Project ID: FC339

MILLION MILE
FUEL CELL TRUCK

U.S. DEPARTMENT OF ENERGY

M2FCT: Million Mile Fuel Cell Truck Consortium

DOE Hydrogen Program
2021 Annual Merit Review and Peer Evaluation Meeting

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

JUNE 9, 2021

M2FCT Consortium - Overview & Relevance

Timeline

- Project start date: 10/01/2020
- Project end date: 09/30/2025

Budget

- FY20 project funding: $10M
  - Planned $1M external partners
  - $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
- 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

Project Goals and Targets

- 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 multicomponent 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
- **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

Target Comparison between Light - and Heavy -Duty

Condition	Traditional	M2FCT Focus
Operating temperature	60 - 80 ℃	~ 90 ℃
Catalyst	Random alloy $Pt_{90}Co_{10}$	Tailored and ordered alloys, annealed Pt
Membrane	Ultra-thin, reinforced with mobile Ce	Stabilized, durable, high selectivity (H+ conductance/H_2 permeance)
Operating voltage	0.6 – 0.9 V	>0.7 V
Durability	5,000 hrs	25,000 hrs
Pressure	150 kPa	250 kPa
Catalyst loading	0.15 g_{Pt}/cm^2	0.3 g_{Pt}/cm^2

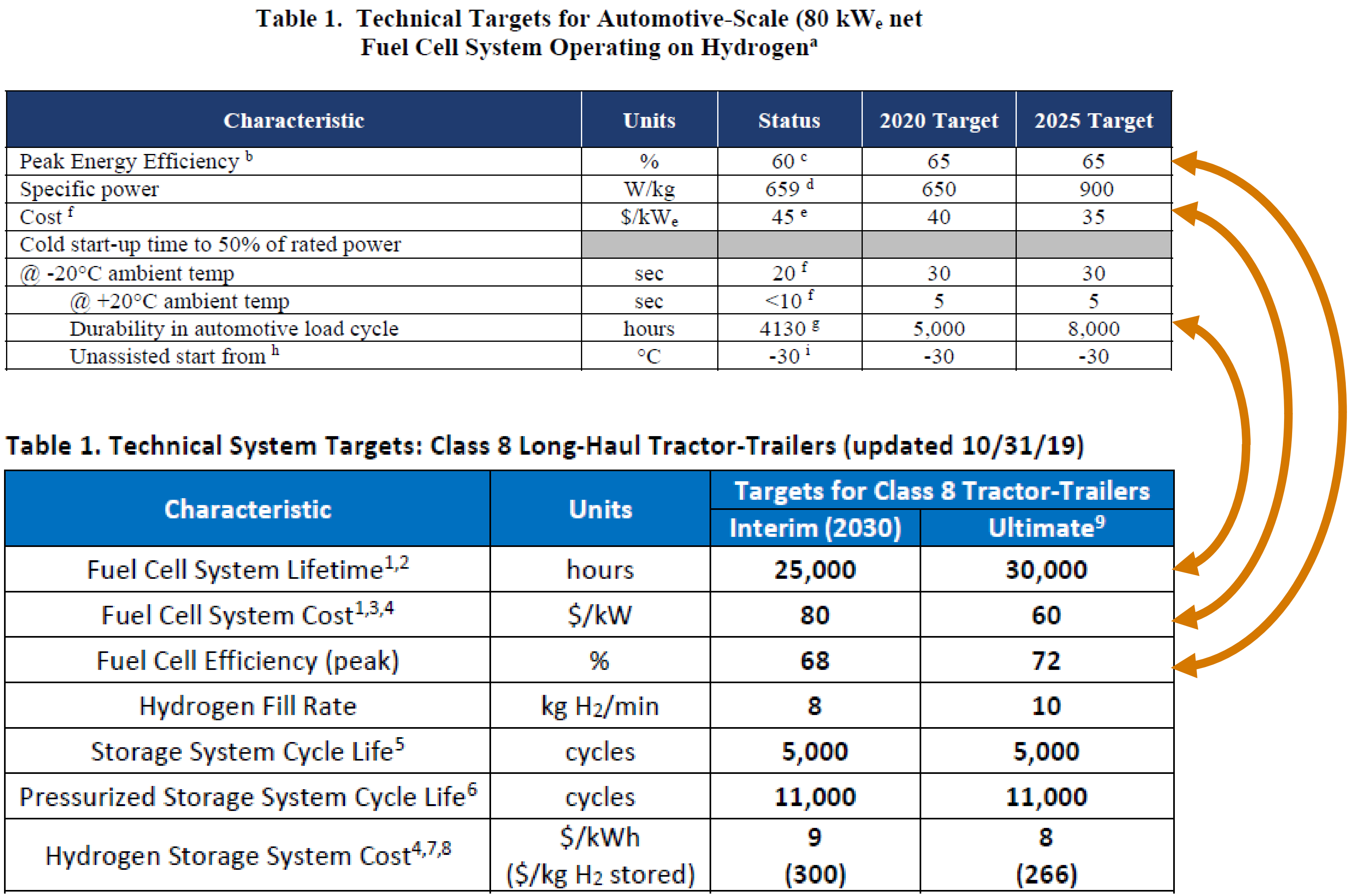


Table 1. Technical Targets for Automotive-Scale (80 kW$_e$ net Fuel Cell System Operating on Hydrogen[a]

Characteristic	Units	Status	2020 Target	2025 Target
Peak Energy Efficiency [b]	%	60 [c]	65	65
Specific power	W/kg	659 [d]	650	900
Cost [f]	\$/kW$_e$	45 [e]	40	35
Cold start-up time to 50% of rated power				
@ -20°C ambient temp	sec	20 [f]	30	30
@ +20°C ambient temp	sec	<10 [f]	5	5
Durability in automotive load cycle	hours	4130 [g]	5,000	8,000
Unassisted start from [h]	°C	-30 [i]	-30	-30

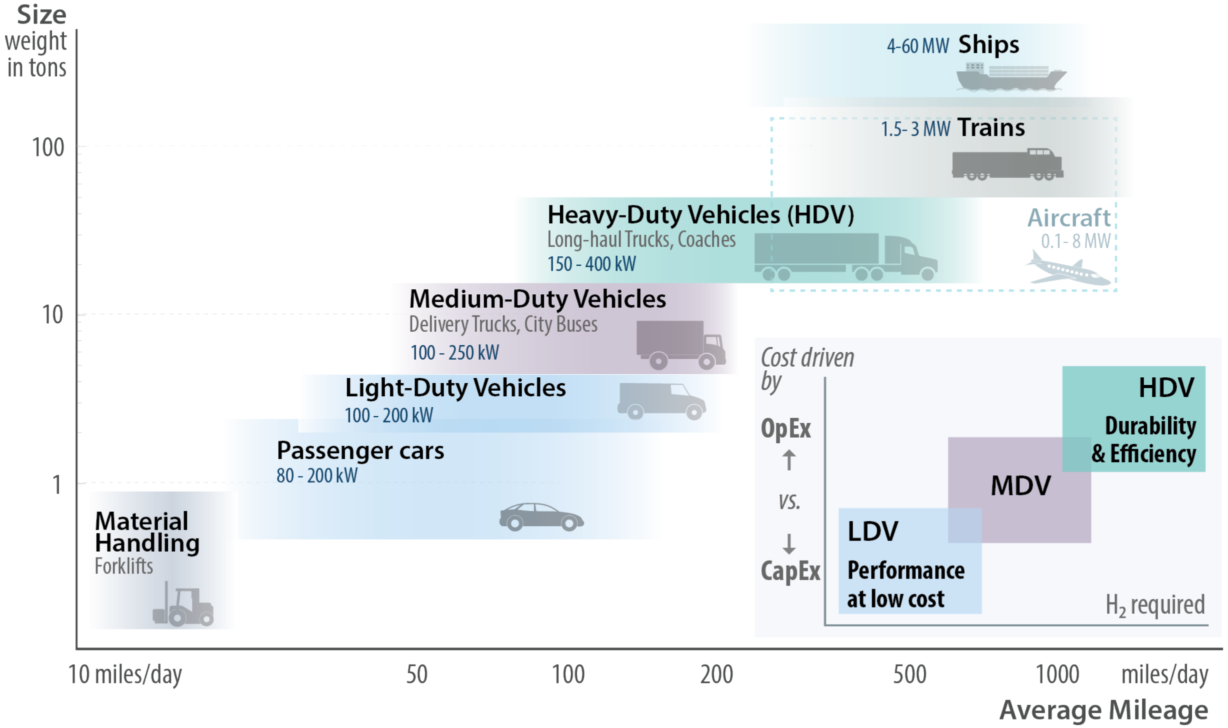
Table 1. Technical System Targets: Class 8 Long-Haul Tractor-Trailers (updated 10/31/19)

Characteristic	Units	Targets for Class 8 Tractor-Trailers	
		Interim (2030)	Ultimate[9]
Fuel Cell System Lifetime[1,2]	hours	25,000	30,000
Fuel Cell System Cost[1,3,4]	\$/kW	80	60
Fuel Cell Efficiency (peak)	%	68	72
Hydrogen Fill Rate	kg H_2/min	8	10
Storage System Cycle Life[5]	cycles	5,000	5,000
Pressurized Storage System Cycle Life[6]	cycles	11,000	11,000
Hydrogen Storage System Cost[4,7,8]	\$/kWh (\$/kg H_2 stored)	9 (300)	8 (266)

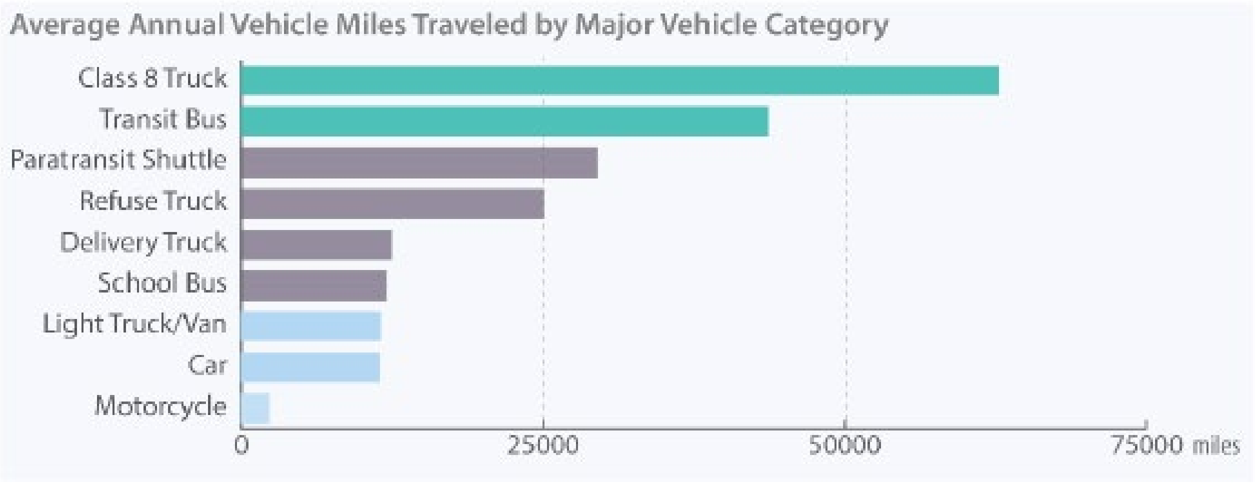
https://www,hydrogen.energy.gov/pdfs/19006_hydrogen_class8_long_haul_truck_targets.pdf

Target Comparison between Light - and Heavy -Duty

Hydrogen Fuel Cell Diversity in Transportation



Average Annual Vehicle Miles Traveled by Major Vehicle Category

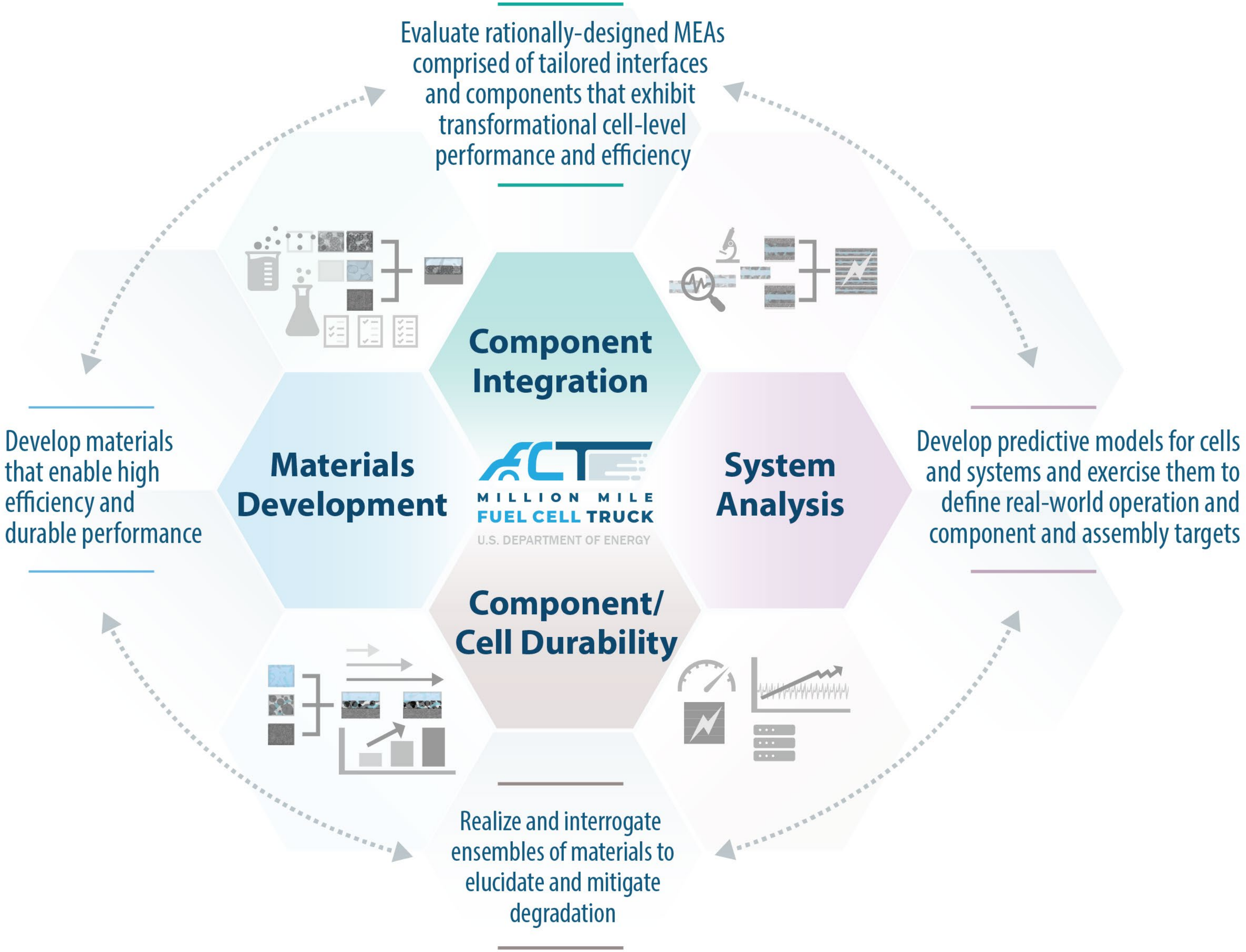


- Heavy-duty market is a critical market for reducing energy consumption and emissions
  - Medium- and heavy-duty trucks consume 25% of the total annual vehicle fuel use and produce 23% of the total CO_2 emissions in the US today
  - Annual freight truck miles traveled is projected to increase by 54% by 2050.
- Decades of development of cost-effective and durable polymer electrolyte membrane fuel cells needs to be leveraged to meet the increased efficiency and durability requirements of the HDV market

David A. Cullen, K. C. Neyerlin, Rajesh K. Ahluwalia, Rangachary Mukundan, Karren L. More, Rodney L. Borup, Adam Z. Weber, Deborah J. Myers & Ahmet Kusoglu, Nature Energy, 2021

M2FCT Approach

Million Mile Fuel Cell Truck (M2FCT) will tackle these challenges through a "team-of-teams" approach featuring main teams in analysis, durability, integration, and 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.



Evaluate rationally-designed MEAs comprised of tailored interfaces and components that exhibit transformational cell-level performance and efficiency

Component Integration

Develop materials that enable high efficiency and durable performance

Materials Development

System Analysis

Develop predictive models for cells and systems and exercise them to define real-world operation and component and assembly targets

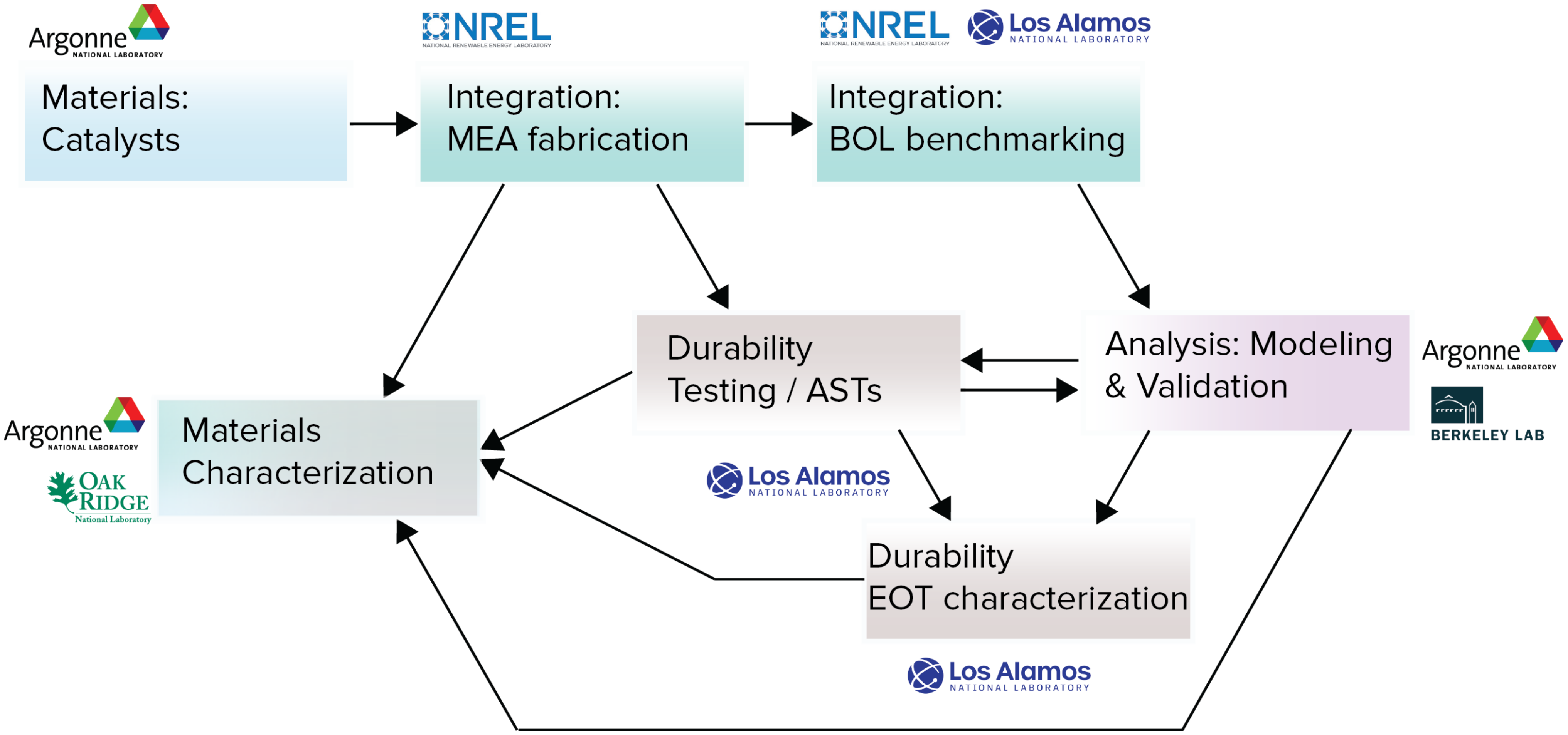
Component/ Cell Durability

Realize and interrogate ensembles of materials to elucidate and mitigate degradation

M2FCT Approach

Integrated consortium

- An example of material and knowledge transfer for catalysts (Materials Development)



Argonne NATIONAL LABORATORY

Materials: Catalysts

NREL NATIONAL RENEWABLE ENERGY LABORATORY

Integration: MEA fabrication

NREL NATIONAL RENEWABLE ENERGY LABORATORY
Los Alamos NATIONAL LABORATORY

Integration: BOL benchmarking

Durability Testing / ASTs

Los Alamos NATIONAL LABORATORY

Analysis: Modeling & Validation

Argonne NATIONAL LABORATORY
BERKELEY LAB

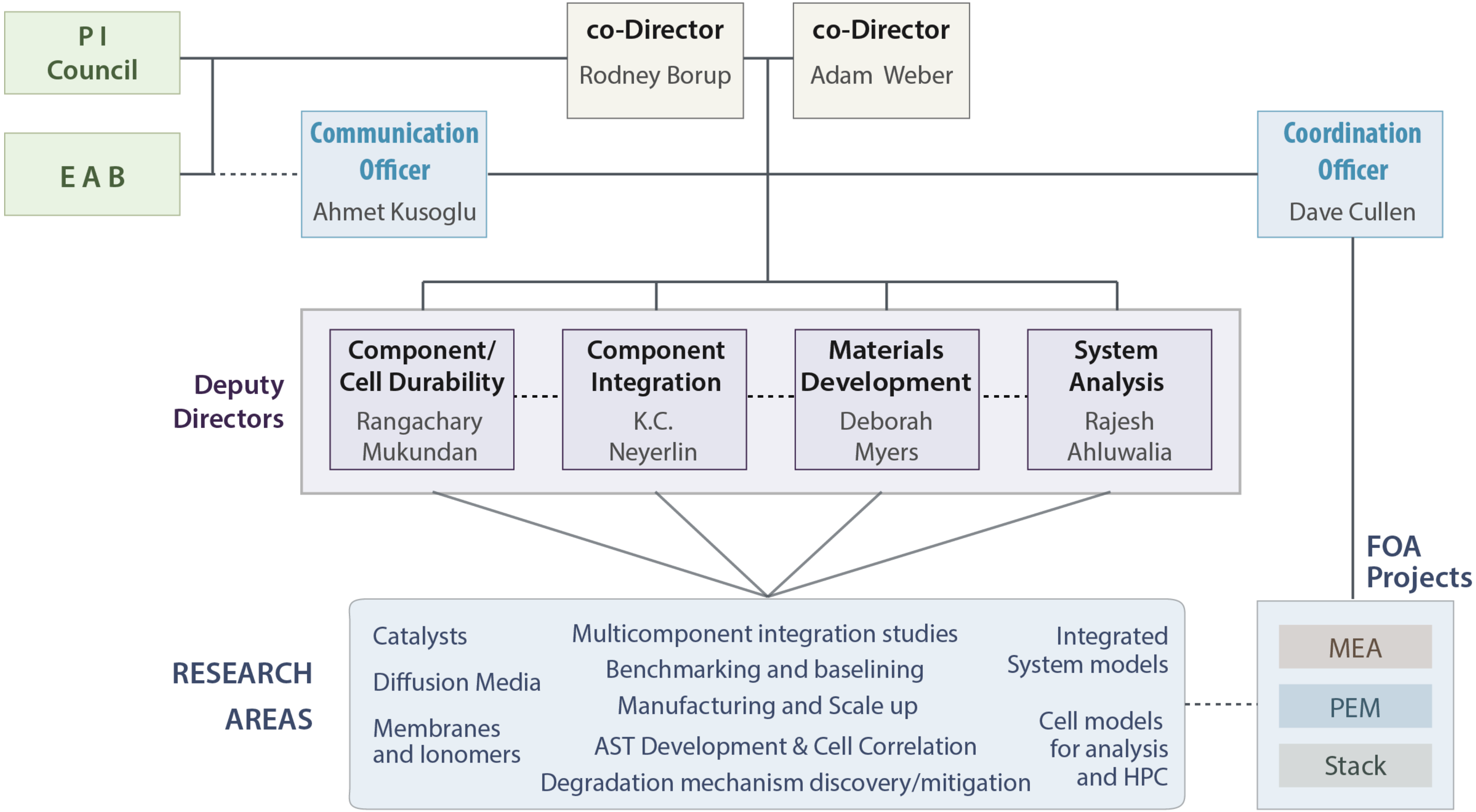
Argonne NATIONAL LABORATORY
OAK RIDGE National Laboratory

Materials Characterization

Durability EOT characterization

Los Alamos NATIONAL LABORATORY

Organization Chart



P I Council

E A B

Communication Officer
Ahmet Kusoglu

co-Director
Rodney Borup

co-Director
Adam Weber

Coordination Officer
Dave Cullen

Deputy Directors

Component/ Cell Durability
Rangachary Mukundan

Component Integration
K.C. Neyerlin

Materials Development
Deborah Myers

System Analysis
Rajesh Ahluwalia

FOA Projects

RESEARCH AREAS

Catalysts
Diffusion Media
Membranes and Ionomers

Multicomponent integration studies
Benchmarking and baselining
Manufacturing and Scale up
AST Development & Cell Correlation
Degradation mechanism discovery/mitigation

Integrated System models
Cell models for analysis and HPC

MEA
PEM
Stack

EAB: External Advisory Board
Amy M. Adams– 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- Hyzon

M2FCT Partners

HD MEA Projects

gm

Carnegie Mellon University

NIKOLA™

HD Membrane Projects

Lubrizol

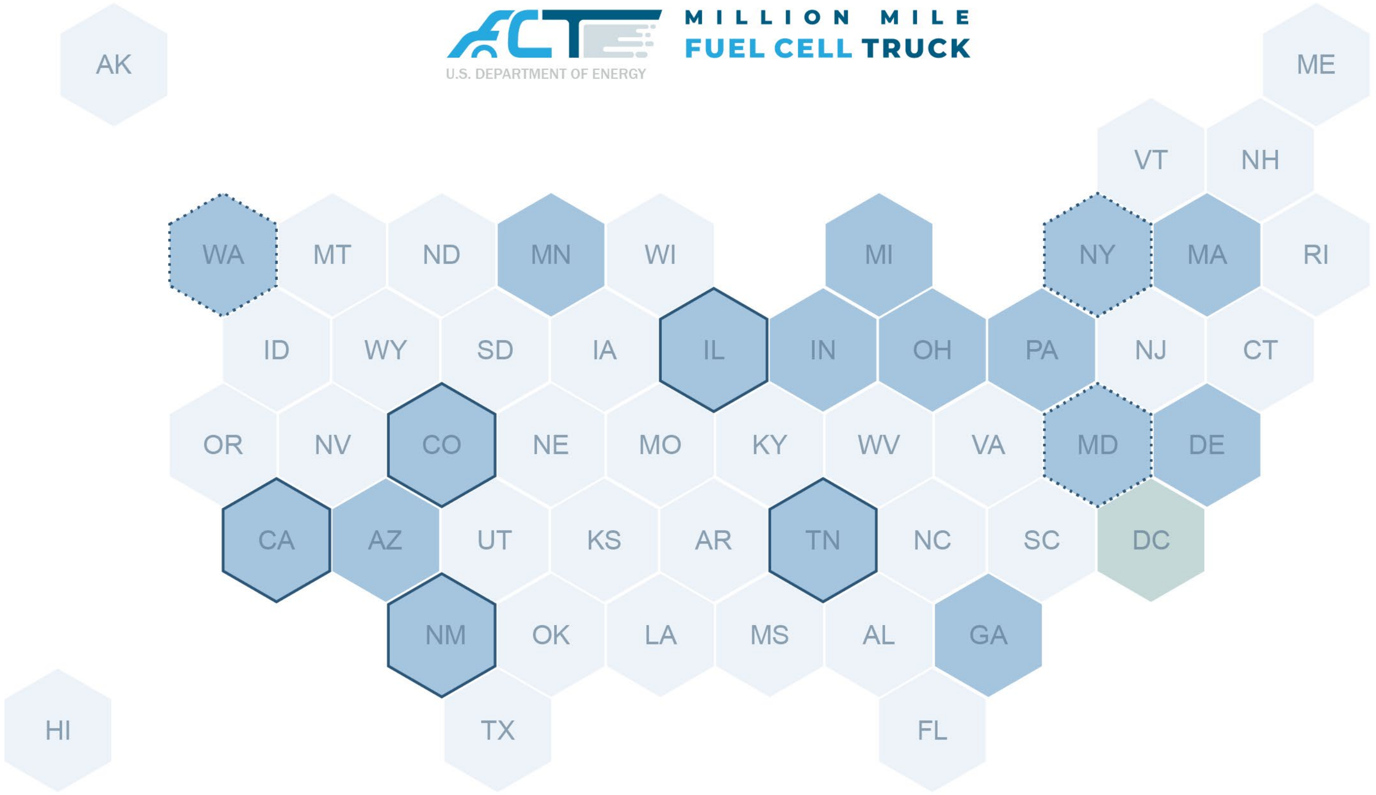
3M

NIKOLA™

HD Stack Projects

Cummins

PLUG POWER



Primary Labs

- LBNL
- LANL
- ANL
- NREL
- ORNL

Partners Labs

- PNNL
- BNL
- NIST

Partners Academia

- Cornell
- Carneige Mellon Univ.
- Colorado School of Mines
- GeorgiaTech
- Northeastern
- University of Tennessee

Partners Industry

- 3M Company
- Akron Polymer Products
- Ballard
- Chemours
- Cummins
- General Motors
- Kodak
- Lubrizol
- Nikola Motors
- Pajarito Powder
- Plug Power

Main Laboratories

Los Alamos NATIONAL LABORATORY

BERKELEY LAB

OAK RIDGE National Laboratory

NREL NATIONAL RENEWABLE ENERGY LABORATORY

Argonne NATIONAL LABORATORY

Affiliate Laboratories

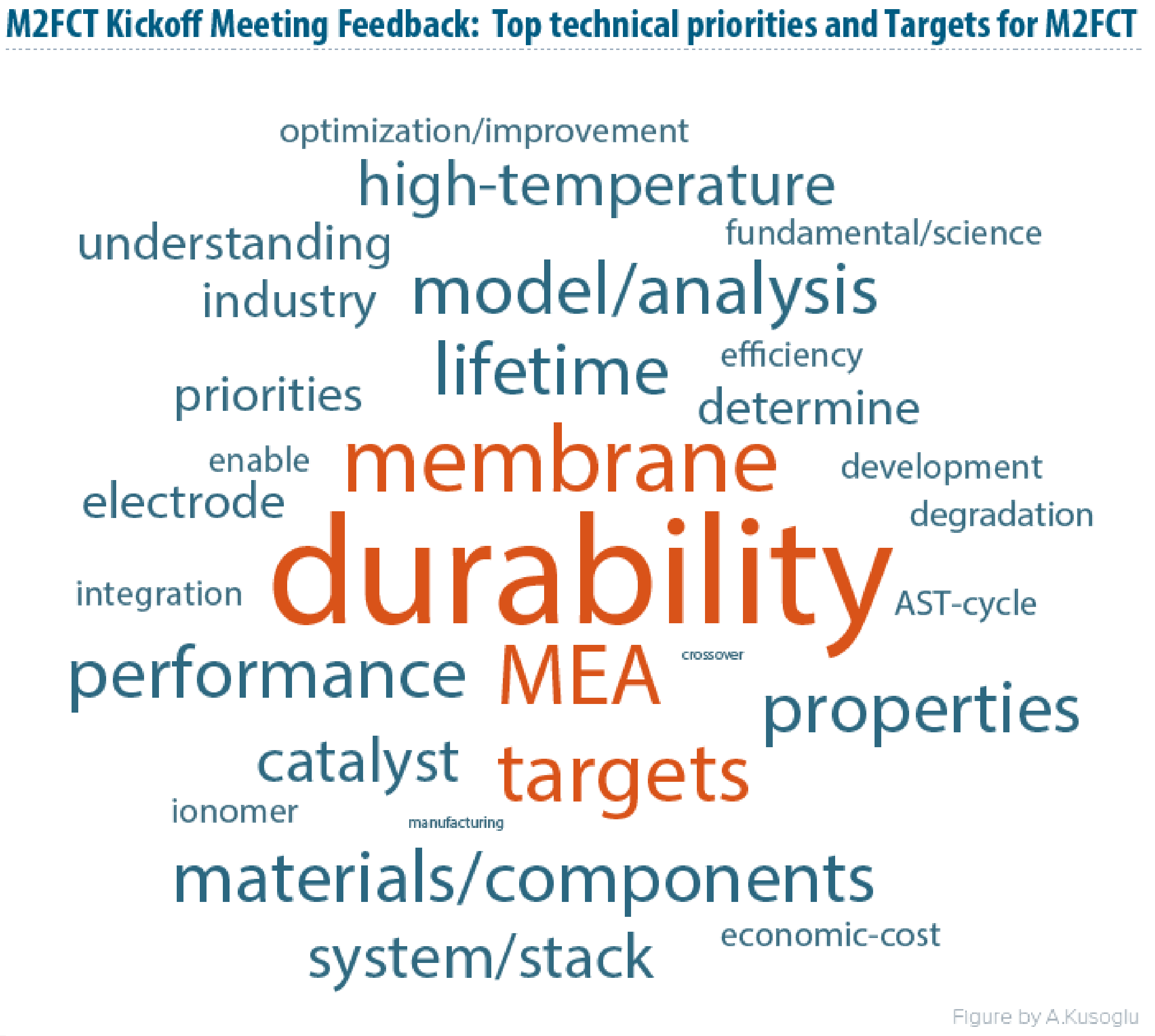
Pacific Northwest NATIONAL LABORATORY

BROOKHAVEN NATIONAL LABORATORY

NIST National Institute of Standards and Technology

M2FCT Kickoff Meeting Feedback

Top technical priorities represented as Word Cloud



M2FCT Kickoff Meeting Feedback: Top technical priorities and Targets for M2FCT

Figure by A.Kusoglu

Font sizes proportional to responses

M2FCT Initial Priorities

1. Analysis ($1.325M)
   1. Integrated system models
   2. Cell models for analysis
   3. Machine Learning/HPC/AI

Highest Emphasis
year 1

2. Materials development ($0.900M)
   1. Catalysts
   2. Membranes and ionomers

Lowest Emphasis
year 1

3. MEA Integration ($2.395M)
   1. Multicomponent integration studies
   2. Benchmarking and baselining
   3. Manufacturing

Medium Emphasis
year 1
(Higher Benchmarking)

4. MEA Durability ($2.755M)
   1. AST Development/cell correlation
   2. Degradation mechanism discovery/mitigation

High Emphasis
year 1

5. Other ($2.625M)
   1. FOA support
   2. Management & Communication
   3. Discretionary funding for new concepts

For funding non-core labs and academia to fill identified portfolio gaps

Work, focus, and specific funding will change over the years

Year 1 Milestones

Establish consistent protocols and baselines between integration and durability that interface with analysis

Milestone Name/Description	Quarter	Status
Work statements finalized with industrial partners related to supporting HFTO-funded heavy-duty projects. (LBNL, LANL, ORNL, ANL, NREL)	Q1	☑
Voltage-loss breakdown methodology developed and utilized in analyzing modeling results for different operating temperatures and humidity. (LBNL, ANL)	Q2	☑
With DOE AST working group, propose new MEA durability ASTs incorporating relevant degradation mechanisms for catalyst, support, electrodes and membrane in a single AST. (LANL, ORNL, ANL, NREL)	Q2	☑
Acceptable transition metal loss from alloy catalysts (% of sulfonic acid sites in ionomer layer) defined with respect to electrode layer losses. (LBNL, LANL, ORNL, ANL, NREL)	Q3	
Establish benchmark performance, durability, and cost of a state-of-the-art MEA in relation to the DOE target of 2.5 kW/g_{PGM} power (1.07 A/cm^2 current density) at 0.7 V after 90,000 cycles of current catalyst AST. (LBNL, LANL, ORNL, ANL, NREL)	Q4	
High throughput, automated particle size/composition measurement demonstrated on MEA cross section encompassing >1000 particles. (ORNL)	Q3	
Deliver MEA for evaluation at NREL with cathode catalyst layer ionomer EW gradient (dual slot-die-coated on both membrane and GDL) with low EW at membrane interface and higher EW at MPL interface that exceeds 1.07 A/cm^2 at 0.7 V. (ORNL, NREL)	Q4	
Demonstration of concept using post-loading of metal oxides onto catalysts synthesized by LANL catalyst team, with catalyst ECSA of no less than 75% of baseline (carbon supported Pt based catalysts). (PNNL)	Q4	
Evaluate performance of intermetallic PtMN/C catalysts in comparison with benchmark performance and durability of a state-of-the-art MEA using commercial catalyst/support in relation to the DOE target of 2.5 kW/g_{PGM} power (1.07 A/cm^2 current density). (BNL, LANL, ANL, ORNL, NREL)	Q4	

Catalyst Materials Development Sub-Tasks

Q6 3/31/2022 Go/No-Go Can be extended to 24 (Q8) months as no-cost extension

Demonstrate $\geq$ 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

Communication & Outreach

Vision: establish M2FCT as the premier research consortium not only at the research front but also for disseminating knowledge

- Outreach
  - Company visits and interactions, international coordination
  - Establish the website and consortium as the go-to place for fuel cells especially for HDV, beyond mere data and publication output
  - Monthly AST Working Group meetings

- In-reach
  - Internal newsletter, use uniform data procedures, storage, and trackers
  - Weekly technical meetings by technical area
  - Biweekly steering committee meetings

- Bring together the fundamentals of fuel-cell research and technology implications

Communication Officer: Ahmet Kusoglu (Berkeley Lab)

Inclusion, Diversity, Equity, Accountability

- Diverse group of researchers within the consortium
- Work with disadvantaged communities including HBCUs, HSIs, community colleges
  - Have various internships for students and targeted discretionary funding
  - Existing collaboration with NNSA to enhance STEM background & education
    - Including set-up of electrochemistry systems at HBCU’s
    - Training, access to cutting-edge research, use of state-of-the-art facilities


Hands-on research


Short Courses/Trainings


On-site support

- Disadvantaged neighborhoods will be favorably impacted with improvements to long-haul trucking corridors and heavy-duty centers (*e.g.*, ports)*
  - Greening of the transportation will greatly improve their local emissions and air and noise pollution

* References: Preble et al. Environ. Sci. Technol. 2019, 53, 24, 14568–14576; Dallman et al. Environ. Sci. Technol. 2013, 47, 23, 13873–13881

M2FCT Consortium

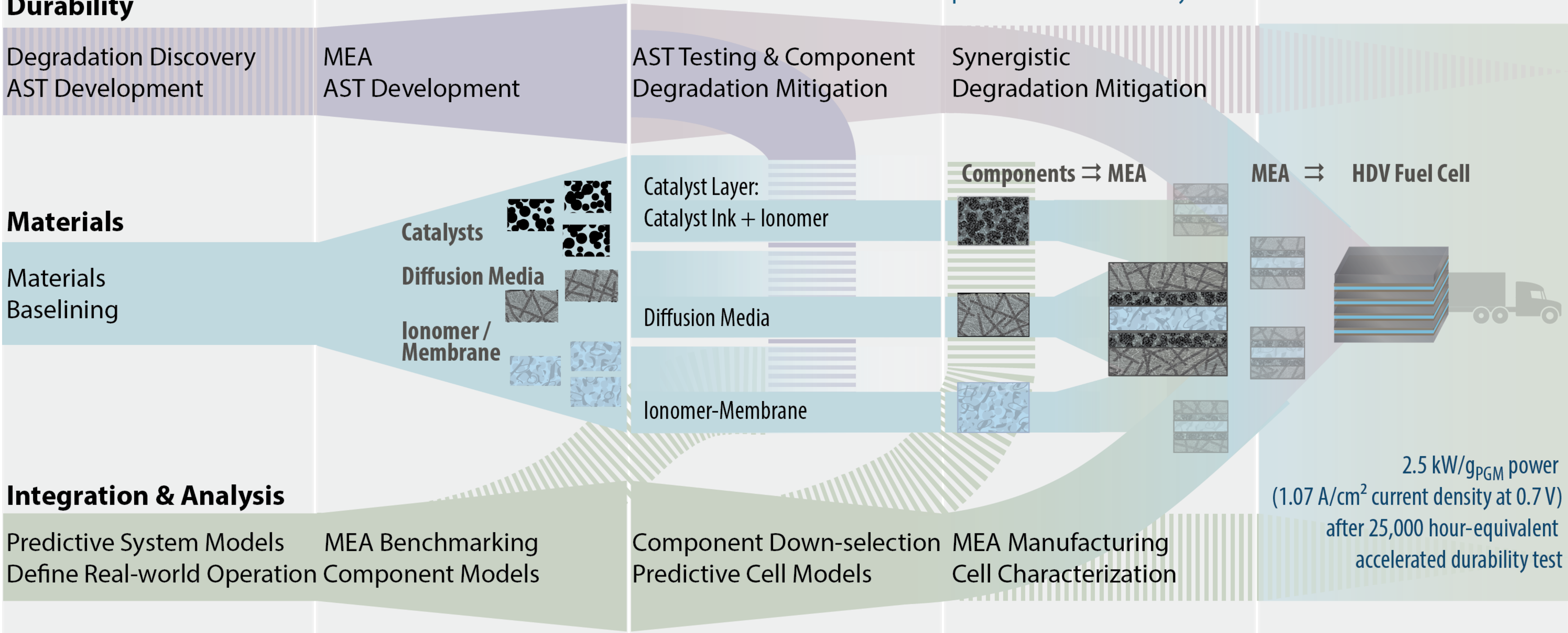
MILLION MILE FUEL CELL TRUCK
U.S. DEPARTMENT OF ENERGY

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

Catalyst Layer:
Catalyst Ink + Ionomer

Diffusion Media

Ionomer-Membrane

Components ⇉ MEA

MEA ⇉ HDV Fuel Cell

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

Integration & Analysis

Predictive System Models
Define Real-world Operation

MEA Benchmarking
Component Models

Component Down-selection
Predictive Cell Models

MEA Manufacturing
Cell Characterization

Establishing Benchmark Material Discovery — Year 1

Material Synthesis and Development for Efficiency — Year 2

Materials Selection, Optimization for Efficiency & Durability — Year 3

Integrated Assembly Testing and Optimization — Year 4

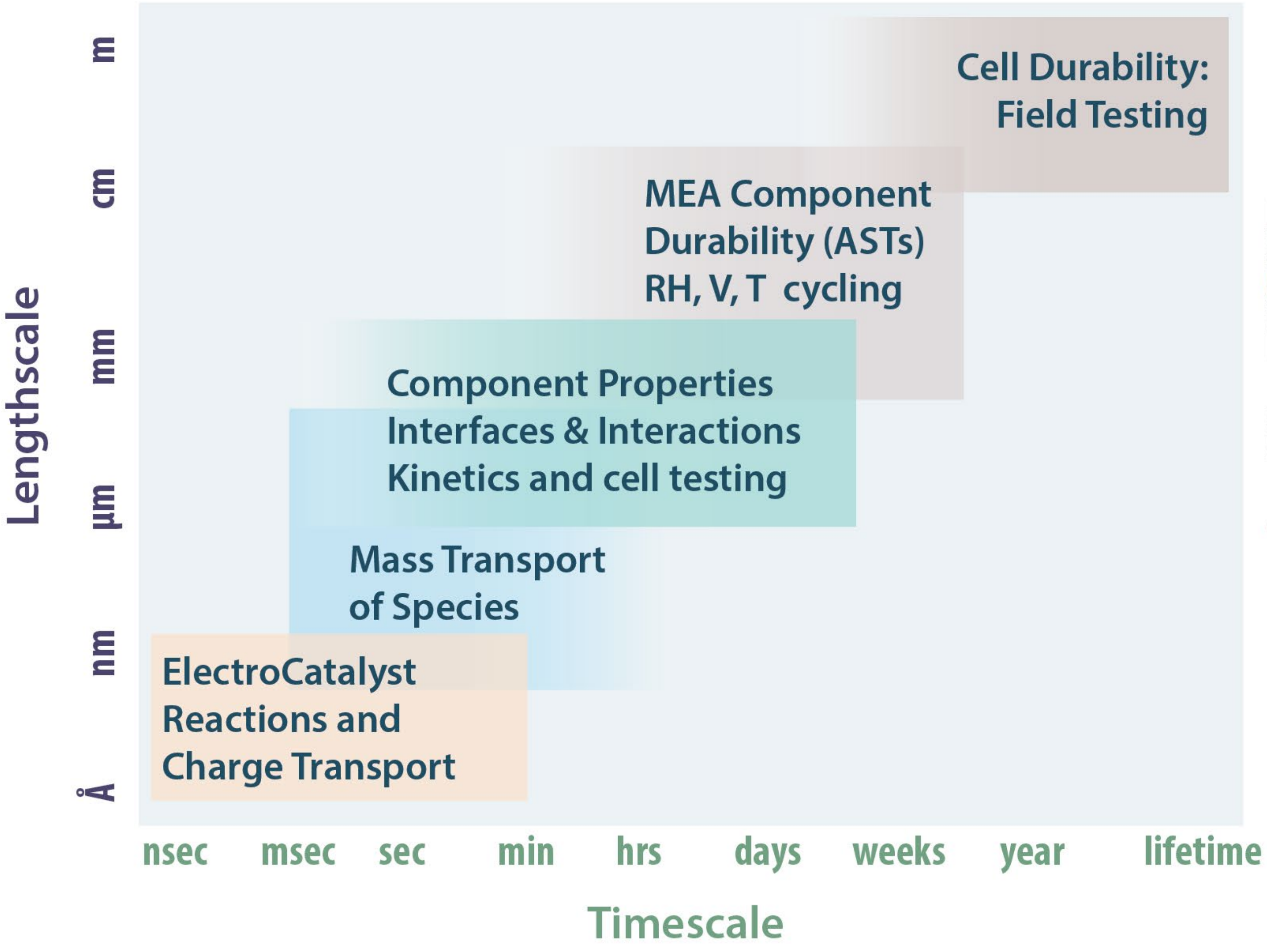
Cell Efficiency and Durability — **Final Target** — Year 5

M2FCT Approach Across Length and Timescale

material development integration, characterization

performance, durability efficiency optimization

fundamental understanding ⟷ technology improvement



images: LANL, NIST

image: ANL

M2FCT Advanced Characterization Approach

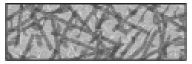
Bipolar plates

Contact Resistance, XPS

Elemental analysis, Coating structure

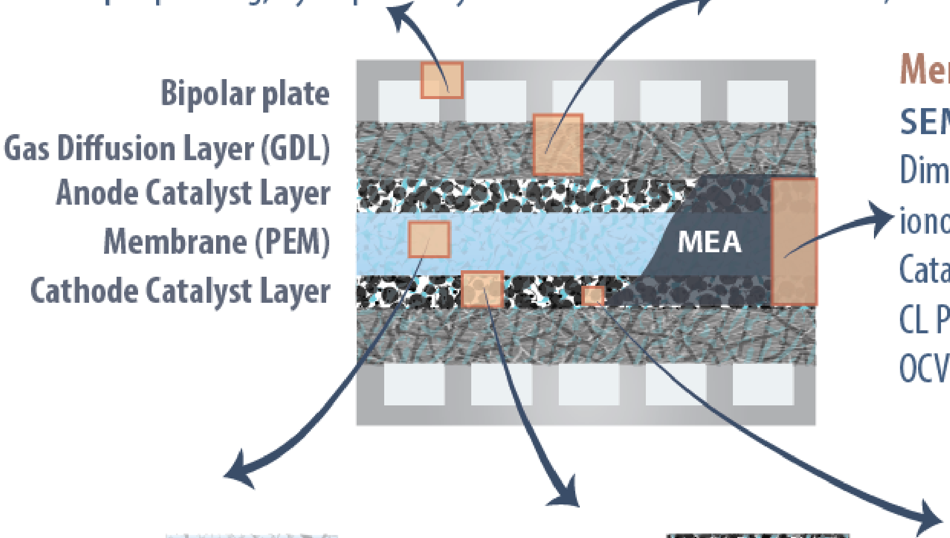
XPS - Depth profiling, Hydrophobicity

GDLs



MIP, BET, XCT, Contact Angle

Porosity, surface area/pore-size distribution, Component Hydrophobicity



Membrane Electrode Assembly (MEA)

SEM/EDAX, TEM, XRF, XCT, Cell Test/ASTs

Dimensions, composition, structure, cross-sections

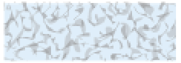
ionomer mapping, Elemental quantitation & mapping

Catalyst particle size and distribution, and transport

CL Performance (O_2/Air Polarization) and durability

OCV decay, Potential Cycling, Accelerated Stress Testing

Ionomer Membrane



FTIR, NMR, Titration, XRF, XRD

DVS, MTS, DMA, SAXS, XAS

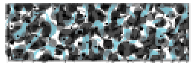
Composition, Eq. weight, Additives

Conductivity, Water & Gas Transport

Mechanical-Chemical Stability

Nanostructure, Morphology, Interface

Catalyst Layer



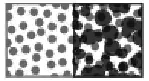
TGA-MS, XCT, AFM, CV

catalyst wt %, I/C ratio, porosity

ionomer-ink interactions, coverage

agglomerates, ECSA and loss

Catalyst and Catalyst Support



SAXS, EXAFS, XRD, NDIR, AST

catalyst particle size and distribution

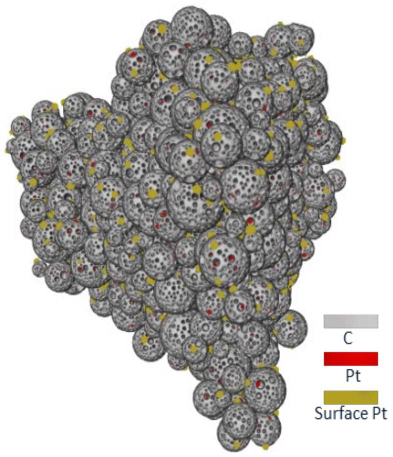
Pt-Pt, Pt-Co bonding distances, Catalyst durability

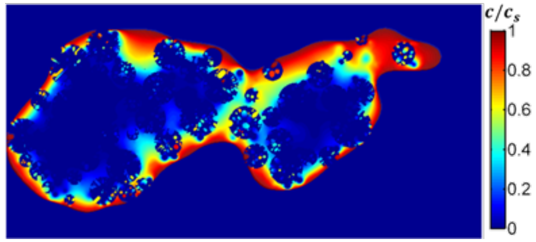
Catalyst support durability, Carbon corrosion

Figure by A.Kusoglu

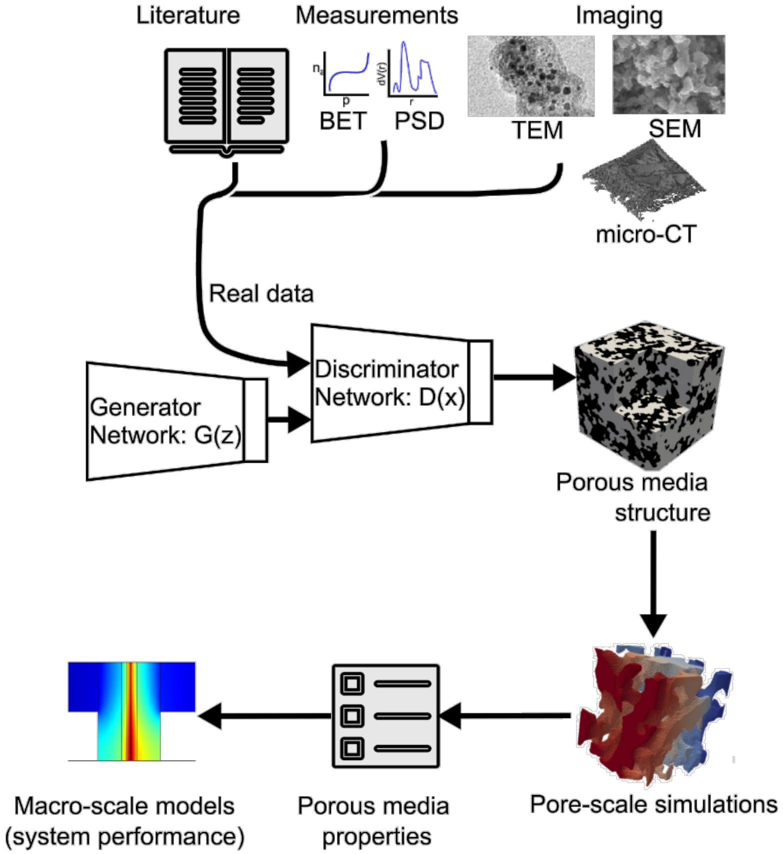
M2FCT Advanced Computing Approach

High performance computing for hi-fidelity and multiscale simulations



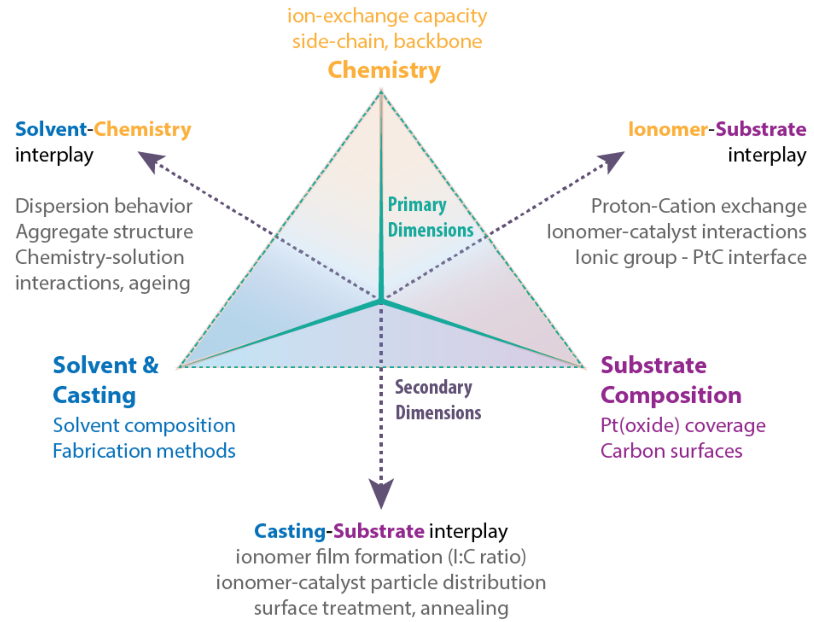


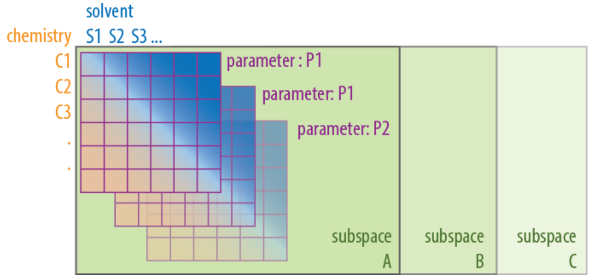
Use HPC to provide data sets for ML studies



Create a "Virtual Fuel Cell" digital twin

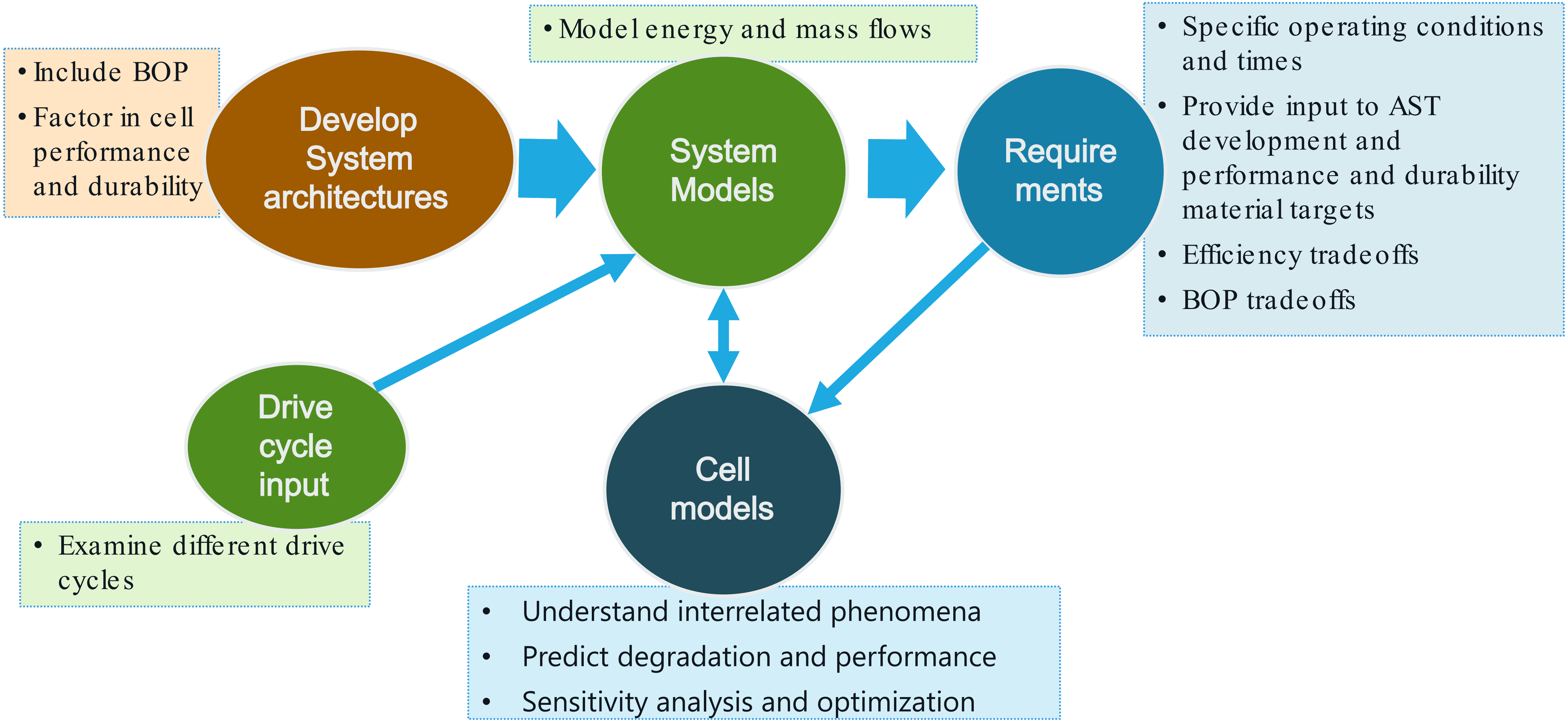
Analyze and collect data sets of membrane properties for ML studies





Analysis

Use cell models and system analysis to inform research



- Include BOP
- Factor in cell performance and durability

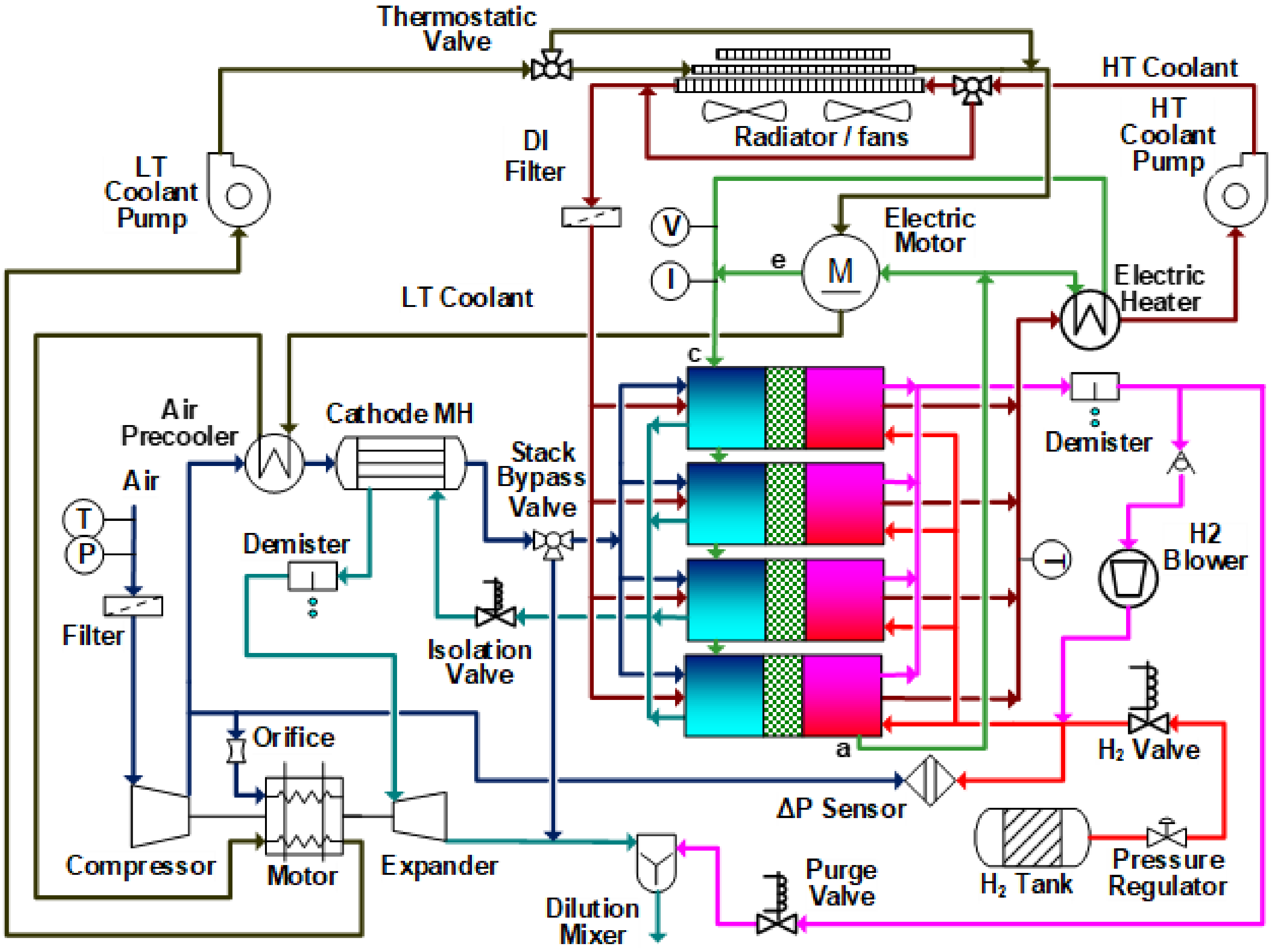
- Model energy and mass flows

- Specific operating conditions and times
- Provide input to AST development and performance and durability material targets
- Efficiency tradeoffs
- BOP tradeoffs

- Examine different drive cycles

- Understand interrelated phenomena
- Predict degradation and performance
- Sensitivity analysis and optimization

Fuel Cell Systems for Medium and Heavy -Duty Trucks



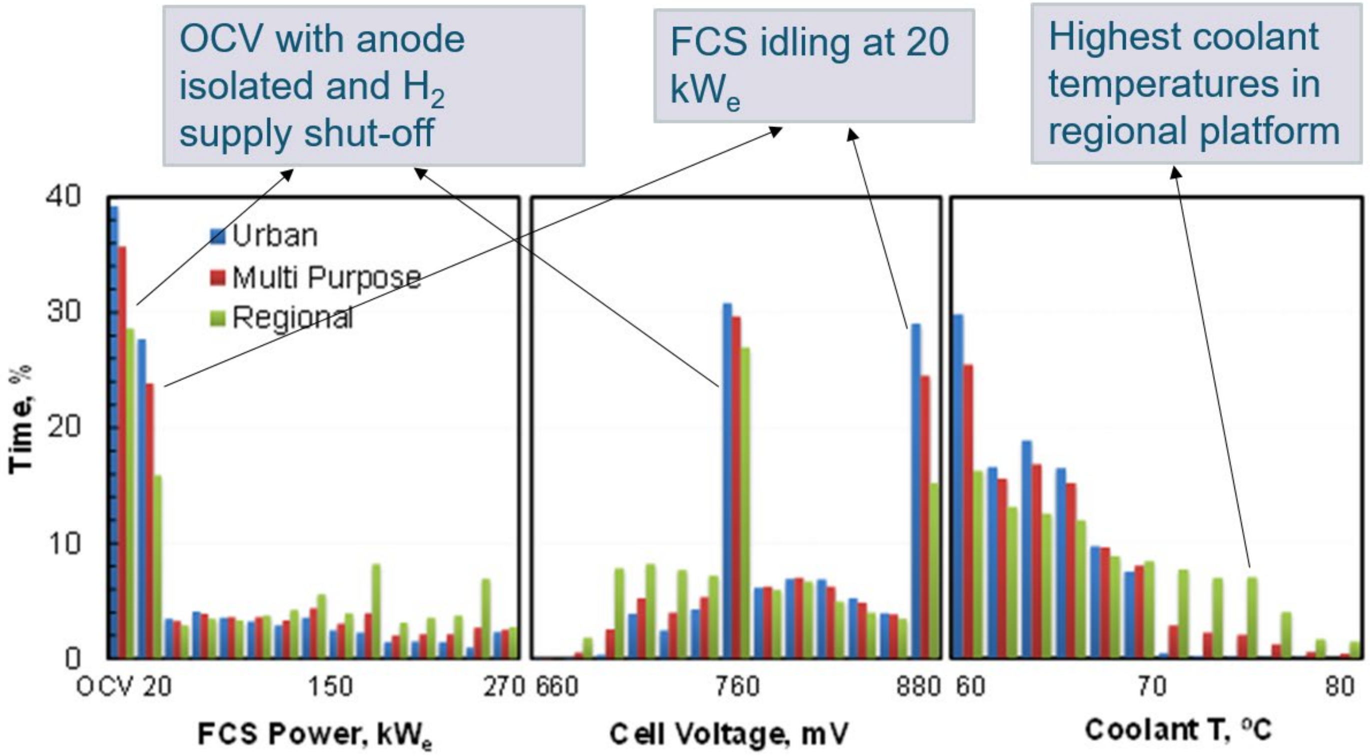
Salient Features

- 275 kW net (35-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 (TBD), 0.05 mg_{Pt}/cm^2
- Membrane: 14 mm, chemically stabilized, mechanically reinforced
- Single air system with expander
- Single anode system with recirculation blower
- Cathode humidifier: No (TBD)
- Rated power conditions at EOL: 2.5 atm, 87-95ºC, 660-700 mV
- Control valves for startup and shutdown, cold start and OCV

David A. Cullen, K. C. Neyerlin, Rajesh K. Ahluwalia, Rangachary Mukundan, Karren L. More, Rodney L. Borup, Adam Z. Weber, Deborah J. Myers & Ahmet Kusoglu, Nature Energy, 2021

Simulated Heavy -Duty Fuel Cell Operation (Class 8 Truck)

Operating Times and Conditions for HDV


OCV with anode isolated and H_2 supply shut-off

FCS idling at 20 kW_e

Highest coolant temperatures in regional platform

Rated power conditions:

- 275-kW FCS
- 0.7 V cell voltage
- 90℃ coolant exit temperature*
- 2.5 atm inlet pressure

*only achieved under sustained high power hill climb

FCS-HDV Duty Cycle Simulation

- Power demand from Autonomie [1] for three HDV vocations on EPA fuel economy cycles
- Conducted drive cycle simulations for fuel cell dominant power train with battery for regenerative breaking, hill climb, and power demand exceeding 275 kW_e
- 20-kW_e FCS idle power clips cell voltage to 880 mV
- OCV controlled to 760 mV by shutting off H_2 supply when FCS power demand is below 20 kW_e
- High temperatures limited by operating fan when coolant exit T exceeds the set target (65°C)
- **Next steps:** Harmonize duty cycles with the 21st Century Truck Partnership

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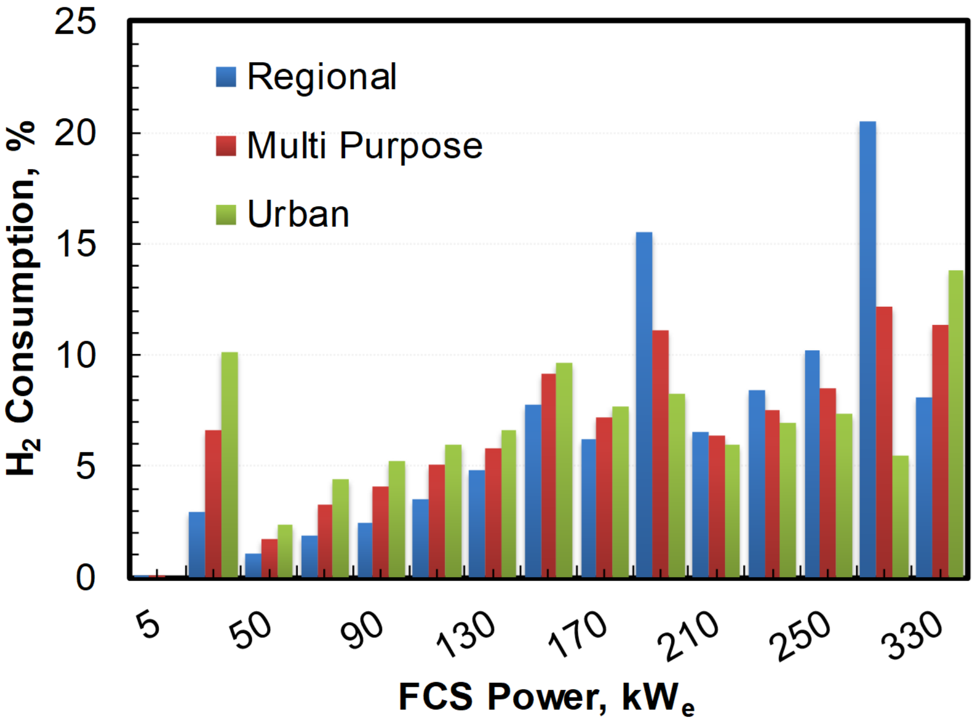
[1] Autonomie simulation results supplied by Ram Vijayagopal, ANL

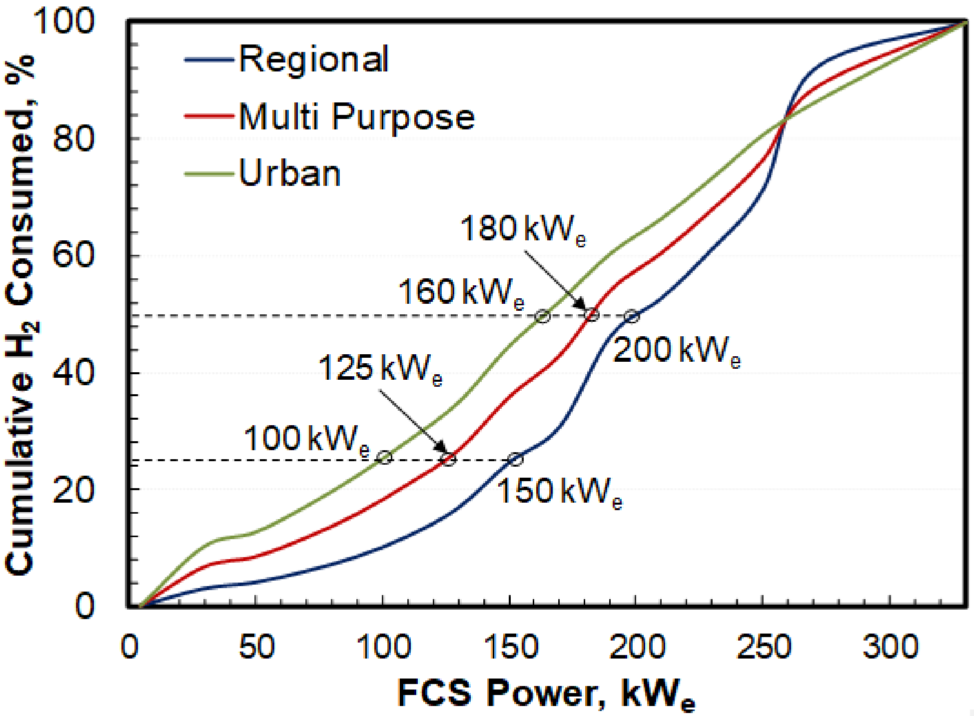
FC System Energy Consumption at Different Power Levels

Key to improving fuel **economy** of Class-8 vehicles is to focus on performance at rated power

- Slightly more H_2 is consumed near rated power in regionals than in multi-purpose and urban vocations
  - H_2 consumption decreases at lower power
  - Peak occurs at FCS idle power

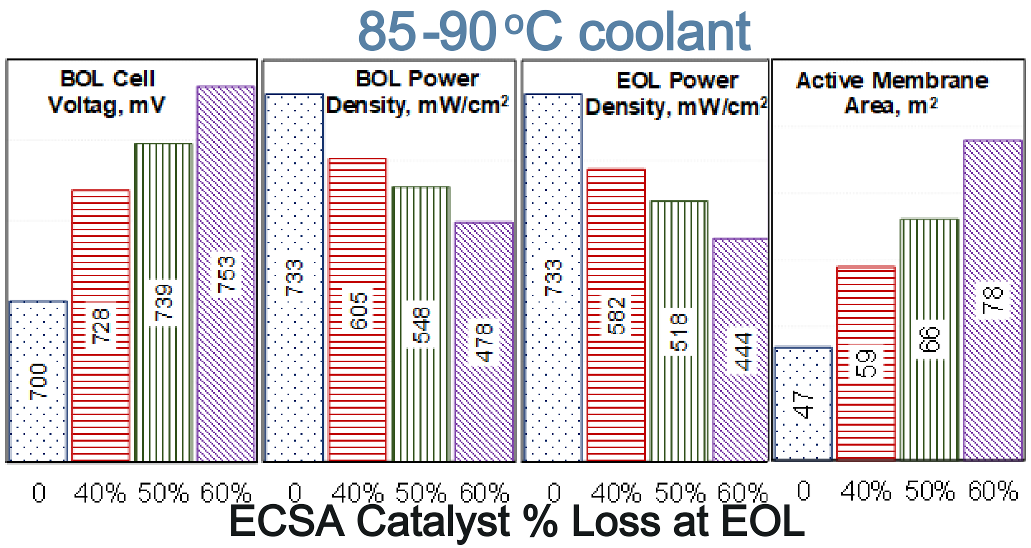


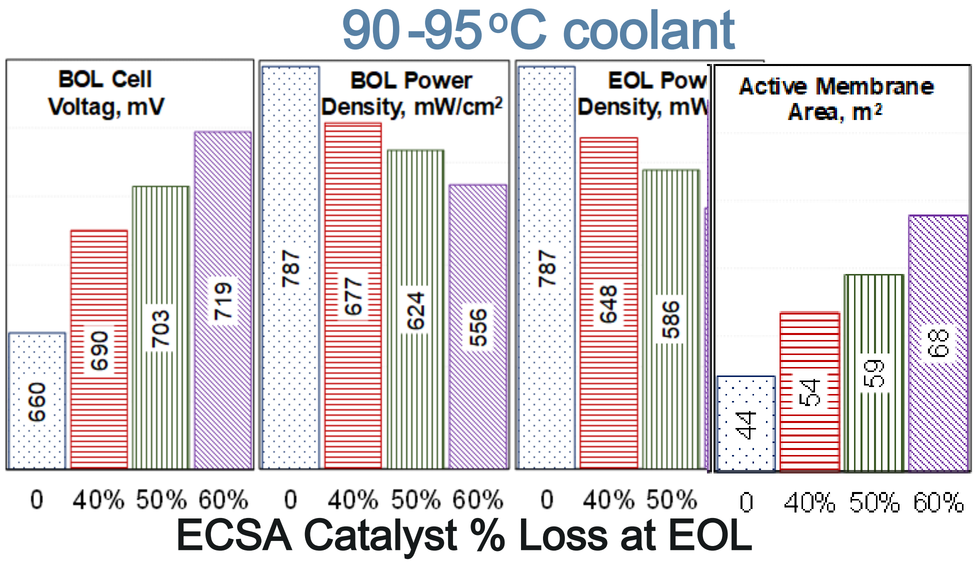
- 50% of H_2 is consumed above 160 – 200 kWe FCS power
- 75% of H_2 is consumed above 150 kWe for regionals, 125 kWe for multi-purpose, and 100 kWe for urban



FCS-HDV Degradation Adjusted Stack Size

Lifetime defined by ECSA loss as marker of aging (275 kW$_e$ at EOL)





- Stack size defined by efficiency target
  - Stack coolant exit temperature at peak power
- Stack active area defined for various catalyst degradation for 700 mV (EOL cell voltage)

Modeled Electrode Degradation Mechanisms

- ECSA loss due to Pt dissolution and growth in particle size
- Degradation in ORR kinetics
- Increase in oxygen transport resistance
- Change in Pt accessibility

- Relaxing EOL cell voltage to 660 mV decreases the initial stack active area by 10-15%
  - Stack coolant exit temperature at peak power, ~ 50 °C higher

2.5 atm, 1.5 SR(c), 0.3 mg/cm^2 Pt, a-Pt/HSC cathode catalyst

Reaching Peak Efficiency Targets: Catalyst Activity and Operating Conditions

- Calculated cell voltages required to reach the target peak efficiencies
  - 860 mV for 63% efficiency
  - Interim: 910 mV for 68% efficiency
  - Ultimate: 956 mV for 72% efficiency
- Determine catalyst mass activities required to reach the target peak efficiencies
  - 0.3 A/mg-Pt for a-Pt/C with 0.25 mg-Pt/cm^2 loading
  - Interim: 1.5 A/mg-Pt (5X status)
  - Ultimate: 7.5 A*/mg-Pt (25X status)

Methods to Reach Target Efficiencies

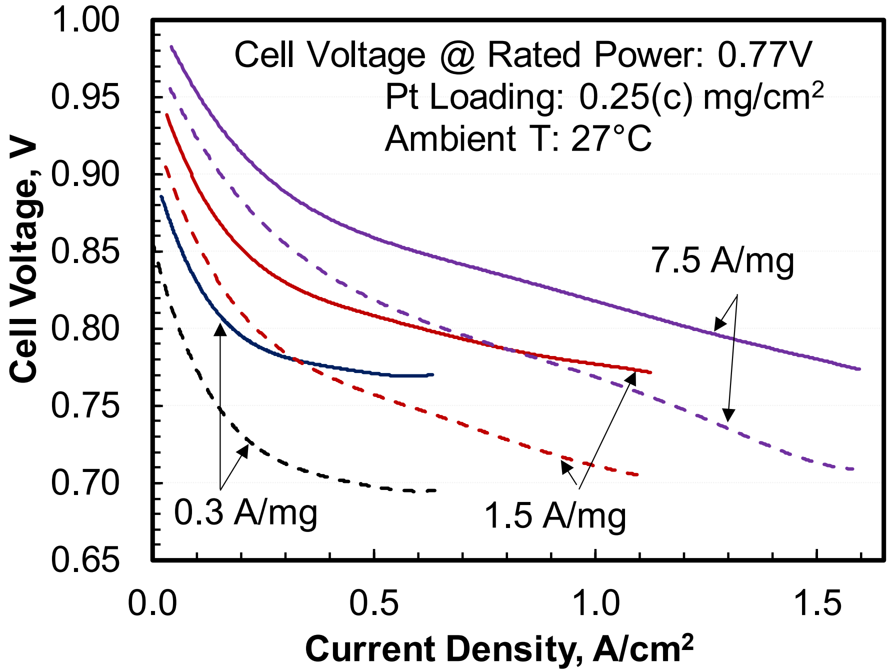
- Increase catalyst MA (new catalysts)
- Low-λ content membranes (new ionomers)
- Increase OCV (decrease H_2/O_2 cross-over)
- Increase kinetics (increase operating temp.)

Peak Efficiency Metrics	Status	Interim	Ultimate
CEM Turndown	10	20	20
Parasitic Losses, %	5	3.4	3.3
Current Efficiency, %	94.7	95.7	96.8
BOL Peak Efficiency, %	63.2	68.5	72.3
EOL Peak Efficiency, %	59.2	65.7	70.3
BOL Cell Voltage, mV	860	910	956
EOL Cell Voltage, mV	809	878	933
BOL Current Density, mA/cm^2	53.4	67.5	91.4
EOL Current Density, mA/cm^2	51.2	63.2	84.5

$$\eta_s = (1-\zeta)\eta_i\left(\frac{E}{\Delta H/F}\right)$$

η_s: Peak efficiency;
ζ: Parasitic loss;
η_i: Current efficiency;
E: Cell voltage;
ΔH: Lower heating value

*T = 65°C
Ambient Outlet Pressure

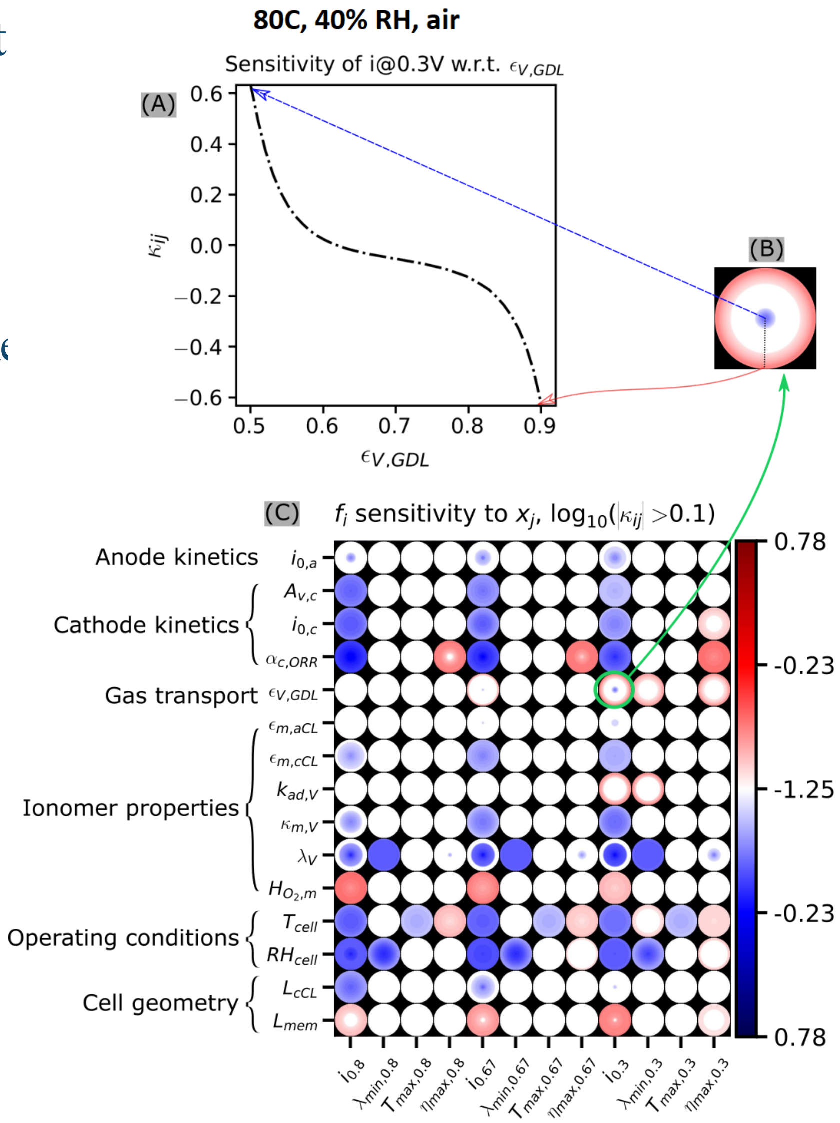


Parameter sensitivity analysis from physics -based model

- How sensitive is cell output $\boldsymbol{f_i}$ to input

$$\kappa_{ij} = \frac{\partial \log f_i(x)}{\partial \log x_j} = \frac{1}{\delta}\left(\frac{f_i(\mathbf{x} + \delta x_j \mathbf{e}_j)}{f_i(\mathbf{x})} - 1\right)$$

- Model Inputs
  - Operating conditions, transport properti Other parameters
- Cell condition variables as outputs
- Multiple operating conditions
- Heatmap shows log of the sensitivity magnitude
  - Red is negative sensitivity and blue is positive
- Crucial parameters at dry conditions (80°C, 40% RH, air)
  - Cathode kinetics and membrane



κ_L: Membrane conductivity in liquid

κ_V: Membrane conductivity in vapor

λ: Membrane water uptake

ξ: Electro-osmotic coefficient

α: Water diffusivity

A_v: Catalyst specific area (anode/cathode)

i_0, α_a, α_c: Exchange current, transfer coefficients

r_{agg}, δ_{agg}: Radius of agglomerate and film thickness

$D_{O_2,m}$: Oxygen diffusivity in ionomer

$D_{ij,bulk}$: Bulk diffusivity

ϵ_G: Gas volume fraction

S_L: Liquid saturation

τ_G: Tortuosity

k_0: Absolute permeability

k_r: Relative permeability

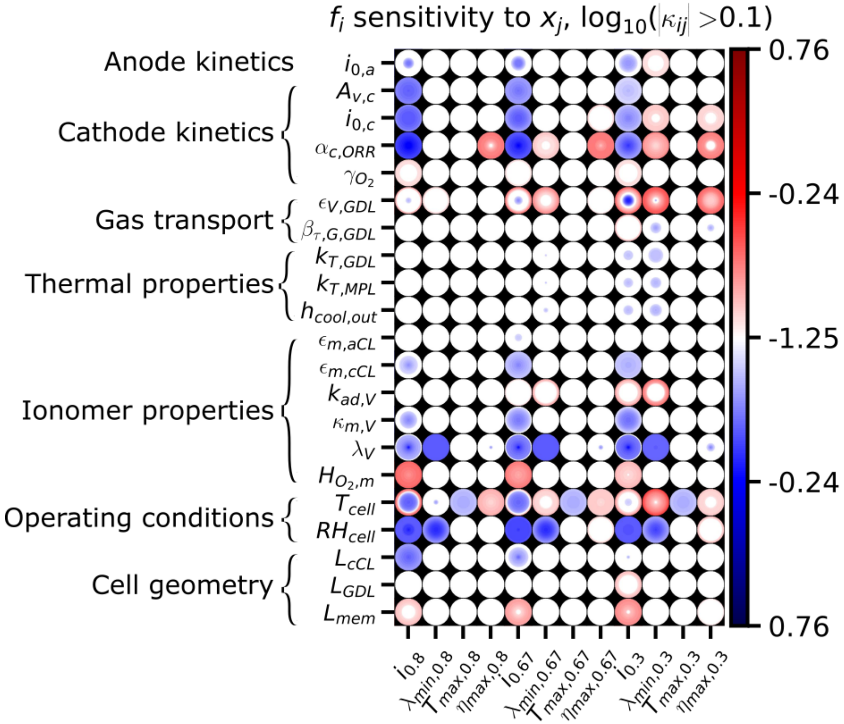
k_T^{eff}: Effective thermal conductivity

σ^{eff}: Effective electronic conductivity

Parameter sensitivity analysis from physics-based model

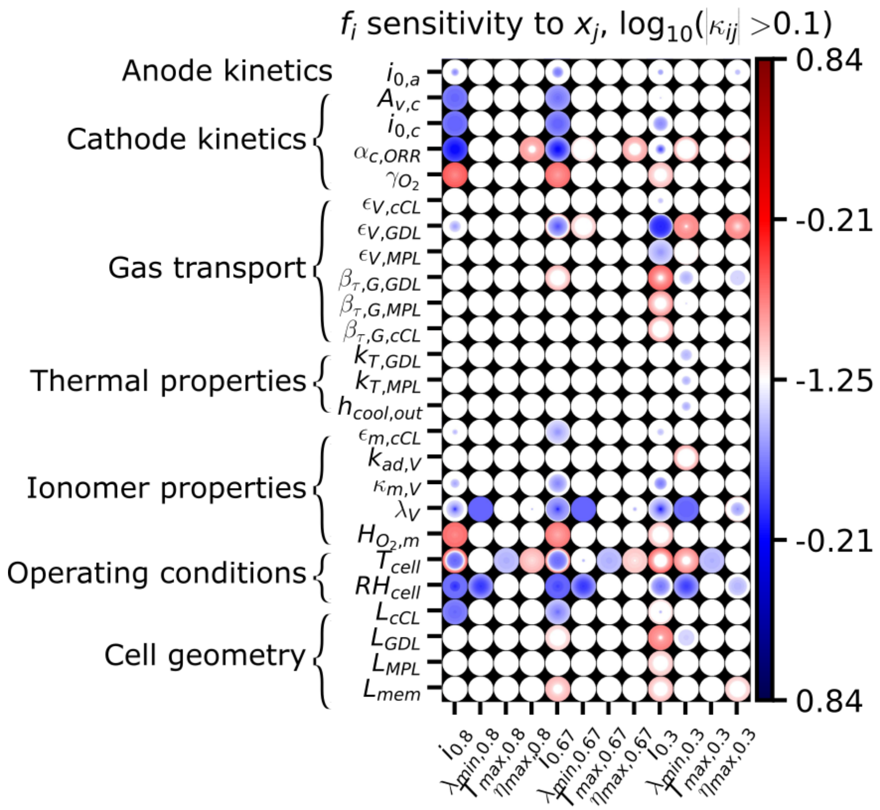
- Crucial parameters at 80°C, 80% RH, air
  - Cathode kinetics parameters: slow ORR
  - Membrane hydration: dry membrane
  - GDL transport: low O_2 partial pressure
- Crucial parameters at 80°C, 80% RH, 5% O_2
  - Cathode kinetics parameters: slow ORR
  - GDL, MPL, CL transport properties: O_2 limited cell

80C, 80% RH, air



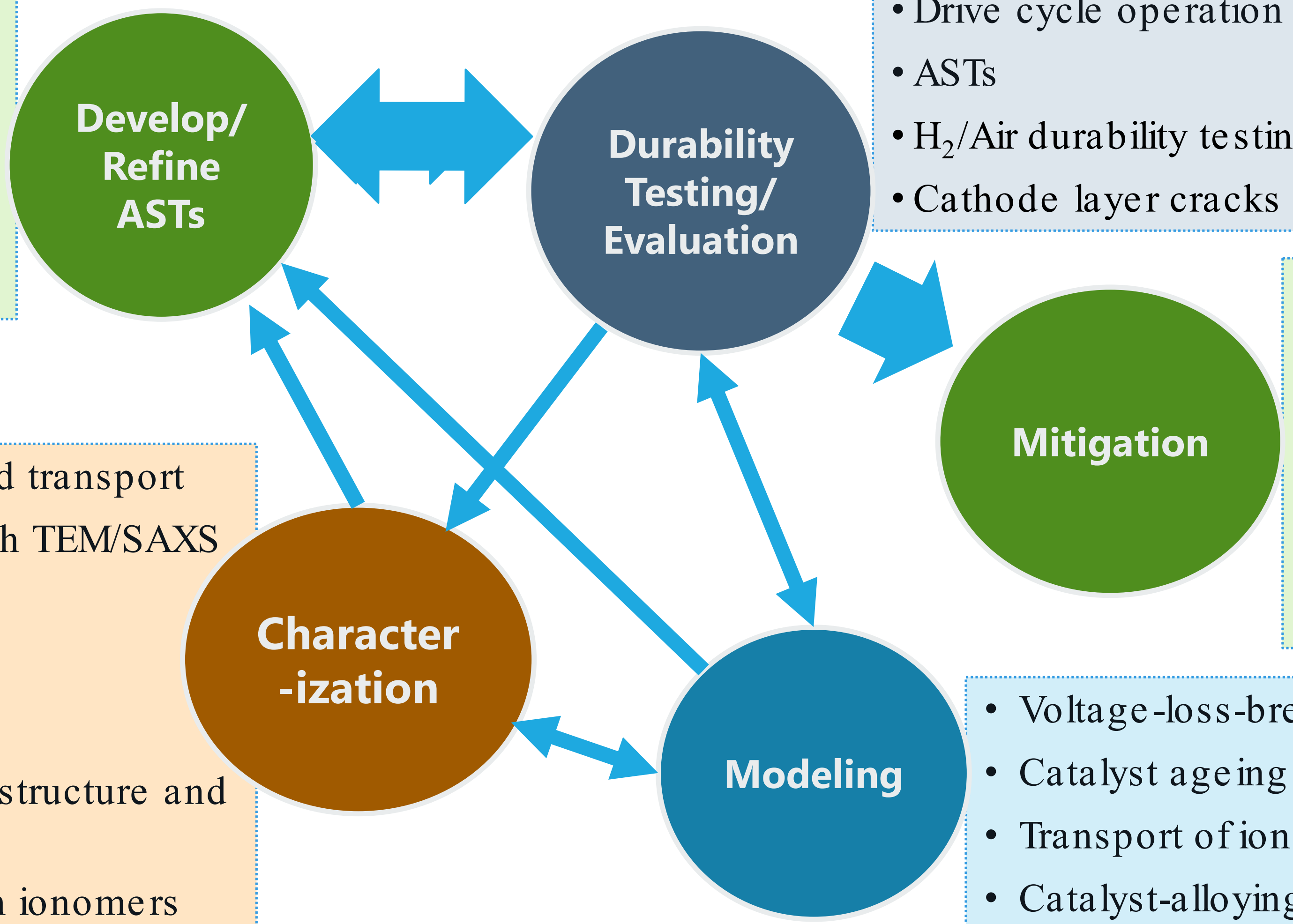
κ_L: Membrane conductivity in liquid
κ_V: Membrane conductivity in vapor
λ: Membrane water uptake
ξ: Electro-osmotic coefficient
α: Water diffusivity
A_v: Catalyst specific area (anode/cathode)
i_0, α_a, α_c: Exchange current, transfer coefficients
r_{agg}, δ_{agg}: Radius of agglomerate and film thickness
$D_{O_2,m}$: Oxygen diffusivity in ionomer
$D_{ij,bulk}$: Bulk diffusivity
ϵ_G: Gas volume fraction
S_L: Liquid saturation
τ_G: Tortuosity
k_0: Absolute permeability
k_r: Relative permeability
k_T^{eff}: Effective thermal conductivity
σ^{eff}: Effective electronic conductivity

80C, 80% RH, 5% O_2



Durability

Understanding, evaluating, and mitigating durability concerns with materials-based solutions



- Mechanistic based ASTs
- Single/multi-mechanism (MEA) ASTs
- Simulate drive cycle

- Drive cycle operation
- ASTs
- H_2/Air durability testing and anode reversal
- Cathode layer cracks

- Catalyst-alloying; particle growth, kinetics losses with leaching
- Catalyst-support corrosion
- Membrane Ce stabilizers
- New materials and integration schemes

- Metal dissolution and transport
- Catalyst ripening with TEM/SAXS
- Catalysts
- Membranes
- GDL/MPL cracks
- Catalyst Layer microstructure and stability
- Cation partitioning in ionomers

- Voltage-loss-breakdown
- Catalyst ageing
- Transport of ions
- Catalyst-alloying; particle growth, kinetics losses with leaching

ASTWG (Working Group) Charter

- 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)
- ASTs to be developed
  - Catalyst
  - Catalyst support
  - Membrane chemical
  - Membrane combined chemical mechanical
  - SD/SU
  - Anode H_2 starvation
  - MEA drive-cycle

Participants

ANL
Ballard
Carnegie Mellon
DOE
GM
LANL
LBNL
Nikola
NREL
ORNL
Cummins
Plug Power
Chemours

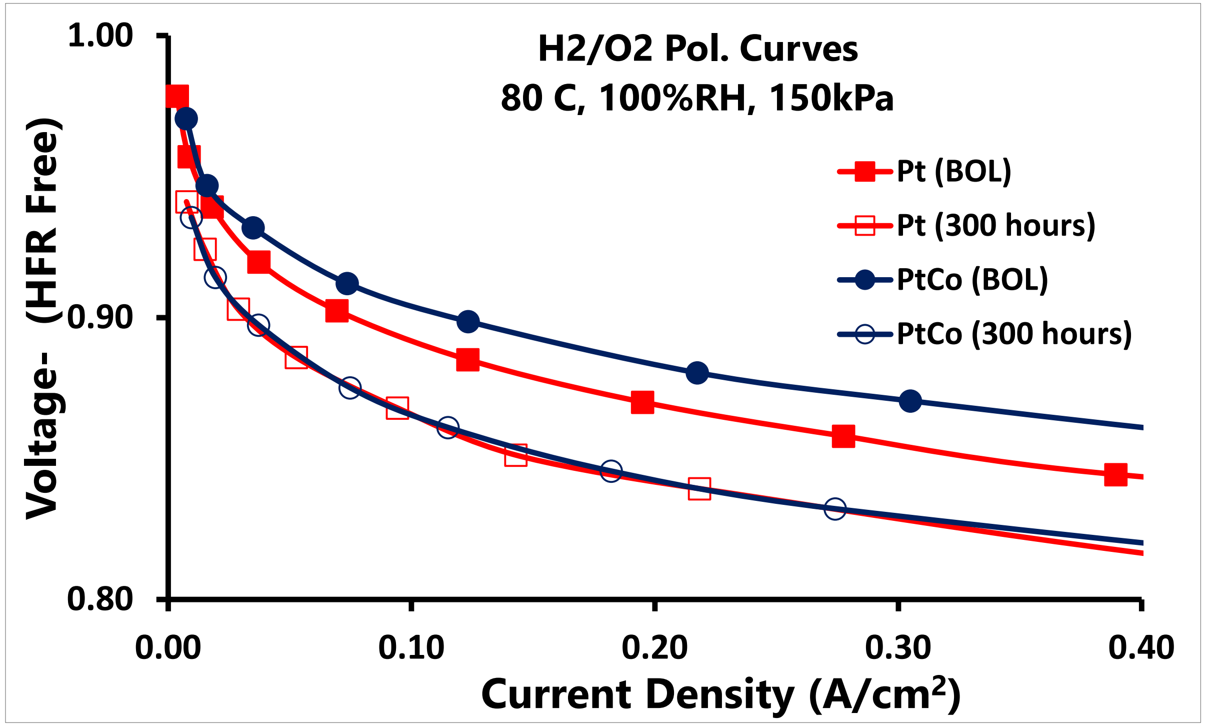
Define the 25,000 hour equivalent AST in the M2FCT 2025 Target

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

https://www.hydrogen.energy.gov/pdfs/19006_hydrogen_class8_long_haul_truck_targets.pdf

ASTWG: Catalyst Durability Protocol

- Develop H_2/Air tests to measure degradation rates under accelerated conditions
- Operate cell @90℃ to accelerate all degradation mechanisms for the longer life time
  - Potential cycling between 0.65V and 0.925V under 100%RH to accelerate catalyst degradation
  - Introduce RH cycling and dry operation to accelerate membrane degradation
- Degradation rates under accelerated conditions will be compared with rates at typical LDV and HDV operating conditions to determine duration of AST to yield 25,000 hours equivalent durability



Pt

Hours	A/mg-Pt
0	0.31
50	0.23
100	0.21
200	0.17
300	0.13

PtCo

Hours	A/mg-Pt
0	0.47
50	0.33
100	0.23
200	0.18
300	0.14

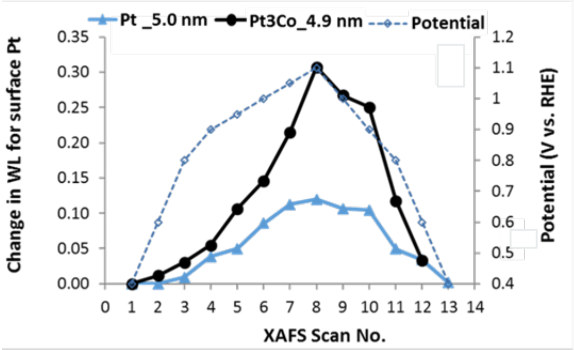
- PtCo has mass activity = 0.47 A/mg_{Pt} at BOL while Pt = 0.31 A/mg_{Pt}
- At 300 hours both have almost the same mass activity of 0.135 A/mg_{Pt}

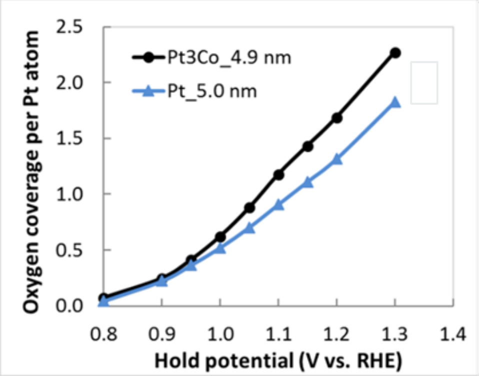
Current plan: Extend out catalyst AST to 90,000 cycles from 30,000 cycles

Time-resolved Measurement of Catalyst Dissolution

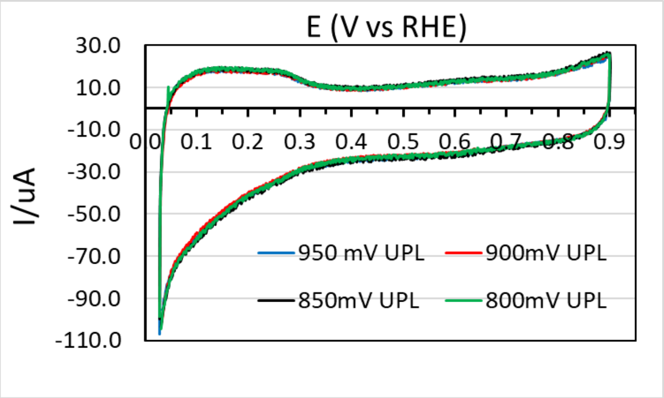
- Dissolution of both Pt and alloying metal measured in aqueous electrolyte during potential control
- Sensitivity at <10 ppt
- Dissolution can be correlated with oxidation state of catalyst using cyclic voltammetry and/or in situ X-ray absorption spectroscopy
- Catalyst degradation mechanisms analyzed using thermodynamic and kinetic modeling
- New cell developed to allow deaeration of electrolyte enabling determination of the effect of dissolved oxygen

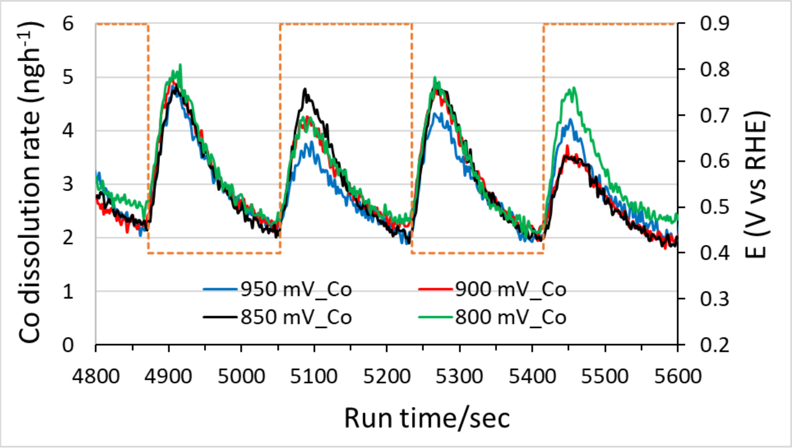
Correlation of Pt and Co loss with Oxide Formation and Reduction





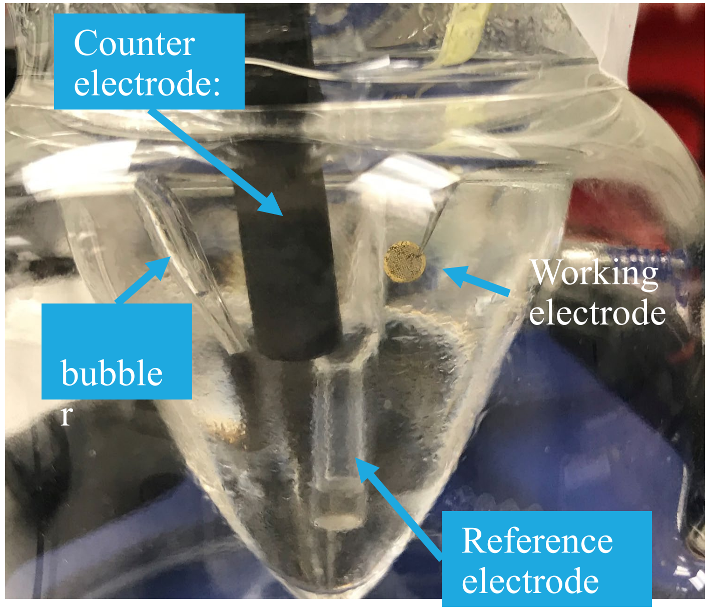
On-line ICP-MS detection of Pt and Co used to determine regions of voltage stability and to elucidate degradation mechanisms



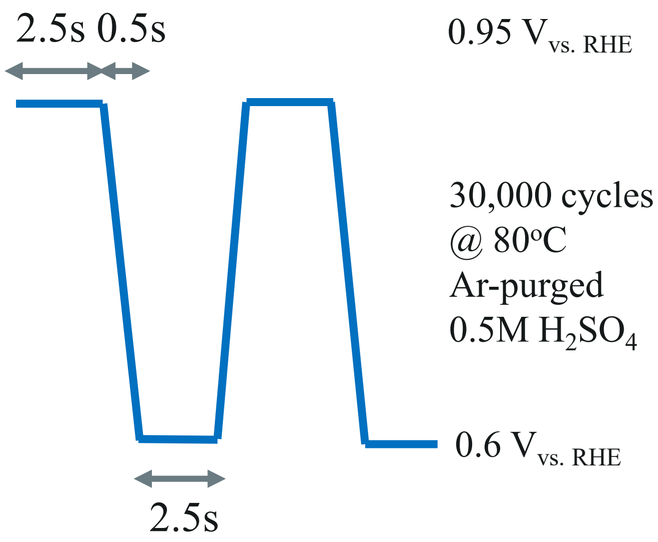


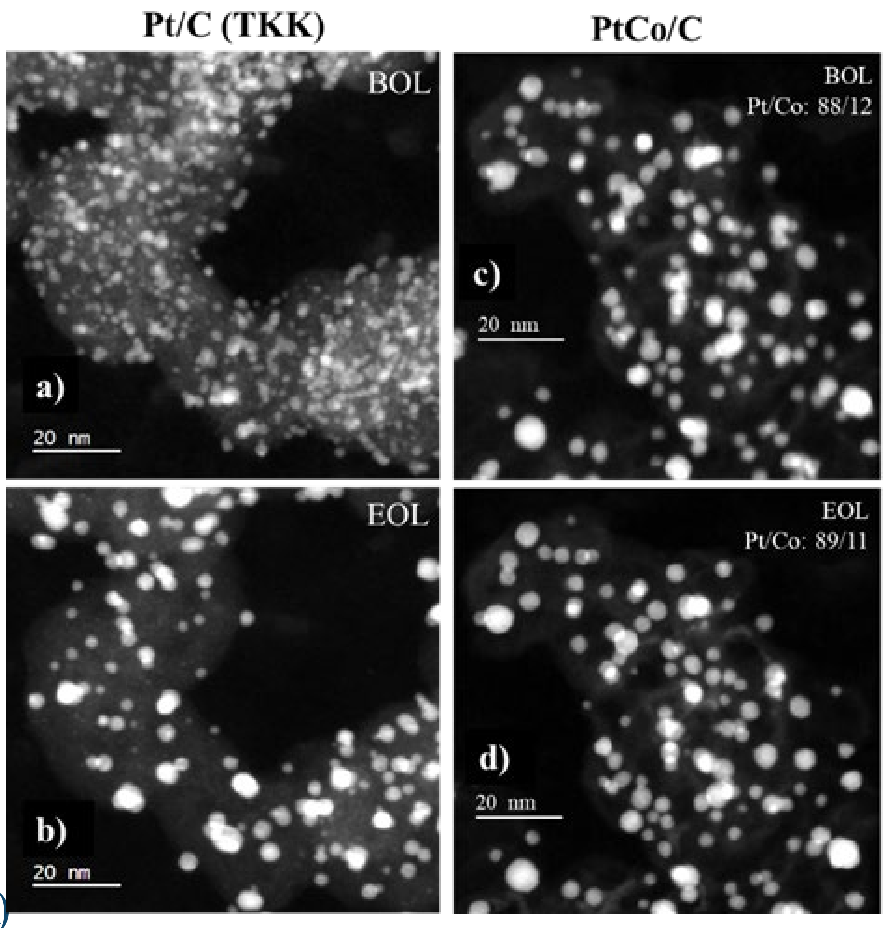
Identical Location (IL) -STEM Capability

a) Experimental Setup



b) DOE Square-wave Protocol

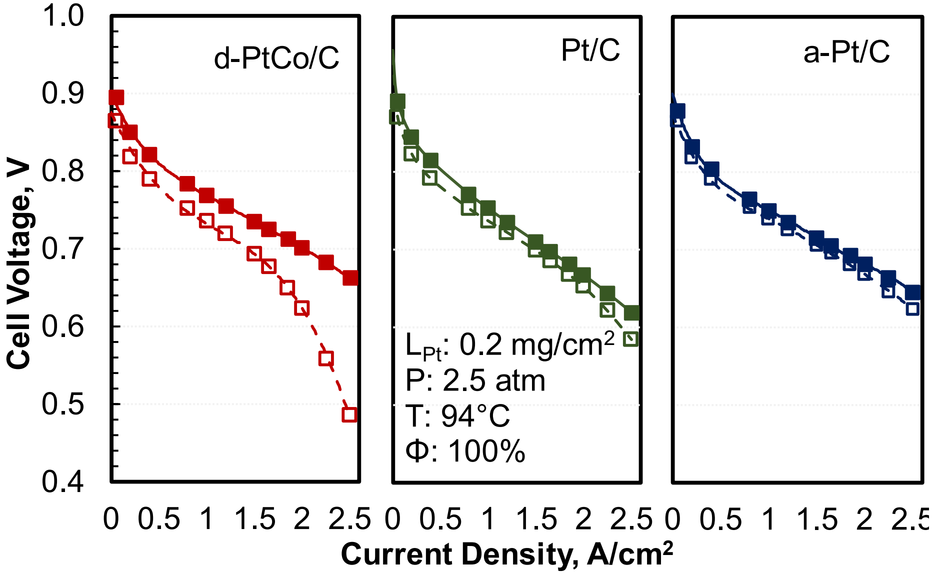


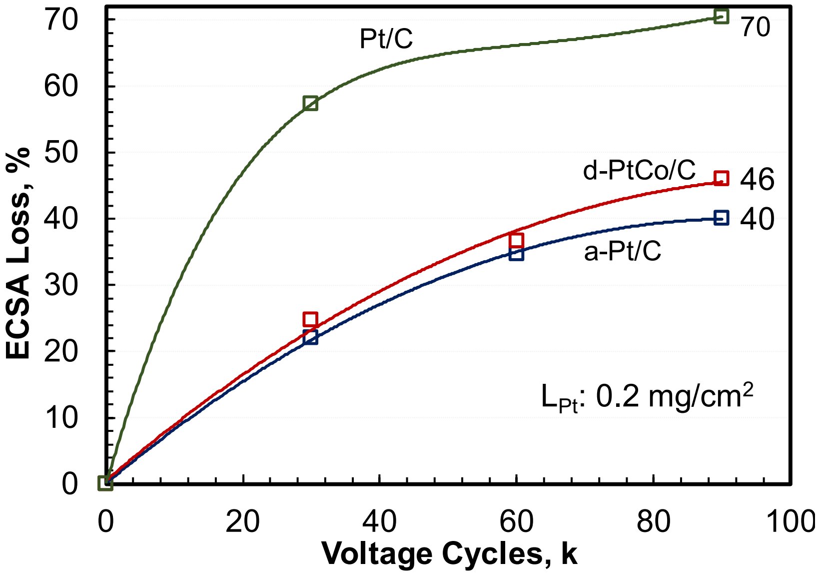


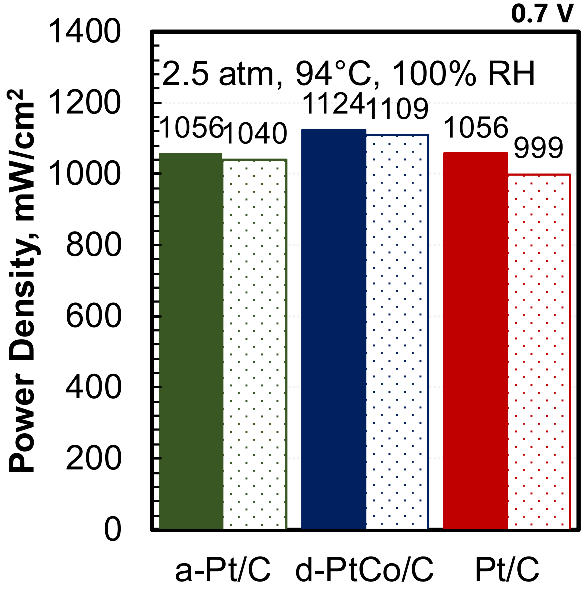
- Morphology and composition changes observed before and after cycling catalyst on Au TEM grid in aqueous three electrode cell
  - Driving primarily particle coalescence degradation mechanism (Pt/C TKK)
  - Little Ostwald ripening observed in IL-STEM tests, in stark contrast to MEA testing of PtCo cathodes

Catalyst Durability Case Study: Modeling

Differential cell performance at BOL and after 90k 0.6-0.95 V AST cycles, 80°C, 100% RH, 0.2 mg/cm^2 cat loading







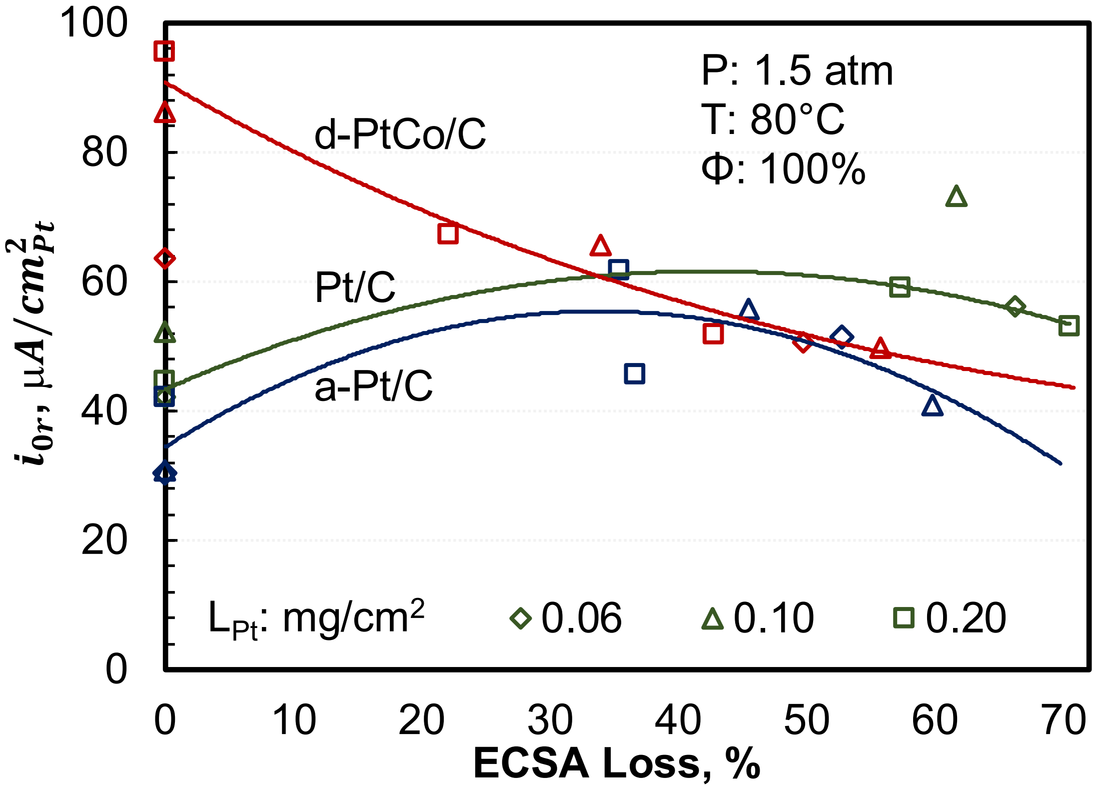
- For high stoics at BOL, d-PtCo/C is superior to a-Pt/C and Pt/C at all current densities
- ECSA loss similar for d-PtCo/C and a-Pt/C but much higher for Pt/C
- Performance loss at high current densities after 90k cycles: d-PtCo/C > Pt/C > a-Pt/C
- Based on differential cell data, all three catalyst systems are potential candidates
- M2FCT target is for integral cell performance at 0.7 V after 25,000 h
  - 2.5 kW/g$_{PGM}$ and 750 mW/cm^2 at 0.7 V after 25,000 h, 2.5 atm, 87^{o}C, 2/1.5 anode/cathode stoichiometry

*Differential cell data from Nagappan Ramaswamy (GM), FC323, 2020 AMR

Catalyst Durability Case Study: Effect of aging

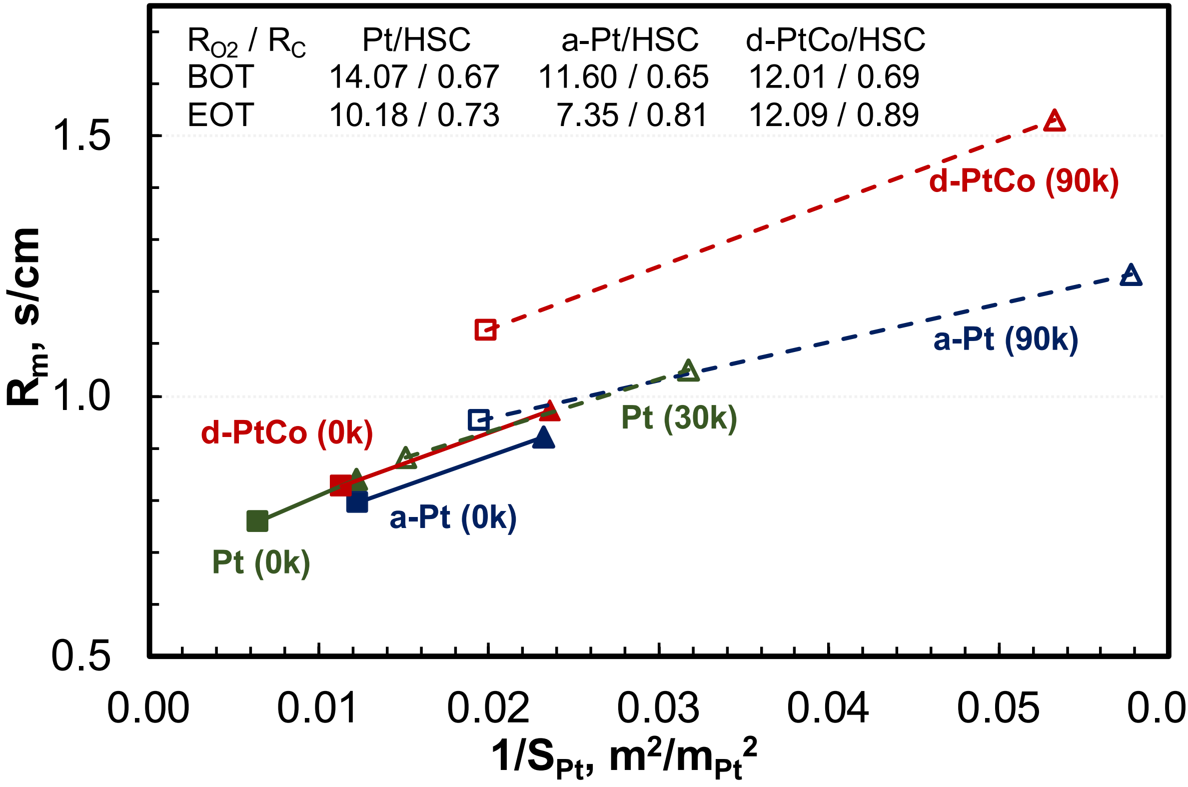
ORR Kinetics

- Fresh Pt/C and a-Pt/C have comparable specific i_{0r} (exchange current density) but Pt/C has higher kinetic activity on g-Pt basis
- Pt/C and a-Pt/C exhibit similar aging of i_{0r}
- d-PtCo/C has higher i_{0r} at BOL but decreases with aging



O_2 Transport Resistance

- Slope R_{O2} and intercept (Micropore resistance (R_c)) change with aging
- Pt/C and a-Pt/C: R_{O2} decreases with aging
  - Redistribution of Pt from pores to surface
- d-PtCo/C: R_{O2} slightly increases w/aging; Co dissolution and poisoning



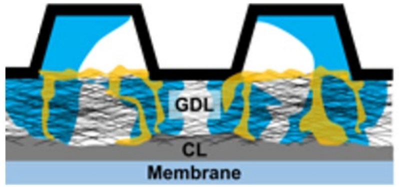
R_{O2} / R_C	Pt/HSC	a-Pt/HSC	d-PtCo/HSC
BOT	14.07 / 0.67	11.60 / 0.65	12.01 / 0.69
EOT	10.18 / 0.73	7.35 / 0.81	12.09 / 0.89

*Differential cell data from Nagappan Ramaswamy (GM), FC323, 2021 AMR

GDLs for Cation Transport Suppression

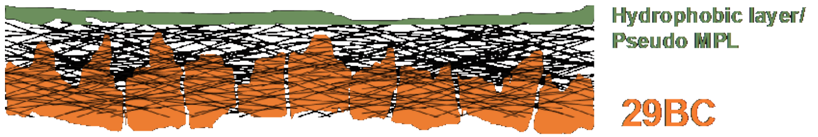
- Cations are present in PFSAs as radical scavengers (Ce and Mn), contaminants (Ca), byproducts of catalyst dissolution (Co and Ni), and from corrosion of BOP components (Fe)
  - Fe transport through GDL (from BPP corrosion or BOP components) problematic for membrane durability

Studied GDL/MPL morphology effect on cation transport



MPLs with cracks and hydrophilic GDLs show enhanced cation transport

GDL modification to suppress the Cation transport rates

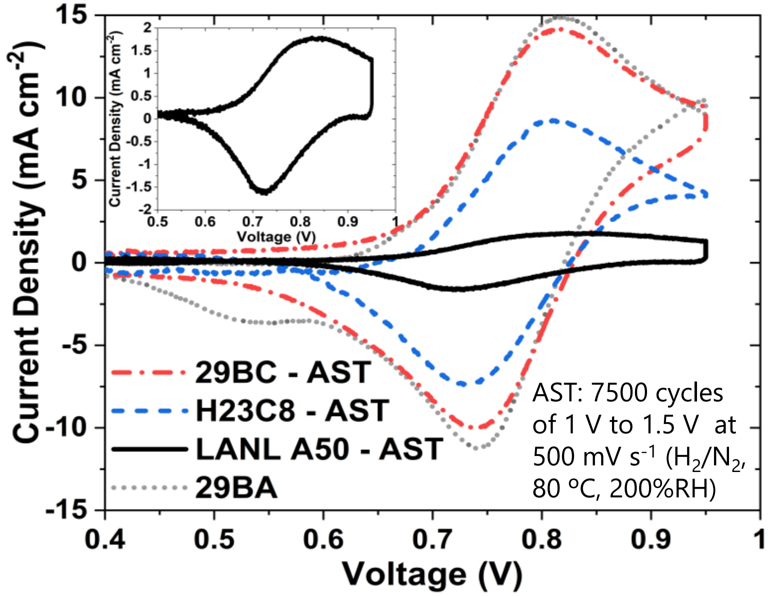


Addition of hydrophobic layer

- Novel GDLs suppress cation transport from the flow field
  - Smaller Fe^{2+}/Fe^{3+} redox couple post- flowfield injection of Fe
  - AST corrodes the 29BC and H23C8 MPL significantly and increases the Fe transport rates

Flowfield injection of Fe to study cation transport

Fe^{2+}/Fe^{3+} redox couple obtained from CV



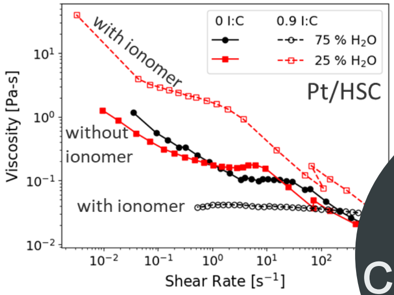
AST: 7500 cycles of 1 V to 1.5 V at 500 mV s^{-1} (H_2/N_2, 80 °C, 200%RH)

- Subtracting the after curve from the before curve to obtain the Fe redox curve
- Peak current used to calculate the Fe loading in the catalyst layer
- Modified 29BC shows significant suppression of Fe transport even after AST

- Babu, S.K., et al. Journal of the Electrochemical Society 168.2 (2021) 024501.

Integration

Glean fundamental information pertinent to the integration of known and novel materials

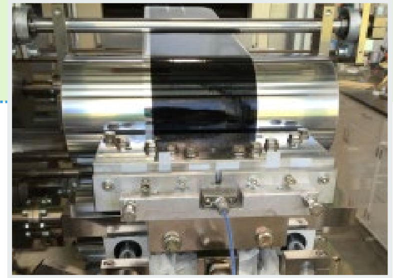


Film and Ink Characterization

- Rheology
- Ionomer aggregation
- Intrinsic interactions and binding

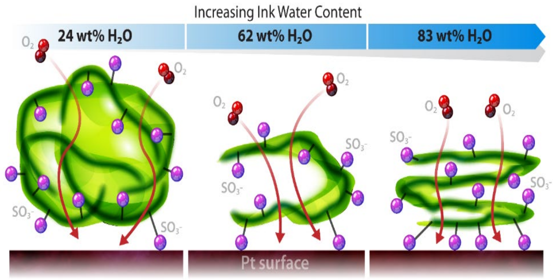
- Time evolution of formation of ionomer and electrodes
- Additives and pore formers
- Manufacturing

Electrode Formation and Design



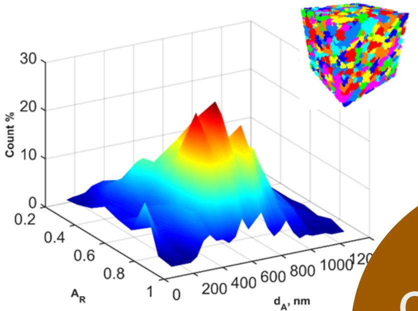
Cell Performance and *In situ* Diagnostics

- Baseline and standardization
- Limiting current
- Ionomer adsorption
- Interfaces



Optimization and Understanding

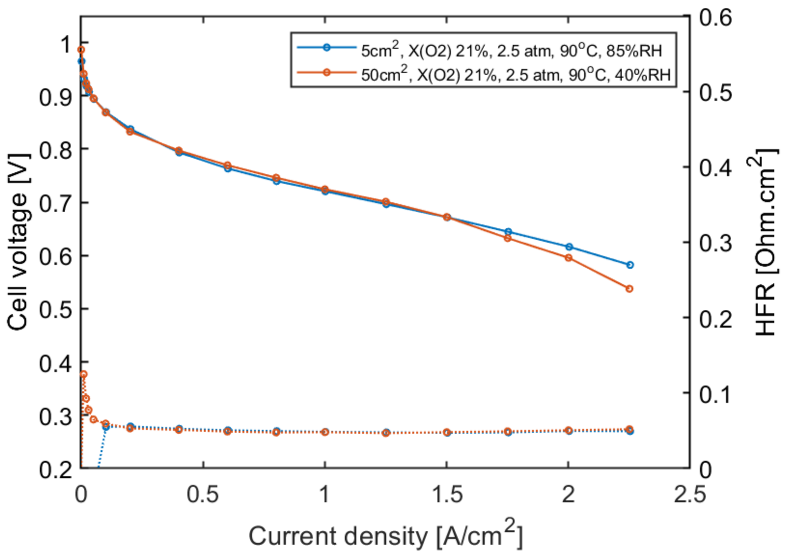
- Multiphysics modeling
- MEAs with novel materials



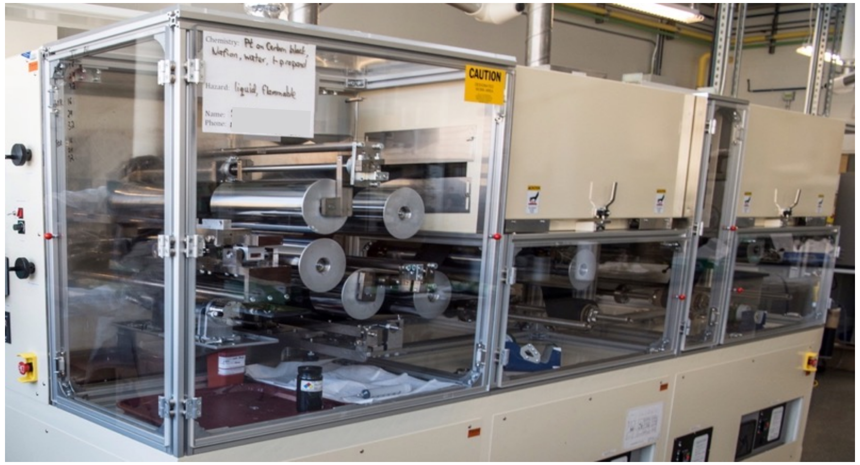
***Ex situ* Component Characterization**

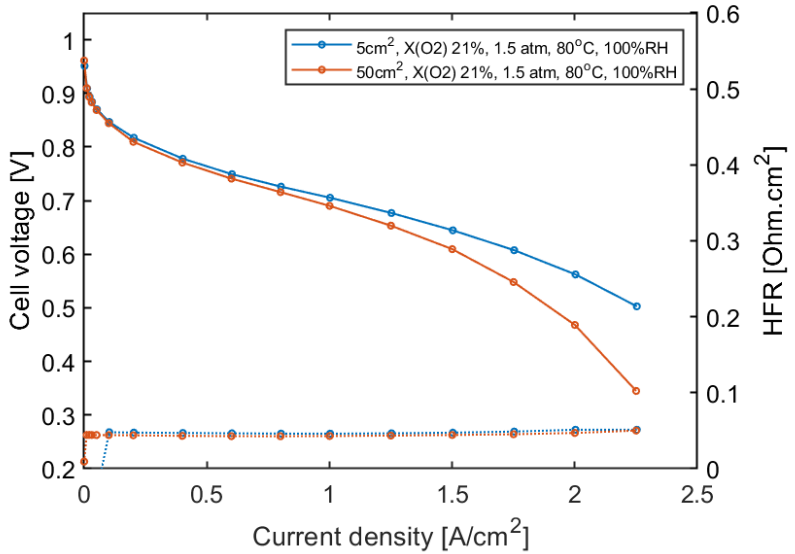
- Ionomer
- GDL/MPL
- Electrode structure

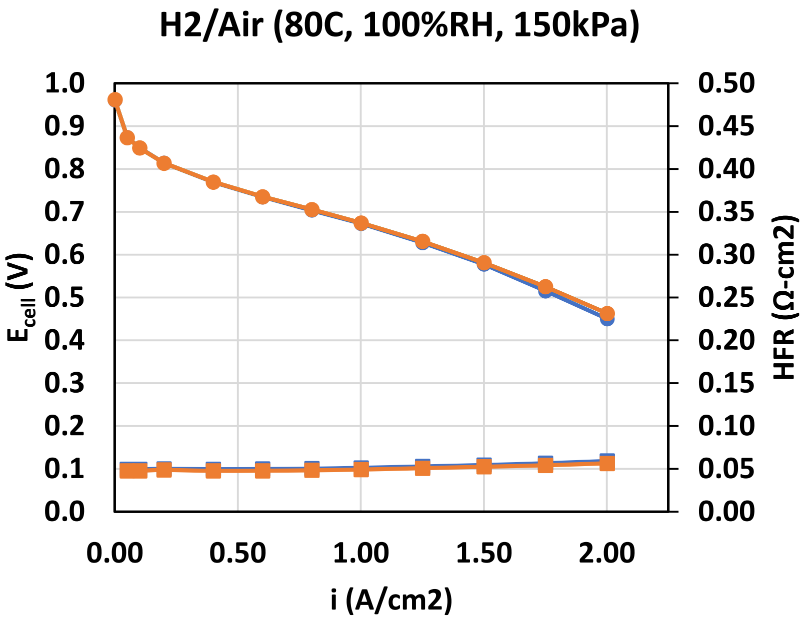
Baselining Roll -to-Roll MEA Performance

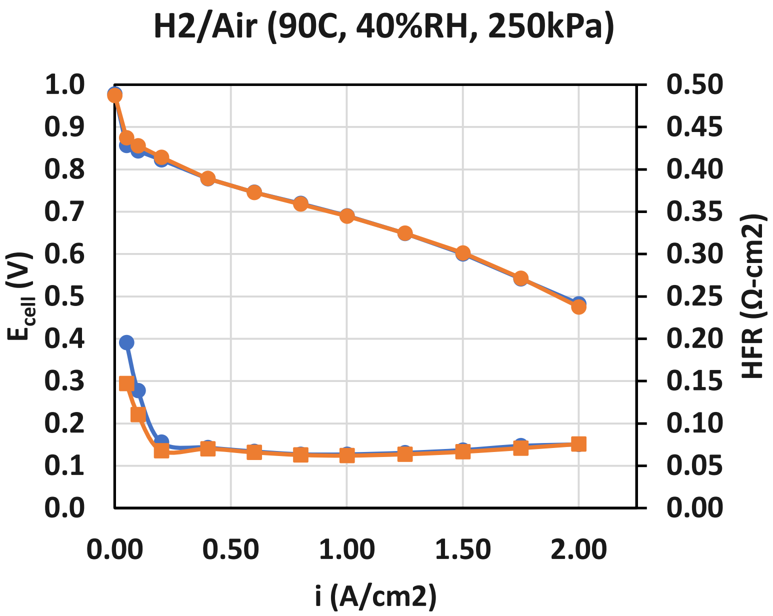


Location	50 cm2	5 cm2
LBNL	2	3
GM	2	3
LANL	2	3
ANL	0	2-3
NIST	2	
CMU	2	2
Nikola	2	
NREL	4	3-4



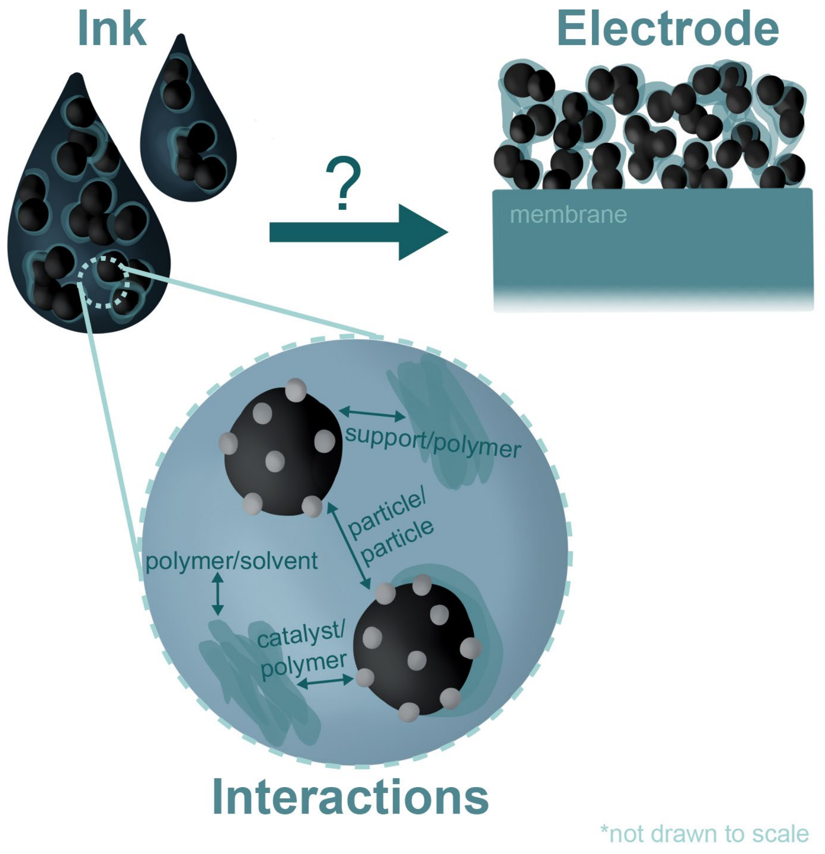



H2/Air (80C, 100%RH, 150kPa)

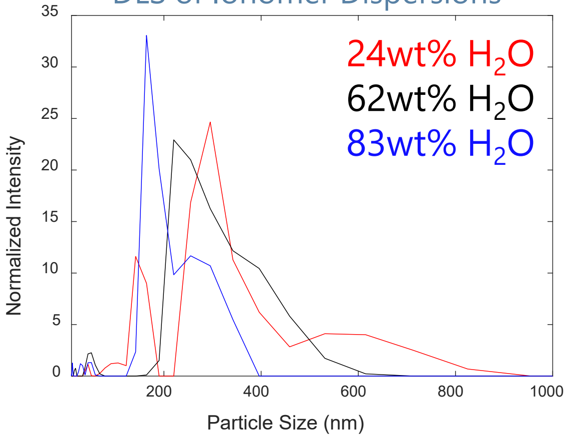

H2/Air (90C, 40%RH, 250kPa)

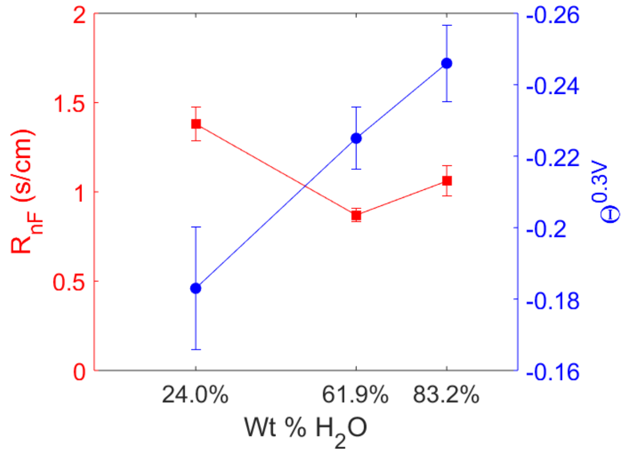
Catalyst -Layer Formation

High-water-concentration dispersions lead to fewer secondary aggregates

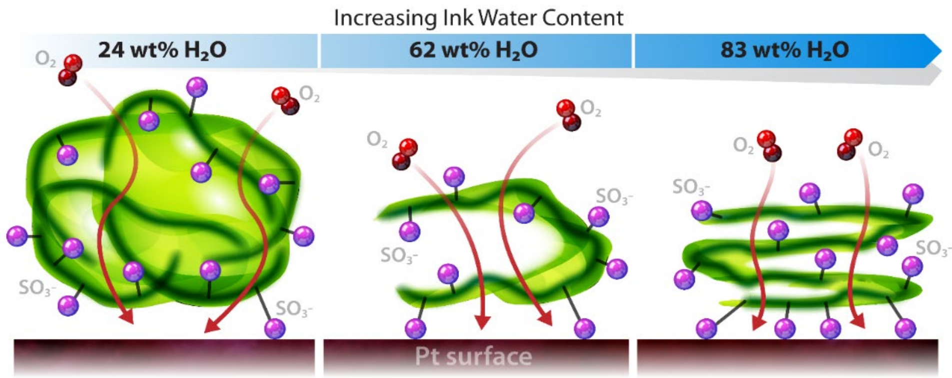


DLS of Ionomer Dispersions





Catalyst/Ionomer Interface

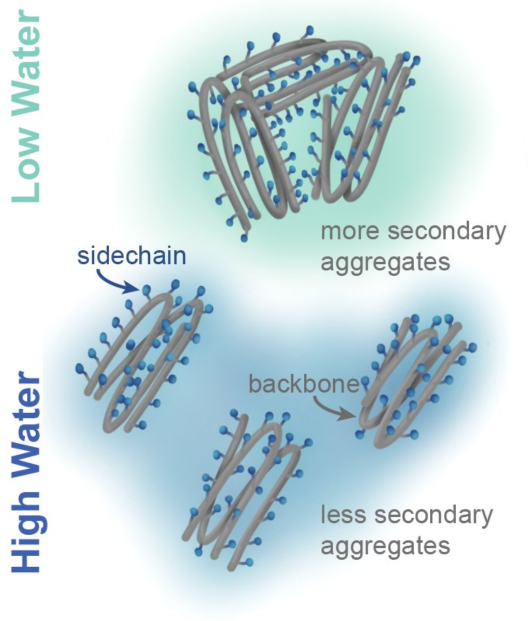


Synergistic conclusions from fundamental and in-situ investigations

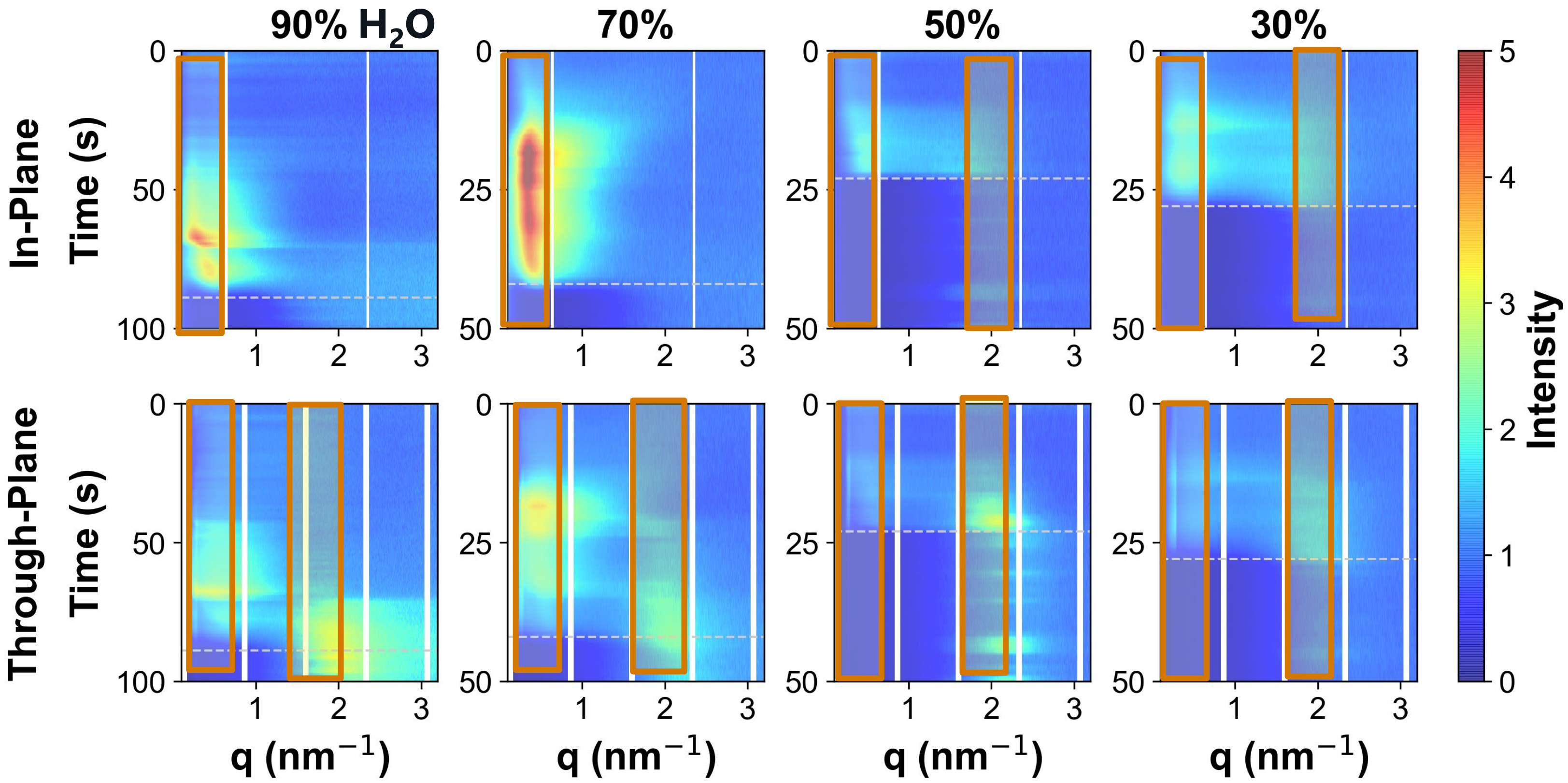
[1] J. Phys. Chem. B, 122 (2018). [2] ACS Appl. Mater. Interfaces 2019, 11, 50, 46953–46964

Catalyst -Layer Formation : PFSAFormation

- High-water-concentration dispersions = more regular arrangement of primary aggregates, fewer secondary aggregates
- Formation of crystalline domains occurs first



Analyzed dispersion structure with Grazing Incidence Small Angle X-Ray Scattering



Berlinger, S. A; Dudenas, P. J.; Bird., A.; Chen, X.; Freychet, G.; McCloskey, B.D; Kusoglu, A.; Weber, A.Z., ACS Applied Polymer Materials (2020).

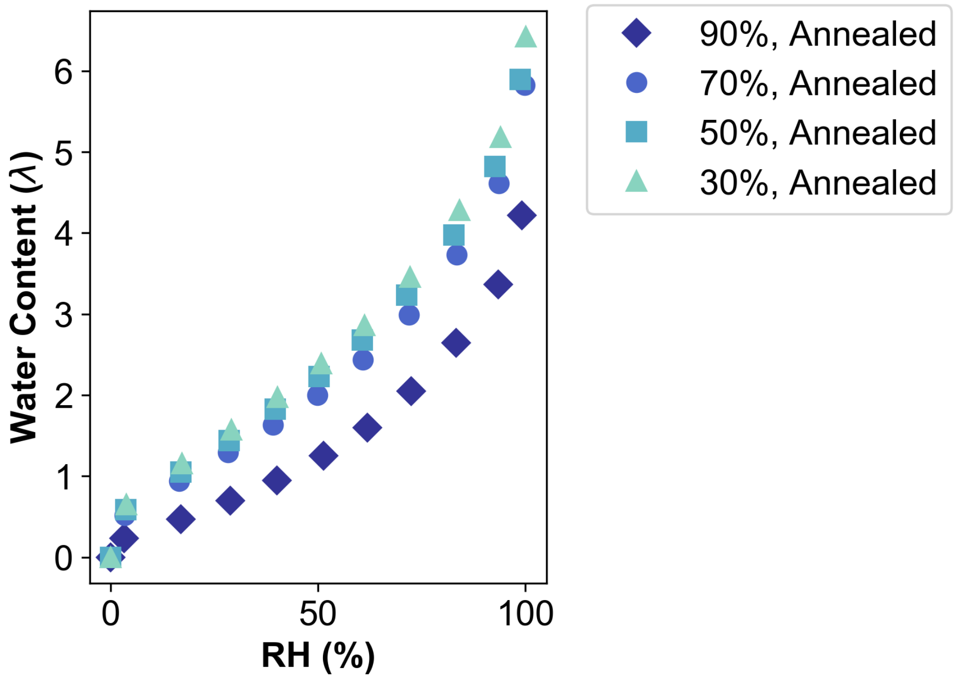
Catalyst -Layer Formation : PFSAFormation

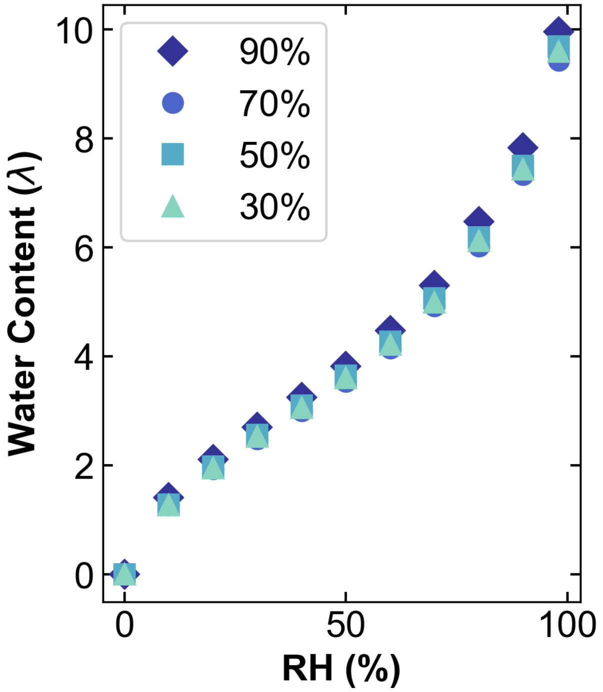
Thin-Films: Dispersion-Cast

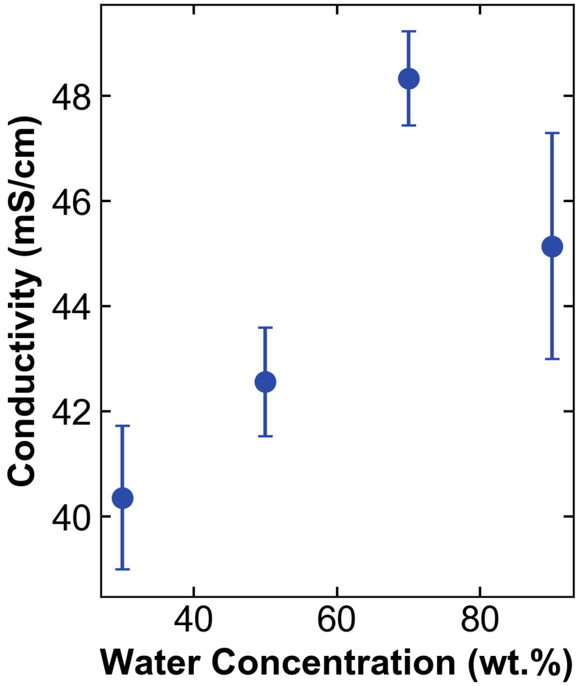
- Annealed films: swelling decreases with increasing dispersion water fraction
  - Formation behavior persists after annealing

Membranes: Dispersion-Cast

- Ionic conductivity increases with dispersion water content
  - Swelling is less sensitive to concentration
  - Impacts the connectivity of conducting domains





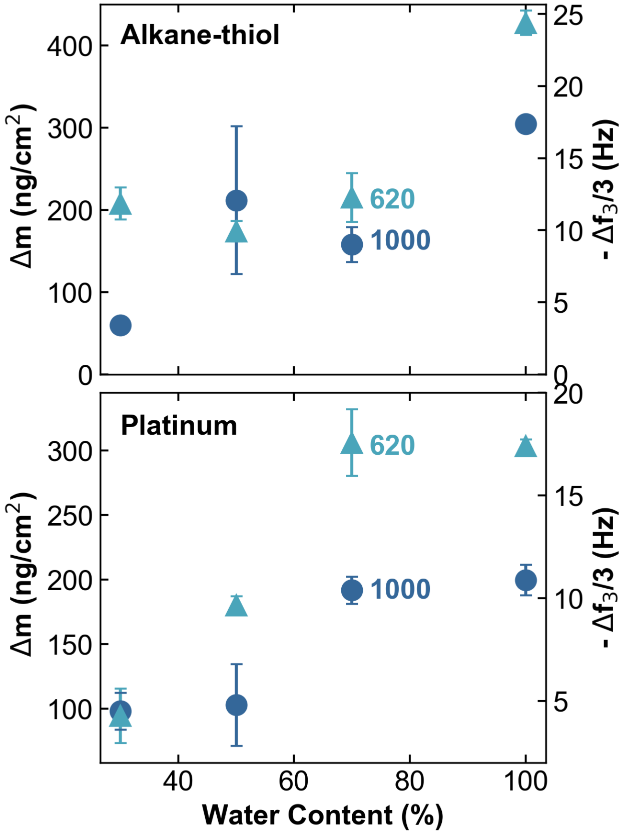


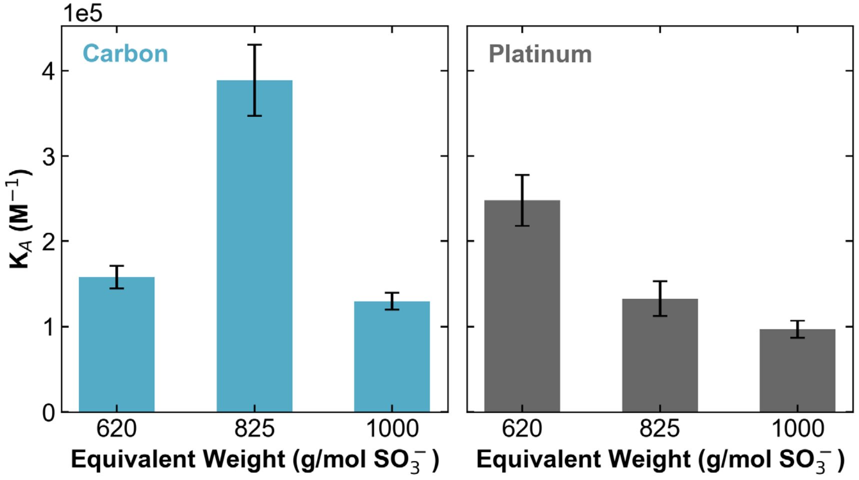
Berlinger, S. A; Dudenas, P. J.; Bird., A.; Chen, X.; Freychet, G.; McCloskey, B.D; Kusoglu, A.; Weber, A.Z., ACS Applied Polymer Materials (2020).

Catalyst -Layer Formation : Ink Interactions

Pt/PFSA interactions are similar to C/PFSA interactions, and governed by a similar mechanism

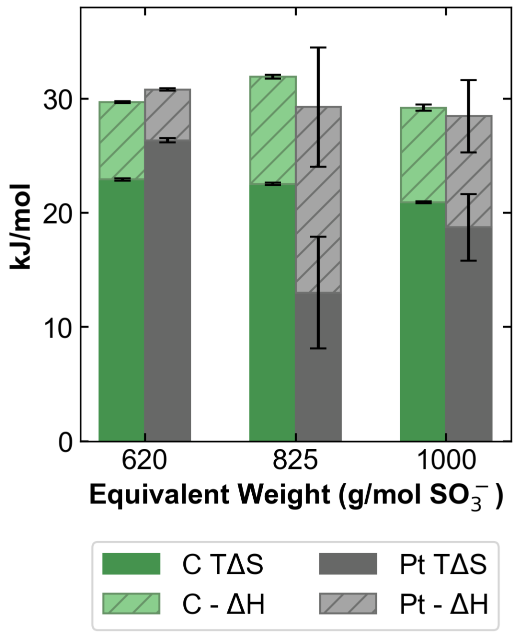
Probing interactions with QCM and ITC





- ITC conducted in 100% water, with Vulcan or Pt black nanoparticles
- ITC trends agree with QCM data (strongest adsorption for 3M 825 for carbon, weaker binding with increasing EW on Pt)
- On both platinum and hydrophobic surfaces, binding is a function of EW, and increases as dispersion water content increases

Binding Mechanism

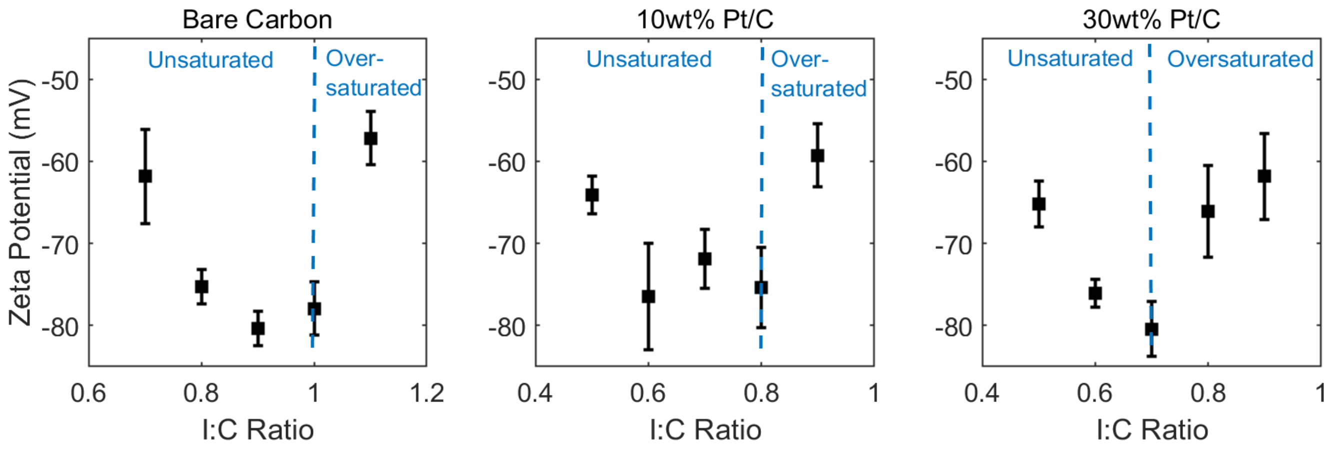


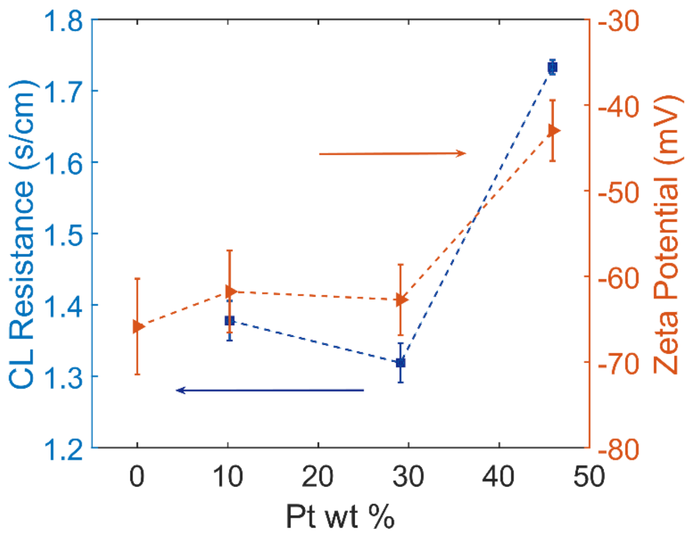
- Binding is entropically dominated on both surfaces during formation: hydrophobic interactions

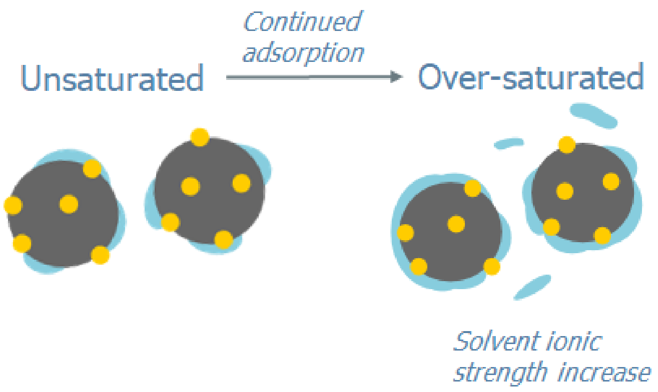
Berlinger, S. A.; McCloskey, B.D.; Weber, A.Z., 10.26434/ChemRXiv.14096990.v1

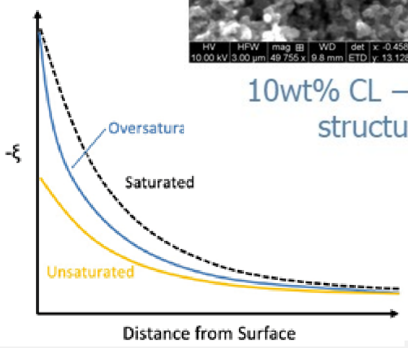
Catalyst -Layer Formation : Ink Interactions

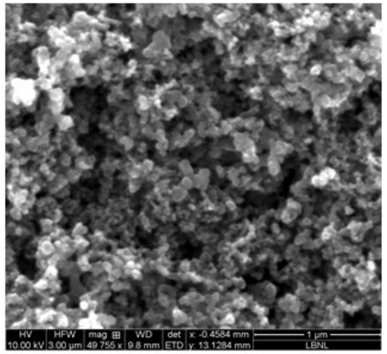
- Carbon/Pt surface areas influence ink interactions and CL performance
  - High I:C results in catalyst layer changes due to excess ionomer impacting ink properties
    - Greater than monolayer type coverage
    - Function of Pt weight percent due to interactions



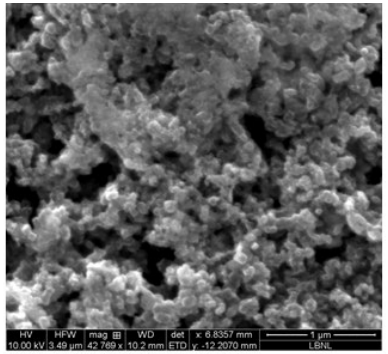








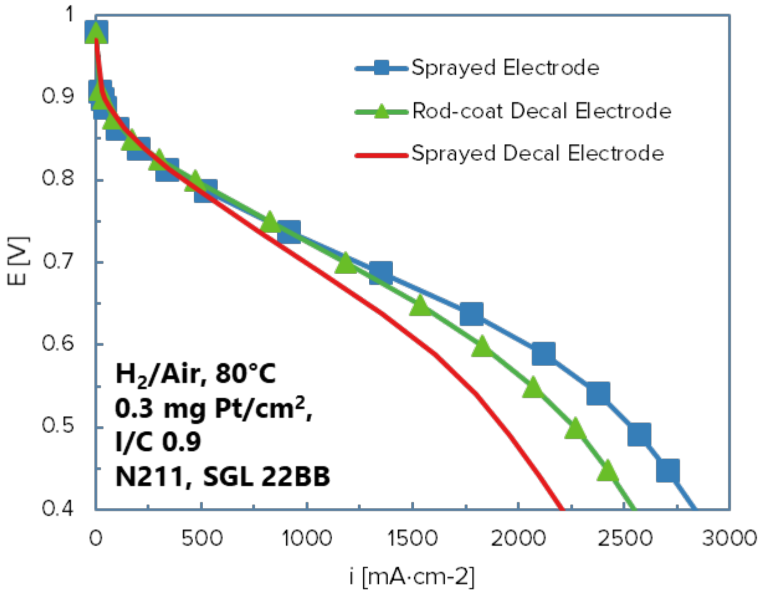
10wt% CL – porous structure

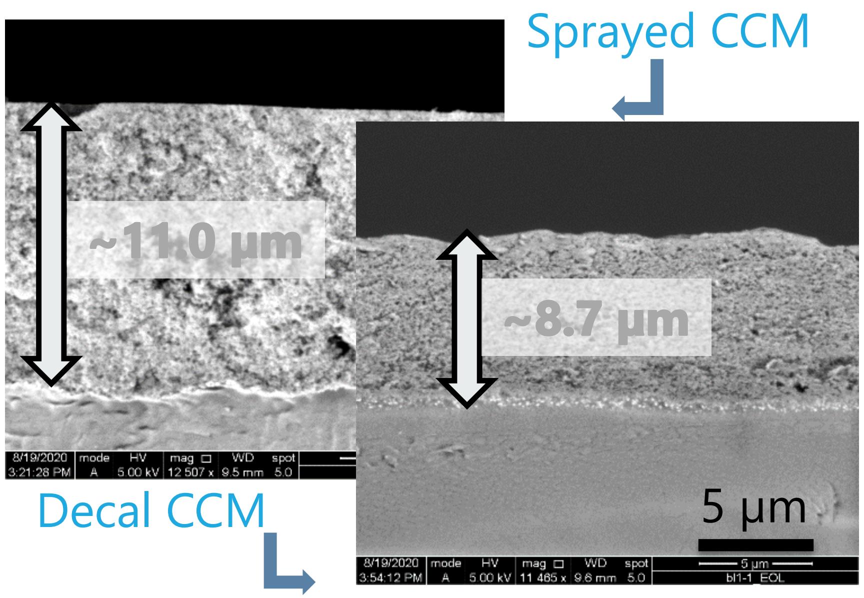


46wt% CL – agglomerated structure

Manuscript Submitted.

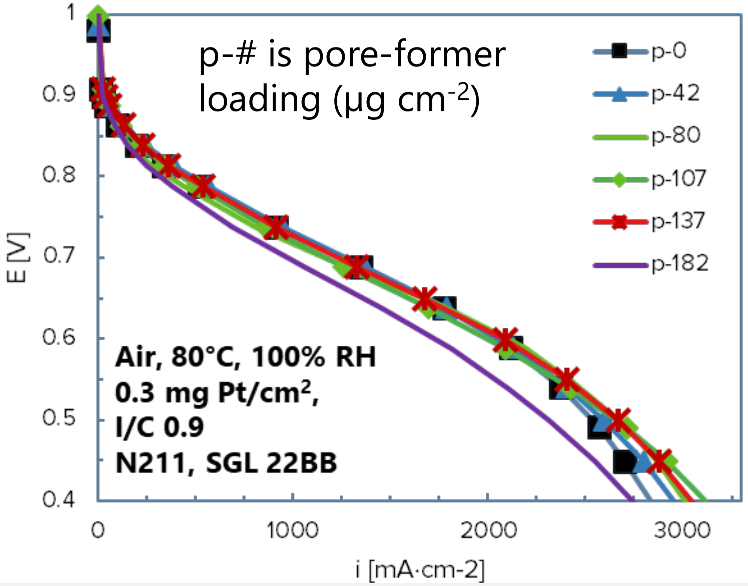
Electrode Structure - Performance

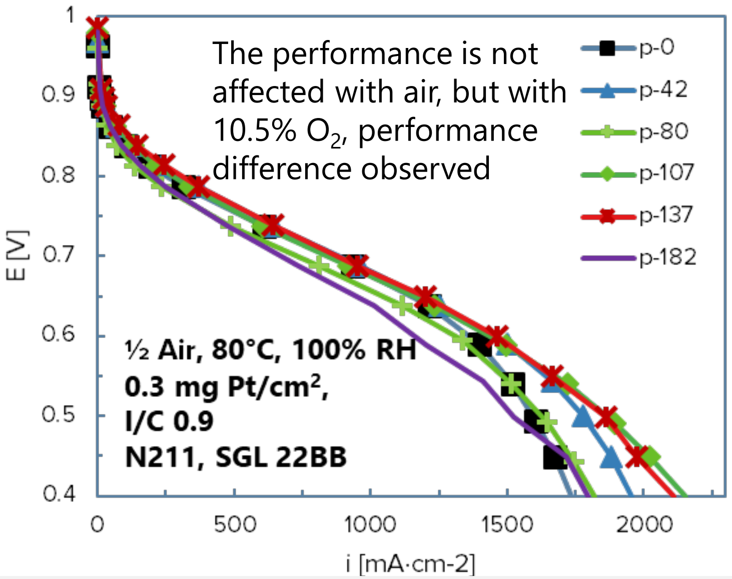




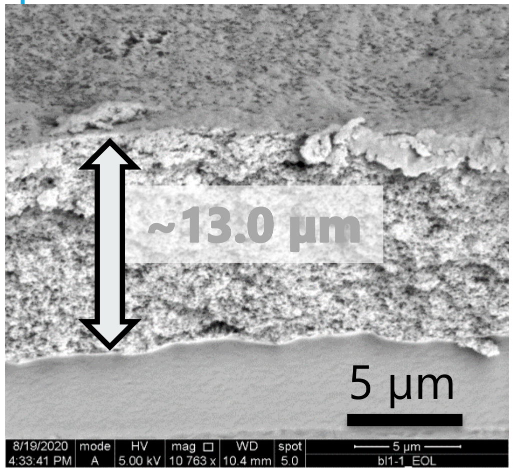
- Sprayed CCM performs best due to high porosity
- Loss in porosity due to compression in decal transferred electrodes causes mass transport losses
- Current focus is on correlating fabrication method and resulting electrode morphology to transport phenomena

Pore formers can be used to tune porosity





P-80 electrode after pore-former removal

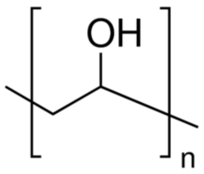


Electrode Structure – Crack Mitigation

Ink Formulation

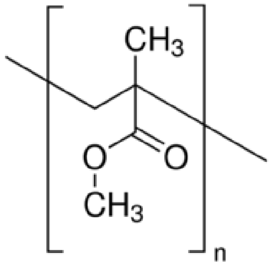
Catalyst wt %	*I/C*	*H_2O wt%*
3.5	*1.00*	*66 or 17*

Additive Details



Poly (vinyl alcohol) – PVA
Molecular Weight: 89-98k

Poly (methyl methacrylate) – PMMA
Molecular Weight: 120k

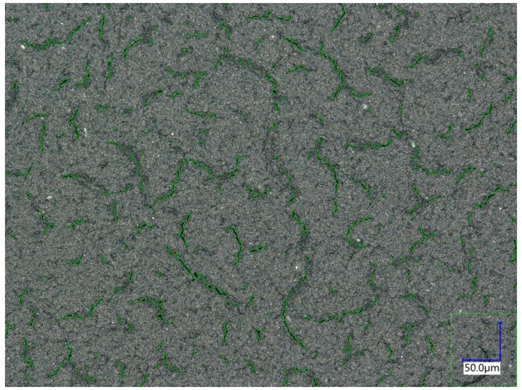


Goal of this study: Utilize additives as means to mitigate catalyst-layer crack formation

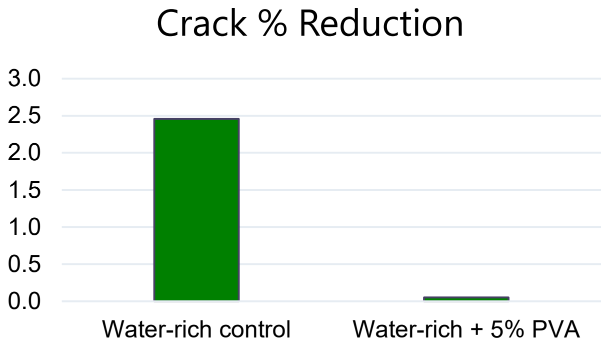
Approach: Insert polymeric additives before the final mixing stage (relative to ionomer mass)

Micrograph (500x) Comparison Utilizing Our Crack Detection Algorithm

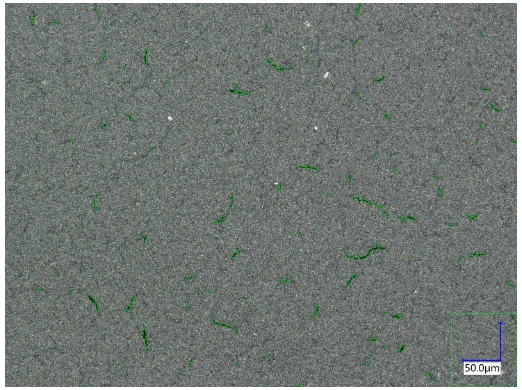
Water-rich control
0.271 – 0.292 mg Pt/cm^2



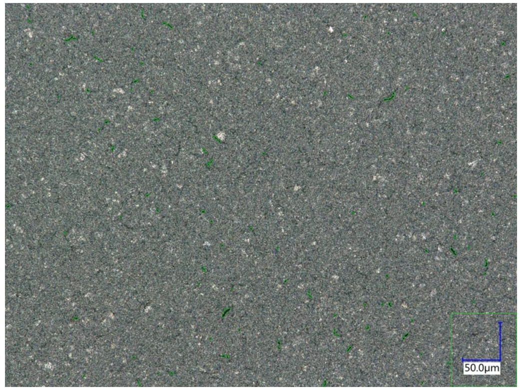
Water-rich w/ 5% PVA
0.273 – 0.280 mg Pt/cm^2

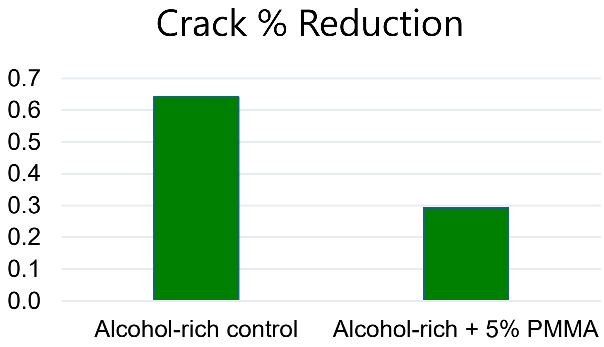


Alcohol-rich control
0.278 – 0.308 mg Pt/cm^2



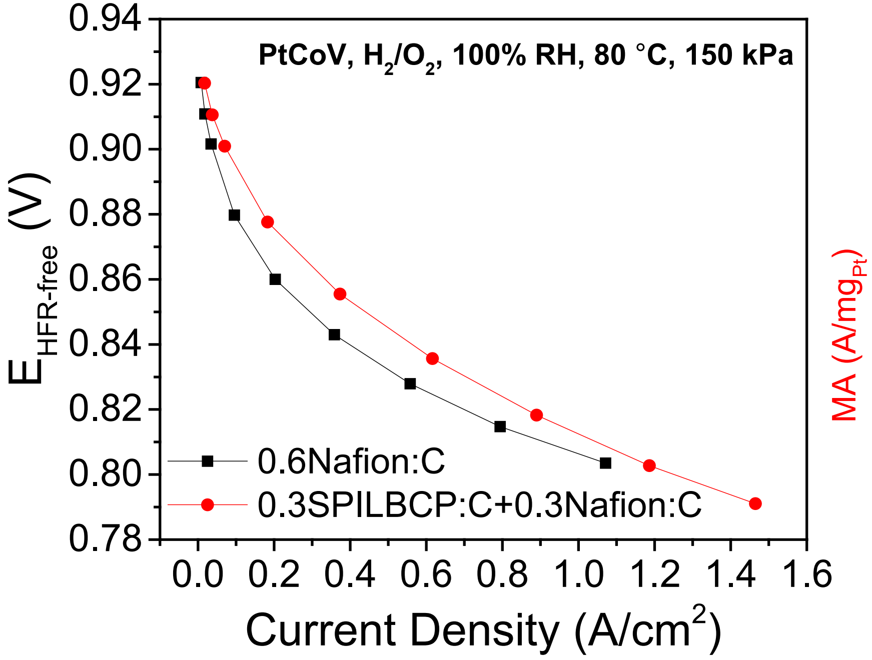
Alcohol-rich w/ 5% PMMA
0.262 – 0.300 mg Pt/cm^2

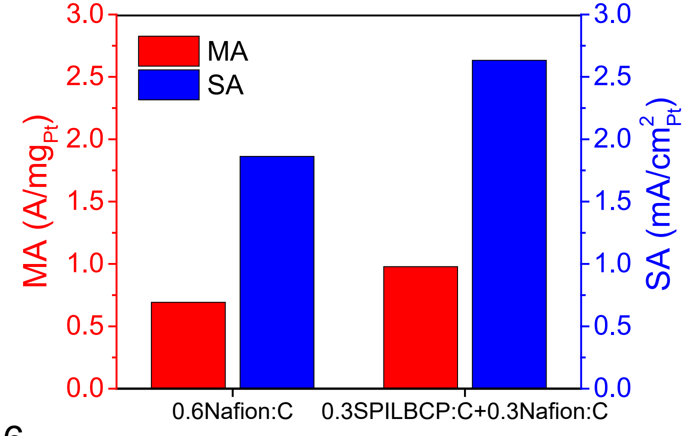




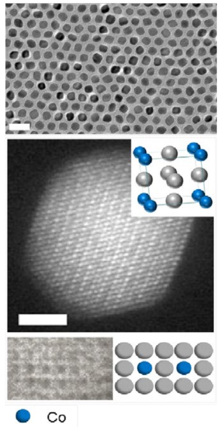
Integrating Novel Materials

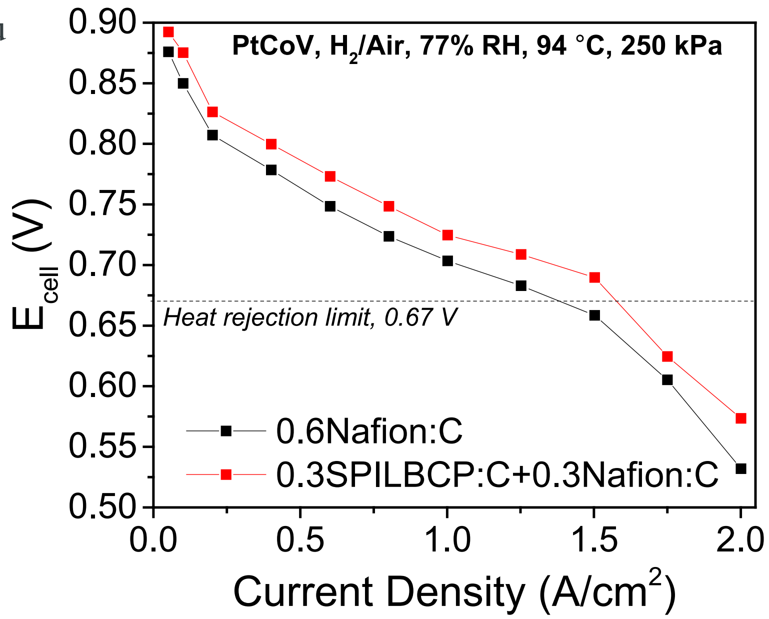
Rated Power Density @ 0.67 V	
0.6Nafion	0.93 W/cm^2
0.3SPILBCP:0.3Nafion:C	1.05 W/cm^2





Ordered inter-metallic PtCo/Vu





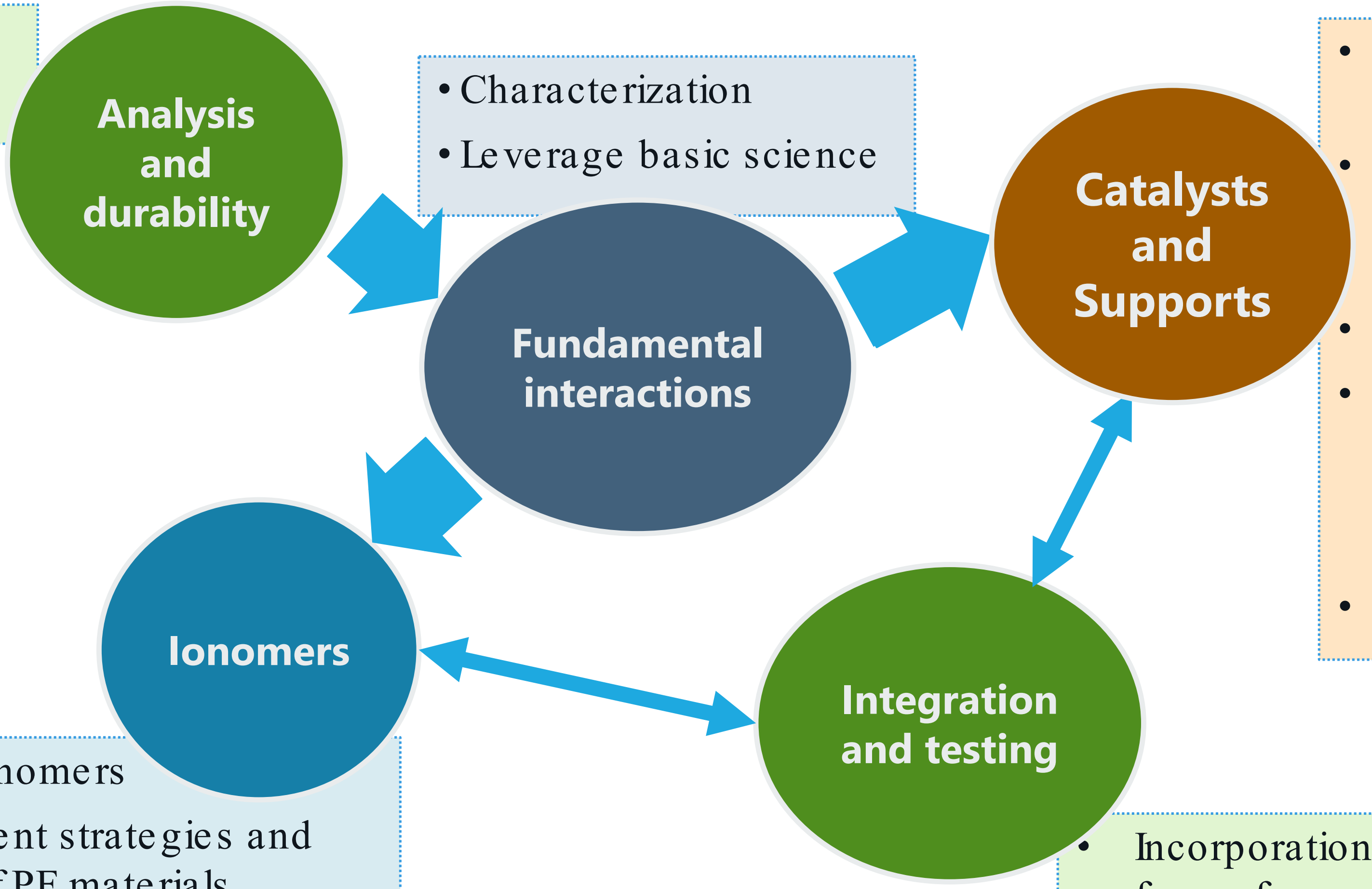
- Ordered inter-metallic PtCo/Vu developed within M2FCT (Stamenkovic)
- Sulfonated polymerized ionic liquid block co-polymers (SPILBCP) from Drexel and Texas A&M University
- 30% increase in mass activity translates to improved efficiency throughout the entire voltage range

- Anode: Pt/HSC (TKK, 46.8 wt%), L = 0.05 mg$_{Pt}$/cm^2, 7w3N, I/C = 0.9
- Cathode: PtCo/V (UCI, 13.1wt%), L = 0.1 mg$_{Pt}$/cm^2, 6w4AcN, I/C = 0.6Nafion:C or 0.3SPILBCP:C+0.3Nafion:C
- 5 cm^2, differential cell

Integration methods based on Neyerlin and coworkers ACS Energy Lett. 5, 1726-1731 (2020)

Materials Development

Synthesizing novel stable and efficient materials for heavy-duty applications



- Design targets
- Operating stressors

- Characterization
- Leverage basic science

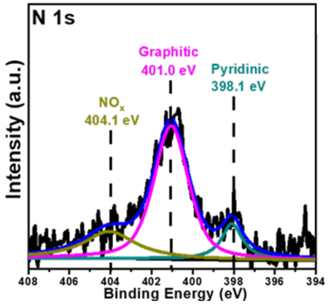
- Pt intermetallics on nitrogen-doped graphitic supports
- Addition of metal oxide (AOx) around Pt and PtM NPs on carbon to stabilize them
- Nitriding of Pt intermetallics
- Control of particle shape, intraparticle composition and structure to inhibit metal dissolution
- Control particle-ionomer interface

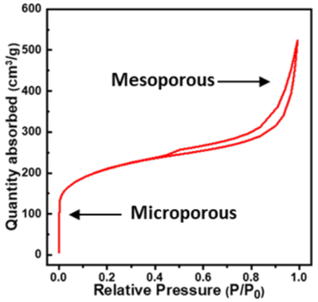
- Novel Perfluoro Ionomers
- Use of reinforcement strategies and characterization of PF materials
- Low molecular-weight oligomers

- Incorporation and testing in MEAs for performance and durability

Structure Engineering for Particles and Supports

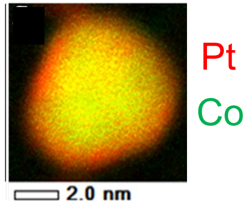
N-doped Porous Graphitic Carbon (NPGC) Support

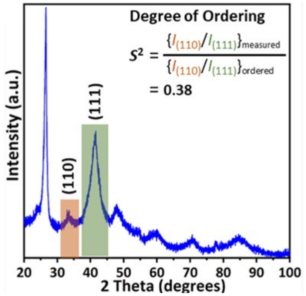


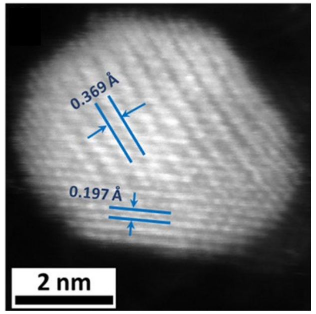


- Uniform nitrogen heteroatoms
- Co-existence of mesopores and micropores
- Pt shell on the L10-CoPt intermetallic core
  - Ordered structure mitigates leaching
  - Strained Pt surface increases activity

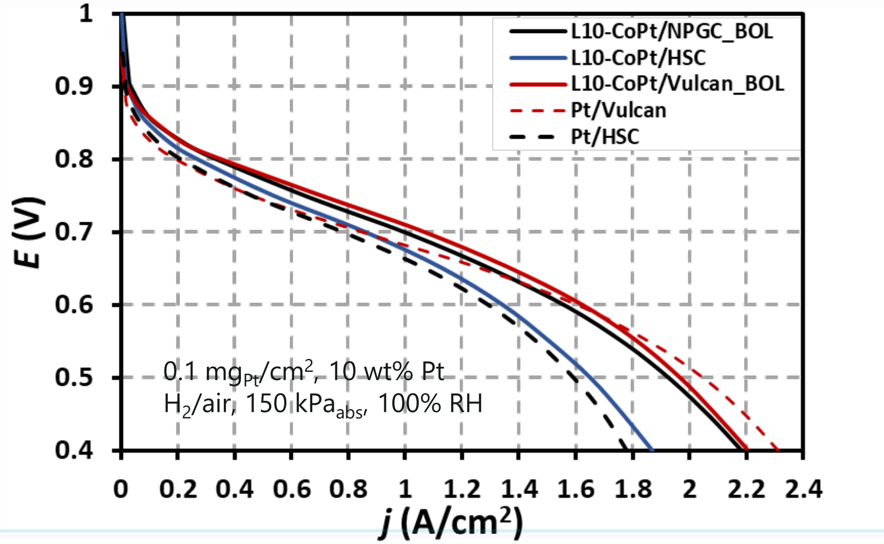
Pt-based Intermetallic Particles



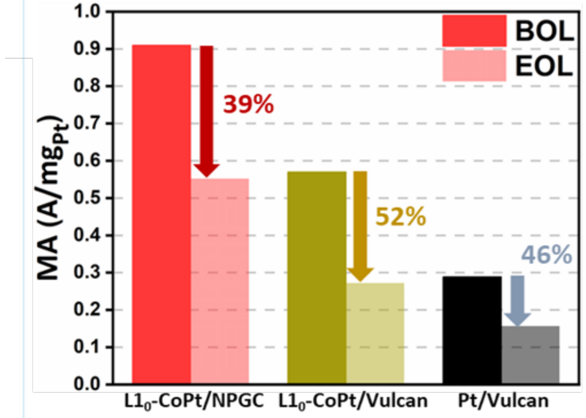


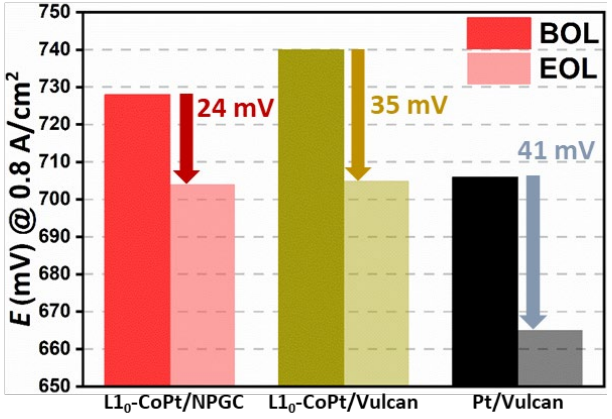


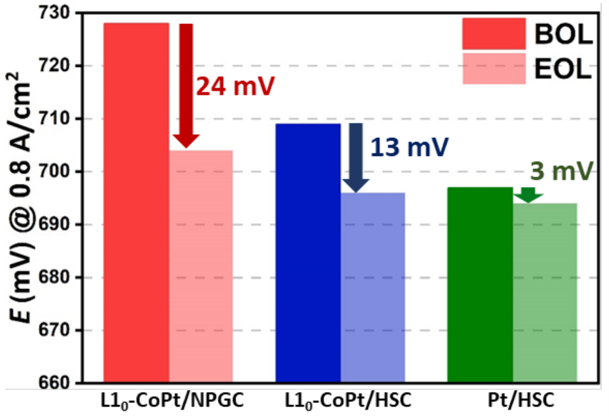
Highly Durable and Active Catalysts



0.1 mg_{Pt}/cm^2, 10 wt% Pt
H_2/air, 150 kPa_{abs}, 100% RH





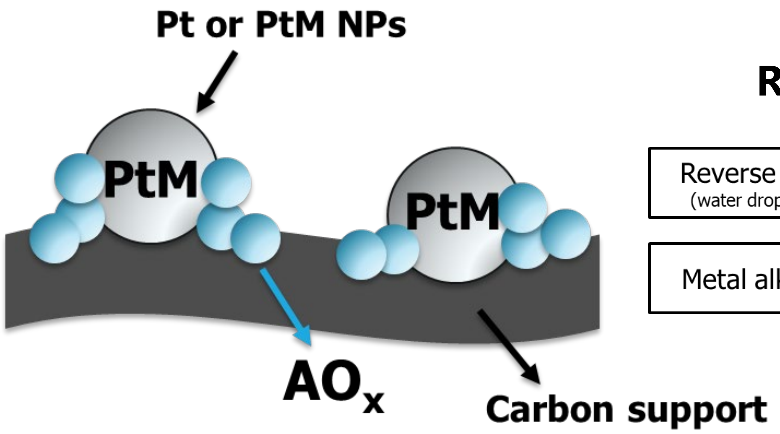


EOL: 30K cycles 0.6-0.95 V

NPGC support possesses an intermediate surface area and porosity compared with the Vulcan carbon support and High -surface area carbon (HSC) support, which renders the catalyst balanced activity, power density, and stability

Oxide Nanoparticles for Stability

- Particle size of 1 to 5 nm can be precisely controlled by using RME method with
- High yield of AOx (through RME) offers the possibility of synthesis scale-up
- Deposition sequence is the key parameter to affect ECSA
  - AOx slightly decreases ECSA and ORR activity – new synthesis to mitigate it



Reverse microemulsion method (RME)

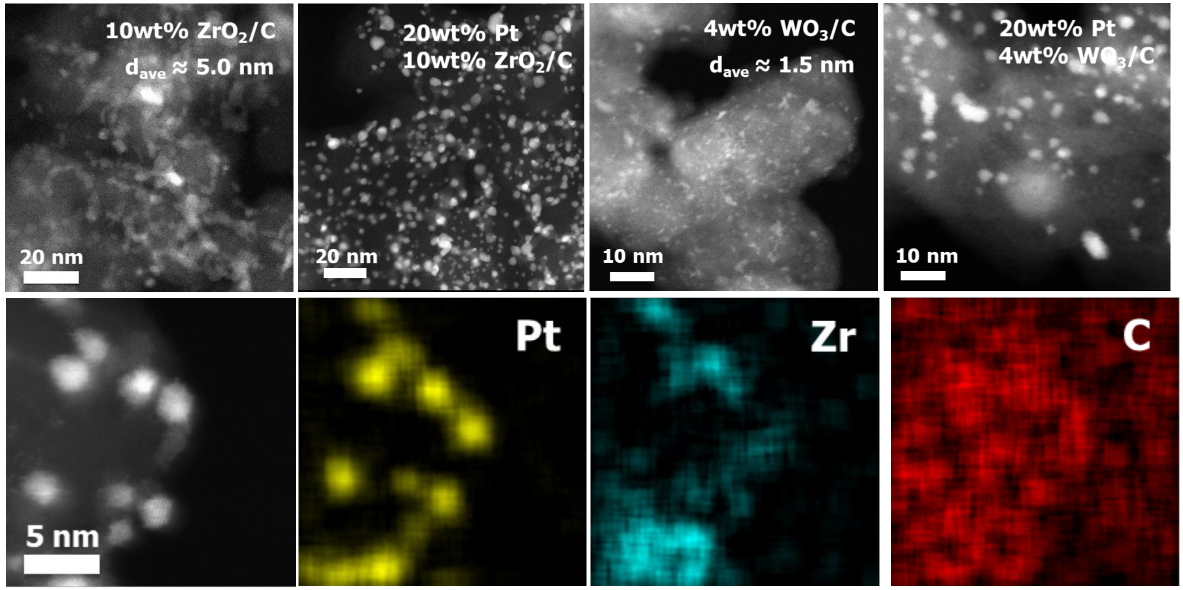
Reverse microemulsion
(water droplet in organic phase)

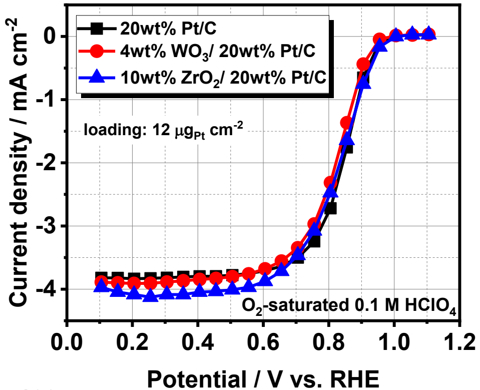
Metal alkoxide solution

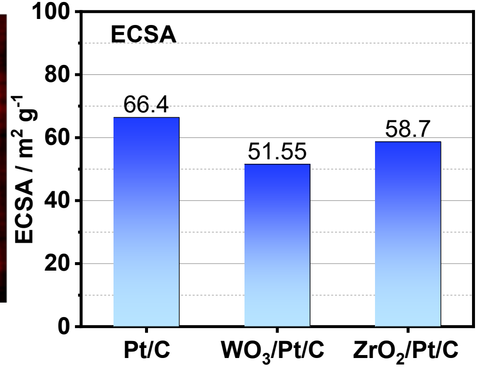
Hydrolysis and aging
(subnano-scaled metal oxide formation)

➡

Deposition and heat-treatment







Intermetallic Synthesis & Characterization

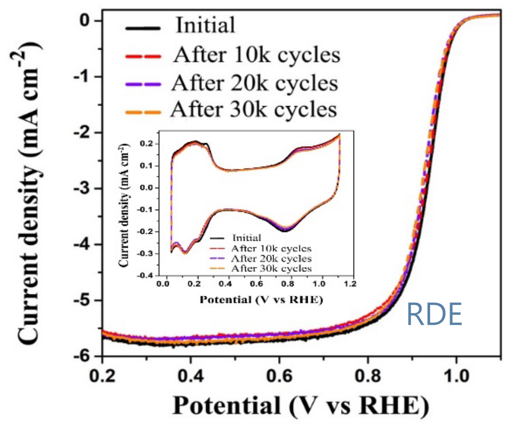
- ORR performance of Intermetallic PtNiN/KB (RDE)

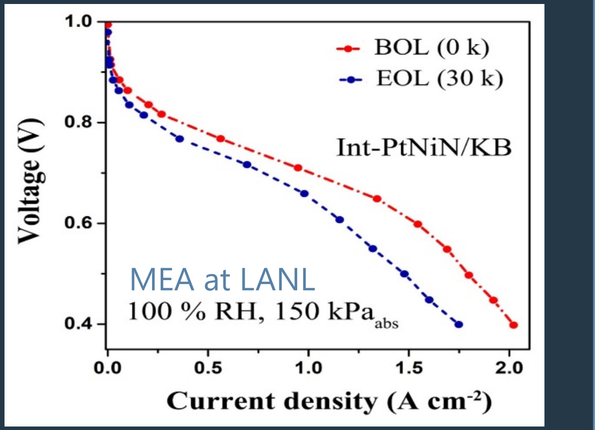
Cycle #	MA (A/mg)	ECSA (m^2/g)	SA (mA/cm^2)	$E_{1/2}$ (mV)
0K	1.83	62.6	2.92	935
30K	1.33 (-27%)	60.3 (-3.7%)	2.21 (-24%)	924 (-11)

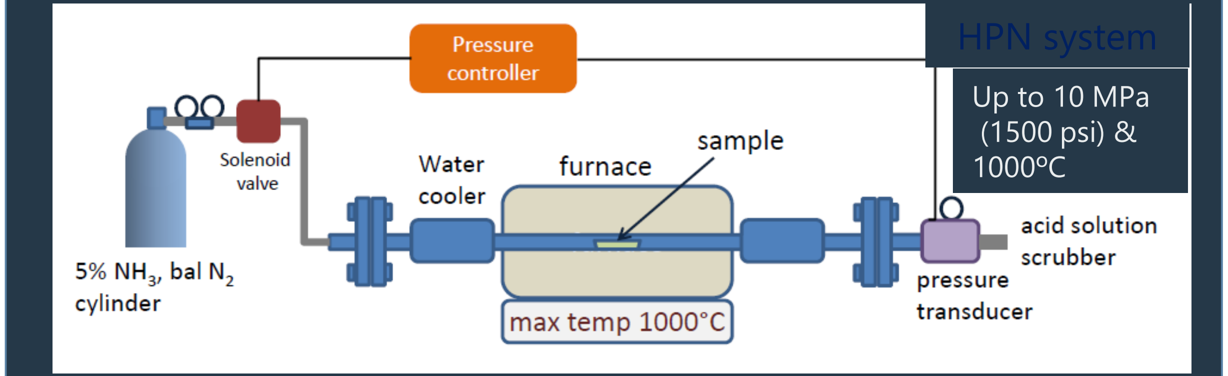
  - MEA: MA (0.49 A/mg), MA loss after 30K (-35%), and current density at 0.8V (348 mA/cm2) exceeded the DOE 2020 LDV targets

- High-pressure nitriding (HPN) - Strategies to further improve the ORR performance
  - Enhanced activity of shape-controlled PtM catalysts by HPN without changing the size and morphology

N-doped Intermetallic PtNiN/KB

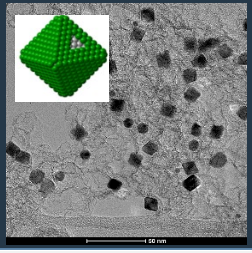


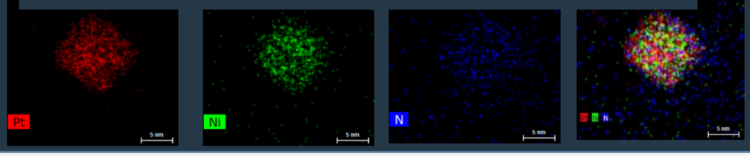




Up to 10 MPa (1500 psi) & 1000°C

N-doped octahedral PtNiN/C by HPN



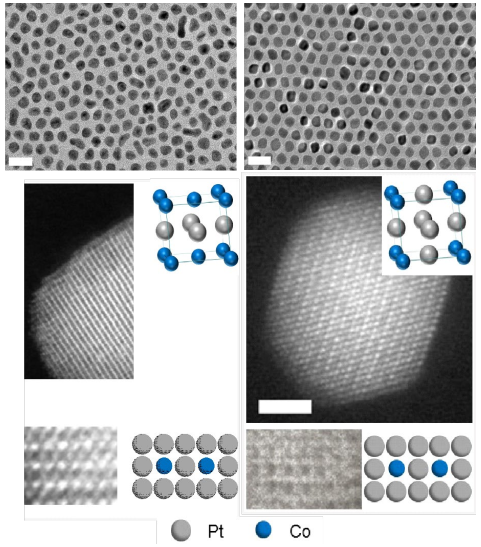


E. Lee, K. A. Kuttiyiel, K.-H. Kim, J. J. Jang, H. J. Lee, J. M. Lee, M. H. Seo, T.-H. Yang, S.-D. Yim, J. A. Vargas, V. Petkov, K. Sasaki, R. R. Adzic, G.-G. Park, ACS Catalysis, accepted.

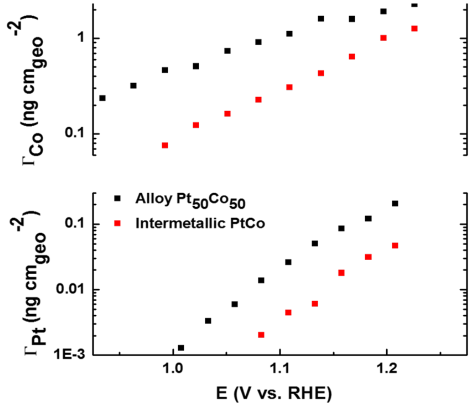
Intra-particle Composition & Structure

- Dissolution of Pt and Co inhibited by formation of ordered intermetallic structure
- Dissolution of Pt inhibited by an Au core underlying Pt skin
- ECSA and MA loss only 20% after 30K AST cycles for PtAu

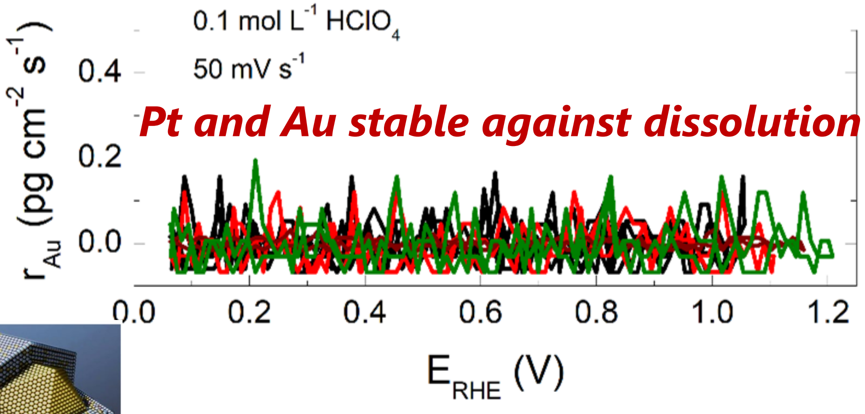
Intermetallic PtCo NPs



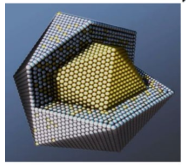
Reduced dissolution of both Pt and Co



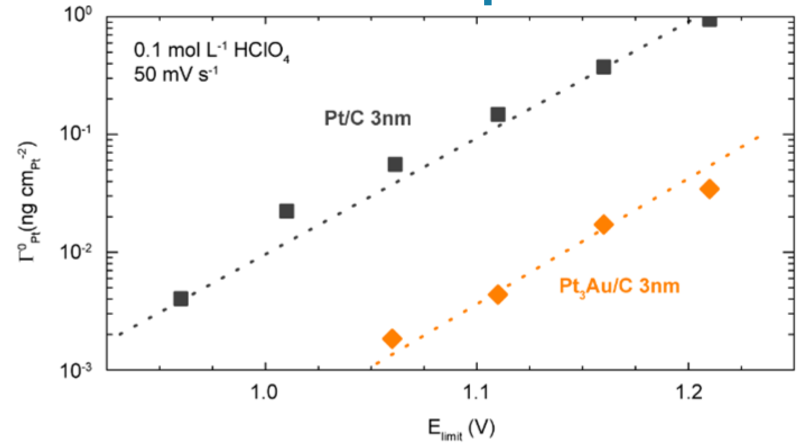
Au Core eliminated Pt dissolution up to 1.2V vs. RHE



Pt and Au stable against dissolution



PtAu nanoparticles



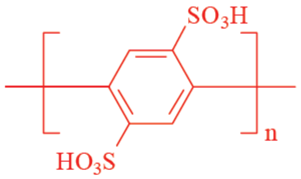
PtAu in MEA:

ECSA and MA loss: ~20% after 30K cycles

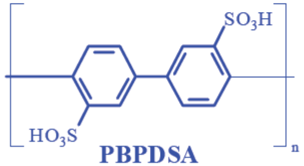
- P.P. Lopes, D. Li, H. Lv, C. Wang, D. Tripkovic, R. Schimmenti, H. Daimon, Y. Kang, J. Snyder, N. Becknell, K.L. More, D. Strmcnik, N.M. Markovic, M. Mavrikakis, & V.R. Stamenkovic, Nature Materials 19, 1207-1214 (2020).

Ionomer Synthesis

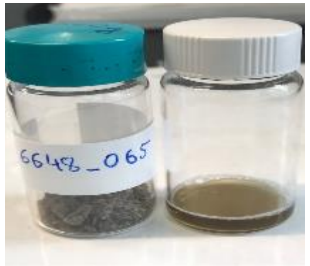
- Development of novel perfluoro chemistries for membranes and ionomers with exceptionally high conductivity, targeting improved selectivity and catalyst/ionomer interactions

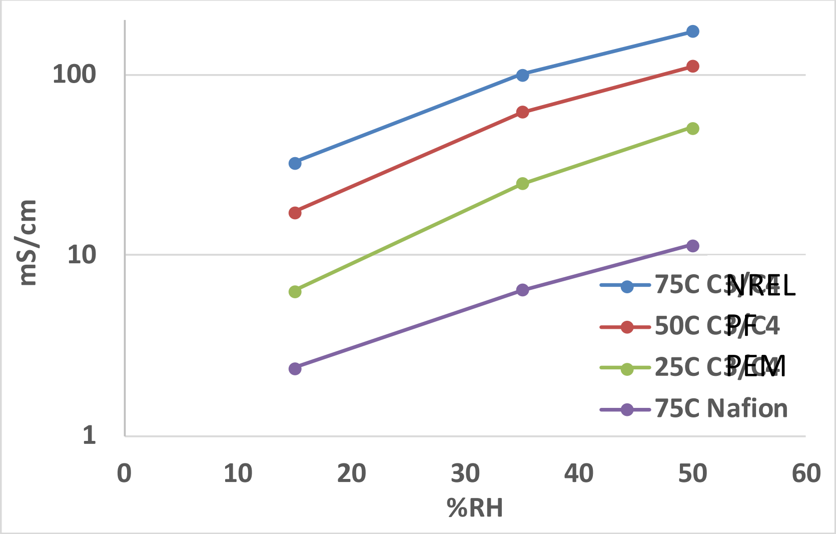


PPDSA



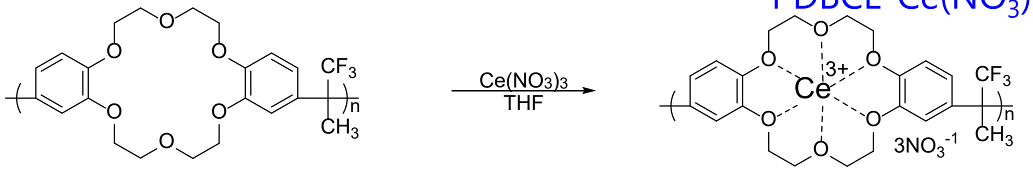
PBPDSA



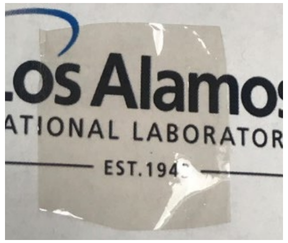


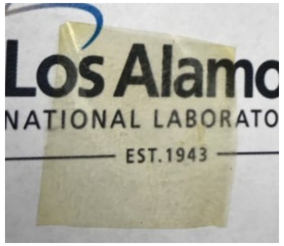
- Synthesis of crow ethers complexes

PDBCE-Ce(NO$_3$)$_3$

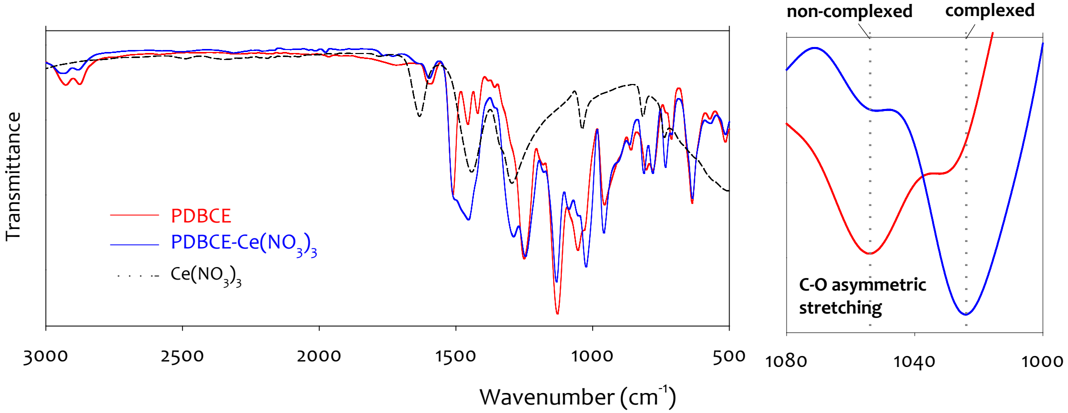


PDBCE





- TGA results indicated the composite polymer contained 38% Ce(NO$_3$)$_3$ (1:1 ratio: 42%)



Collaborations: M2FCT Partners

2019 Commercial Trucks FOA

HD MEA Projects

gm
Carnegie Mellon University
NIKOLA™

2020 H2@Scale FOA

HD Membrane Projects

T
Lubrizol
3M
NIKOLA™

HD Stack Projects

Cummins
PLUG POWER

Status of Interactions

- Virtual meetings held between M2FCT steering committee and 7 MEA/Membrane projects
  - Updates/introductions to projects
  - Discussion 2021 FOA support needs
- All nine FOA projects presented at M2FCT kickoff meeting
  - Cummins and Plug Power manufacturing projects interested in testing M2FCT MEAs
- NDAs executed with GM, Nikola, and CMU
  - NDAs in progress with membrane and manufacturing projects
- FOA project folders hosted on M2FCT google drive
  - Presentations with statements of work.
- Searchable PI and Capabilities list being generated for M2FCT website
- Quarterly meetings to be held between FOA Coordination Officer and FOA PIs
  - Discuss current FOA support efforts and challenges
  - Effort captured in quarterly reports

Also have collaborations through the discretionary funding

Collaborations: Non-FOA activities

Entity	Scope of collaboration
AvCarb	R2R gas diffusion electrode fabrication
3M	Ionomer materials and discussions for ionomer studies
Drexel University	Sulfonated ionic liquid block co-polymer
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

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

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 lifetime prediction with heavy-duty materials and operating conditions
    - Refine existing LDV ASTs
    - Develop new ASTs/protocols specific for HDV
  - Propose new protocols in collaboration with ASTWG by end of FY21
  - Electrode stability
  - Membrane and ionomer durability with additives
  - High-temperature operating time effect on durability
    - Membrane
    - Catalyst
  - Understand long-term durability effects on other components (GDL, contamination, reversible degradation, carbon corrosion at operating potentials)

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

- Catalysts & Catalyst supports
  - Pt-Co Intermetallics
  - Metal oxide-metal-carbon junction to stabilize PtM NPs catalysts
  - Nitrogen-Doped PtMN Catalysts and Supports
  - Tailored Pt nanomaterials, supports, and interfaces
- Membranes & Ionomers
  - High-conductivity Novel Perfluorinated Ionomers
  - Low Molecular Weight Oligomeric Electrode and Membrane Ionomers
  - Composite PFSA Membranes and Ionomer EW and Side-Chain Chemistry
- Other components (GDLs, Bipolar plates…)
- Material and characterization studies

Planned activities include discretionary funding for additional collaborators on identified gaps and needs

Summary

- **Relevance/Objective:**
  - Optimize performance and durability of fuel-cell components and assemblies
  - Emphasis to support heavy-duty transportation applications
- **Approach:**
  - Synergistic combination of modeling and experiments to develop materials, optimize component properties, behavior and phenomena
    - Analysis
    - Durability
    - Materials development
    - Integration
- **Technical Accomplishments:**
  - Analysis of operating conditions, performance & efficiency
  - Durability measurements at projected heavy-duty loadings
    - Development of heavy-duty related ASTs
  - Catalyst Inks to Performance and Catalyst Layer (CL) Analysis:
    - Ink composition-solvent studies, cell testing, impact of fabrication, membrane and ionomer characterization, cell and CL modeling
  - Material Developments
    - Higher mass activity/durable catalyst
    - Advanced ionomers and membranes for durable high-temperature operations
- **Future Work:**
  - Emphasis on efficiency and durability
  - Continue to develop the knowledge base to improve catalyst layer structures and component integration

Who is M2FCT? National Lab Contributors

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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
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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

Technical Backup and Additional Information

Technology Transfer Activities

- Copyright of microelectrode drawings
  - R&D 100 award
- Engaging with various stakeholders and OEMs including FOA projects and others as noted
- Publications, presentations, discussions
- Outreach through website development

Publications

- Katzenberg, Adlai, Debdyuti Mukherjee, Peter J Dudenas, Yoshiyuki Okamoto, Ahmet Kusoglu, and Miguel A Modestino. "Dynamic Emergence of Nanostructure and Transport Properties in Perfluorinated Sulfonic Acid Ionomers." *Macromolecules* 53.19 (**2020**) 8519-8528.
- R. K. Ahluwalia, X. Wang, L. Osmieri, J-K Peng, C. F. Cetinbas, J. Park, D. J. Myers, H. T. Chung, K. C. Neyerlin, "Stability of Atomically Dispersed Fe–N–C ORR Catalyst in Polymer Electrolyte Fuel Cell Environment" *Journal of Electrochemical Society*, 168 (**2021**) 024513
- Berlinger, Sarah A, Peter J Dudenas, Ashley Bird, Xunkai Chen, Guillaume Freychet, Bryan D McCloskey, Ahmet Kusoglu, and Adam Z Weber. "Impact of Dispersion Solvent on Ionomer Thin Films and Membranes Impact of Dispersion Solvent on Ionomer Thin Films and Membranes." *ACS Applied Polymer Materials* 2.12 (**2020**) 5824 - 5834.
- Ehlinger, Victoria M, Andrew R Crothers, Ahmet Kusoglu, and Adam Z Weber. "Modeling proton-exchange-membrane fuel cell performance/degradation tradeoffs with chemical scavengers." *Journal of Physics: Energy* 2.4 (**2020**) 044006.
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- Van Cleve T.; Wang G., Mooney M., Cetinbas C.F., Kariuki N., Park J., Farghaly A., Myers D., Neyerlin K.C. "Tailoring electrode microstructure via ink content to enable improved rated power performance for platinum cobalt/high surface area carbon based polymer electrolyte fuel cells." *Journal of Power Sources*, 482, **2021**, 228889.
- X. Zhao, H. Cheng, L. Song, L. Han, R. Zhang, G. Kwon, L. Ma, S. Ehrlich, A. I. Frenkel, J. Yang, K. Sasaki, H. L. Xin, Rhombohedral Ordered Intermetallic Nanocatalyst Boosts the Oxygen Reduction Reaction, *ACS Catalysis*, 11 (2021) 184-192; DOI:10.1021/acscatal.0c04021.
- Abir, Muhammad; Hussey, Daniel S; Khaykovich, Boris; "Design of Neutron Microscopes Equipped with Wolter Mirror Condenser and Objective Optics for High-Fidelity Imaging and Beam Transport". *Journal of Imaging*, 6(10) p. 100 (**2020**).
- Bethune, Keith; St-Pierre, Jean; LaManna, Jacob M; Hussey, Daniel S; Jacobson, David L; "Contamination Mechanisms of Proton Exchange Membrane Fuel Cells-Mass Transfer Overpotential Origin". *The Journal of Physical Chemistry C*, v. 124(44), p.24052 (**2020**).
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- Hussey, DS; Abir, M; Cook, JC; Jacobson, DL; LaManna, JM; Kilaru, K; Ramsey, BD; Khaykovich, B; "Design of a neutron microscope based on Wolter mirrors". *Nuclear Instruments and Methods in Physics Research Section A*, v. 987, p. 164813 (**2021**).

Progress toward DOE Targets or Milestones

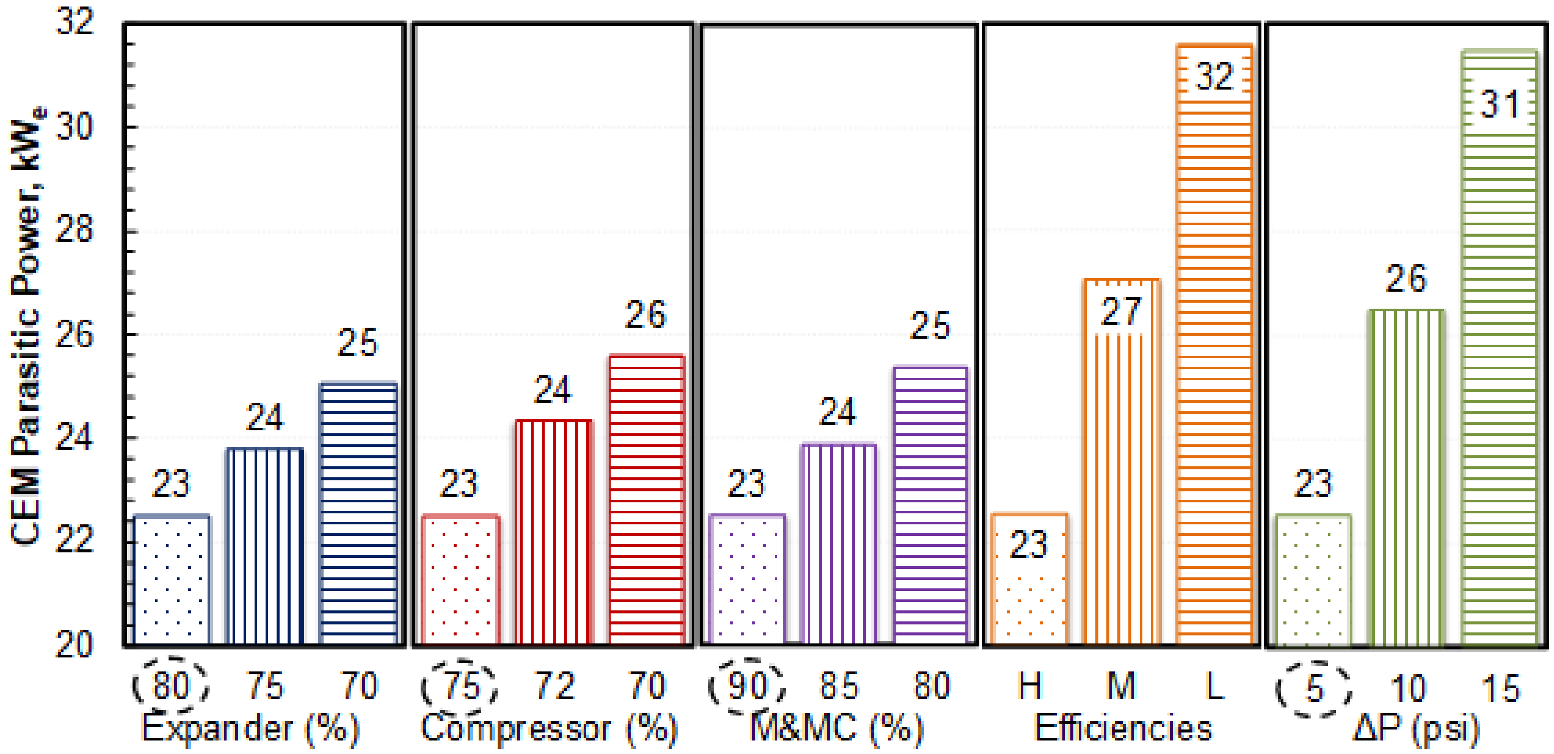
M2FCT 2025 Target

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

- Existing baseline efforts with new materials and also development of AST and test protocols mean that it is too early yet to judge where we are at in terms of this metric

FCS for Medium and Heavy-Duty Trucks: Air Management

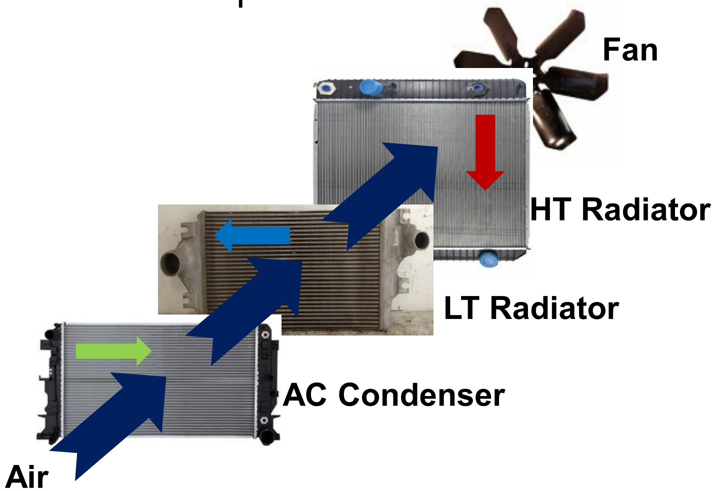
	Units	High (H)	Medium (M)	Low (L)
Compressor Efficiency	%	75	72	70
Expander Efficiency	%	80	75	70
Combined Motor & Motor Controller Efficiency (M&MC)	%	90	85	80
Pressure Drop: Compressor Outlet to Expender Inlet (∆P)	psi	5	5	5



- Feasible to have a single air system with <27 kW_e parasitic power if H or M component efficiencies can be achieved
- Recommended performance targets to DOE for FOA on air management system for HD trucks

FCS Heat Rejection on Hill Climb

Modeled system equivalent to radiator fan for 450-hp diesel engine, 52°C air-to-boil temperature

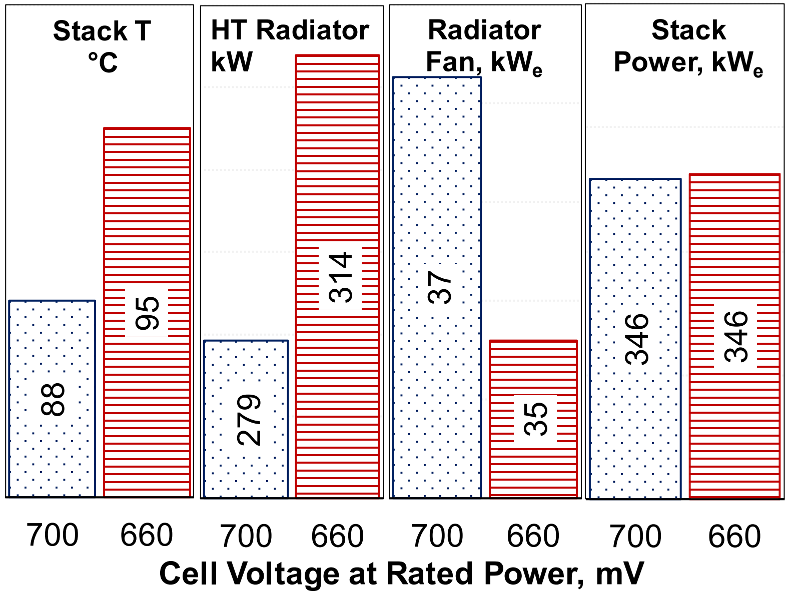


Study Conclusions

Study parameters: Hill climb at 30 mph, 6% grade, 20-min duration, 25°C ambient temperature

1. Largest FCS for which waste heat can be rejected using radiators in a 450-hp diesel trucks: 275 kW_e net
2. Smallest battery operated in charge depleting mode during hill climb: 35 kWh
3. Lowest cell voltage at EOL for exit coolant temperature below 95°C: 660 mV

Heat Exchangers	Dimensions and Details	Heat Loads	Radiator Fan 37 kW
HT Radiator	40″ (W) x 42″(H) x 2″ (D) Fins: louvered, 12-fpi, 10-mm height Tubes: 2-mm height	280 kW	Vehicle Speed: 30 mph
LT Radiator	40″ (W) x 35″ (H) x 2.5″ (D) Fins: louvered, 8-fpi, 20-mm height Tubes: 10-mm height	49 kW	Ambient T: 25°C
AC Condenser	40″ (W) x 28″ (H) x 0.75″ (D) Fins: plain, 12-fpi, 10-mm height Tubes: 2-mm height	12 kW	Air Flow Rate: 9.3 kg/s



Parameter sensitivity analysis from cell model

- Summary of most crucial model parameters
  - Need to be measured with better accuracy and certainty
  - Guide for experimentalists

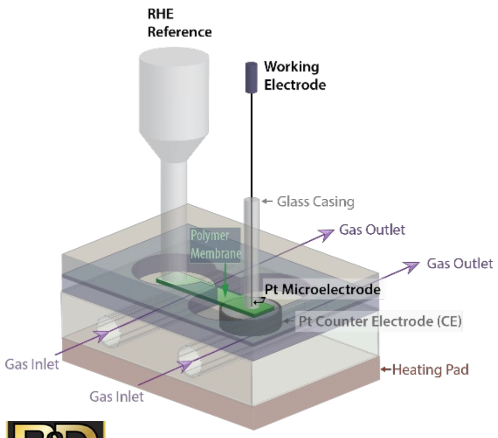
Type	Operating conditions	Most crucial model parameters
Dry	80°C, 40% RH, air	ORR α_C, Ionomer water uptake, ECSA, membrane conductivity
Normal	80°C, 80% RH, air	ORR α_C, Ionomer water uptake, GDL porosity, ECSA
Oxygen starved	80°C, 80% RH, 5% O_2	ORR α_C, Ionomer water uptake, ECSA, oxygen reaction order
Flooded	80°C, 100% RH, air	CL saturation curve, ionomer contact angle, ORR α_c, GDL porosity & tortuosity

Analyzing Transport Properties Using Microelectrodes

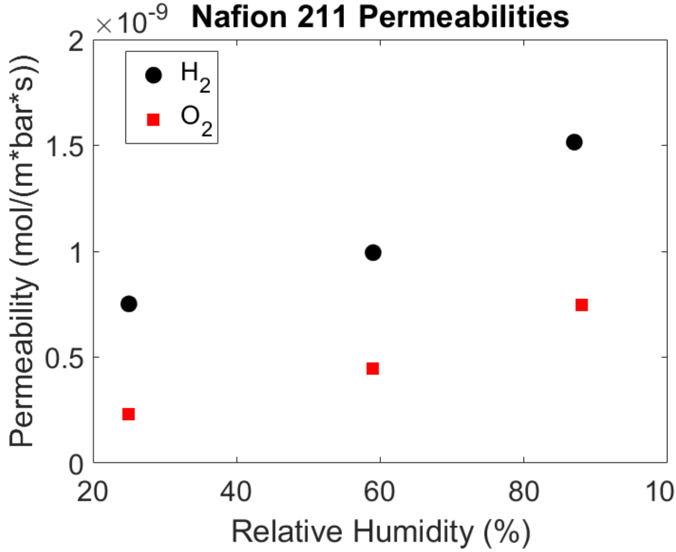
- Developed method for determining gas solubility and diffusivity in ionomer membranes
  - Multi-regime analysis used to extract parameters
    - More accurate than previous methods of calculation
    - Long-time: Numerical model for complicated geometry, extracts permeability of membrane
    - Short-time: Cottrell equation modified to account for higher initial surface area, uses permeability to extract D
    - Cottrell Equation[1]:
    - $P = DK_g$ can then be used to determine the solubility, K_g

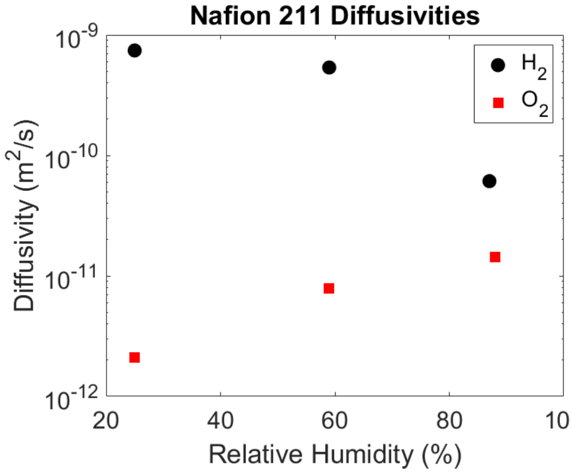
$$I = \frac{nF\pi^{0.5} P C_g R^2 \boldsymbol{R_F}}{D^{0.5} t^{0.5}}$$

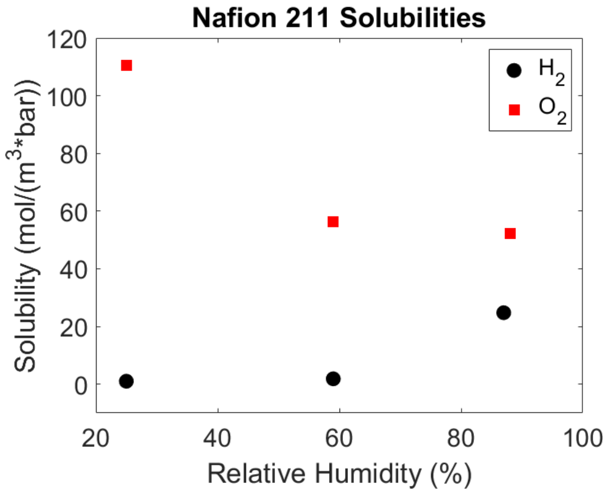
- Data demonstrate different trends for hydrogen and oxygen in terms of diffusivities



R&D 100 WINNER







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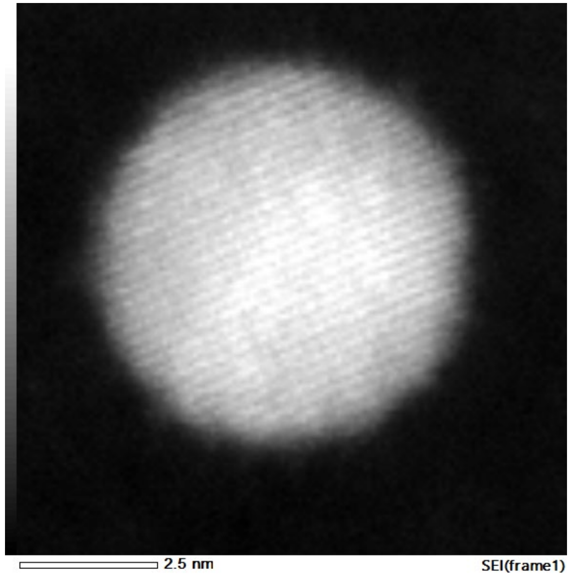
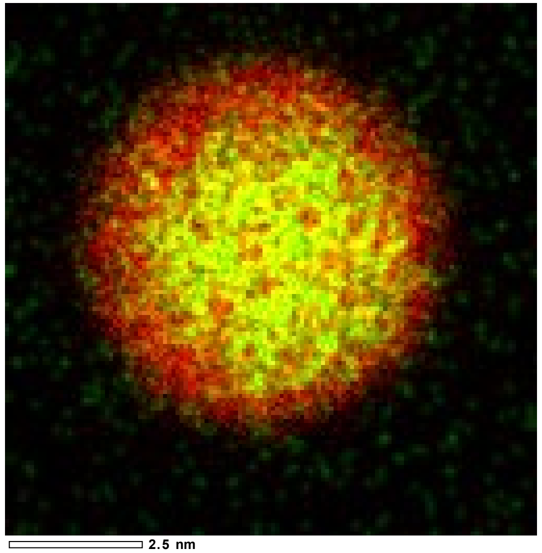
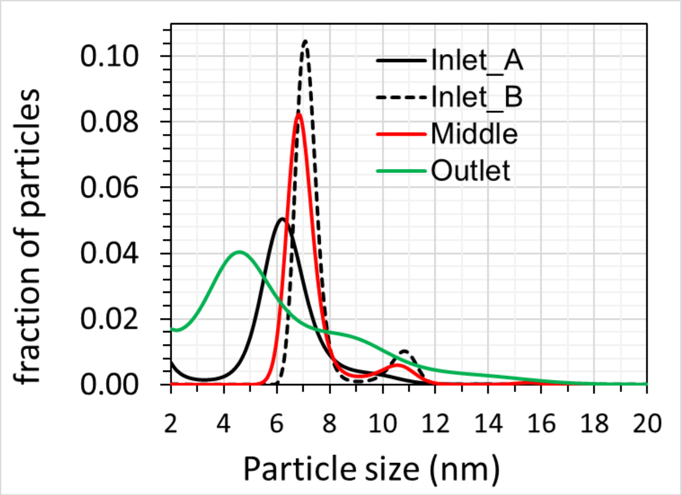
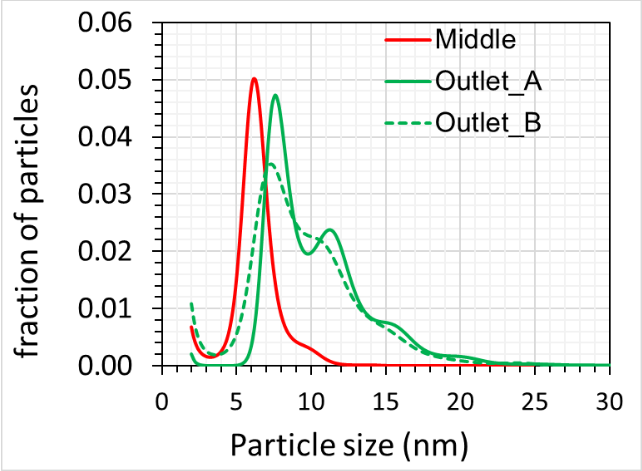
[1] A. Parthasarathy, *Journal of The Electrochemical Society*, 138, 916 (1991).

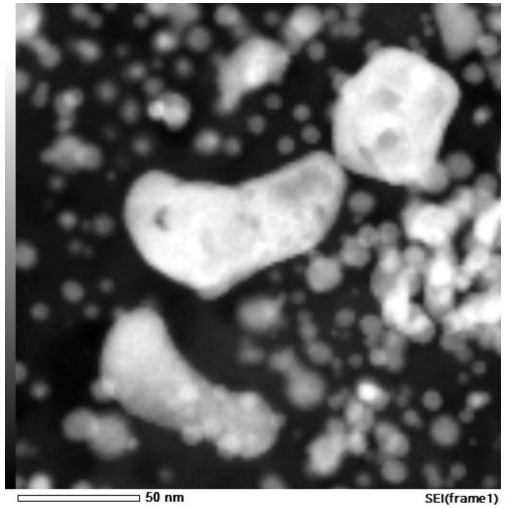
Catalyst Durability Case Study

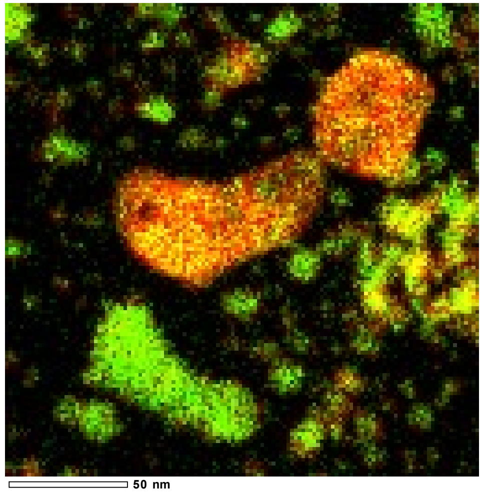










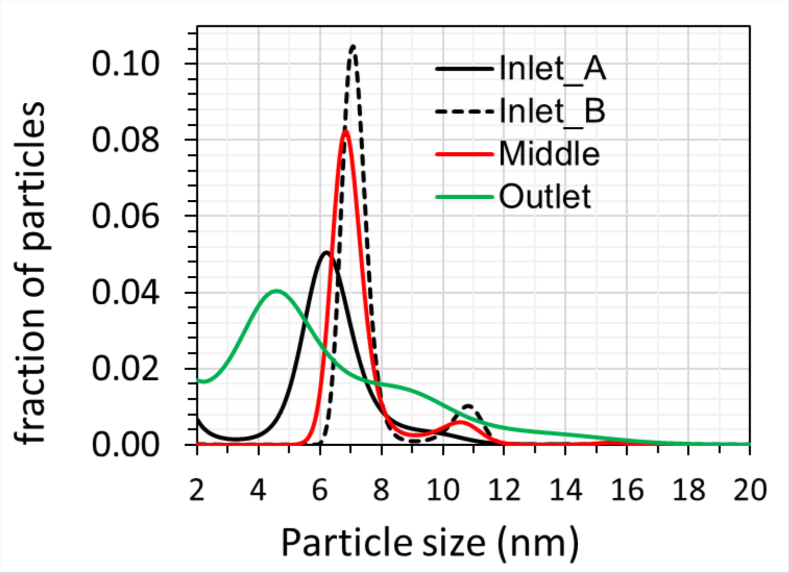


- Shell thickness is pretty significant (> 1nm) for the 4-7nm particles
- Particles 10-50nm tend to be more Co rich than smaller particles, but with even thicker shells
- Some of the larger particles have a spongy structure (catastrophic Co loss) and are very Pt-rich
- USAXS and WAXS provide global analysis
- Significant particle size growth with more growth at the outlet
- Significant Co leaching observed

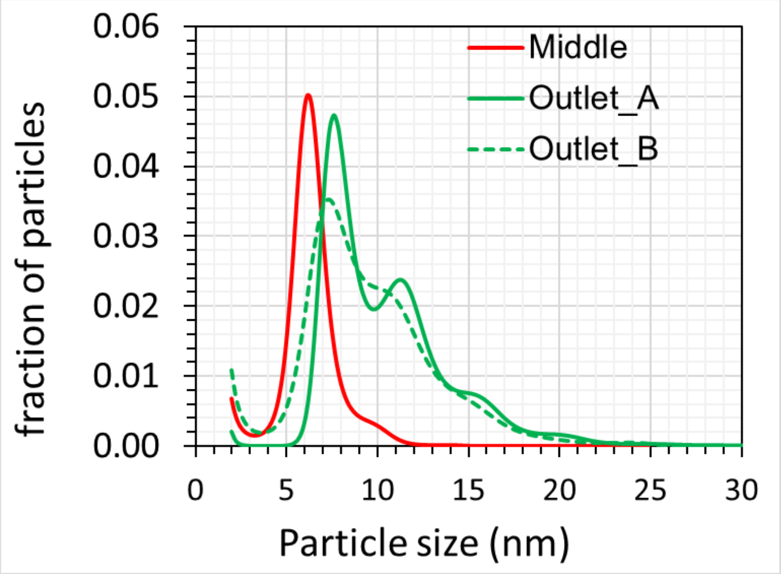
ASTWG

- USAXS and WAXS provide global analysis
- Significant particle size growth with more growth at the outlet
- Significant Co leaching observed

Pt:- PSD



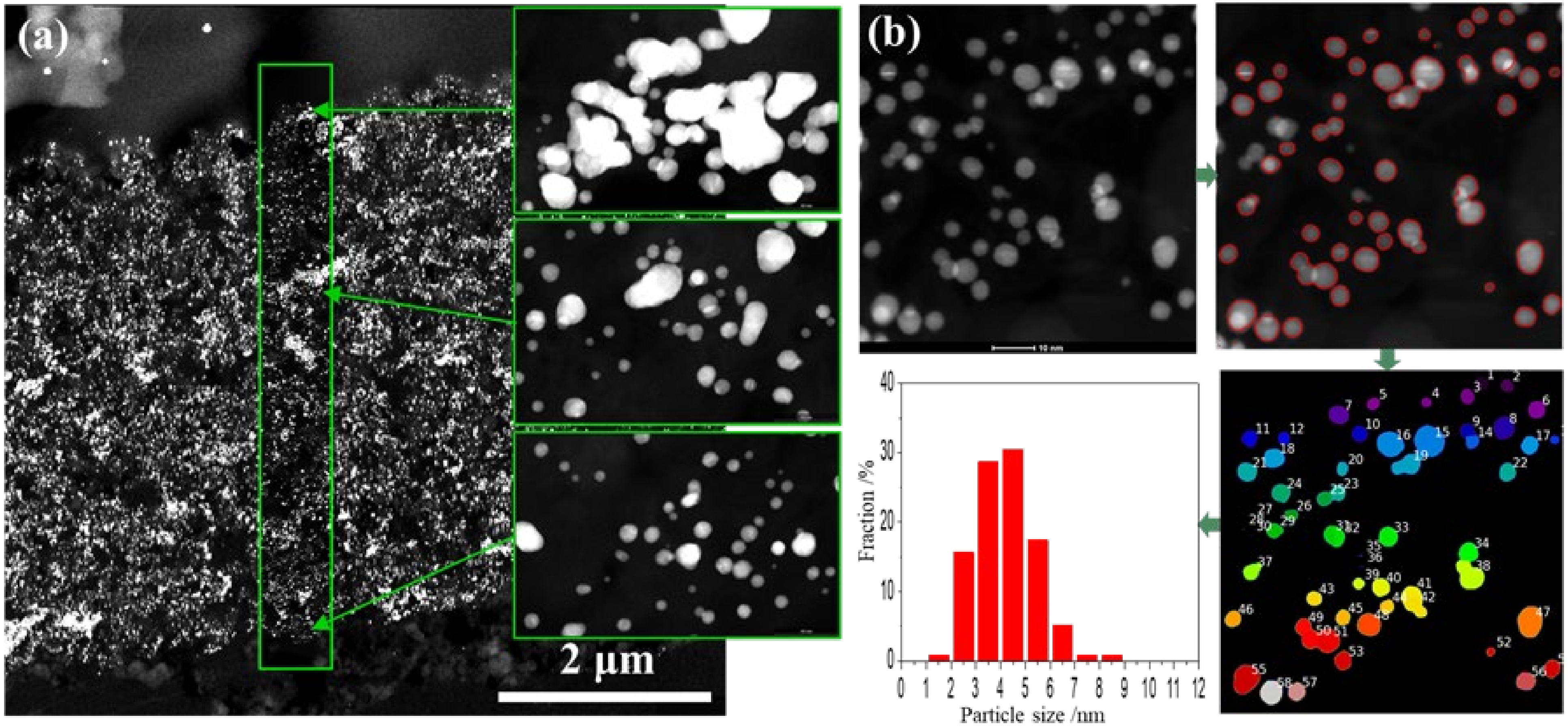
PtCo PSD



	USAXS		WAXS Peak (220)
Sample	particle size (nm)	Stdev (nm)	Particle size (nm)
Inlet A	6.53	1.35	10.01
Inlet B	7.53	1.19	10.92
Middle	7.34	1.36	10.40
Outlet	6.42	3.33	[a]12.21
Outlet			[a]70.11
Outlet			[b]16.57

	USAXS		WAXS Peak (220)	
Sample	particle size (nm)	Stdev (nm)	Particle size (nm)	% composition Pt/Co
Middle	6.53	1.36	4.85	92.1/7.9
Middle			14.59	100/0
Outlet_A	10.53	3.46	4.47	94.5/5.5
Outlet_ A			15.35	100/0
Outlet_ B	9.86	3.4	4.50	93.6/6.4
Outlet_B			14.21	100/0

Automated data acquisition and analysis in STEM

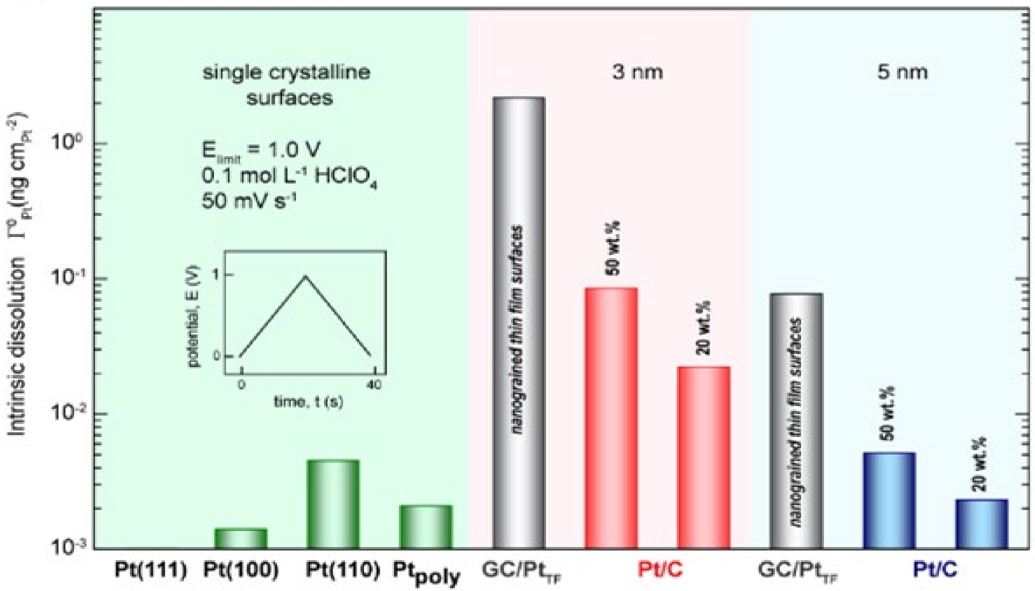


- Automated image acquisition software purchased for FEI Talos TEM
  - Instrument runs autonomously overnight acquiring dozens of images and EDS maps
  - Custom python codes under development for handling these large data sets for automated particle size and ionomer distribution measurements

Approach: Suppressing Pt Dissolution to Enhance Durability

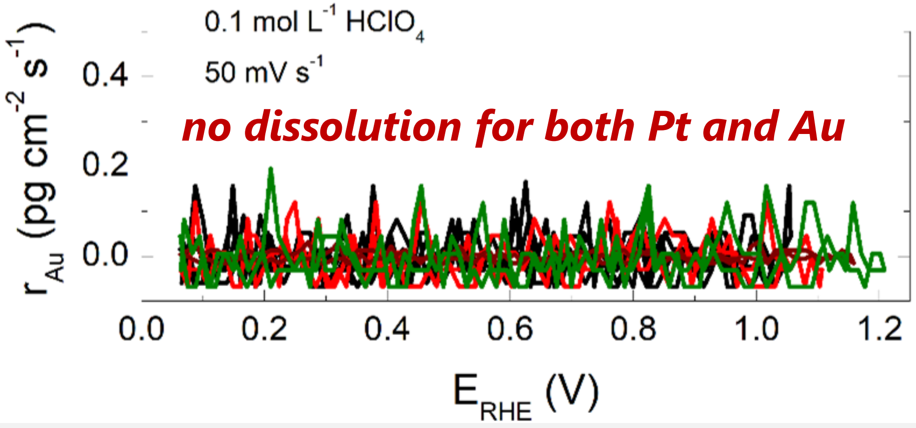
ANL / UC Irvine: Vojislav Stamenkovic

Establishing the baseline for Pt dissolution



intrinsic Pt dissolution normalized to Hupd

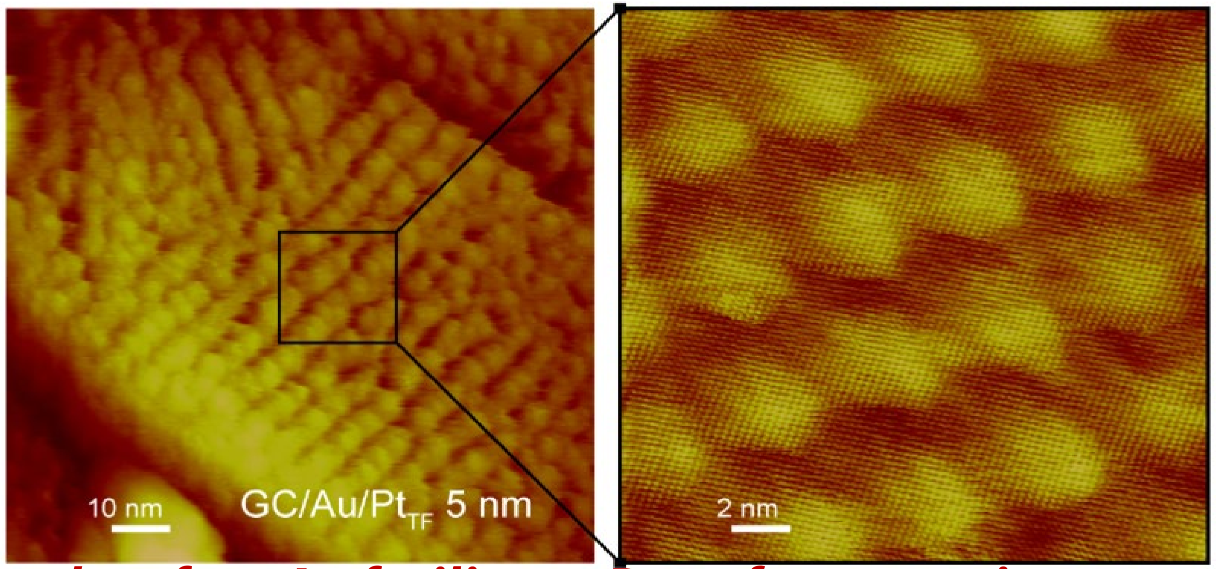
Eliminated Pt dissolution up to 1.2V vs. RHE



no dissolution for both Pt and Au

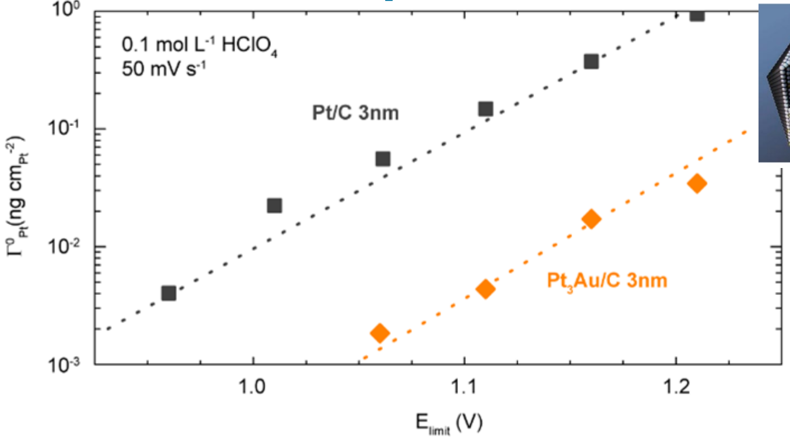
Foundation of the mechanism to eliminate Pt dissolution

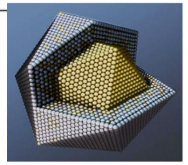
PtAu thinfilms



subsurface Au facilitates Pt surface texturing towards (111)

PtAu nanoparticles





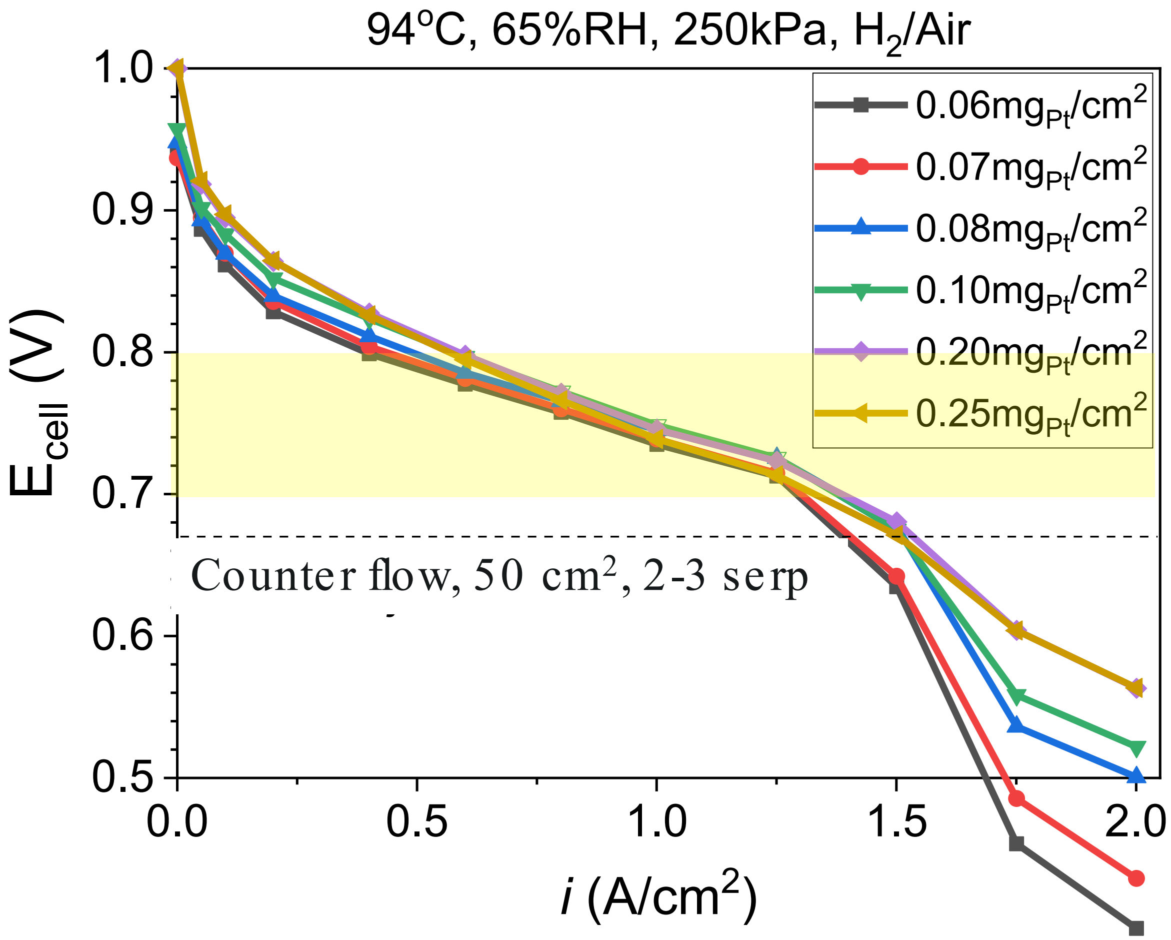
PtAu in MEA:

ECSA and MA loss: ~20% after 30K cycles

Pt Loading

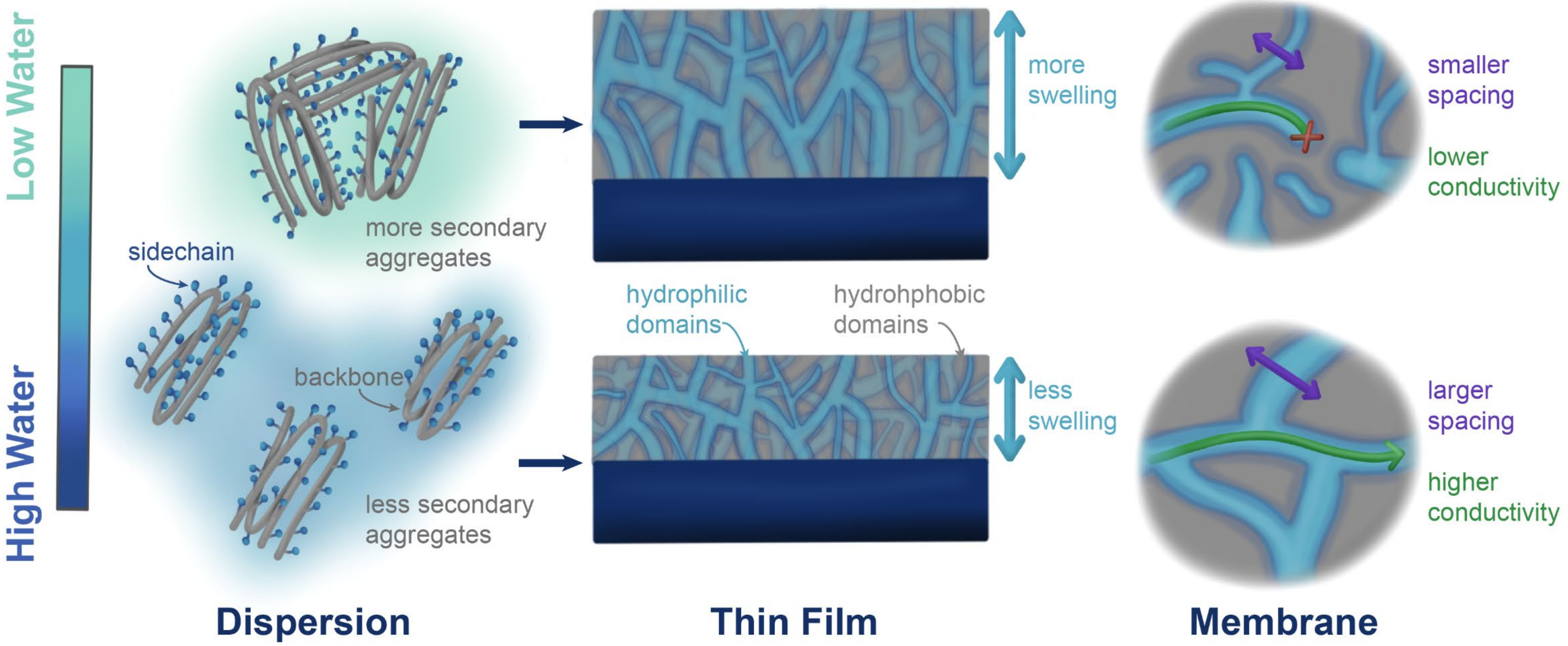
- Kinetic improvements with increased loading from 0.06 to 0.25
- In high H2 consumption region there is currently minimal improvement with increased Pt loading
- More baseline data coming with 50 wt% PtCo/HSC (Umicore) and 50 wt% Pt/HSC (TKK)
    - Need both materials and integration solutions
    - Need to keep metal (Pt) wt% high
        - Keep carbon loading below ~0.3 mg/cm²

- 30 wt% PtCo/HSC Umicore



Dictating Thin films/ Membrane Properties

Nafion films/membranes prepared from high water concentration dispersions have more interconnected and ordered networks



ACS Applied Polymer Materials (2020).

Ionomer EW trends validated via *ex-situ* experiments

Results confirm the impact of EW on ionomer thin-film permeability and catalyst poisoning

Confirm direct correlation between *ex-situ* model experiments and *in-situ* performance trends

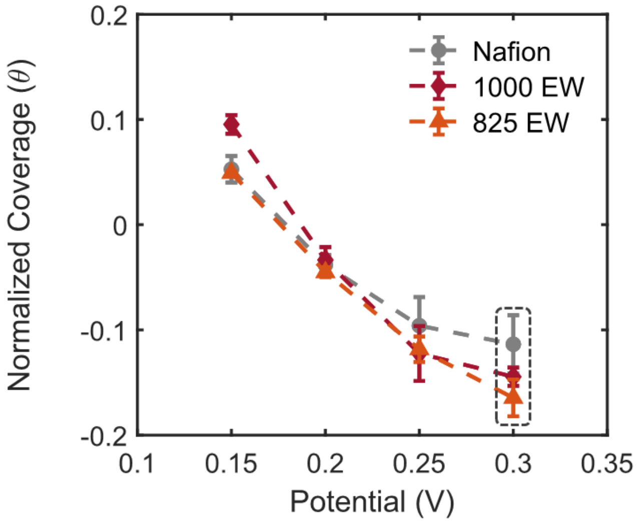
- Catalyst poisoning trends

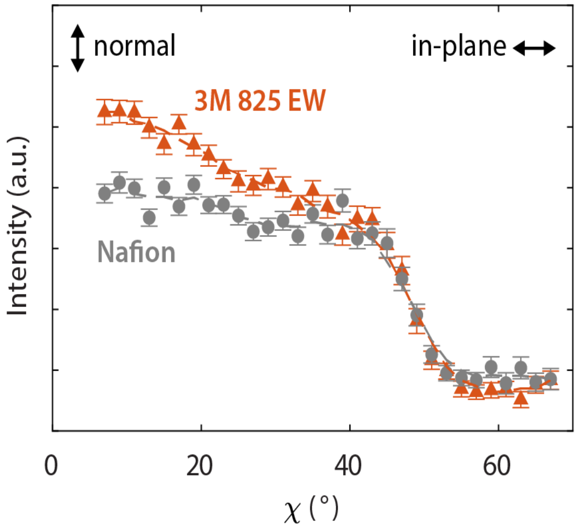
CO displacement: Higher anion adsorption with low EW ionomers

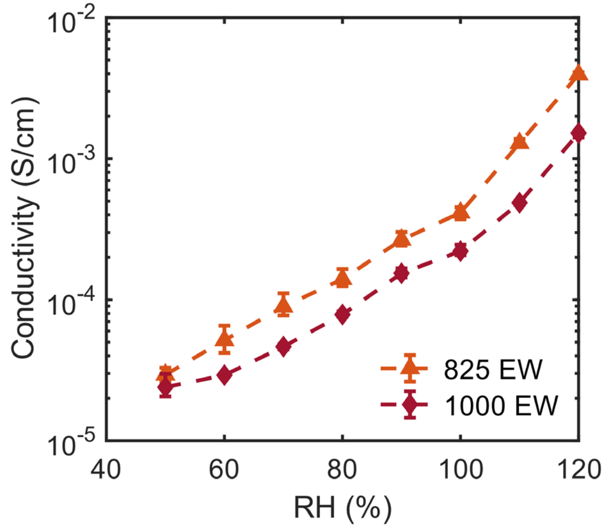
GISAXS: Low EW ionomer have higher intensity at low χ – higher anisotropy

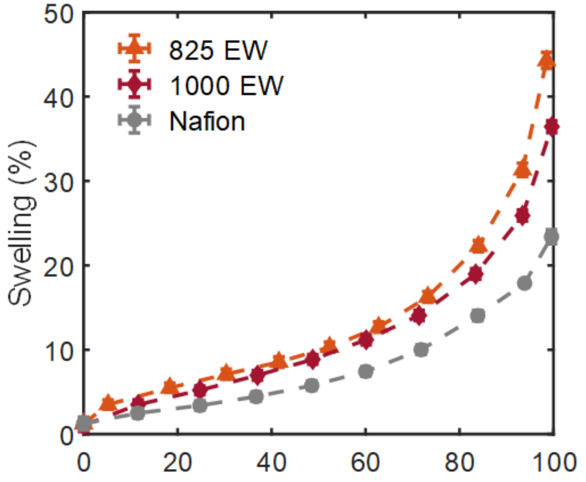
- Thin-film Permeability trends

In-situ (conductivity) and ex-situ (ellipsometry) measurements confirm higher water uptake with low EW

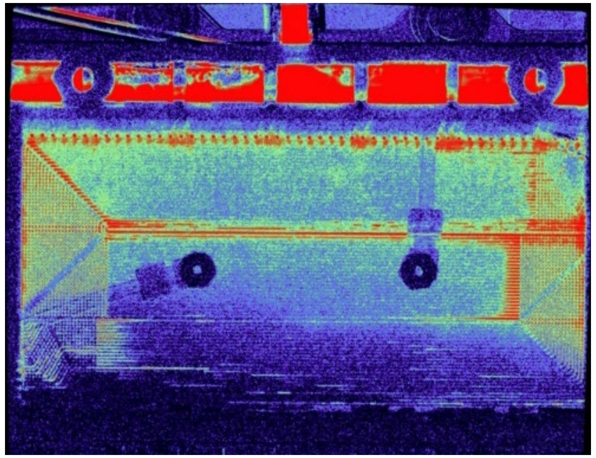


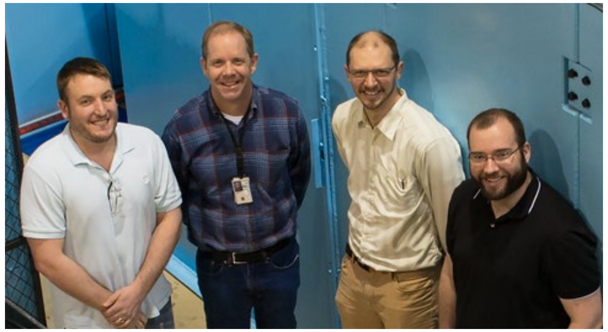




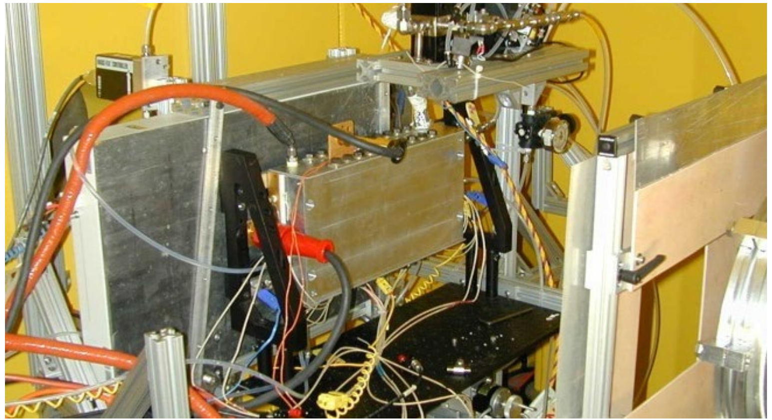


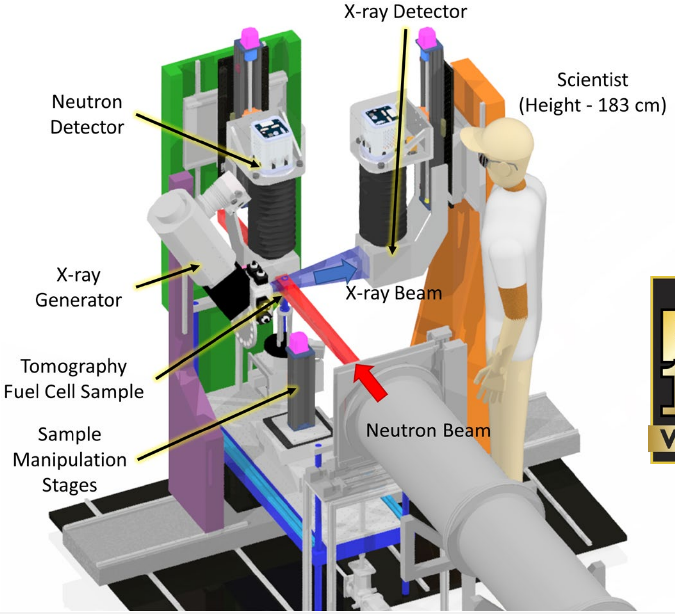
Neutron Microscopy of Water Transport Phenomena


Operando images of the Mirai


NIST neutron imaging team: Eli Baltic (left), David Jacobson, Daniel Hussey (PI), and Jacob LaManna (right)

- Neutrons readily penetrate several cm of common metals, but have high sensitivity to water
- NIST's neutron and X-ray tomography (NeXT) system will be upgraded fall 2021 (lower right)
  - Higher energy and finer spot sized X-ray source extend application range
  - Neutron detector with improved time and spatial resolution
  - Dose reduction algorithms enable 1 hour acquisition times for 3D water content
- A neutron microscope is anticipated for the end of FY2022
  - With spatial resolution ~3 µm, and time resolution 1 s, this will nearly raise neutron imaging to the level of synchrotrons


"Plug and Play" support for full scale test sections for testing M2FCT materials



2018 R&D 100 WINNER

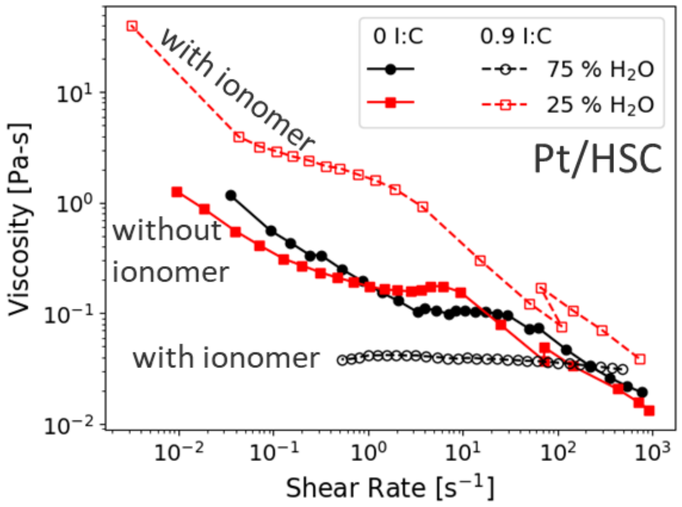
M2FCT Component Integration - Manufacturing

- Understand impact of ink formulation and processing on (a) coating quality (b) electrode structure (c) performance and durability
- Develop correlations between catalyst/ink parameters and ink macro behaviors (e.g., rheology)
- Understand impact of MEA/component defects on durability (and durability studies)

Explore novel electrode structures to improve durability

- Ink additives to modify porosity, structural integrity, and cracking
- Multilayer structures

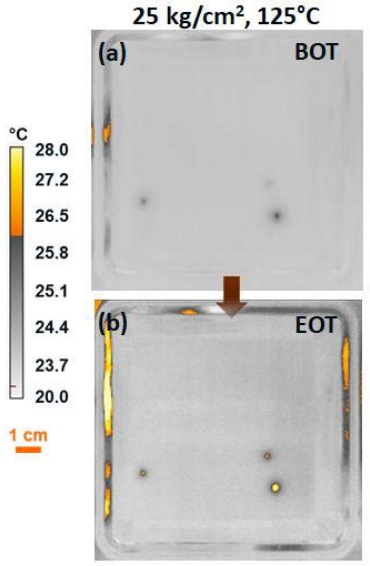
Develop and demonstrate methods for in-process ink QA

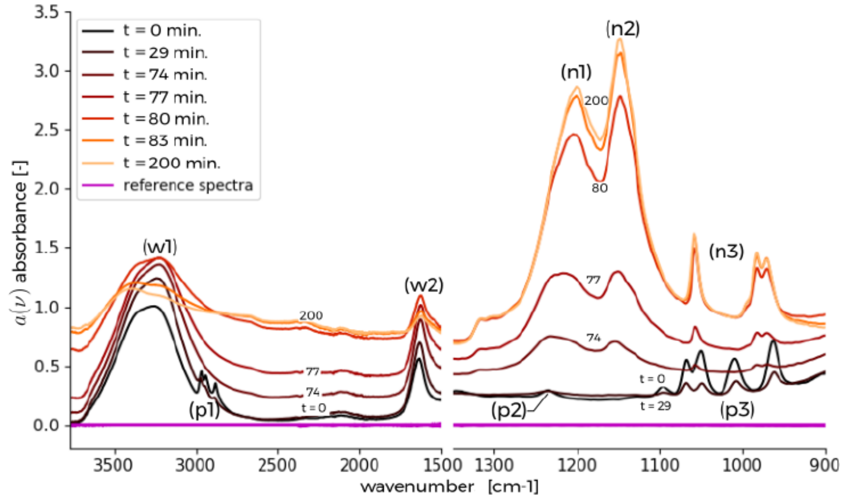


Ink property and rheology characterization

Scalable electrode processing

Unique spatial tools to study defects

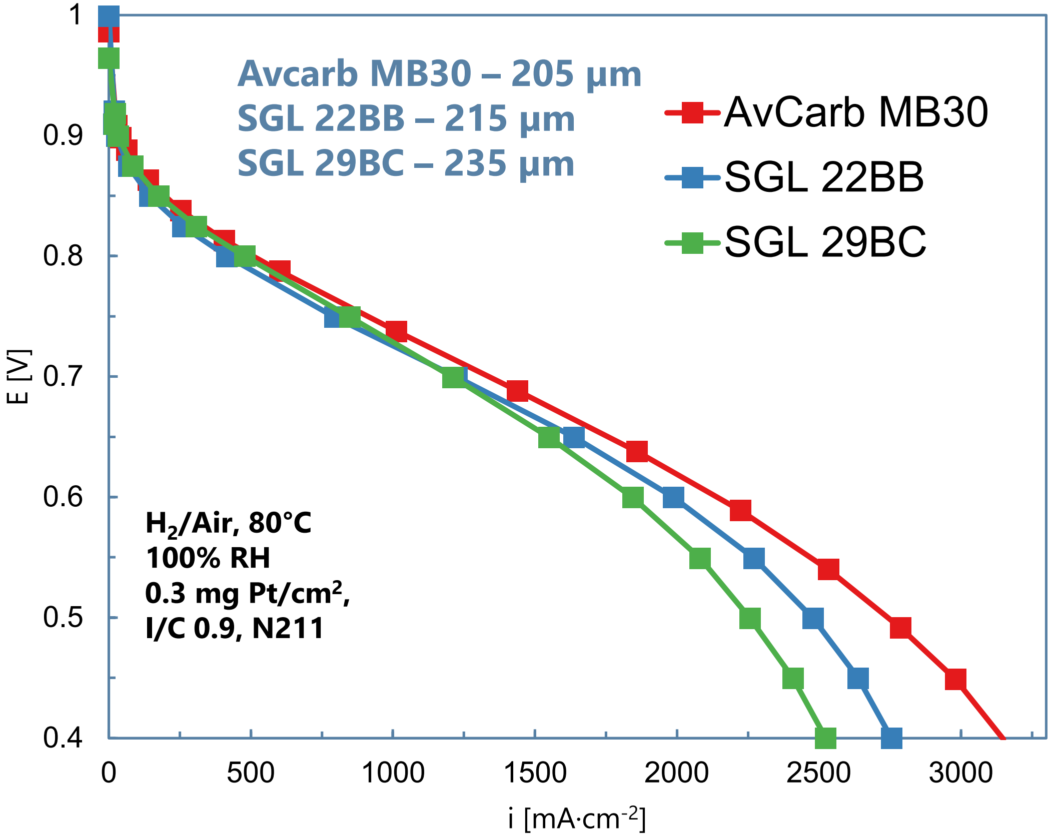




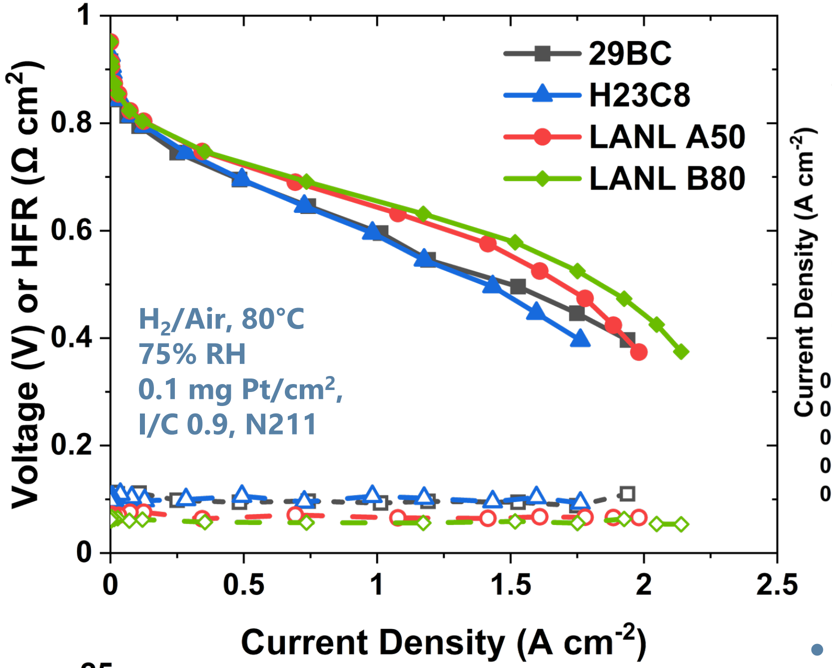
Spectroscopic metrology tools for inks and electrodes

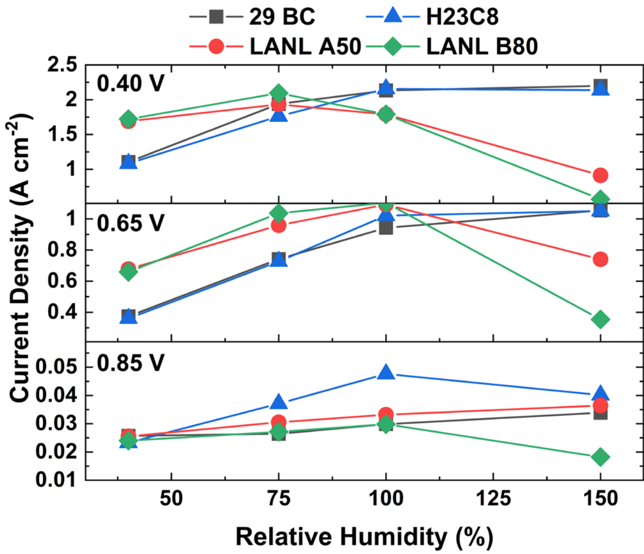
Appl. Mater. Interfaces. 10 (2018) 43610–43622

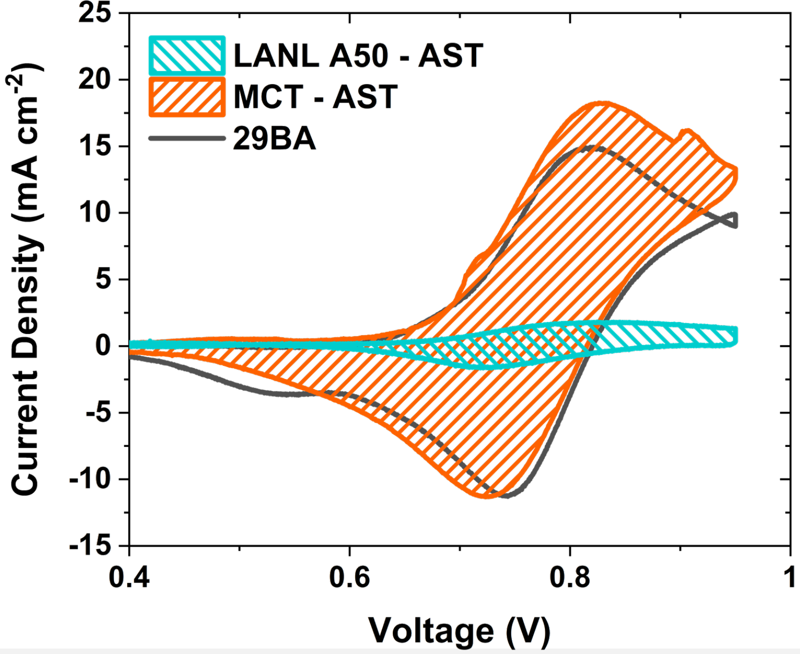
GDL Integration



- AvCarb MB30 GDL showed the best performance of the three GDLs compared here.
- Current focus is on understanding the GDL/MPL morphology effect on performance



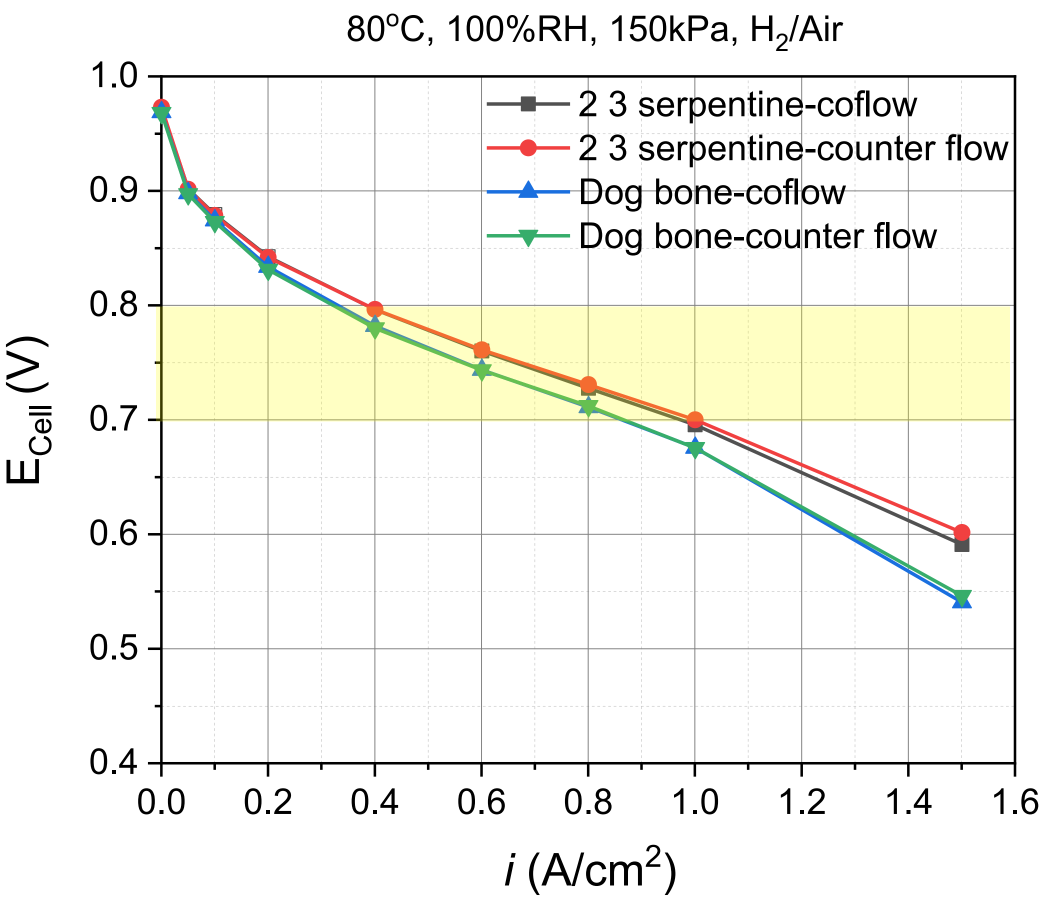




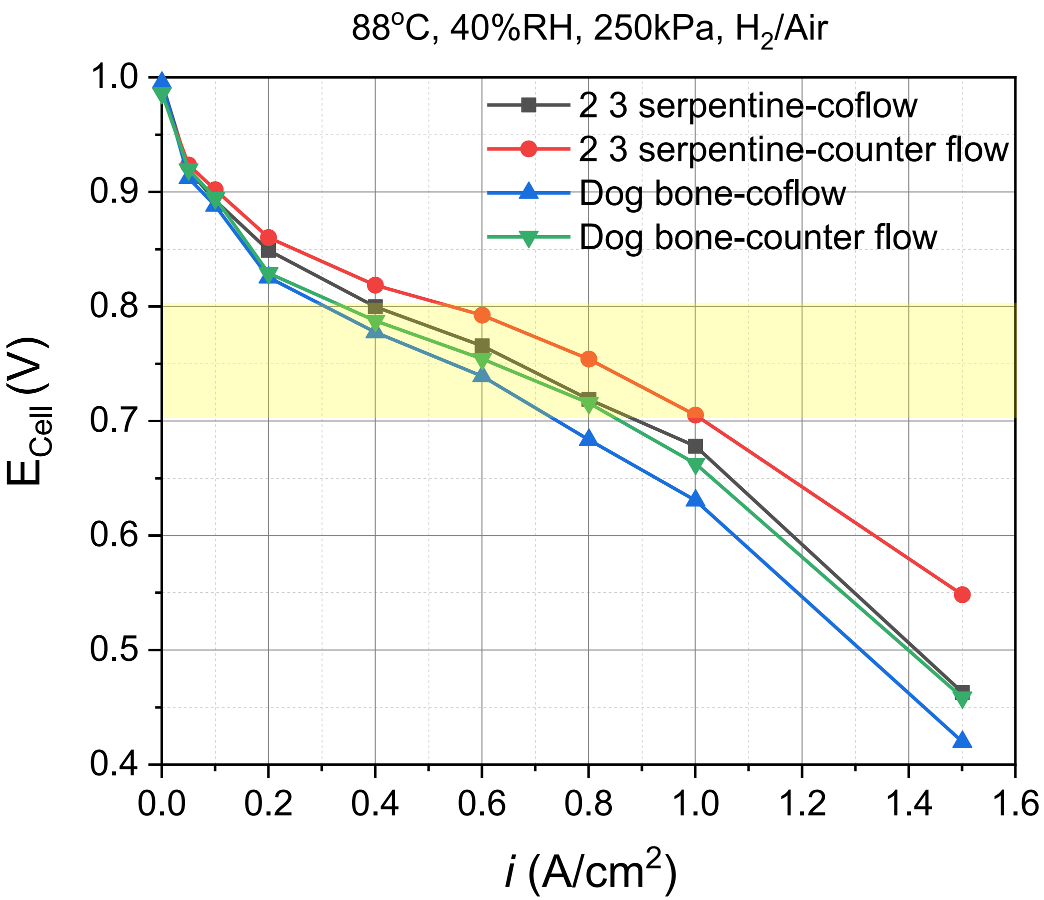
- Novel GDLs suppress cation transport from the flow field – smaller Fe^{2+}/Fe^{3+} redox couple
- Novel GDLs increase performance at low RH in differential cell testing

Journal of the Electrochemical Society 168.2 (2021) 024501.

Flow Fields & Orientation



- flow orientation didn't affect the performance
- minimal impact of flow field



- 20-40 mV improvement from flow orientation
- ~20-40 mV from flow field

- Need to establish common platform for materials down selection assessment