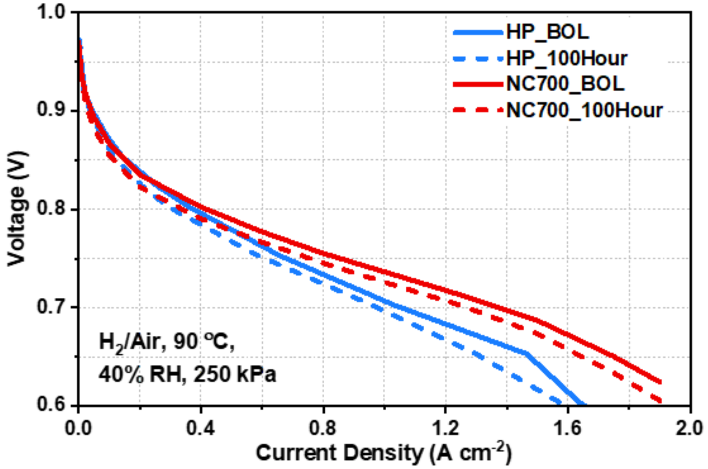

## Nafion® HP membrane shows significant thinning (≈ 15%) with MEA protocol



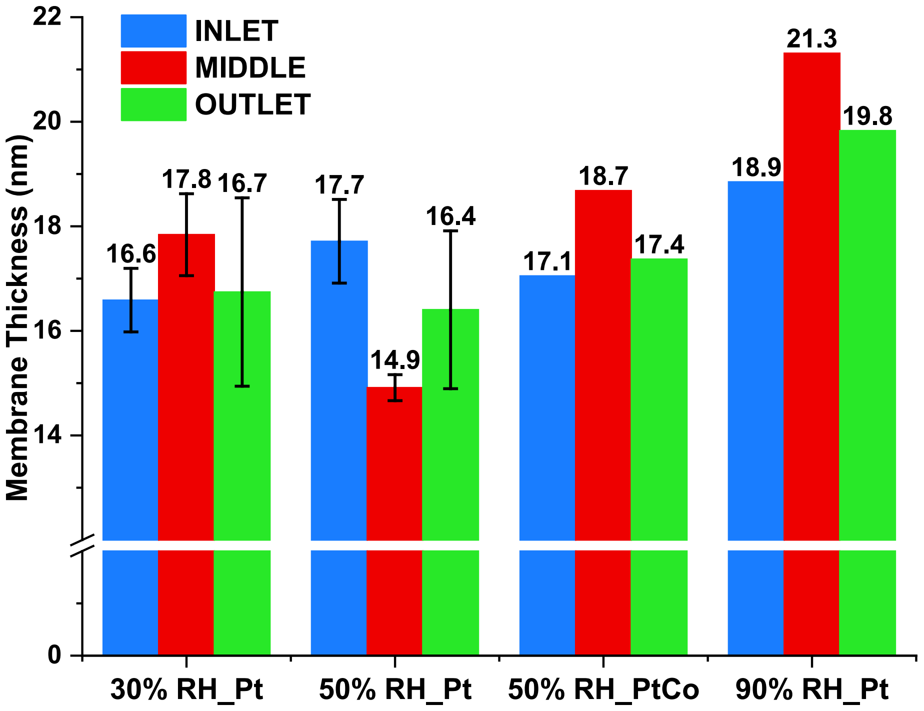

## Pt30 0690 PtCo alloy catalyst meets target at higher loadings (0.28mg/cm²)

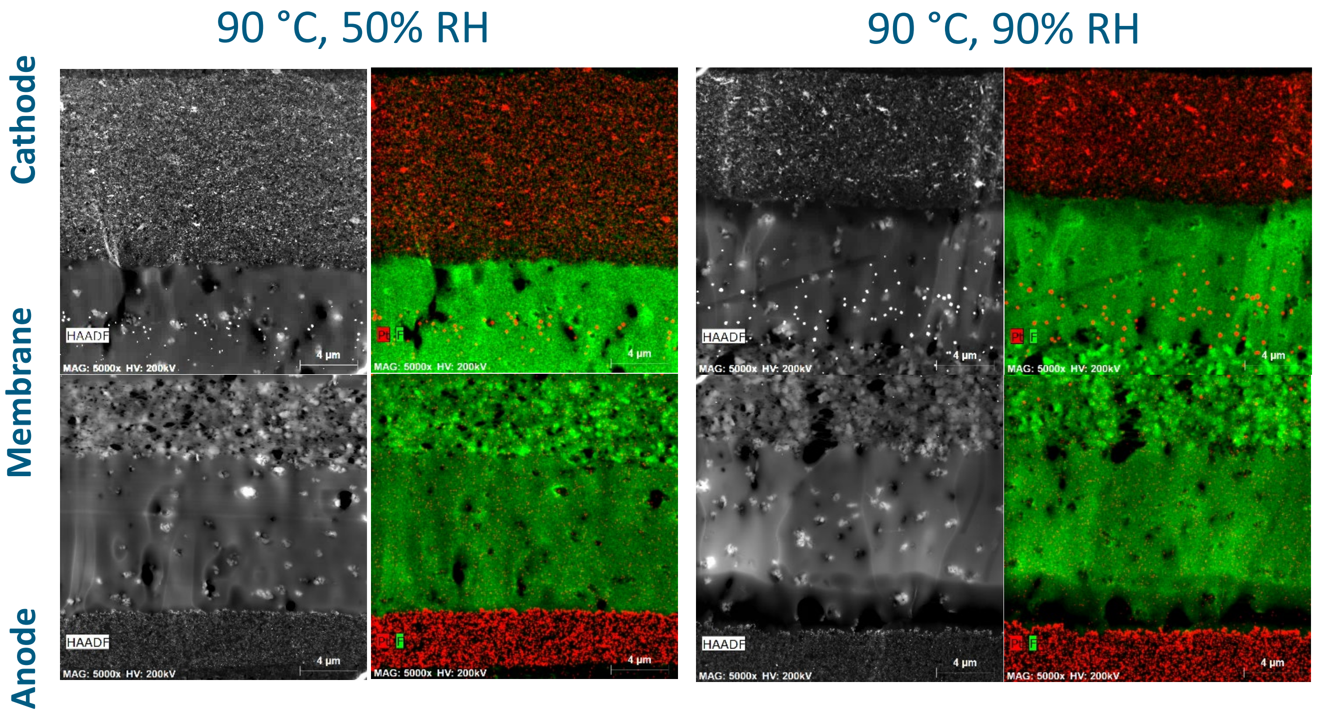


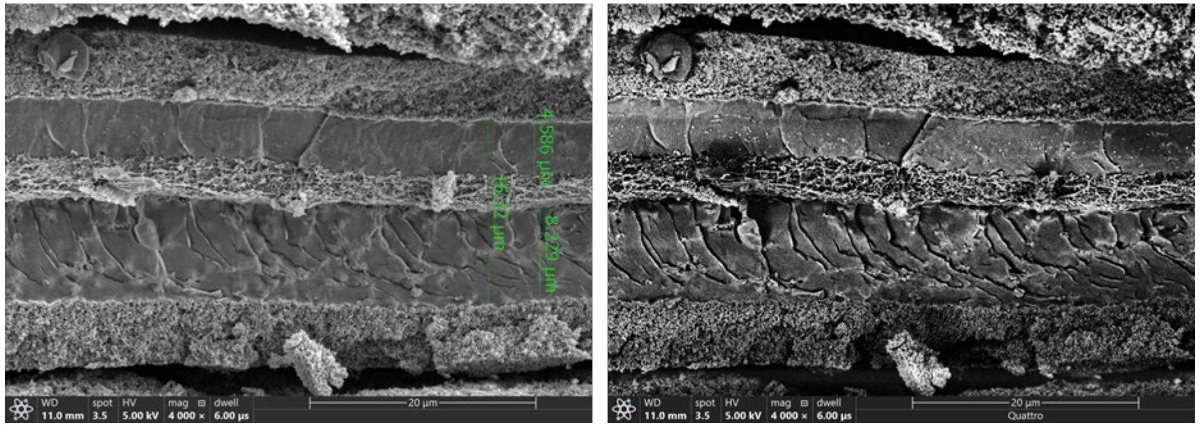

## Nafion® NC700 shows better BOL performance Durability testing in progress



# M2FCT MEA AST

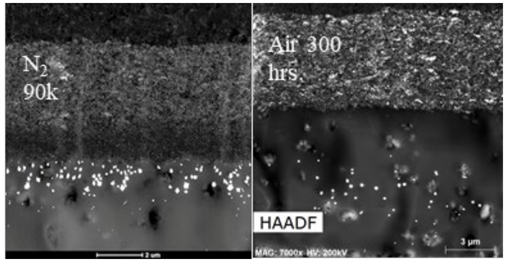


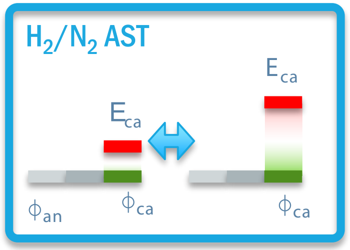



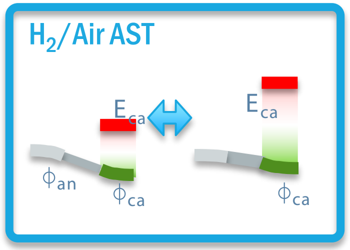




- Membrane thinning for both 30% and 50% RH is more obvious than 90%RH
- Membrane thinning is more at the cathode side and at the inlets and outlets
- 50% RH shows similar Pt band location

# Comparison of Catalyst Degradation – Potential and Environment

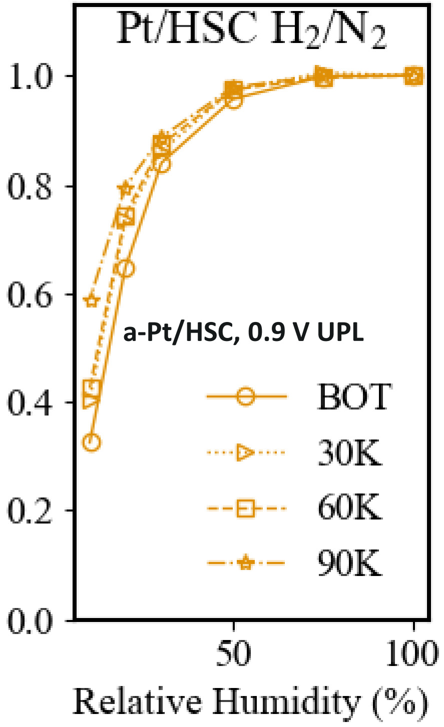


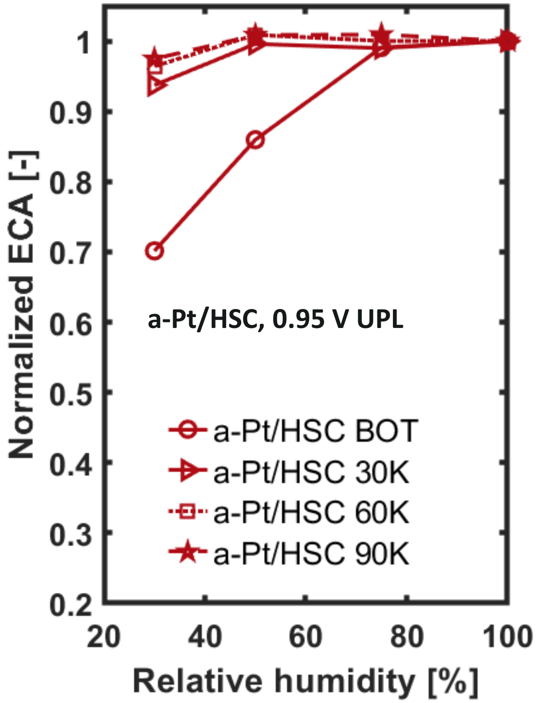




Reference: [3] Implementation of Automated Electron Microscopy, Haoran Yu, Dave Cullen, Michael Zachman, and Shawn Reeves

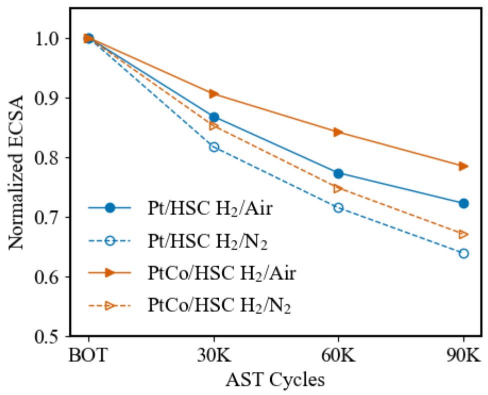
## Catalyst accessibility, Upper potential limit effect

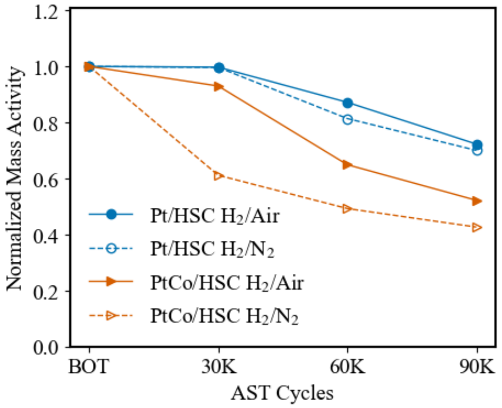


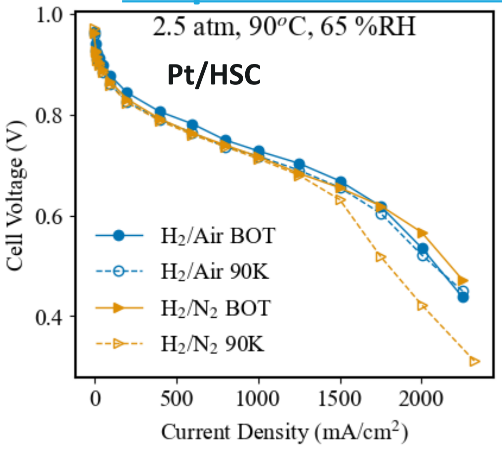

## Mass activity and ECSA loss, $H_2/N_2$ vs. $H_2$/Air AST

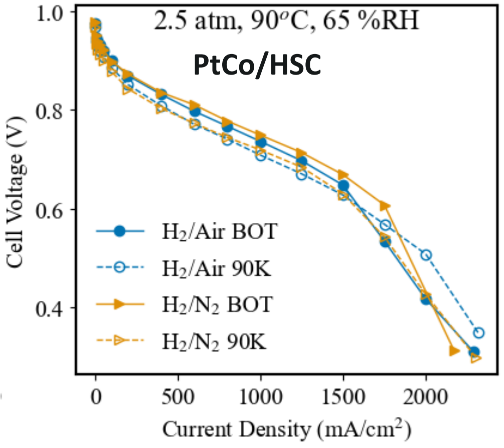
- AST cycle; H2/N2 or H2/Air (1 slpm and 2.5 slpm) with 0.675 – 0.9 V cycling




## Air pol curves $H_2/N_2$ vs. $H_2$/Air AST



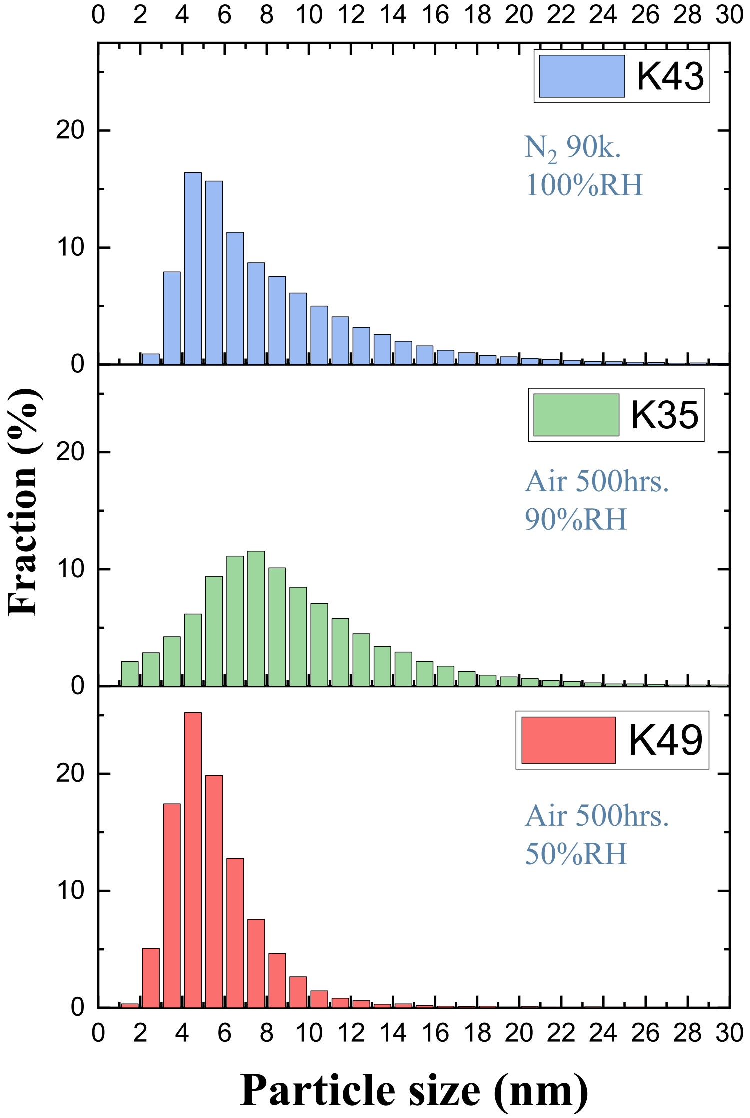




- With 0.9 V UPL, both Pt/HSC and PtCo/HSC samples show less catalyst dissolution compared to 0.95 V UPL
- Both PtCo/HSC and Pt/HSC sample degradation under 0.675 – 0.9 V AST potential window is faster in $H_2/N_2$ environment compared to $H_2$/Air environment.

# Catalyst particle coarsening

## 50%RH shows less coarsening but more Co dissolution than 90%RH

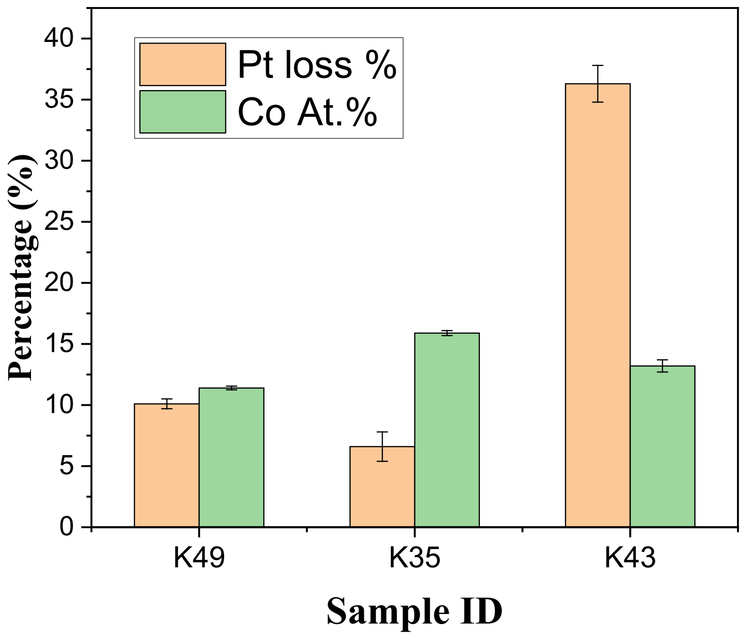
| **Catalyst** | **PtCo/C** | **PtCo/C** | **PtCo/C** |
|---|---|---|---|
| **MEA ID** | **K49** | **K35** | **K43** |
| **Relative humidity (%RH)** | **50** | **90** | **100** |
| AST | Air 500hrs. | Air 500hrs. | $N_2$ 90k cycles |
| Analyzed area ($\mu m^2$) | 1.6 | 9.7 | 18.1 |
| Total number of particles analyzed | 7146 | 22,340 | 43,738 |
| **Median (nm)** | **5.1** | **8.2** | **6.8** |
| Percentage 25%-75% (nm) | 4.1-6.5 | 6.0-11.2 | 5.0-10.0 |
| No. Particles ≤10 nm (%) | 95 | 67 | 75 |
| No. Particles >10 nm (%) | 5 | 33 | 25 |
| Particle volume ≤10 nm (%) | 56 | 17 | 19 |
| Particle volume >10 nm (%) | 44 | 83 | 81 |
| Geometric Surface Area ($m^2/g_{Pt}$) | 32.8 | 21 | 22 |
| **Co at.% (EDS mapping of full electrode cross section)** | **11.4±0.2** | **15.7±0.2** | **13.2±0.5** |
| **Pt loss %** | **10.1±0.4** | **6.6±1.2** | **36.3±1.5** |

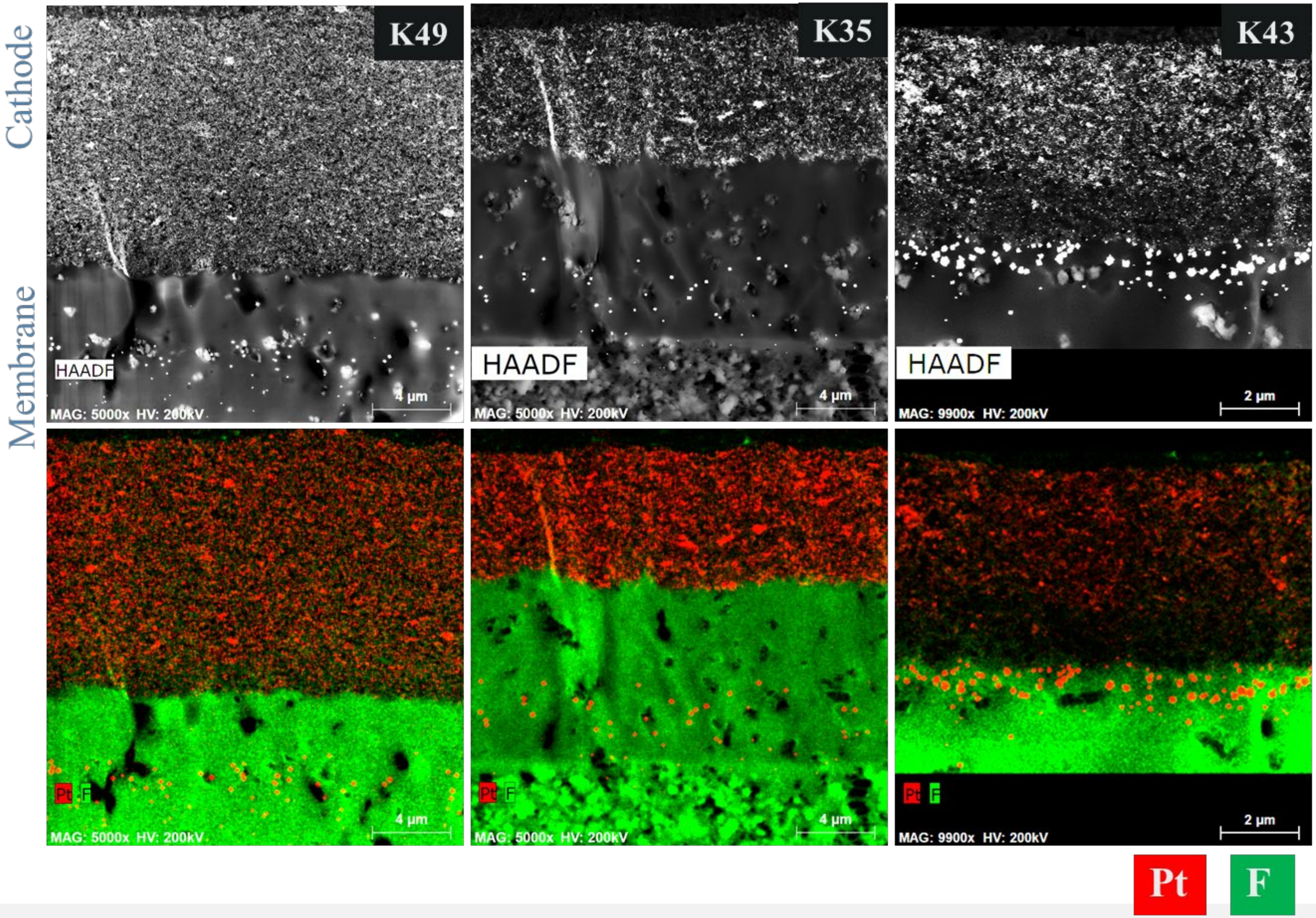


# Catalyst Dissolution and Migration

## 50%RH shows similar Pt band location and Pt loss% as 90%RH

**K49 – PtCo, 90$^o$C, 50%RH, Air 250 kPa.**
**K35 – PtCo, 90$^o$C, 90%RH, Air 250 kPa**
**K43 – PtCo, 80$^o$C, 100%RH, $N_2$ ambient P**

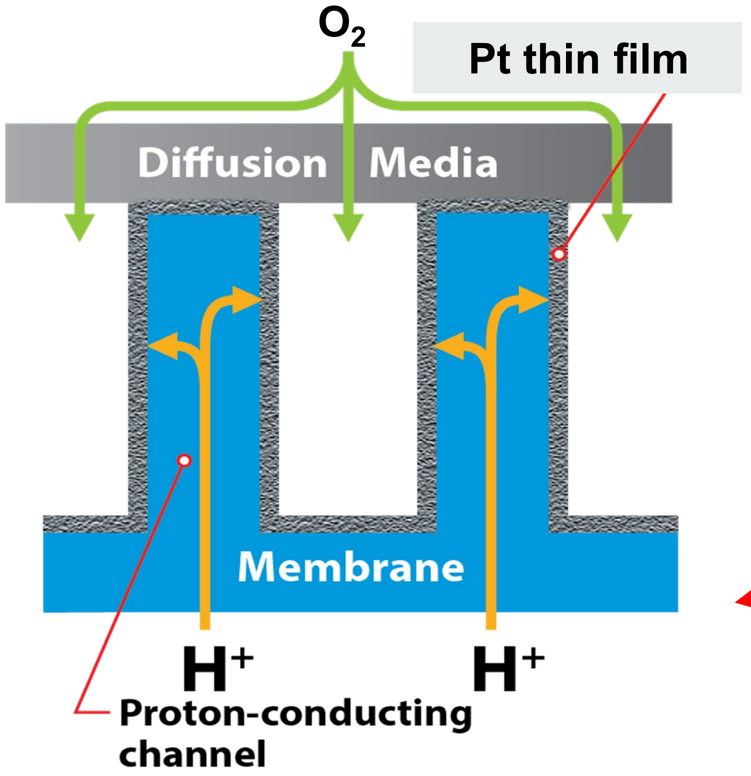


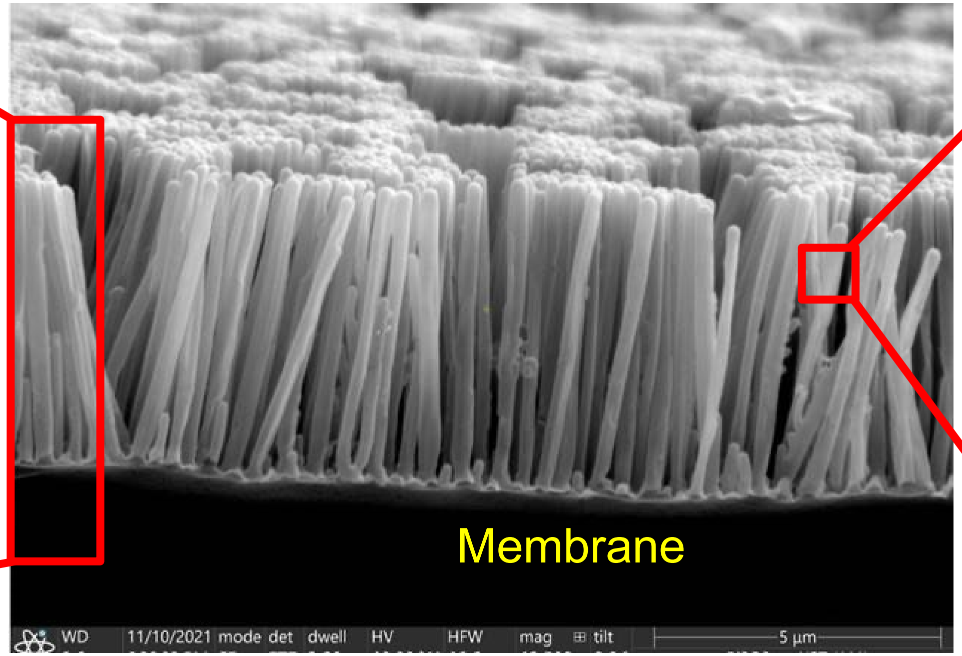



# Catalyst Thin-Film Electrodes

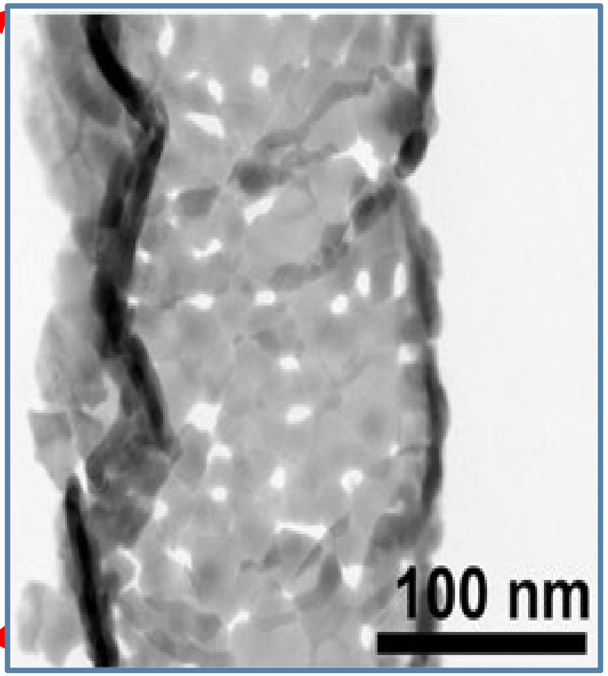
## Co-Axial Nanowire Electrode (CANE)

- Ionomer-free exterior: prevents catalyst poisoning and $R_{O2}$
- Ionomer nanowires: facile $H^+$ transport and high roughness
- **Thin-film:** reduced catalyst corrosion
- Carbon-free electrode: reduced support corrosion



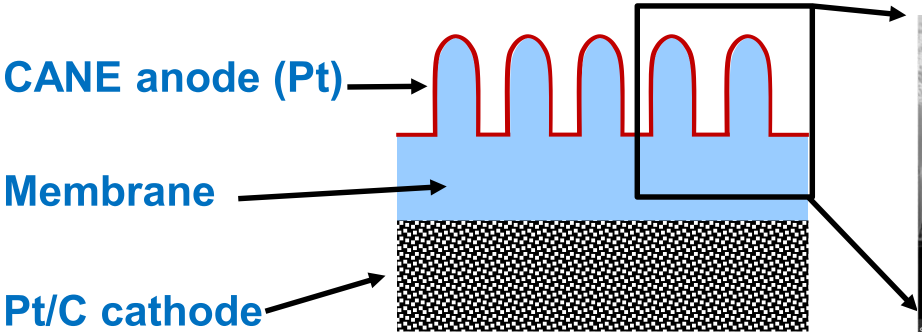

SEM image of CANE electrode



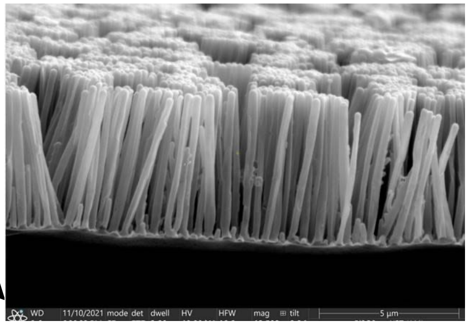

Cross section – TEM image

# Reversal Tolerant Anodes based on Thin Film Catalyst

- CANE consists of Pt thin film supported on high surface area Nafion nanowires, enabling carbon free electrodes
- Anode reversal test: drew 0.2 A/cm$^2$ from cell, switched from **$H_2$** to **$N_2$** at anode, monitored time to drop cell voltage to -2V.
- **CANE** anodes show **0%** performance loss at 0.7 V, while **Pt/C** anodes lose **86%** after a few reversals.
- **Thin film electrode architecture provides exceptional anode reversal tolerance**

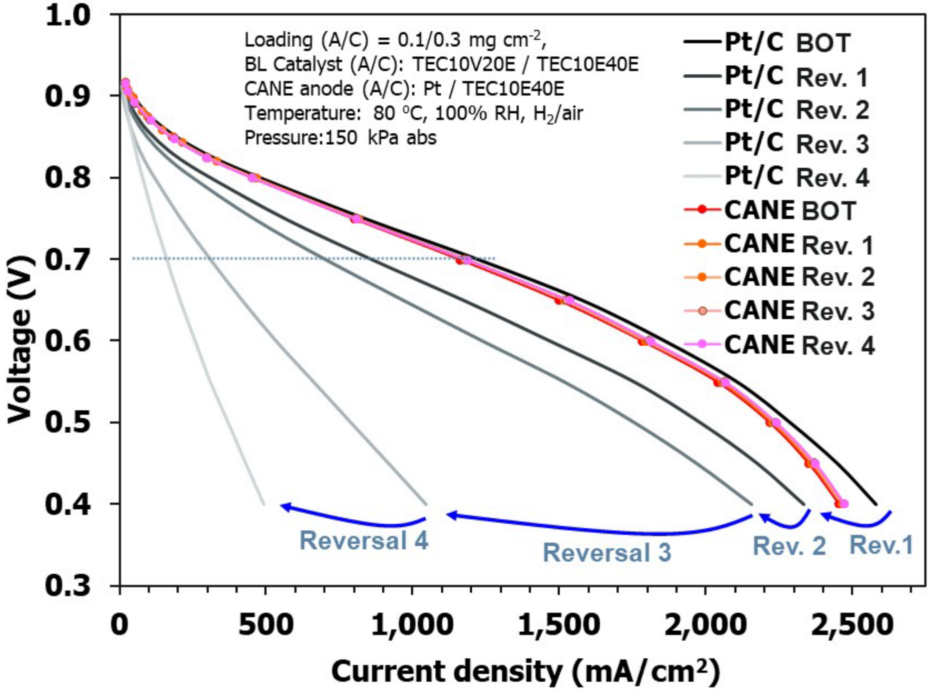



**Schematic of MEA with CANE anode**

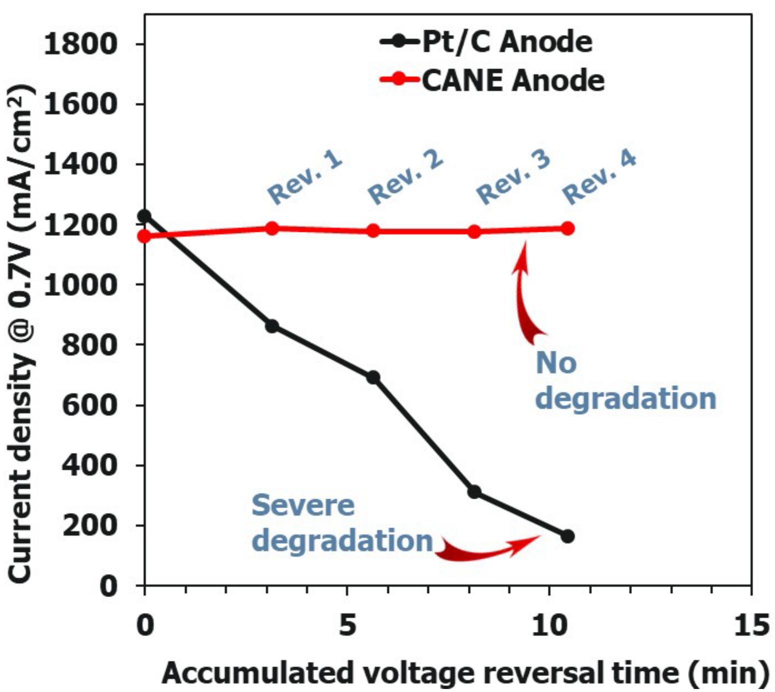


**SEM image of CANE**

**Fuel cell performance with anode reversal**



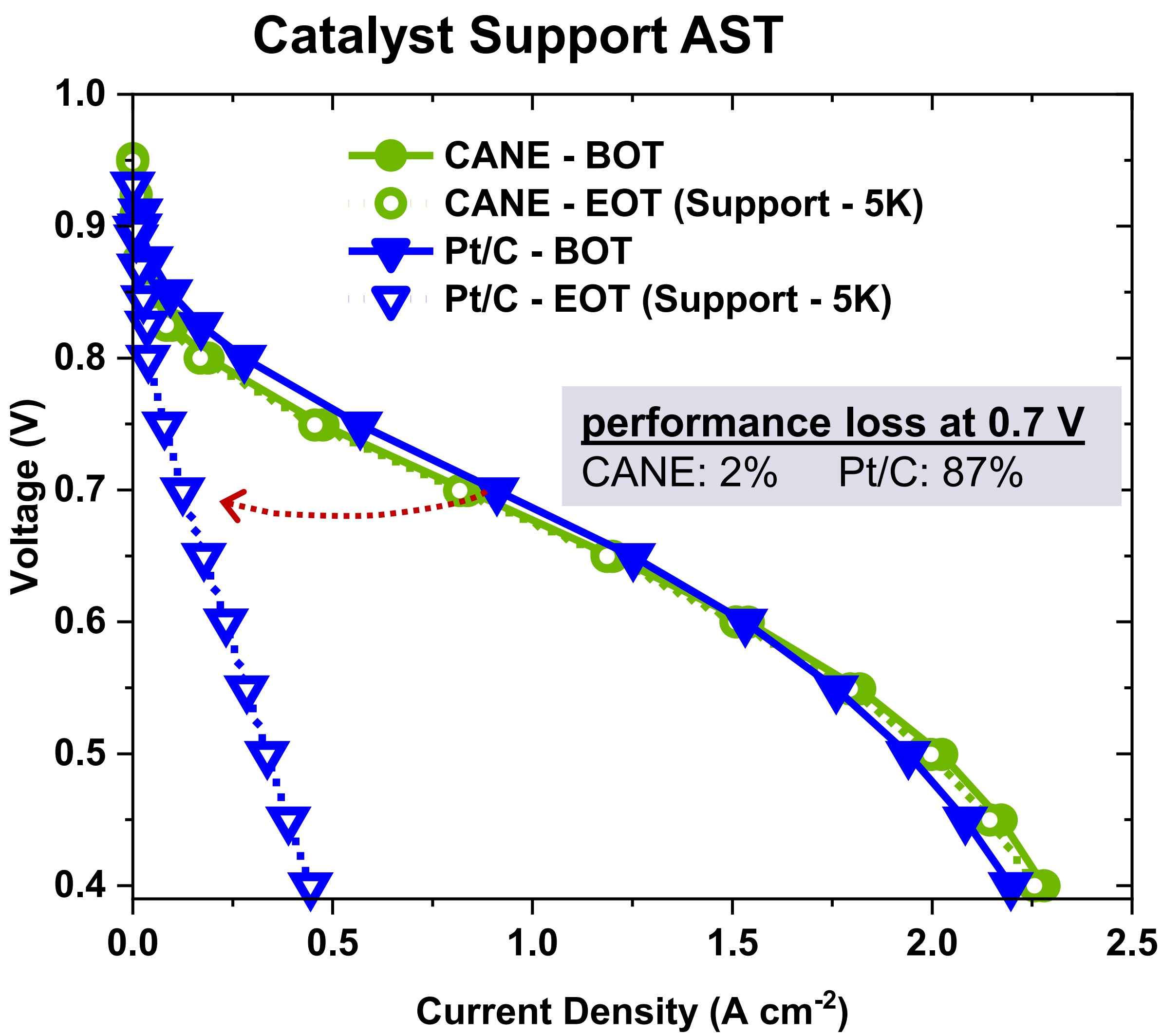

**Current density @ 0.7V vs reversal time**

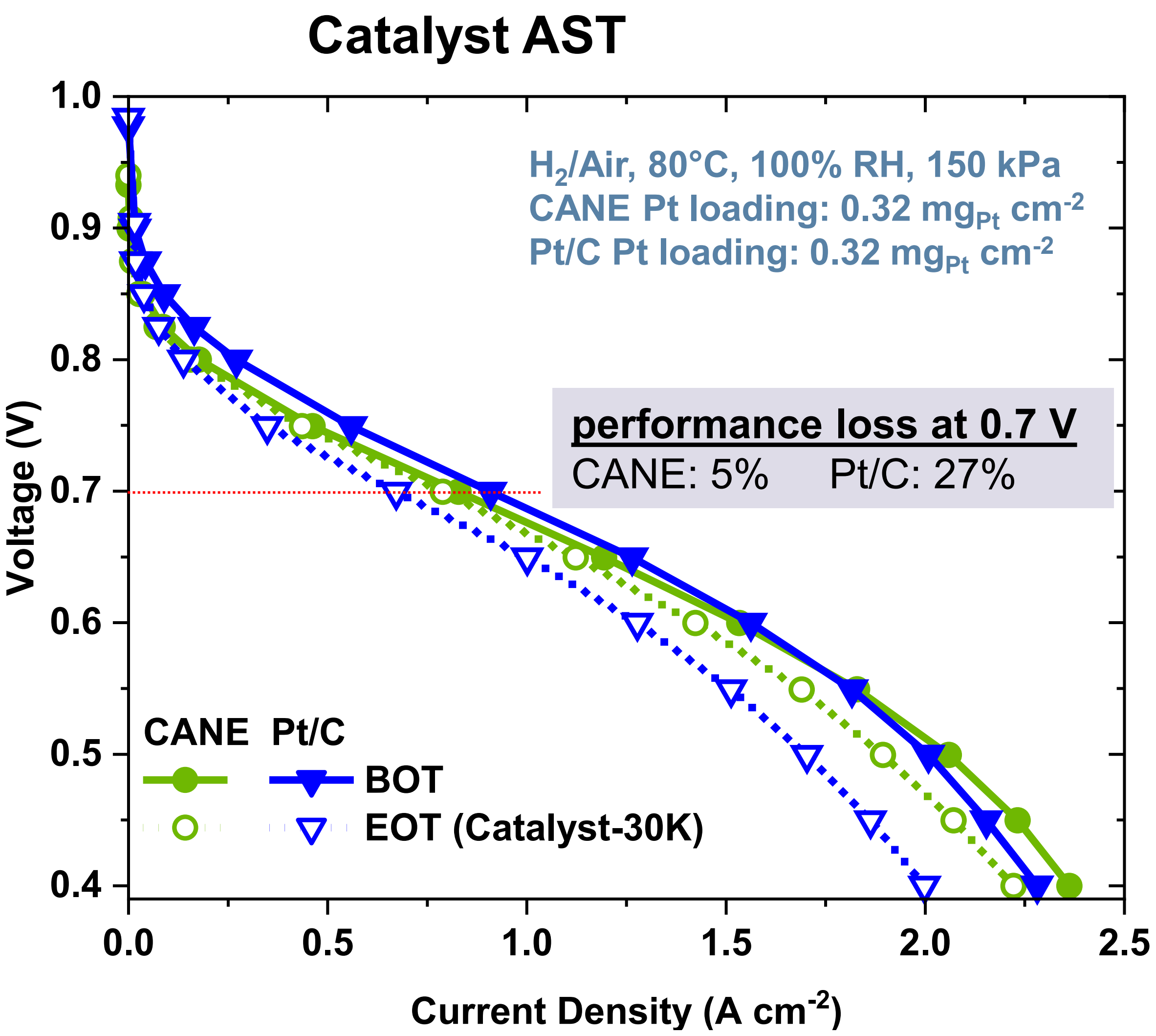





W. P. R. Liyanage, S. Komini Babu, J. S. Spendelow, in preparation

# CANE (Co-Axial Nanowire Electrode): Durability

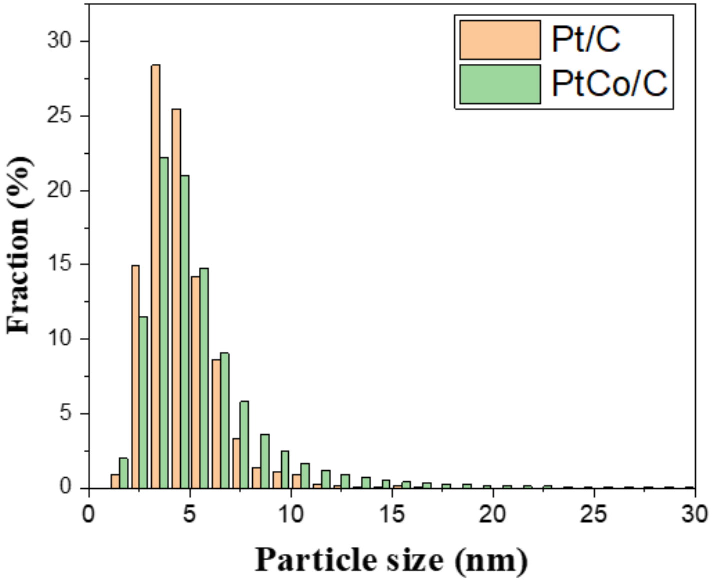


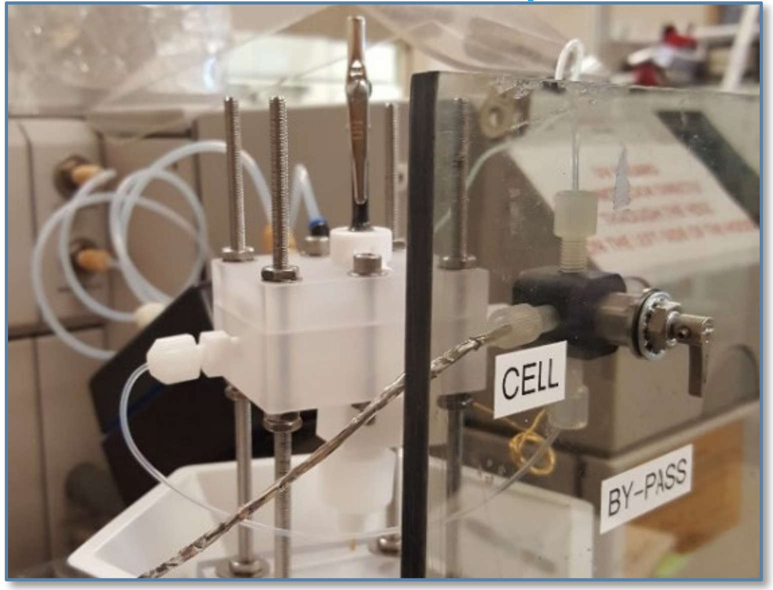

**Thin film architecture provides exceptional durability**

# Understanding Cathode Catalyst Degradation Mechanisms via Time-resolved ICP-MS measurements

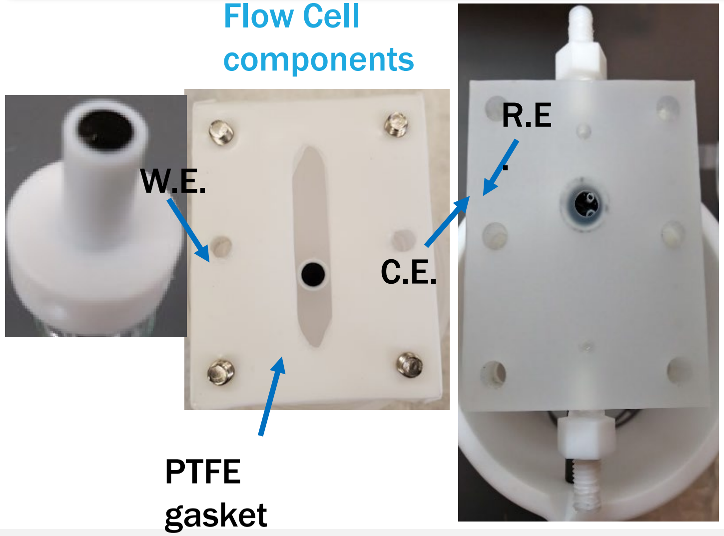
- Determine the potential and potential profile dependence of the dissolution of the metallic components of cathode catalysts
- Data used to develop models that can be used to explain and predict catalyst losses over time
- Catalysts
  - Umicore, PtCo (50wt% on HSC), ~ 5 nm
  - Umicore, Pt (50wt% on HSC), ~ 5 nm
  - M2FCT-developed catalysts



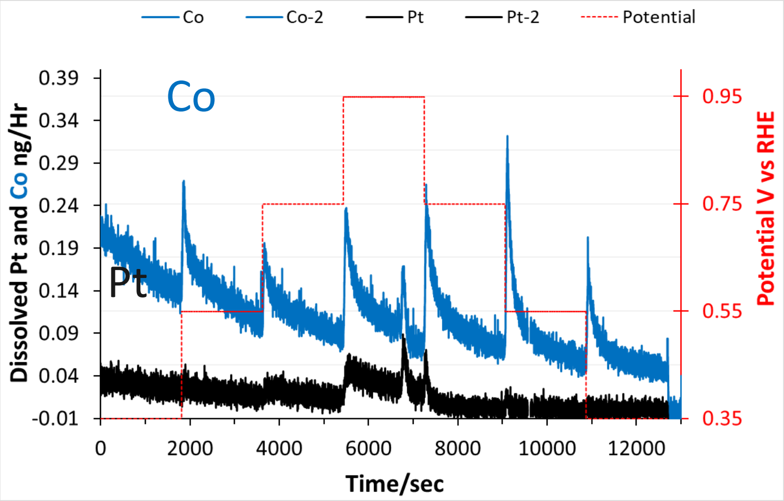

Electrochemical Flow Cell Coupled to ICP-MS




Flow Cell components



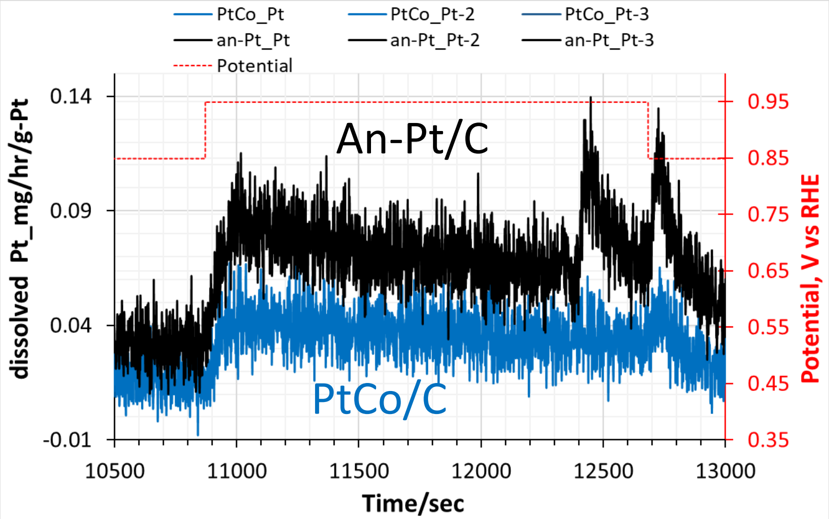
# Potential Dependence of Dissolution from An-Pt/C and PtCo/C



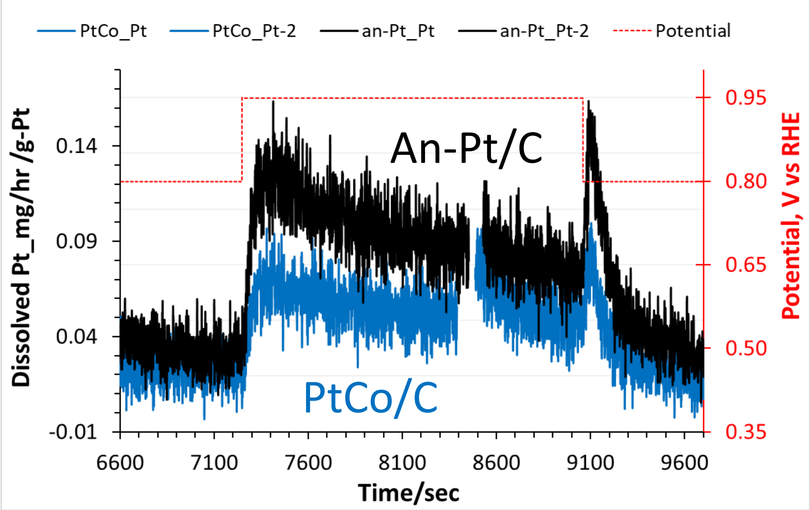

- Co dissolution from PtCo/C (Umicore Pt50 0690):
  - Occurs at all potentials
  - Is accelerated during increasing and decreasing potential steps
  - Has rates >3x higher than Pt
- Pt dissolution from An-Pt/C (Umicore Pt50 0550) and PtCo/C:
  - Occurs during positive potential steps to potentials as low as 0.55 V
  - Is higher for An-Pt/C than for PtCo/C (when normalized to Pt weight)
    - corresponding with smaller particle size for Pt/C

## Pt dissolution

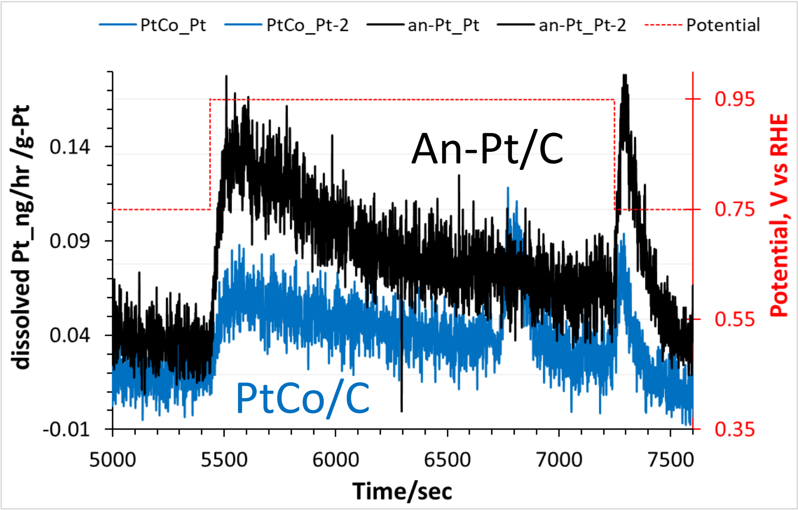
0.85 V to 0.95 V (100 mV) step




0.8 V to 0.95 V (150 mV) step




0.75 V to 0.95 V (200 mV) step

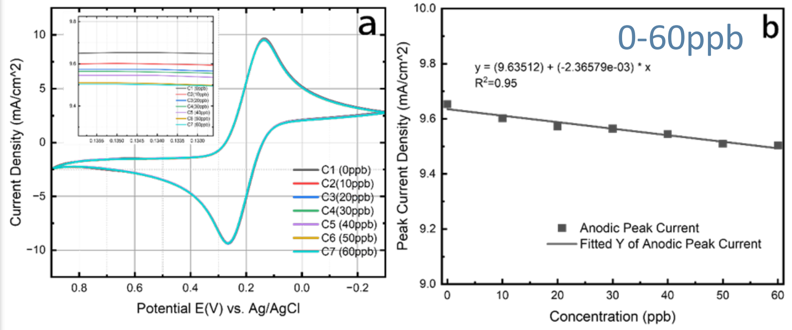


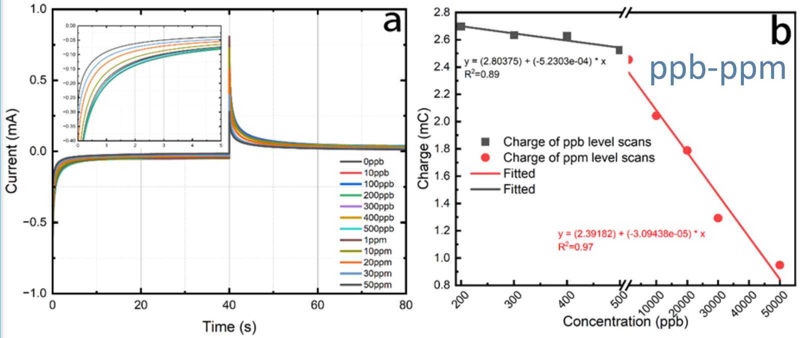
# Real-Time Continuous Monitoring of Ionomer Degradation

## Ion-Sensitive Field-Effect Transistor (ISFET) Microsensors

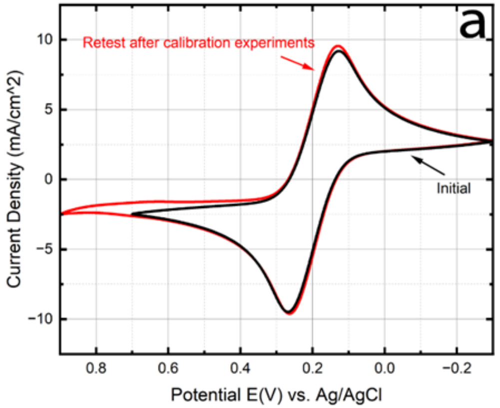
### Sensitivity

- -2.3uA/ppb
- Detection of limit: 10ppb

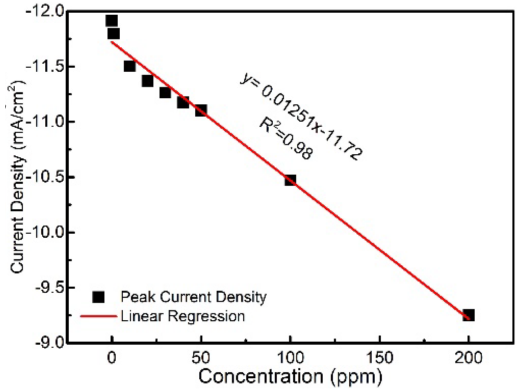



### Stability




- Retest after 100 days




### State-of-The-Art Comparison

- Real-Time Continuous Detection of FER for PFSA-based Membranes
- Develop Sensing Arrays to Compare with Hydrocarbon Membranes
- Benchmark Sensors during Fuel Cell Testing ASTs

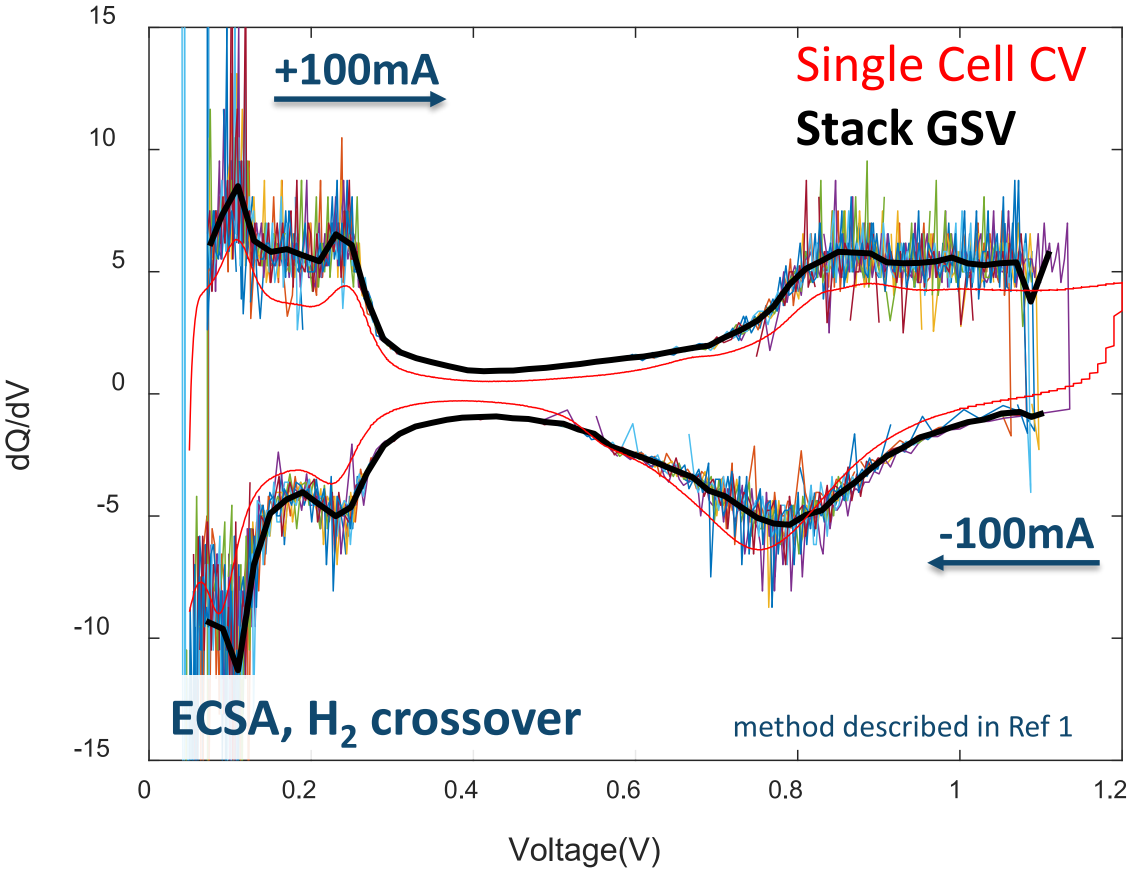
| Traits | This work | Hanna Inst. F- Combination Ion Selective Electrode | Dionex DX-120 Ion Chromatographer |
|---|---|---|---|
| Sensitivity | 10ppb | 20ppb | 5ppb |
| Cost | < $100 | ~$700 | $15,000 - $20,000 |
| Portability | Portable | Not portable | Not Portable |
| Commercialization for FC trucks | Viable | Potentially viable | Not Viable |
| Remote Data Collection | Yes | No | No |
| User-Friendly | Yes | Yes | No |
| In-situ real-time $F^-$ detection | Yes | No | No |

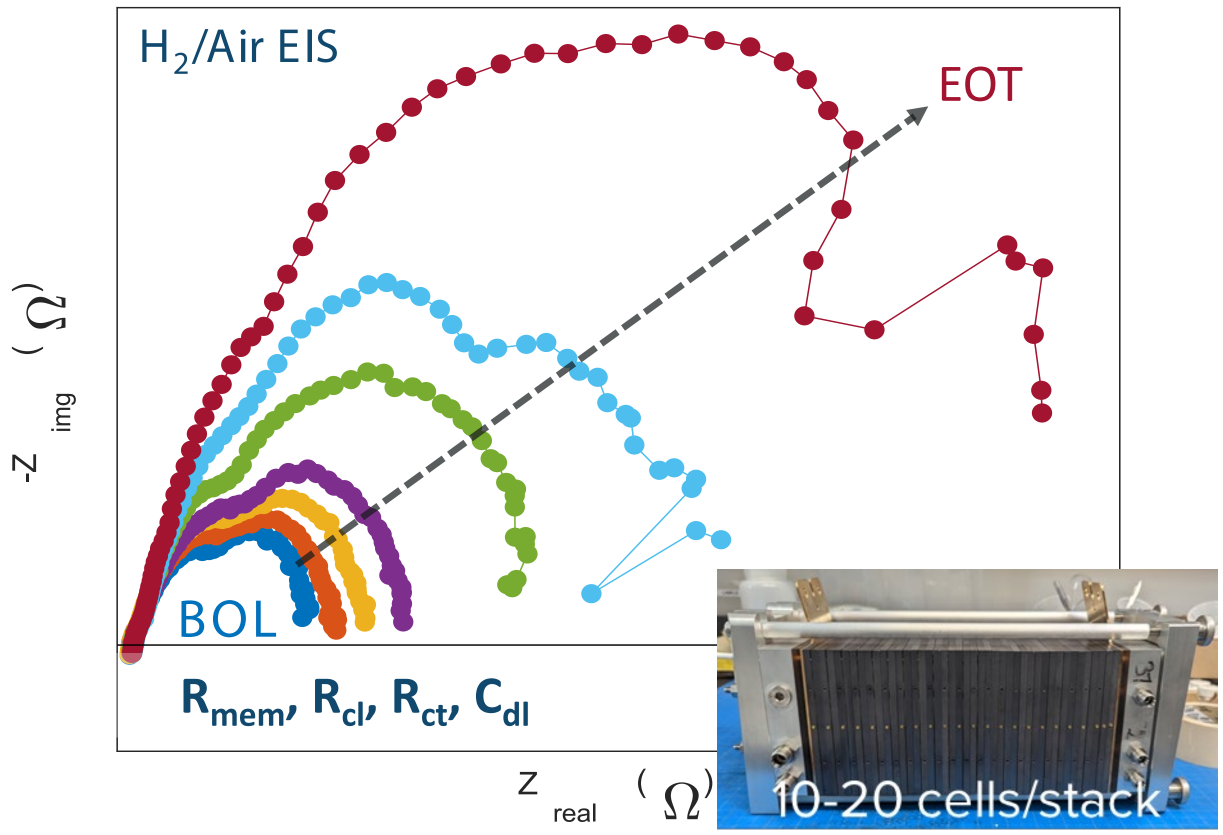
$F^-$ ISFET



D. Dong* et al., ECS Sensors Plus. 1(3) 2022, 035601

# Short Stack Testing with Diagnostics



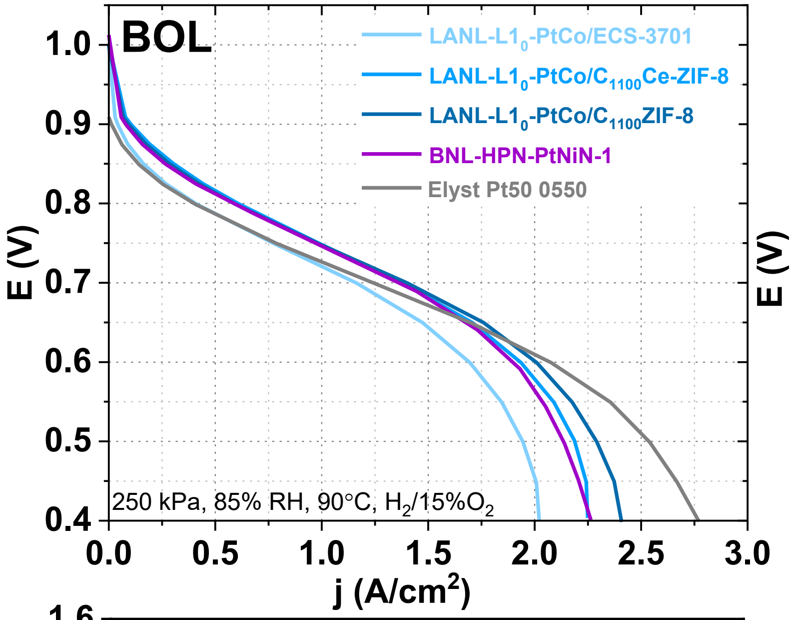

- Simultaneously measure single cell ECSA and H2 crossover on stack using current control
- Individual cell H2/Air AC Impedance
- Track many MEA properties over AST

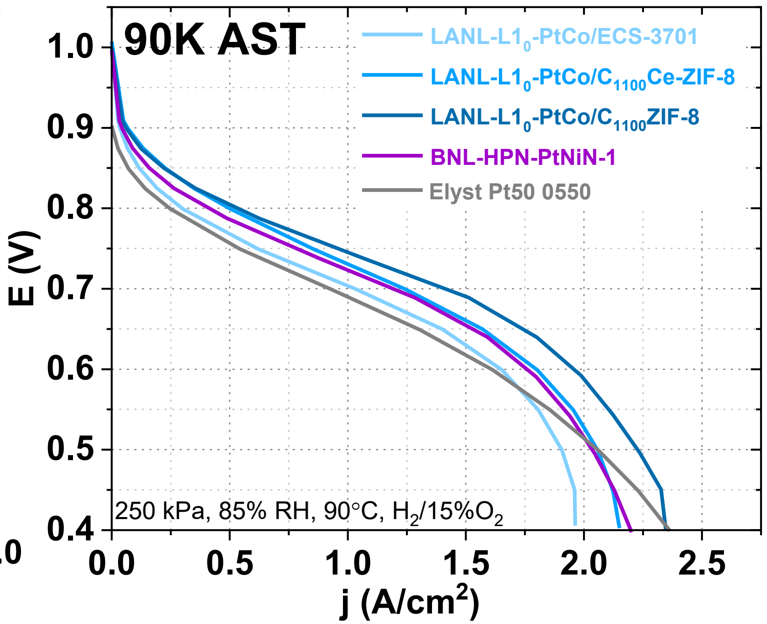
## High throughput MEA testing and characterization on short stack

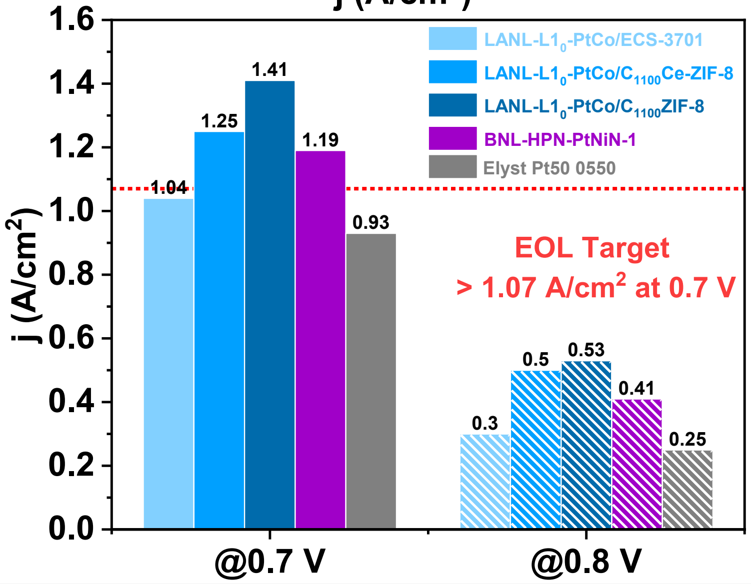
MILLION MILE
FUEL CELL TRUCK
U.S. DEPARTMENT OF ENERGY

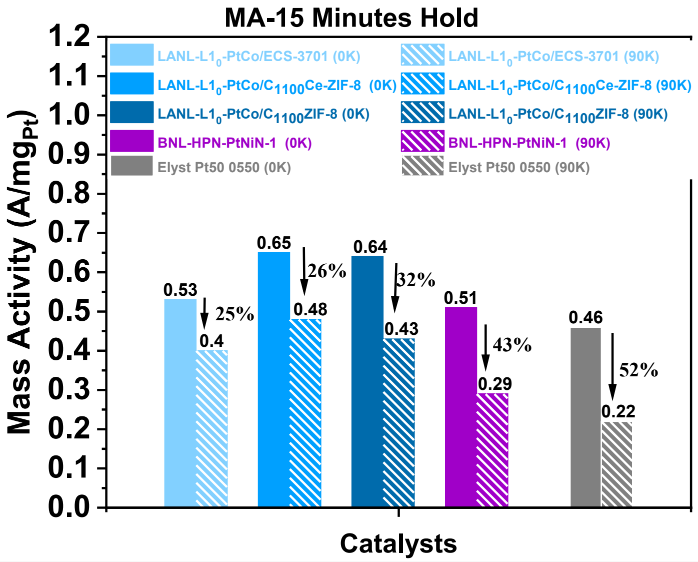
# Materials Development

# Multiple M2FCT Fuel Cell Catalysts with High Durability



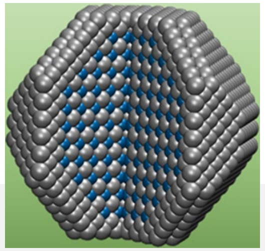

- M2FCT labs have identified multiple paths to high durability:
  - Combining intermetallic PtCo with advanced supports
  - High-pressure nitriding of PtNi
- M2FCT target calls for 2.5 kW/gPGM (1.07 A/cm$^2$ with 0.3 mgPGM/cm$^2$) at 0.7 V after 25,000 hour-equivalent accelerated durability test
- New PtCo and PtNiN catalysts provide 2.8-3.3 kW/gPGM (1.19-1.41 A/cm$^2$) after 90K catalyst AST cycles*

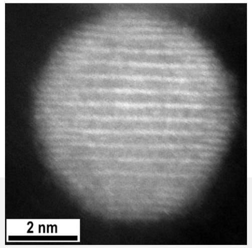
*90K cycles from 0.6-0.95 V with 0.5 s ramp and 2.5 s soak. 90K catalyst AST cycles are considered to represent the catalyst degradation component of the durability test, but degradation of support, membrane, and ionomer are not captured in this AST.

Anode/cathode loading = 0.05/0.25 mg$_{PGM}$/cm$^2$; NC700 membrane

# Nitrided PtNi Intermetallics with Improved Performance/Durability

- Optimized intermetallic PtNiN/C catalysts to improve ORR performance by:
  - Increasing N-content in cores by high-pressure nitriding (HPN) at 900 psi (*F, G*)
  - Increasing the total metal loading from 42 wt.% to 52 wt. % (40~42% Pt, 10~12% Ni) (*G, I*)
  - Thickening Pt shell by acid-wash treatment (6 h in 0.1 M $H_2SO_4$ at 60°C; SA) (*H, I*)
- All the catalysts showed higher current density at 0.8 V than reference a-Pt/C at BOT
- The catalysts with higher Pt loading (42%, H, I) showed low durability
- The effect of acid washing is still unclear (G, I)





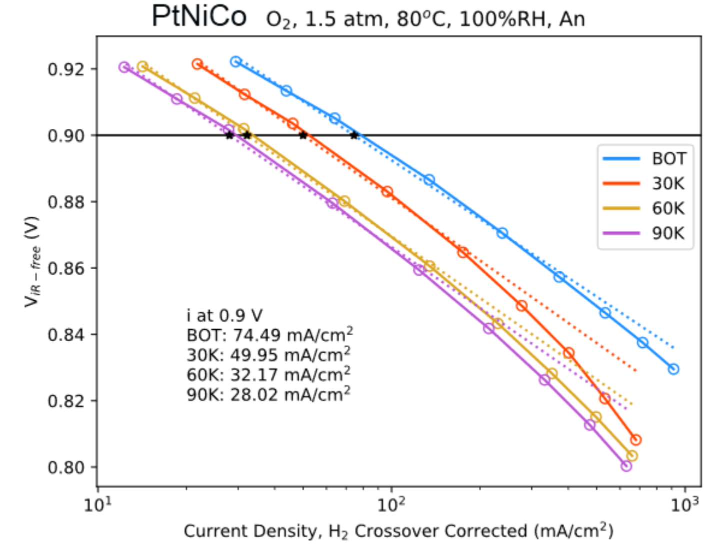

## Beginning of Test (BOT) and Post-AST (90K) $H_2$/Air

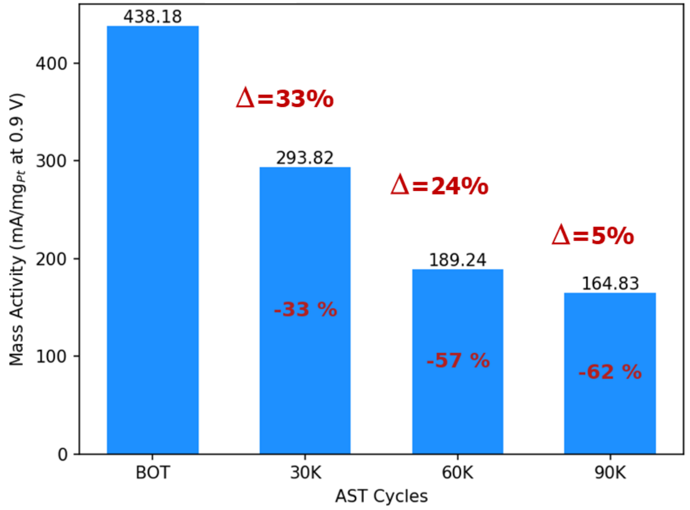
| | Catalysts | Cycles | *i* at 0.8V (A/cm²) |
|---|---|---|---|
| F | HPN-Int-PtNiN-40% (900 psi) (4.5 nm) | 0K | **0.377** |
| | | 90K | 0.129 |
| G | SA-HPN-Int-PtNiN-40% (900 psi) (4.3 nm) | 0K | **0.414** |
| | | 90K | **0.143** |
| H | Int-PtNiN-52% (4.2 nm) | 0K | **0.457** |
| | | 90K | 0.122 |
| I | SA-Int-PtNiN-52% (4.0nm) | 0K | **0.467** |
| | | 90K | 0.108 |
| Ref | a-Pt/C (4~5 nm) | 0K | 0.32 |
| | | 90K | 0.14 |

*90K cycles from 0.6-0.95 V with 0.5 s ramp and 2.5 s soak. 90K catalyst AST cycles are considered to represent the catalyst degradation component of the durability test, but degradation of support, membrane, and ionomer are not captured in this AST

# Intermetallic Ternary Alloy PtCoNi/C Improved Performance/Durability

## $H_2/O_2$ Performance - Mass Activity



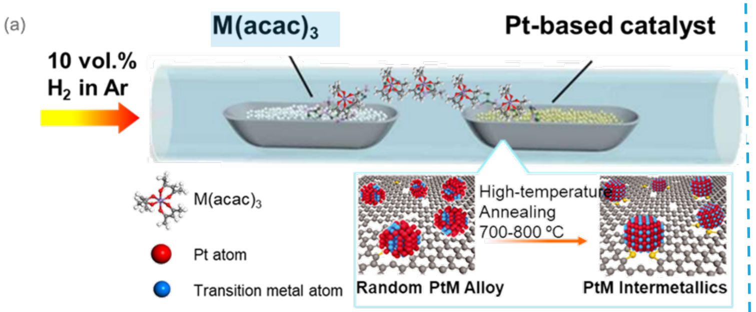

## $H_2$/Air Performance: BOT – 90K cycles

| Test Conditions | BOT Current Density at 0.7 V ($mA/cm^2$) | BOT Current Density at 0.8 V ($mA/cm^2$) | 30K Current Density at 0.7 V ($mA/cm^2$) | 30K Current Density at 0.8 V ($mA/cm^2$) |
|---|---|---|---|---|
| 1.5 atm, 80 °C, 75 %RH | 740.00 | 255.08 | 567.96 | 157.10 |
| **1.5 atm, 80 °C, 100 %RH** | **879.52** | **319.84** | **708.76** | **191.46** |
| 2.5 atm, 90 °C, 65 %RH | 905.51 | 327.23 | 710.58 | 220.12 |
| 2.5 atm, 90 °C, 85 %RH | 1116.12 | 390.62 | 886.87 | 272.41 |

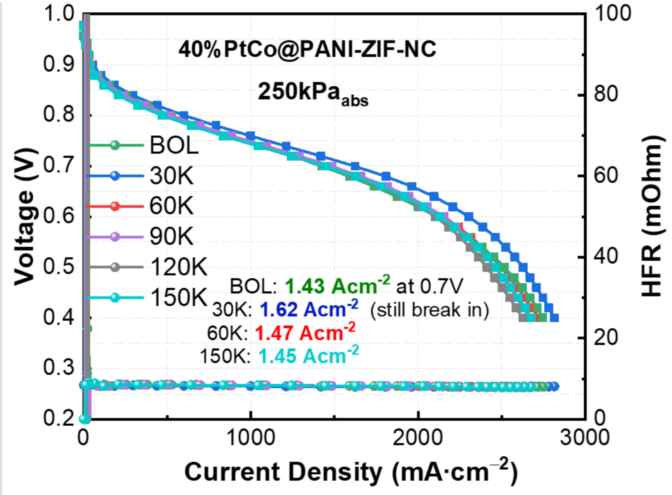
| Test Conditions | 60K Current Density at 0.7 V ($mA/cm^2$) | 60K Current Density at 0.8 V ($mA/cm^2$) | 90K Current Density at 0.7 V ($mA/cm^2$) | 90K Current Density at 0.8 V ($mA/cm^2$) |
|---|---|---|---|---|
| 1.5 atm, 80 °C, 75 %RH | 488.30 | 111.68 | 451.84 | 99.67 |
| **1.5 atm, 80 °C, 100 %RH** | **666.63** | **154.61** | **634.11** | **138.76** |
| 2.5 atm, 90 °C, 65 %RH | 595.95 | 169.10 | 539.93 | 152.55 |
| 2.5 atm, 90 °C, 85 %RH | 785.56 | 207.64 | 736.78 | 189.02 |

- Achieved go/no-go (>138 mA/cm2 H2/Air performance at 0.8 V after 90K AST cycles) with intermetallic ternary alloy PtCoNi/C
  - Selected PtNiCo/C utilized minimal amount of Pt on support (total metal loading 15%) while benefiting from intermetallic structure
  - Performance passing Go/No-Go achieved with 0.17 mgPt/cm2 cathode loading
- Electrode structure will be further optimized to enable higher cathode loadings/higher performance

# Chemical Vapor Deposition Approach to Preparing PtCo Catalysts

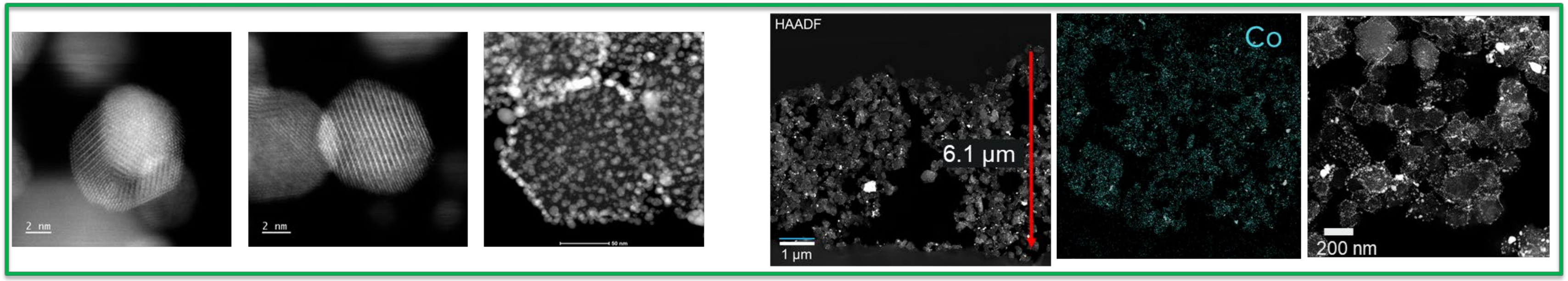



*$Co(acac)_3$: Tris(acetylacetonato)cobalt(III); melting point: 210-213 °C*

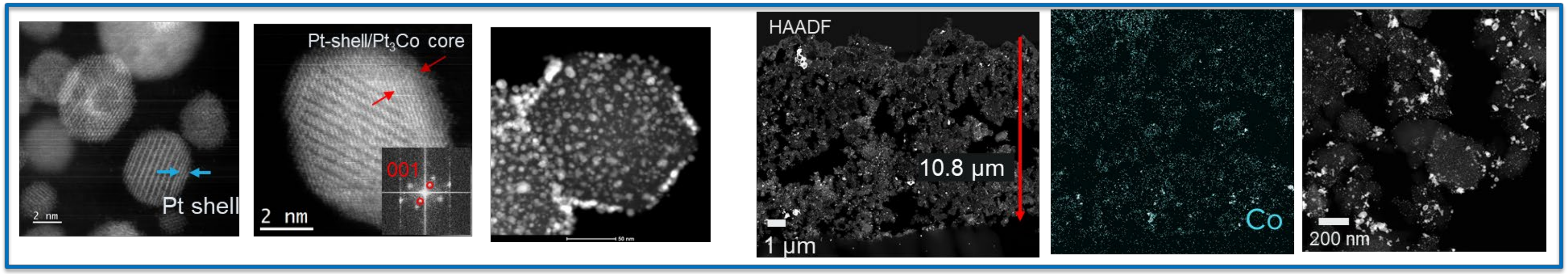



*0.6V-0.95V, $H_2/N_2$, 150K AST (6 s/cycle)*
*Pt loading at the cathode: **0.25** $mg_{Pt}/cm^2$; 5 $cm^2$ differential cell at 80 °C; $H_2$: 500 sccm ; air: 2000 sccm; Gore membranes; HOPI*

## 40PtCo@PANI-ZIF-NC BOL

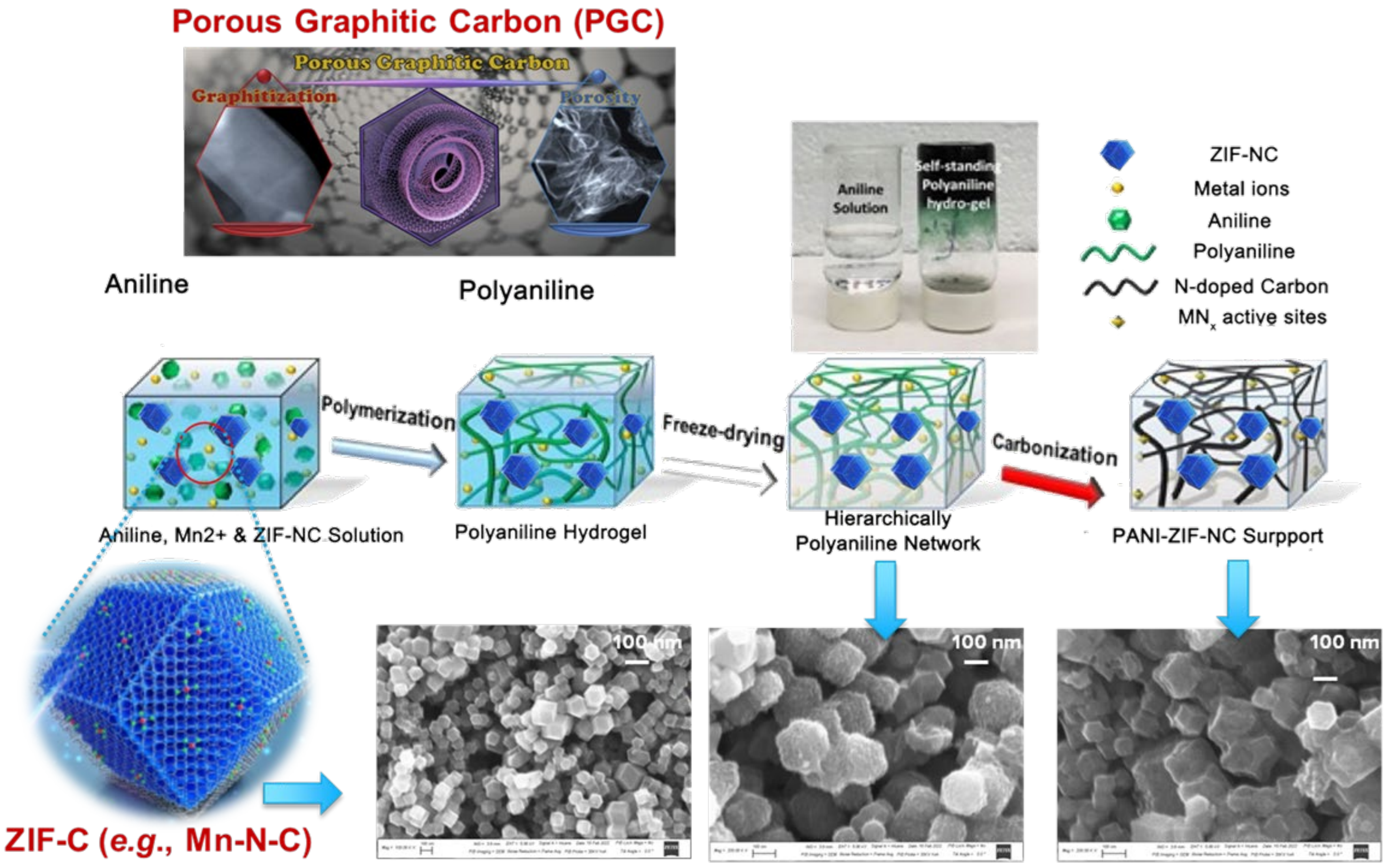



## 40PtCo@PANI-ZIF-NC-700-EOL

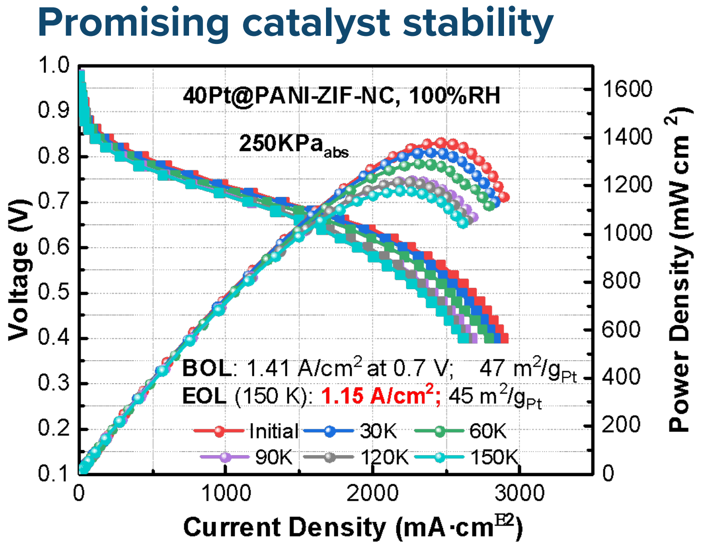


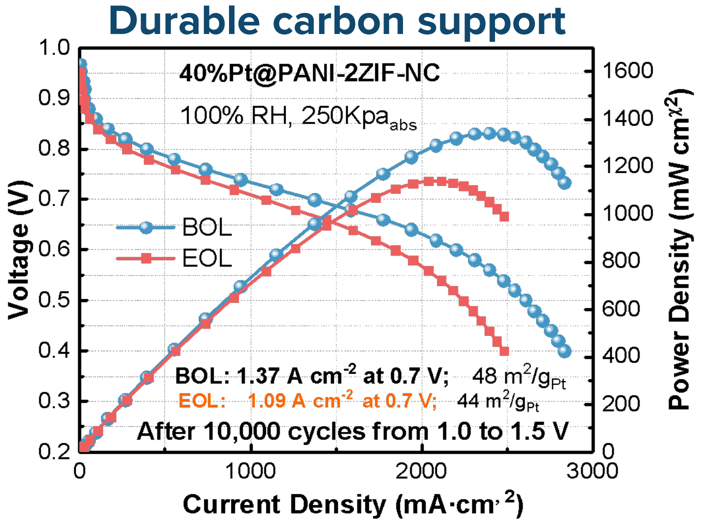

- The CVD-derived PtCo catalyst presented promising durability during the long-term AST (0.60-0.95 V, $H_2/N_2$, 80°C, 100% RH).
- The L12- $Pt_3Co$ intermetallic structure was well maintained, with superlattice spot of inner $Pt_3Co$ observed.
- Insignificant Pt NP agglomeration; Co content in the PtCo catalyst retained well.
- The cathode layer becomes more porous, likely exposing more Pt atoms

# Polymer/ZIF-Derived Hybrid Carbon Support with Improved Durability




- Optimal balance among graphitization degree (defined 2D peak in Raman), high surface area (BET area: ~670 m2/g), and mesopores of the hybrid support promote Pt catalyst and support stabilities

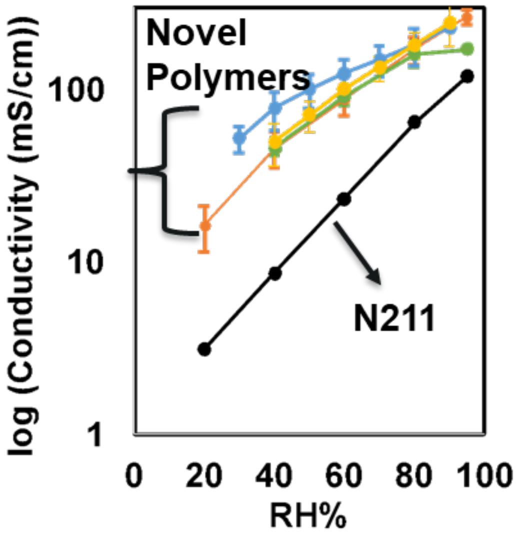



*0.6V-0.95V, $H_2/N_2$, 150K AST (6 s/cycle)*
*Pt loading at the cathode: **0.25** mg$_{Pt}$/cm$^2$; 5 cm$^2$ differential cell at 80 °C; $H_2$: 500 sccm ; air: 2000 sccm; Gore membranes; HOPI*

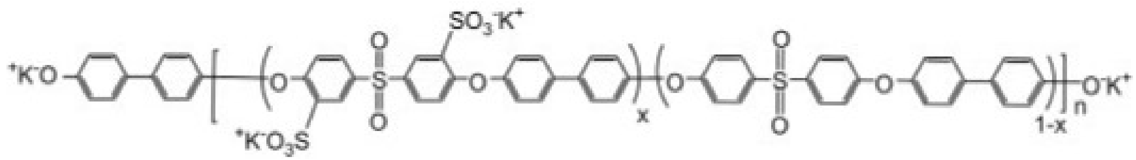
# Novel Cation Conducting Polymer Electrolytes

## Polymers with perfluoro or partially fluoro chemistry

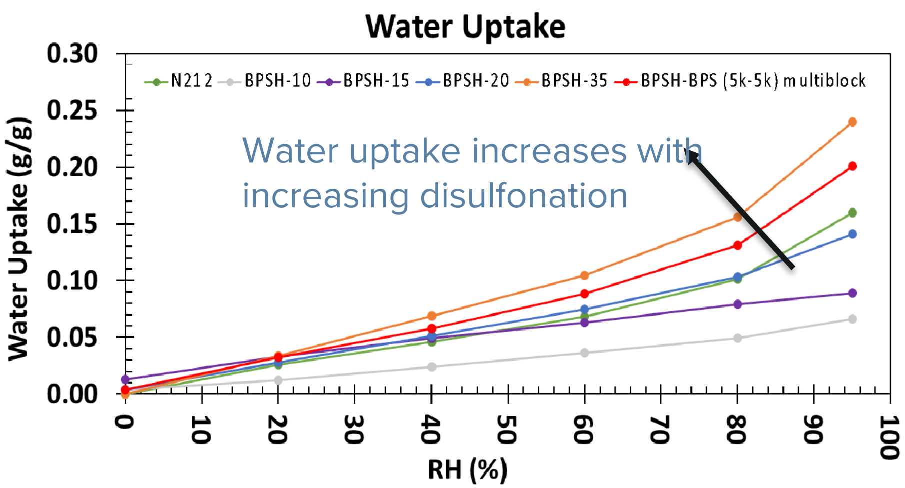


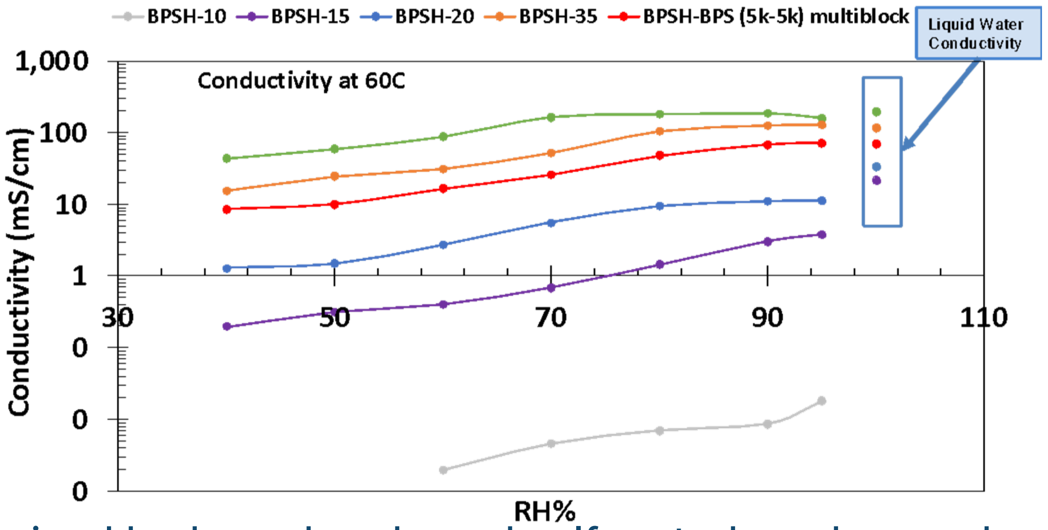

- In progress - improving mechanical properties and limit water swelling

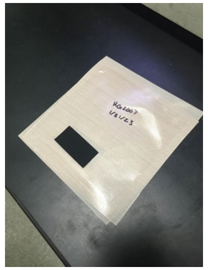
## Hydrocarbon based Polymer membrane & ionomers



Disulfonated biphenol based poly(arylene ether sulfone)
BPSH-XX where X= degree of disulfonation



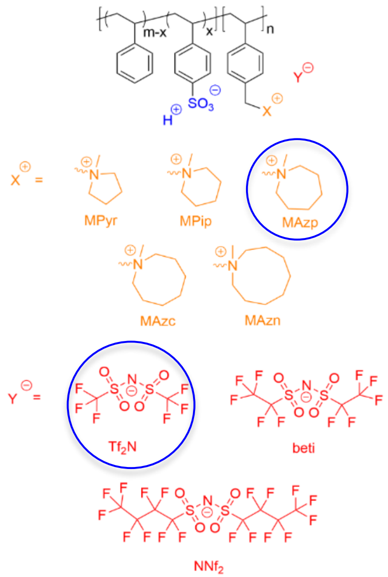



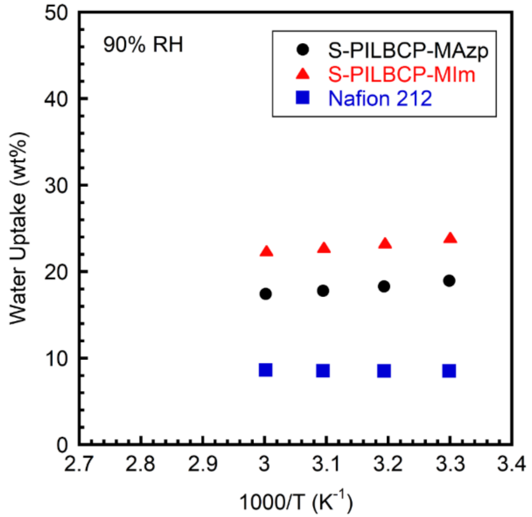
| Ink Formulation | Target % |
|---|---|
| BPS-50 Polymer | 0.5 |
| IPA | 10 |
| Water | 90 |

- Synthesized hydrocarbon based sulfonated random and multiblock copolymers at NREL with varying degree of disulfonation
  - Established successful formulation for preparing hydrocarbon-based ionomer ink (higher sulfonation helped)
- In progress- multiblock synthesis as a function of block lengths, IEC & developing structure-property & further optimization of ionomer ink formulation

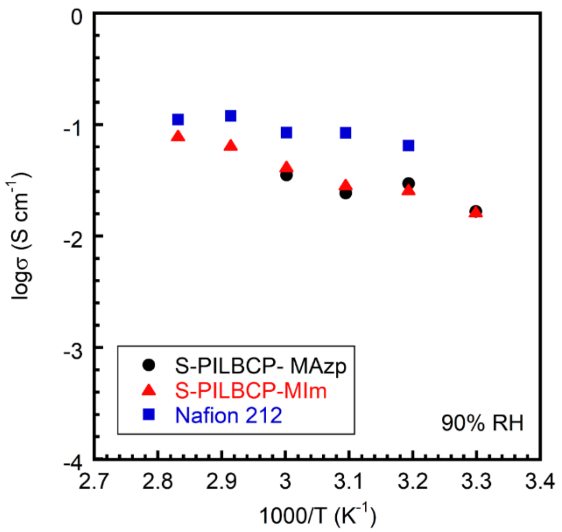
# Advanced PILBCP Ionomer Composites

Gen II S-PILBCP
Poly(S-PILBCP-Mazp)

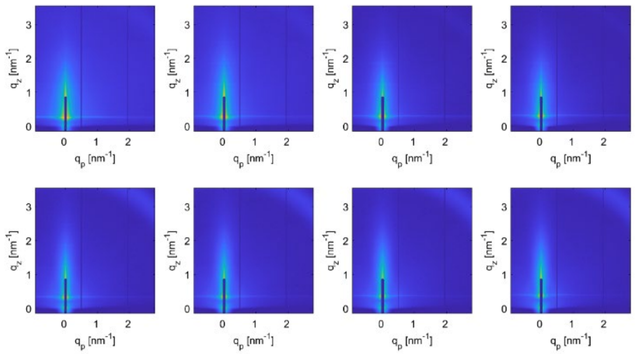



**Water uptake at 90% RH**
S-PILBCP-MIm – Gen I
S-PILBCP-MAzp – Gen II




**Ionic Conductivity at 90% RH**
S-PILBCP-MIm – Gen I
S-PILBCP-MAzp – Gen II

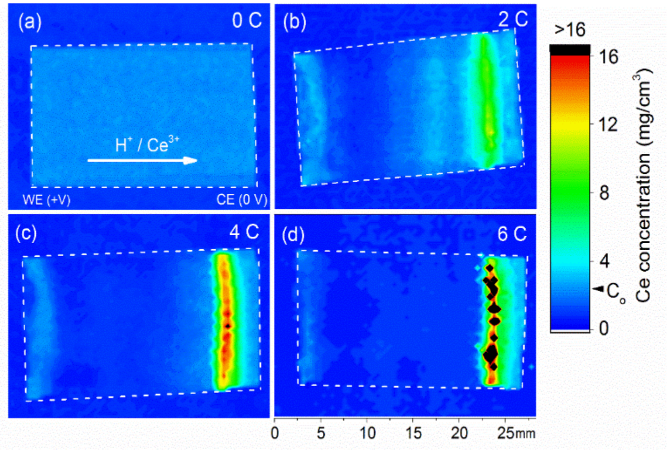


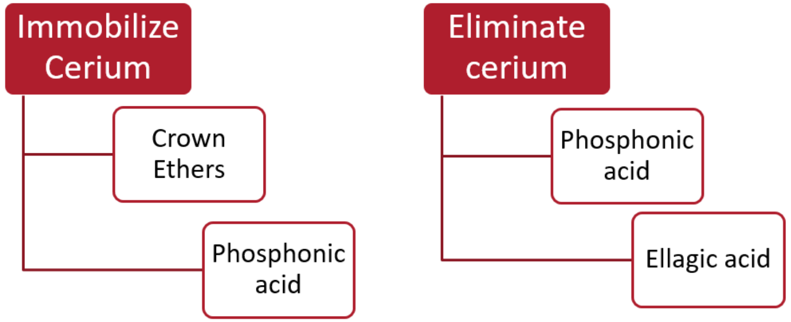

**GISAXS**
Domain Spacing
As-synthesized: 12-15 nm
GISAXS: 5.77 ± 0.027 nm
(Film thickness: 27 nm)

PILBCP: polymerized ionic liquid block co-polymer
**Sulfonated-block:** Ionic conductivity
**Ionic Liquid-block :** $O_2$ permeability, kinetic effects

- Gram-scale synthesis of poly(S-PILBCP-Mazp)
  - Water uptake and conductivity comparable to Gen I and Nafion
  - Phase separation maintained after solubilization and recasting
- Objectives for Next Generation S-PILBCPs
  - Improve $O_2$ permeability of S-PILBCP with greater free volume in IL-block
  - Increase dispersibility in aqueous-based inks
  - Production scale-up

# Improving Membranes by Stabilizing Cerium




## Phosphonic Acids

C8 (n=6), C10 (n=8)

$CF_3(CF_2)_nCF_2\text{-}PO_3H_2$

ODPA (n=6), PDPA (n=10)

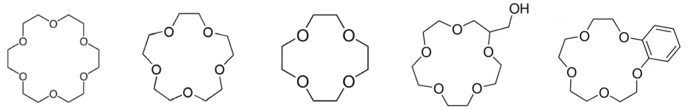
$PO_3H_2\text{-}CH_2(CH_2)_nCH_2\text{-}PO_3H_2$

PFOPA (n=5), FHPA (n=11)

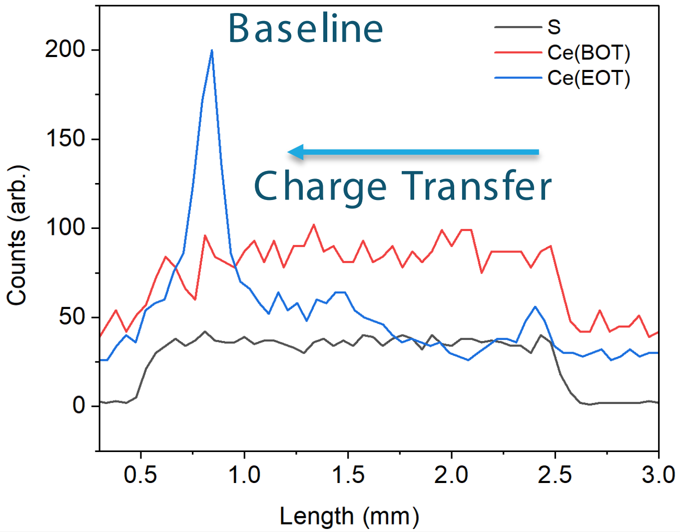
$CF_3(CF_2)_nCH_2\text{-}CH_2\text{-}PO_3H_2$

## Crown Ethers

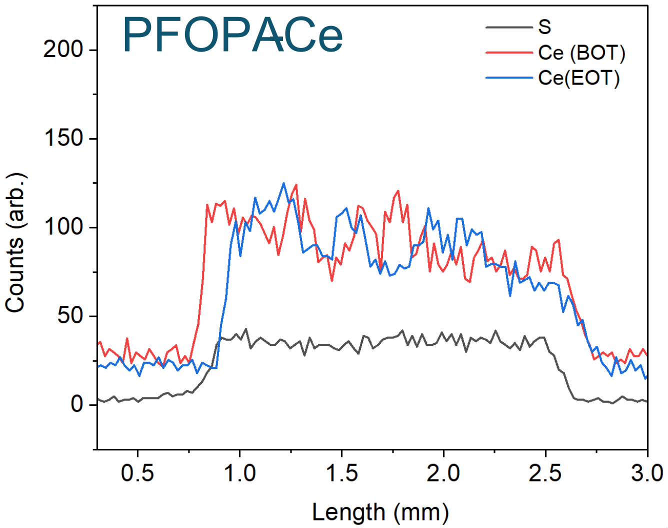
18C6 15C5 12C4 A-15C5 B-15C5



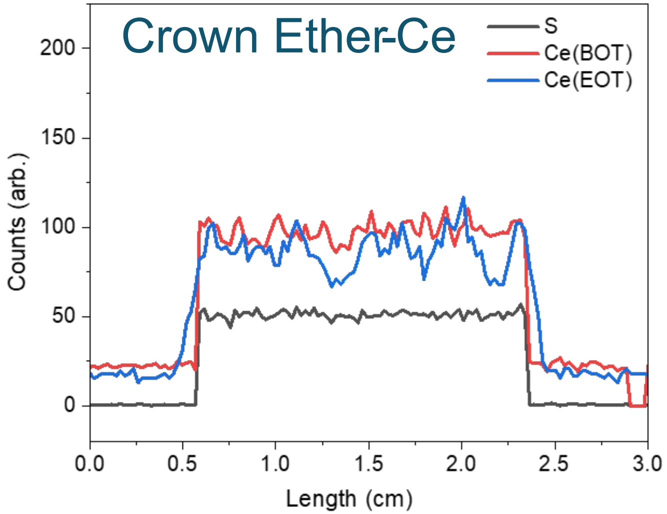

### Cerium elemental profile before and after migration




### Cerium elemental profile with Phosphonic Acid (PFOPA)




### Cerium elemental profile with CRE (15C5)

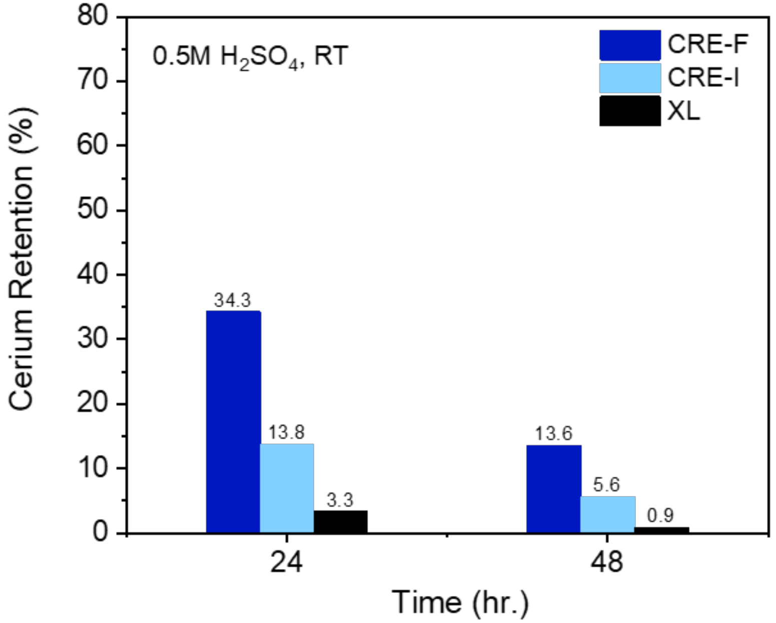




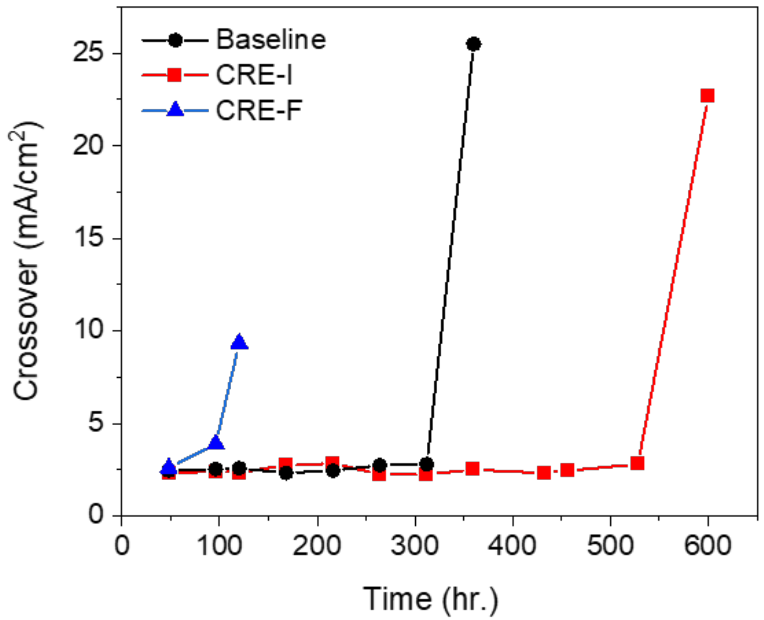

Tanya Agarwal, et al., Available at SSRN 4355106; Tanya Agarwal, et al., 2023, *Journal of Power Sources*, 554, 232320

# Improving Membrane Durability by Stabilizing Cerium: Crown Ethers

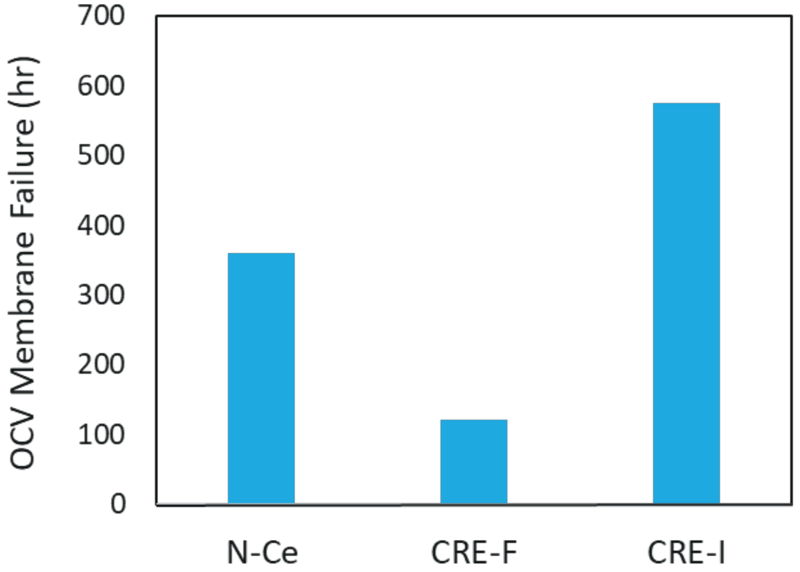
## Cerium Retention in Acidic Solution



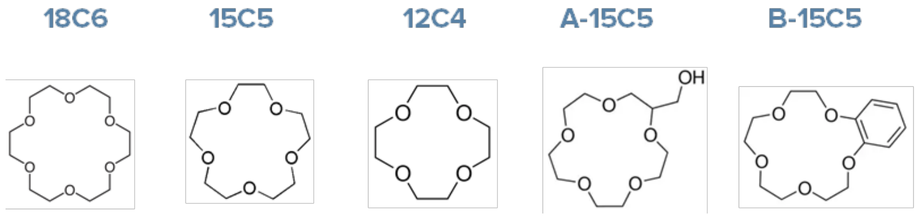

## Hydrogen crossover change during OCV hold testing (90ºC, 30% RH)




## OCV hold Time to Failure




- 15C5 was found to demonstrate the best Ce retention of various Crown Ethers
- Bonding the CRE to the side chain improves Ce Retention
- Mixing Crown Ethers into the membrane improved Durability

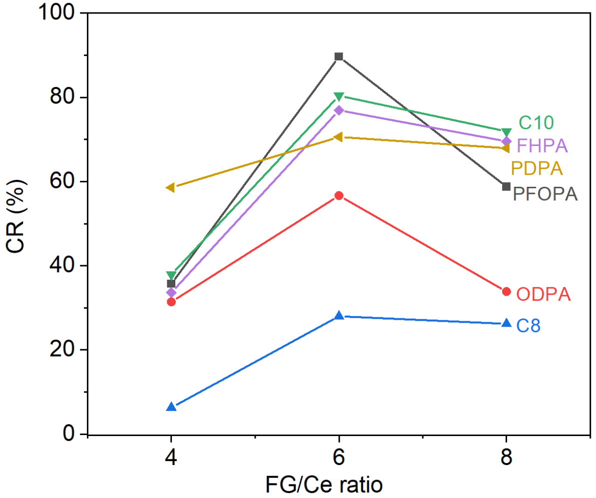



**CRE-I:** Membranes incorporated with crown ether by mixing (15C5)
**CRE-F:** Membrane with crown ether covalently bonded to the side chain (15C5)
**XL:** Commercial membranes from Chemours

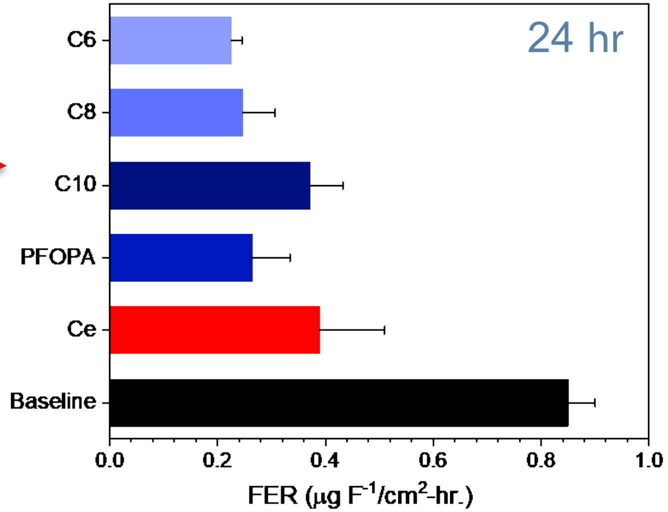
# Improving Membrane Durability

## Phosphonic Acids work for Ce stabilization and Radical Scavenging

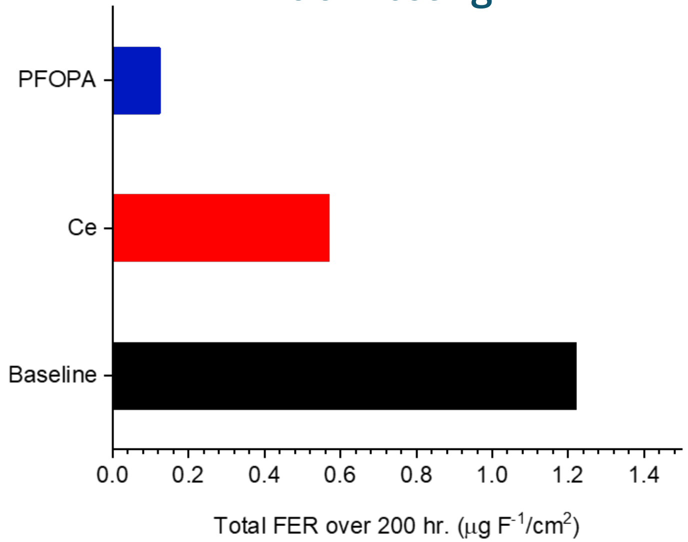
### Cerium Retention (CR) for various phosphonic acid length and pKa



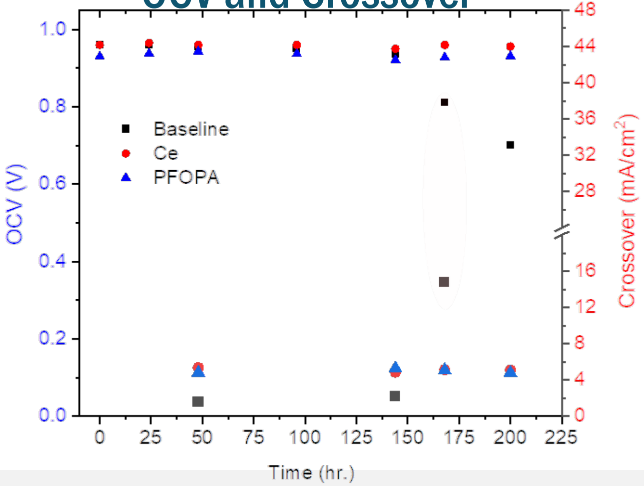

### Fluoride emissions from Fenton test for membranes




### Fluoride emissions MEA during OCV Testing




### OCV and Crossover




- Phosphonic Acids can be effective at stabilizing Ce
- Length of Phosphonic Acid important for stabilizing Ce
- PFOA found to reduce Total Fluoride emission compared to Ce
- Phosphonic acids have their own radical scavenging ability without Ce
- Best durability performance was found with PFOPA (no Ce)

C8 (n=6), C10 (n=8) ODPA (n=6), PDPA (n=10) PFOPA (n=5), FHPA (n=11)

$CF_3(CF_2)_nCF_2\text{-}PO_3H_2$  $PO_3H_2\text{-}CH_2(CH_2)_nCH_2\text{-}PO_3H_2$  $CF_3(CF_2)_nCH_2\text{-}CH_2\text{-}PO_3H_2$



Agarwal, T., Sievert, A., Babu, S.K., Adhikari, S., Prasad, A.K., Advani, S.G., Hopkins, T., Park, A., Kim, Y.S. and Borup, R., Available at SSRN 4355106.

# Collaborations: Non-FOA activities

| Entity | Scope of collaboration |
|---|---|
| **Pajarito Powder** | Catalyst durability testing |
| **Pusan National University (Seung Geol Lee)** | DFT calculations |
| **CEA-Grenoble** | Electron tomography collaboration and exchange of materials (follow-on to Embassy Science Fellow Program |
| **Fraunhofer ISE, Colorado School of Mines, University of Connecticut (OREO International Collaboration)** | Collaboration specifically to bring together the differentiated coating capabilities between NREL and ISE and have a unique and consistent set of characterization tools between the two universities |
| **3M Company** | NREL and 3M collaborated in development of novel materials. 3M provided NREL, at no cost, with several critical components needed for synthesis of targeted cationic ionomers. |
| **UCI/NFCRC HIMaC2/UCI** | MEA fabrication and testing XRF, MEA |
| **Hahn-schickard and Simon Fraser University (S. Holdcroft)** | Hydrocarbon ionomers for thin film studies have been provided |

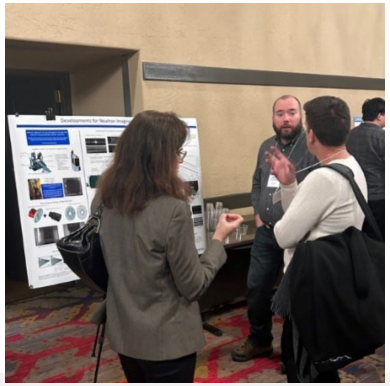
| Entity | Scope of collaboration |
|---|---|
| **Ionomr** | Hydrocarbon membranes durability testing, and samples being provided |
| **University of Minnesota** | Having discussions related to film stress measurements in drying catalyst layer films |
| **Giner Inc.** | Providing the ZIF-derived PGM-free Mn-N-C carbon-supported Pt catalyst to Giner for an SBIR project.<br>Electron microscopy support for SBIR project via CNMS user program |
| **pH Matter** | Supplied catalyst powders and tested MEAs for microscopy evaluation |
| **University of Montpellier (IMMORTAL)** | Electron microscopy characterization of Pt-Rare Earth Metal (REM) alloy catalysts |
| **Bar-Ilan University** | Doped carbon supports |
| **University of Delaware (Ajay Prasad, Suresh Advani)** | Radical Scavenger Development |

# Inclusion, Diversity, Equity, Accountability

## Outreach and Workforce Development

- M2FCT hosted UGS, GRA interns and DOE SCGSR Fellows to gain hand-on experience working with fuel cell systems and materials and learn about hydrogen technologies
- Multiple MSI students with M2FCT
- Internship programs (K12 and SULI) for summer 2022
- Discretionary projects includes three MSIs (UCI, UCM, FIU)
- Defined multiple projects staffed and coordinated with MSIPP and M2FCT
  - Ce migration with GM
  - BPP corrosion with Treadstone
  - Negotiating with others

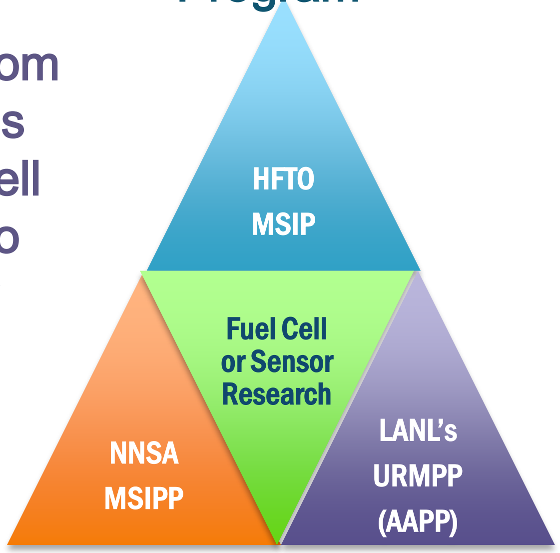
Industry, Students and Lab Staff



MSIP Students Attending M2FCT Meeting Feb 2023


M2FCT working with Minority Serving Institution Partnership Program

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




MSIPP

# Future Work

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

- FOA Support
  - Continue and expand support for existing and future FOA projects
- Analysis
  - Refine models, characterization, and diagnostics for heavy-duty operating conditions
  - Define operating conditions efficiency and durability trade-offs as well as total cost of ownership for 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
  - Cross-correlations of metadata for material, integration and AST studies and feature importance analysis for operation and material parameters

- Material Development
  - Scale-up synthesis of best-of-class catalyst and integrate into large active area MEAs (50 cm2)
  - Improve catalyst activity and durability
    - Nitride PtNi intermetallics
    - PtCo intermetallic catalyst
    - Multi-metallic Pt-based high entropy alloys
    - PtNiCo/C
    - Metal oxide loading on carbon supports
    - MOF-based carbons
    - PtCo intermetallics melamine and melamine-based polymers
  - Ionomer, Membranes
    - Develop non-fluorinated hydrocarbon ionomers;
    - Develop organic radical scavenger additives.
    - Explore grafting, cross-linking, block co-polymer and cathode catalyst layer fabrication strategies to address water solubility of high IEC ionomers
    - Evaluate effect of block length and ion-exchange capacity of multi-block co-polymers on ionomer properties and electrode structure

# Future Work

- Integration
  - Baseline SOA
    - Establish benchmark performance and cost of state-of-art PEM and MEA
    - Trade off with Pt loading and TCO, contrast with advanced electrode layers
  - Integrate newly developed materials into optimized MEA structures
    - Integrate new catalysts with advanced electrode designs to improve performance and durability
    - Evaluate and compare effect of catalyst loading on performance and durability for conventional and next generation electrodes
  - Catalyst layer studies
    - Catalyst ink to structure formation models
    - Low-dose, cryo-electron microscopy imaging of ionomer distribution in epoxy-free electrodes with varying support and ionomer type.
    - Explore various methods to form uniform thickness, crack-free electrodes
  - Electrode Development
    - Enhance activity of coaxial nanowire electrodes by thin films of Pt alloys
    - Evaluate effects of Pt electrochemical surface area and loading on performance and durability of coaxial nanowire electrodes
  - Transport property measurements
    - Characterize Nafion/HOPI dispersions in by SANS
  - Multi-scale cell models that account for pore-level transport in GDLs, MPLs, and CLs

- Durability
  - Evaluate materials with differing durability using the newly developed H2/Air MEA AST and refine MEA AST
    - Durability test of 50cm$^2$ MEAs by integrating components developed by M2FCT under the newly developed M2FCT MEA AST
    - Durability study of baseline MEAs based on the Chemours NC700 and the newly developed M2FCT MEA AST
    - Assessment on the durability of MEAs with varying crack-based irregularities associated with catalyst and microporous layers
    - Performance and durability of MEAs using short stack testing platform including ECSA and impedance diagnostics
  - Quantify Ionomer/membrane durability
    - Perform micro-electrode and micro-cavity-electrode studies to illustrate role of ionomer in catalyst durability
    - Perform fatigue testing of membranes and MEAs (before and after durability testing) to assess membrane state of health
    - Screen performance and durability of membranes with a focus on hydrocarbon material integration
  - Modeling
    - Develop EIS model to track catalyst layer ionomer resistance (both H+ transport and O2 transport) during durability measurements
    - Detailed multiscale physics-based model of Pt dissolution and movement throughout the cell

# Summary

- Relevance/Objective
  - Optimize performance and durability of fuel-cell components and assemblies for heavy-duty applications
- Approach
  - Synergistic combination of modeling and experiments to develop materials, optimize component properties, behavior and phenomena
- Technical Accomplishments
  - Analysis of operating conditions, performance & efficiency for different systems
    - 175kW and 275 kW comparison
  - Durability measurements at projected heavy-duty loadings
    - Developed heavy-duty related MEA AST
    - Evaluated the role of catalyst layer cracks on catalyst durability
    - Improvements to membrane durability
    - Stabilized radical Scavengers; Preventing Fe migration with Crack free MPL.
  - Integration
    - Established baseline performance for 90k AST cycles for 3 catalysts
    - Showcased electrode structures to enable improved performance and durability
    - Mitigated crack formation across multiple material sets
    - Established methodology for ECA extraction from stacks
  - Material Developments
    - Higher mass activity/durable catalyst
    - Advanced ionomers and membranes for durable high-temperature operations
- Future Work
  - Develop the knowledge base and implement towards novel integration and improved durability and support FOA projects

# Who is M2FCT? National Lab Contributors

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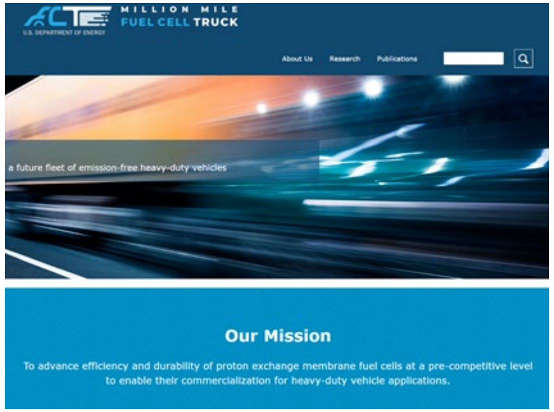
## Main Page

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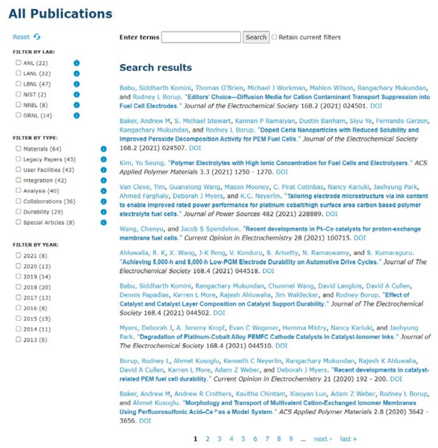


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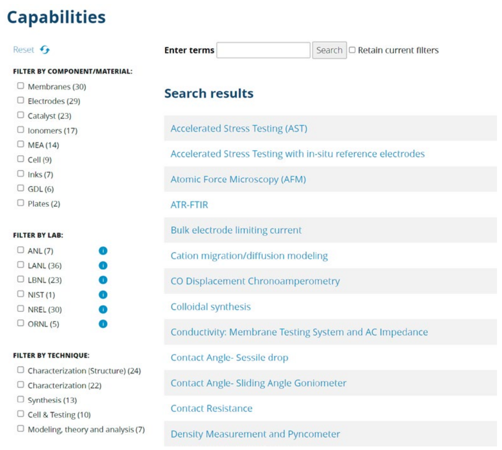
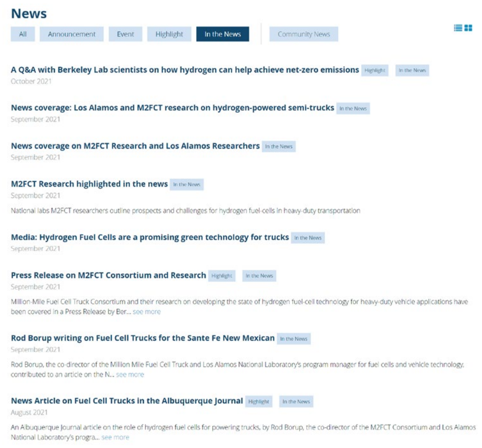



## News

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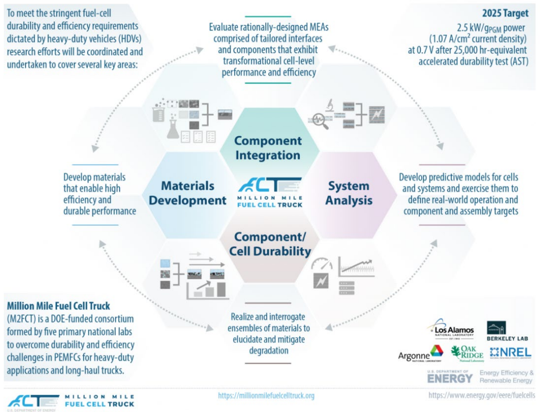
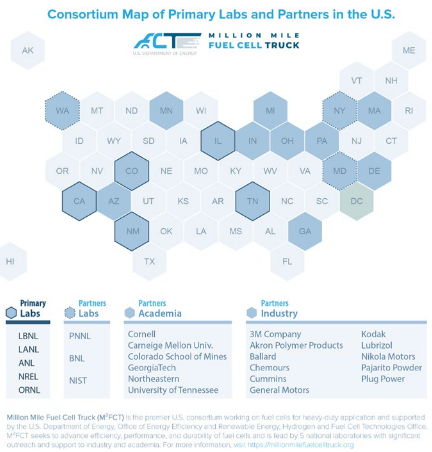
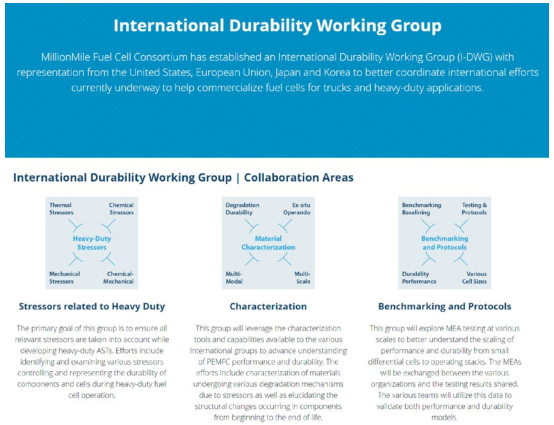


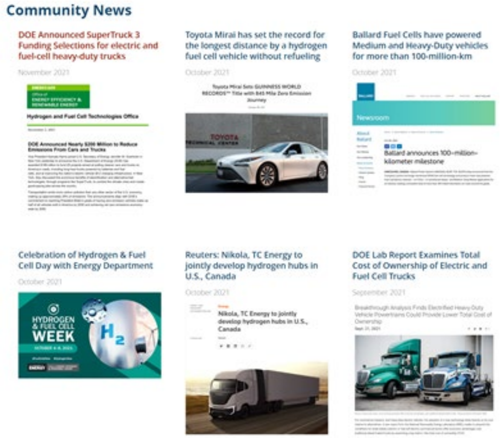

## People and Partners




## Outreach: Working Groups




## Community News



# Acknowledgements

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## 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