

Fuel Cell System Modeling and Analysis

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Project ID: FC017

This presentation does not contain any proprietary, confidential, or otherwise restricted information.

Timeline

- Start date: Oct 2003
- End date: Open
- Percent complete: NA

Barriers

- B. Cost
- C. Performance
- E. System Thermal and Water Management
- F. Air Management
- J. Startup and Shut-down Time, Energy/Transient Operation
- This project addresses system, stack and air management targets for efficiency, power density, specific power, transient response time, cold start-up time, start up and shut down energy

Budget

- FY17 DOE Funding: \$500 K
- Planned DOE FY18 Funding: \$250 K
- Total DOE Project Value: \$250 K

Partners/Interactions

- Eaton, Ford, Honeywell, UDEL/Sonijector
- SA, Aalto University (Finland)
- 3M, Ballard, Johnson-Matthey Fuel Cells (JMFC), UTRC, FC-PAD, GM
- IEA Annex 34
- Transport Modeling Working Group
- Durability Working Group
- U.S. DRIVE fuel cell tech team

Objectives and Relevance

Develop a validated system model and use it to assess design-point, part-load and dynamic performance of automotive (primary objective) and stationary (secondary objective) fuel cell systems (FCS)

- Support DOE in setting technical targets and directing component development
- Establish metrics for gauging progress of R&D projects
- Provide data and specifications to DOE projects on high-volume manufacturing cost estimation

Impact of FY2018 work

- Projected $46.0^* \pm 0.7$ $\$/kW_e$ FCS cost at 500,000 units/year and 8.5 ± 0.4 kW_e/g_{Pt} FCS Pt utilization with SOA d-PtCo/C cathode catalyst, reinforced 14- μm 850 EW membrane, and $Q/\Delta T = 1.45$ $kW/^\circ C$ constraint
- Verified that the SOA catalyst system can achieve 1180 ± 55 mW/cm^2 stack power density exceeding the target at low Pt loading (0.125 $mg-Pt/cm^2$ total)
- Projected <5% penalty in power density if the cathode humidifier is removed, and ~15% penalty if stack inlet pressure is lowered to 2 atm from 2.5 atm
- Showed that parasitic power approaches 25 kW_e if the compressor discharge pressure is raised to 4 atm
- Modified the reference system configuration to include valves and controls for protected shutdown, safe startup from sub-freezing temperatures, and limiting cell voltage to 0.85-0.875 V during idle

*51 $\$/kW_e$ at 100,000 units/year; Q: Stack heat load; ΔT : Stack coolant exit T – Ambient T

Approach

Develop, document & make available versatile system design and analysis tools

- GCtool: Stand-alone code on PC platform
- GCtool-Autonomie: Drive-cycle analysis of hybrid fuel cell systems

Validate the models against data obtained in laboratories and test facilities inside and outside Argonne

- Collaborate with external organizations

Apply models to issues of current interest

- Work with U.S. DRIVE Technical Teams
- Work with DOE contractors as requested by DOE

1	Evaluate the advantages of operating PEMFC stack at elevated pressures up to 4 atm using 2-stage centrifugal compressors.	12/17
2	Determine the comparative performance of PEMFC stacks with and without cathode humidifier.	03/18
3	Evaluate the performance and durability of MEAs with de-alloyed PtCo cathode catalyst on high surface area carbon with tailored pore size distribution relative to the targets of 0.44 A/mg-PGM mass activity, 1000 mW/cm ² at rated power, 300 mA/cm ² at 800 mV, and 5000 h lifetime.	06/18
4	Update the performance and cost of an automotive fuel cell system with an advanced low-PGM catalyst relative to 2020 targets of 65% peak efficiency, Q/ΔT of 1.45 kW/K, and \$40/kW cost.	09/18

Technical Accomplishments: Summary

Stack: Collaboration with FC-PAD and GM in obtaining data to develop and validate model for pressures up to 3 atm

- State-of-the-art (SOA) dispersed de-alloyed PtCo/C catalyst systems
- De-alloyed PtCo/C catalyst system: durability on drive cycles

Air Management: Investigating integrated air management system with high speed centrifugal compressors and expanders including cooling requirements of motor and airfoil bearings (AFB) and two-stage compressors (Honeywell patent)

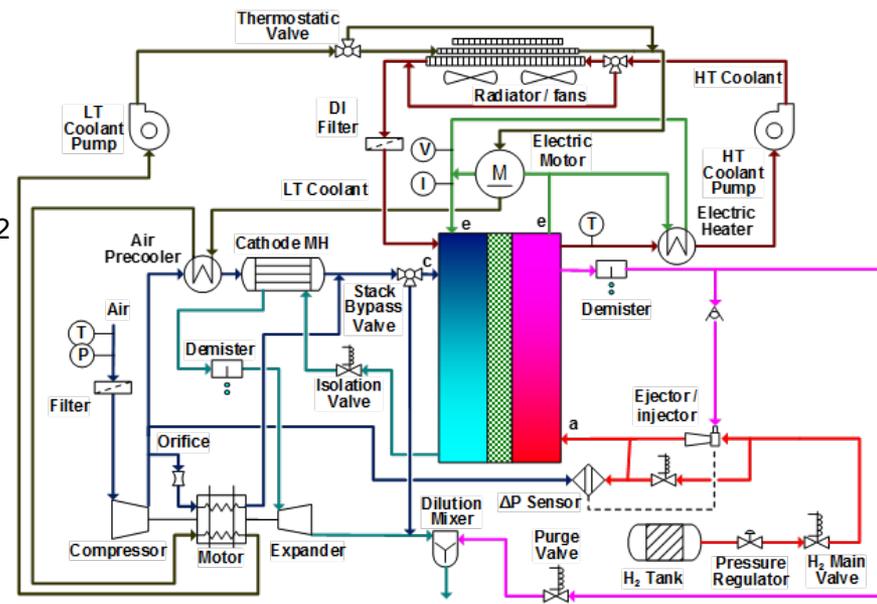
Water Management: Optimizing cost of integrated PEFC stack and cross-flow humidifier

- Investigating FCS performance without cathode humidifier (dispersed catalyst electrodes)

Fuel Management: Evaluating the performance of anode system with a pulse injector in lieu of H₂ recirculation blower (collaboration with Ford & UDEL)

Thermal Management: Optimizing system performance and cost subject to $Q/\Delta T$ constraint

System startup and shutdown: Modified reference system to incorporate controls for protected shutdown, safe startup from sub-freeze temperatures, and limiting cell voltage at idle

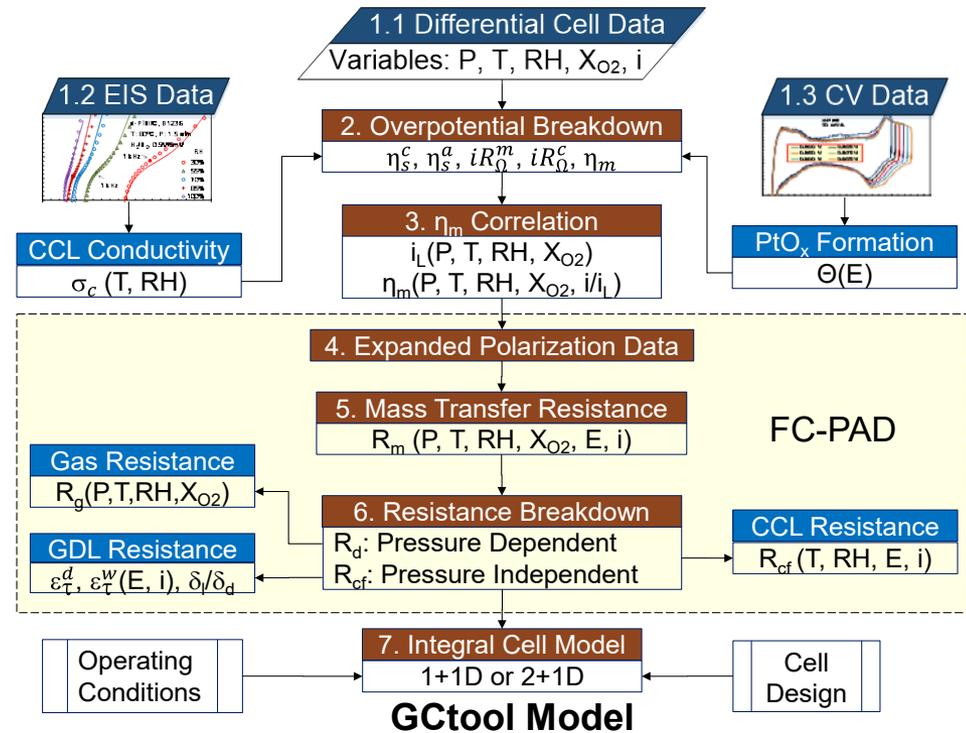
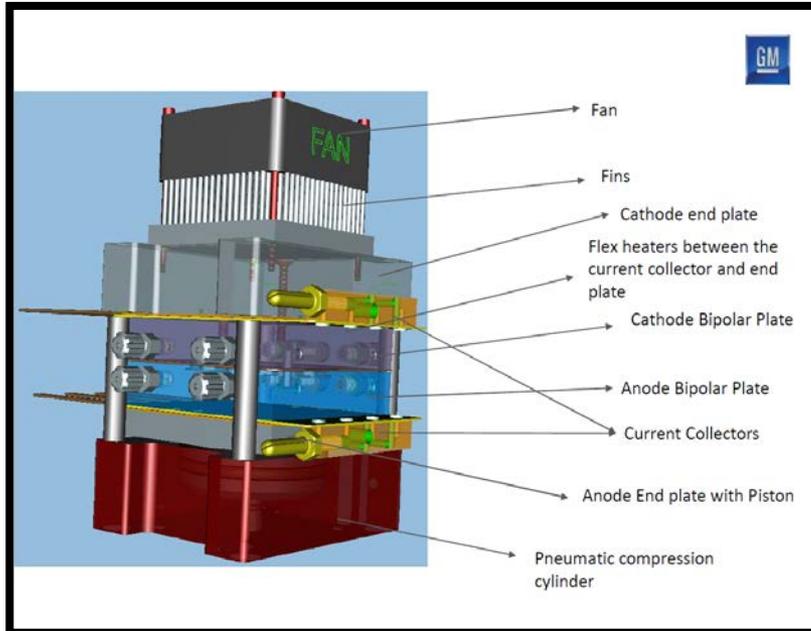


Argonne 2018 FCS Configuration

Model Framework and Single Cell Hardware

US-EU Differential Cell Hardware*

- P: 1-3 atm; T: 45-95°C,
- RH: 30-150%; X(O₂): 1-21%
- **Random Tests:** Semi-statistical randomized with multiple variables, forward scans, 3-min hold at each cell voltage
- **Controlled Tests:** Model guided single-variable with some two-variable tests, forward scans, 3-min hold at each cell voltage

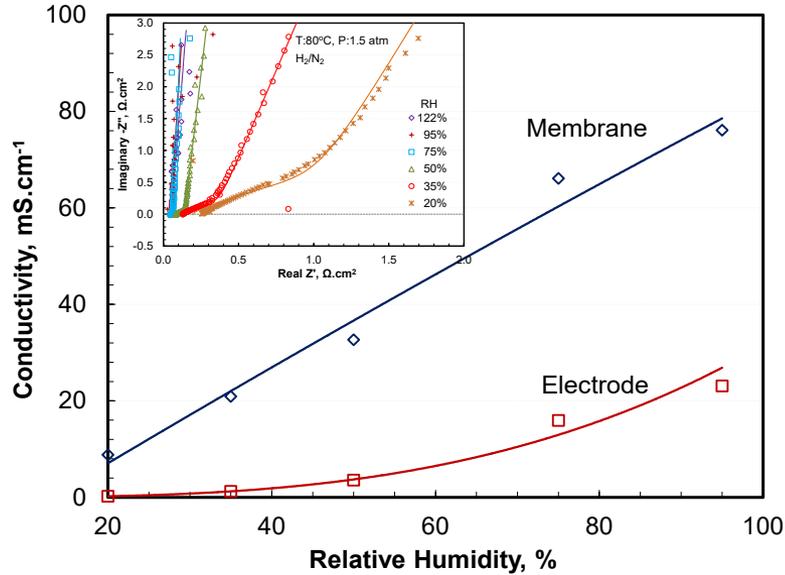


Representative State-of-the-Art Low-PGM MEA		
	Cathode	Anode
Catalyst	d-Pt ₃ Co/C	Pt/C
Catalyst Support	HSAC	Vulcan
Ionomer Equivalent Weight	950	950
Pt Loading	0.1 mg/cm ²	0.025 mg/cm ²
ECSA	45 m ² /g	60 m ² /g
Electrode Thickness	7 μm	5 μm
Diffusion Medium Thickness	200 μm	200 μm
Membrane	18 μm Reinforced	

Kinetics of ORR on d-PtCo/C Catalyst

Electrode Resistance

- Electrode (σ_c) / membrane conductivities (σ_m) from Galvanostatic impedance data in H_2/N_2



Distributed ORR Kinetic Model

- For Tafel kinetics, the ORR and CCL Ohmic overpotentials are separable

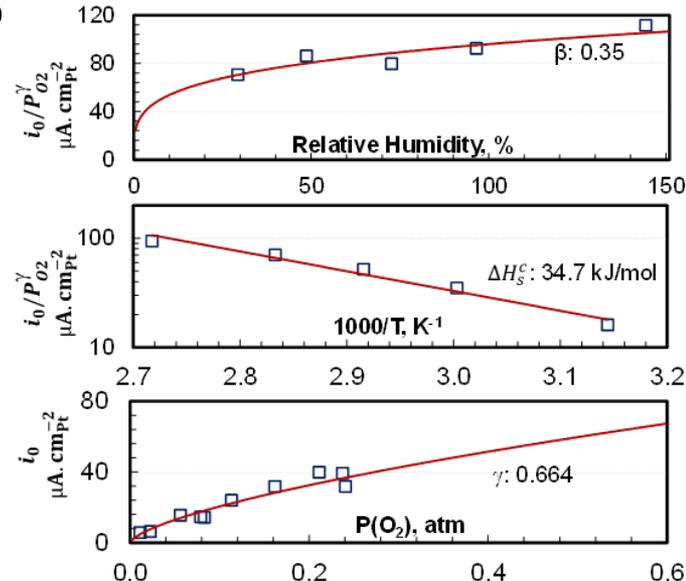
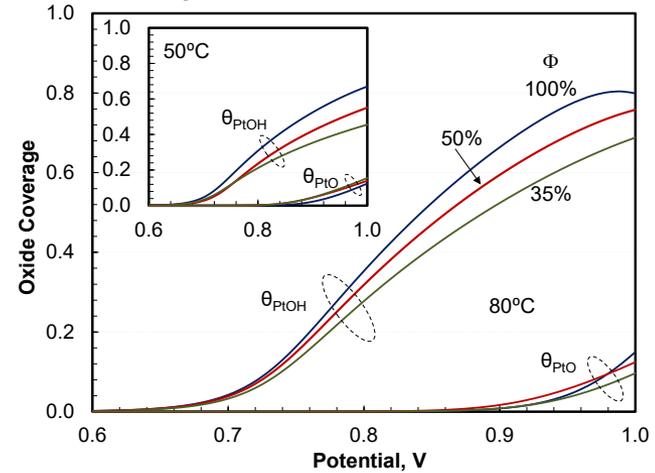
$$\eta_c = \eta_s^c + iR_\Omega^c \left(\frac{i\delta_c}{b\sigma_c} \right)$$

$$i + i_x = i_0 S_{Pt} (1 - \theta) e^{-\frac{\omega\theta}{RT}} e^{\frac{\alpha n F}{RT} \eta_s^c}$$

$$i_0 = i_{0r} e^{-\frac{\Delta H_S^c}{R} \left(\frac{1}{T} - \frac{1}{T_r} \right)} P_{O_2}^\gamma \Phi^\beta$$

PtO_x Formation on d-PtCo/C Catalyst

- Transient solid solution model from measured CV reduction charge after hold for 12 s to 2 h at constant potential¹



Kinetic data from controlled tests



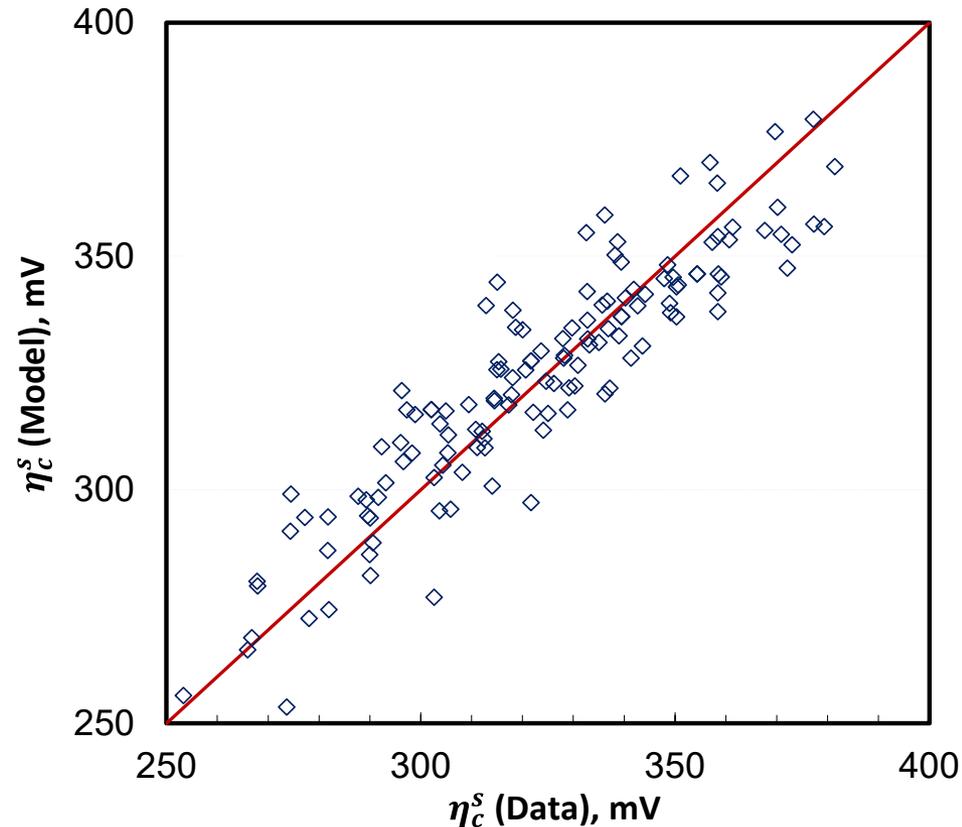
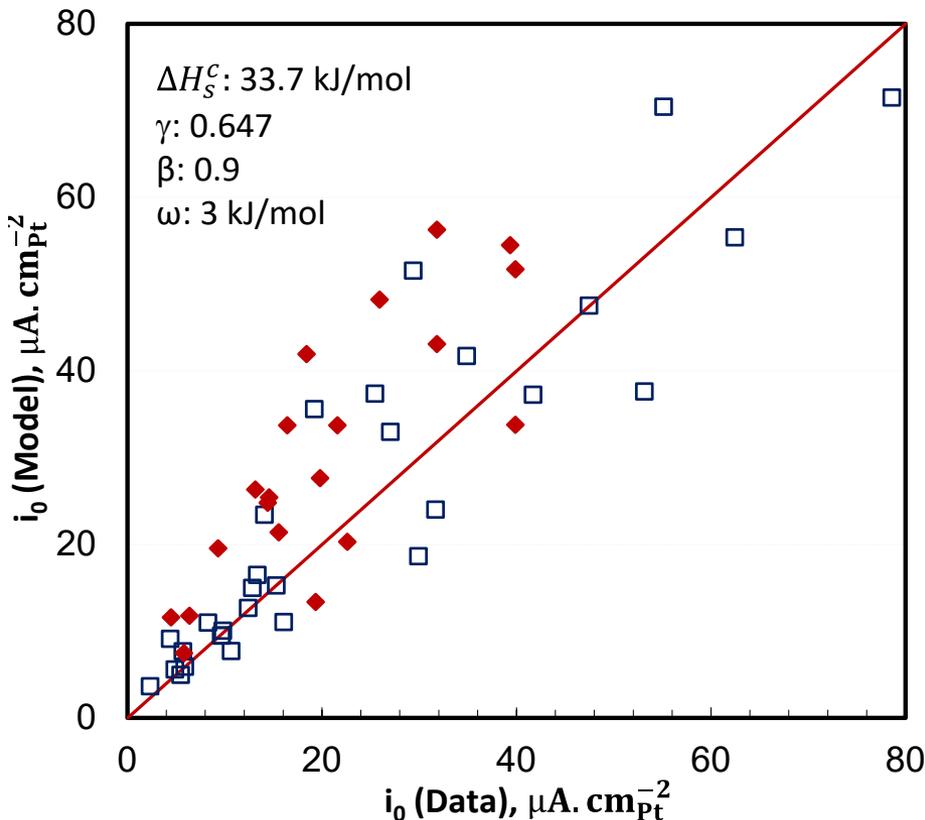
Calibration of ORR Kinetic Model*

i_0 calibration: 1 data point per pol curve

- Data: $i + i_x = i_0 S_{Pt} (1 - \theta) e^{-\frac{\omega\theta}{RT}} e^{\frac{\alpha n F}{RT} \eta_s^c}$
- Model: $i_0 = i_{0r} e^{-\frac{\Delta H_s^c}{R} \left(\frac{1}{T} - \frac{1}{T_r}\right)} P_{O_2}^\gamma \Phi^\beta$

η_s^c calibration: all data in kinetic region

- Data: $\eta_s^c = E_N - E - iR_\Omega^m - iR_\Omega^c$
- Model: $\eta_s^c = b \left[\ln \left(\frac{i + i_x}{i_0 S_{Pt}} \right) - \ln(1 - \theta) + \frac{\omega\theta}{RT} \right]$

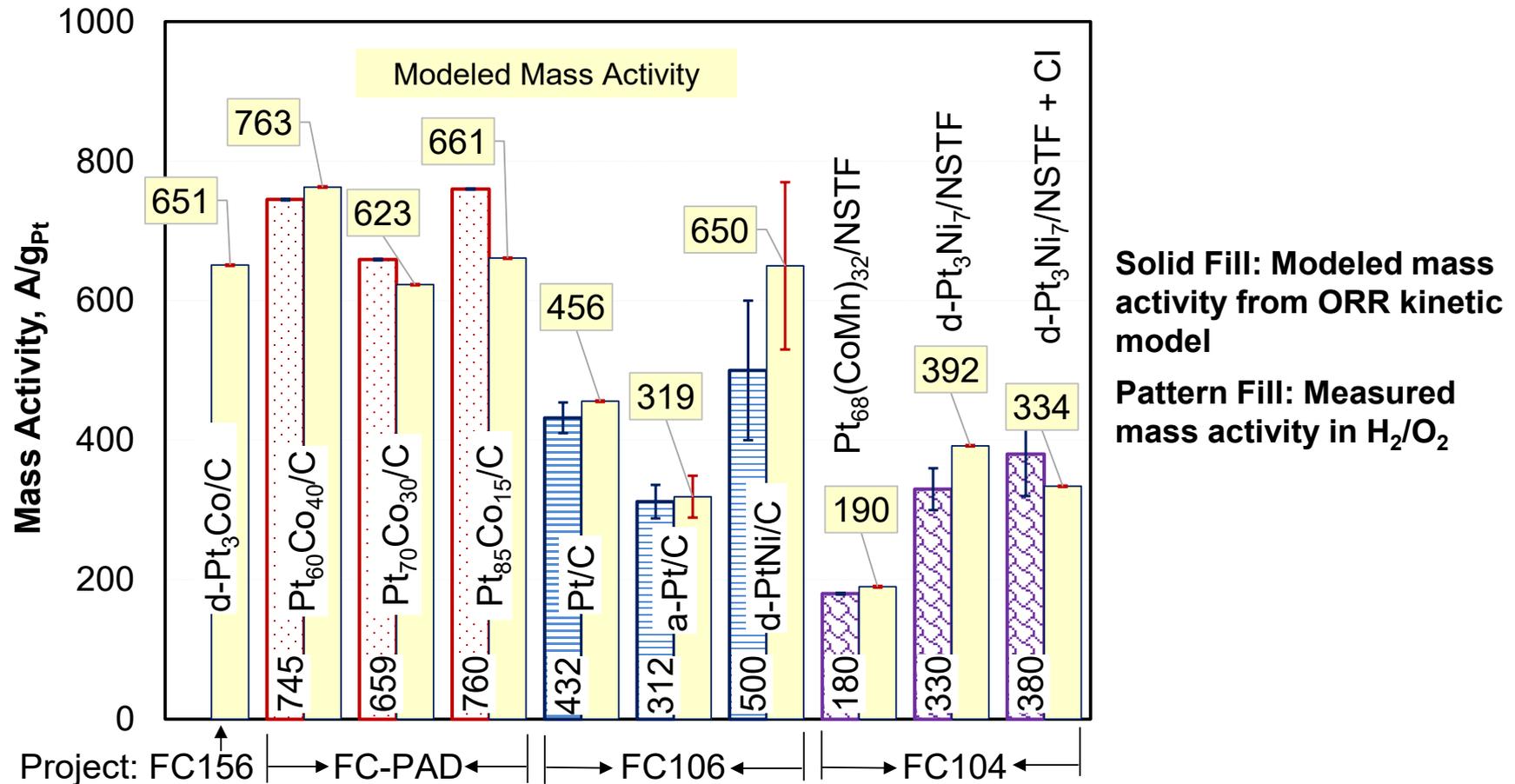


Open symbols: Random tests (BOT); Closed symbols: Controlled tests (EOT: end of test)

*Kinetic model based on random test data; BOT: Beginning of tests; EOT: End of tests

Mass Activity for ORR on d-PtCo/C Catalysts

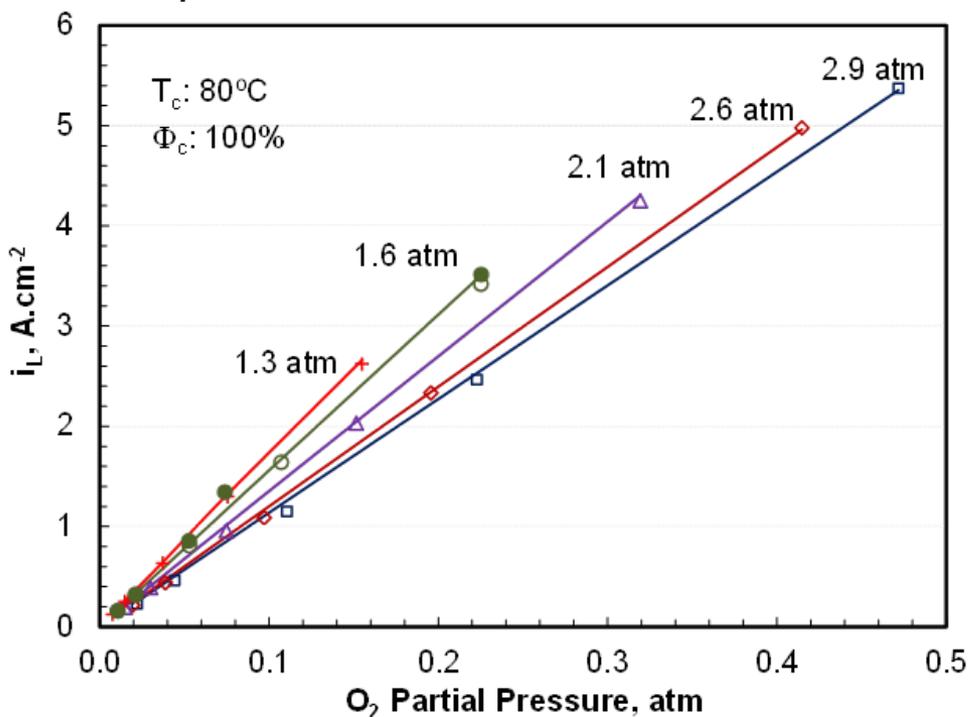
- d-PtCo/C has 2X modeled mass activity of a-Pt/C that has nearly the same particle size
- d-PtCo/C and d-PtNi/C alloy have comparable mass activities
- Both low-PGM alloy catalysts (d-PtNi/C and d-PtCo/C) meet the mass activity targets of 440 A/g_{Pt}



Oxygen Mass Transfer: Limiting Current Density

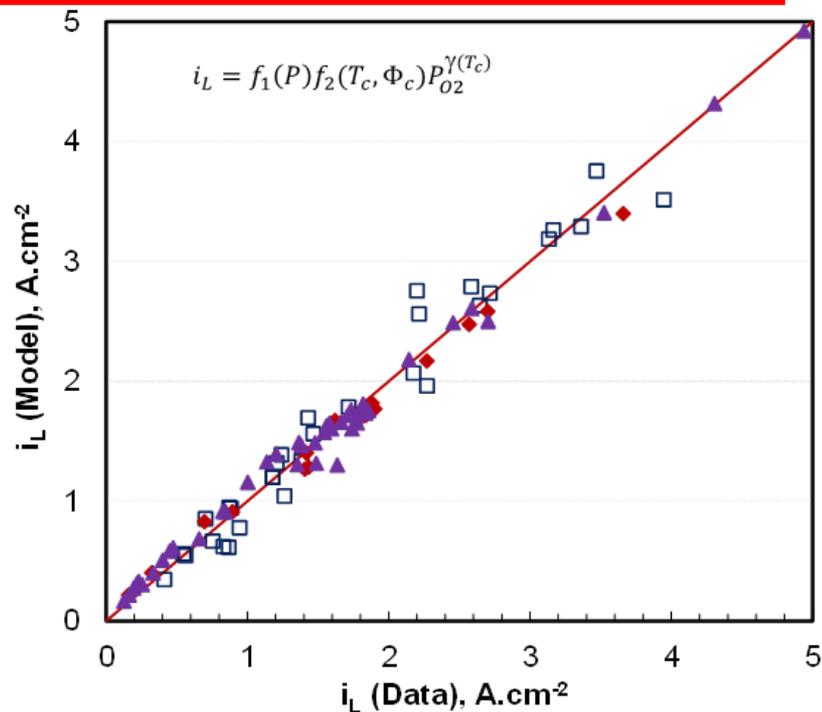
Determined limiting current density (i_L) and correlated mass transfer overpotential (η_m) with reduced current density (i/i_L)

- $\eta_m = E_N - E - iR_{\Omega}^m - \eta_c - \eta_a$
- i_L defined as current density at which η_m equals 400 mV



Symbols

T: Bipolar plate temperature; T_c : CCL temperature
 Φ : Gas channel RH; Φ_c : CCL RH



Model Variables

- P : Pressure
- P_{O_2} : O_2 partial pressure in gas channel
- T_c : CCL temperature, function of T , E_N , E , and i
- Φ_c : CCL RH, function of Φ , i , and water transport across membrane

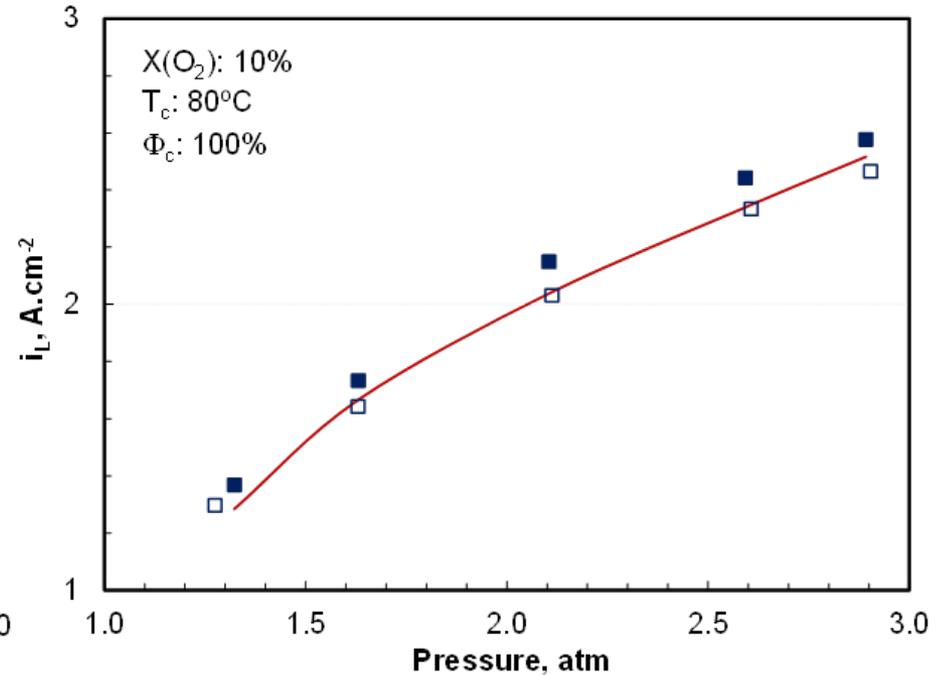
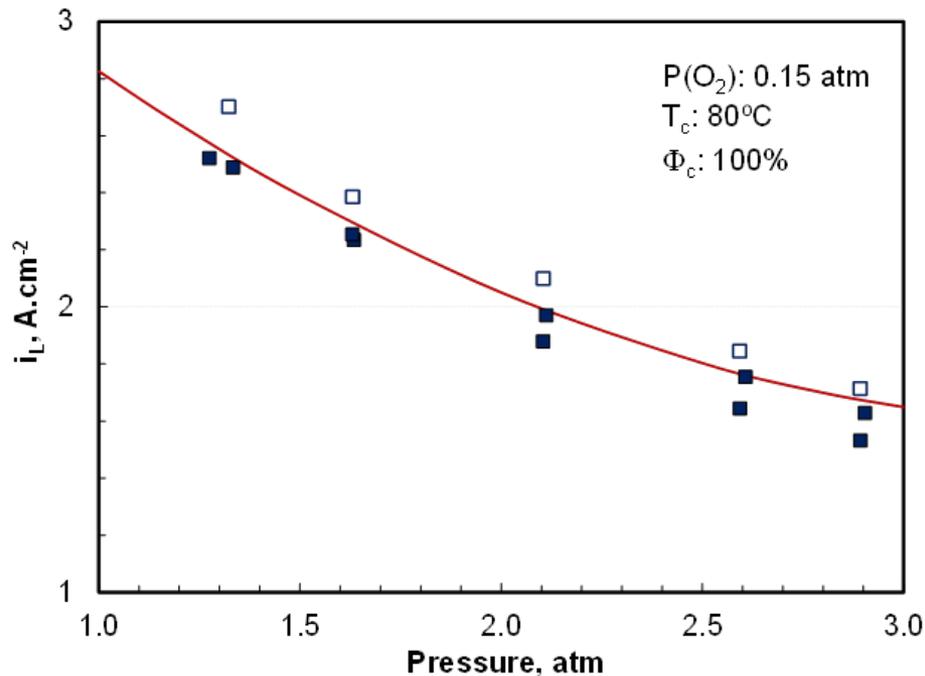
Limiting Current Density: Effect of Pressure

At constant $P(O_2)$, i_L decreases with increase in P because of the inverse dependence of O_2 gas phase diffusivity on P .

- The decrease in i_L is less than proportional to $1/P$, implying that non-Fickian diffusion controls mass transport resistance

At constant $X(O_2)$, i_L increases with increase in P because of higher $P(O_2)$.

- The increase in i_L is somewhat less than proportional to $P(O_2)$, implying that mass transport resistance also increases with $P(O_2)$.



The plots include the experimental data for i_L at different P , $X(O_2)$, $T=80^\circ\text{C}$, and $\Phi=100\%$. For comparison, the model was used to rescale the i_L data to $P(O_2)=0.15$ atm or $X(O_2)=10\%$, $T_c=80^\circ\text{C}$, and $\phi_c=100\%$.

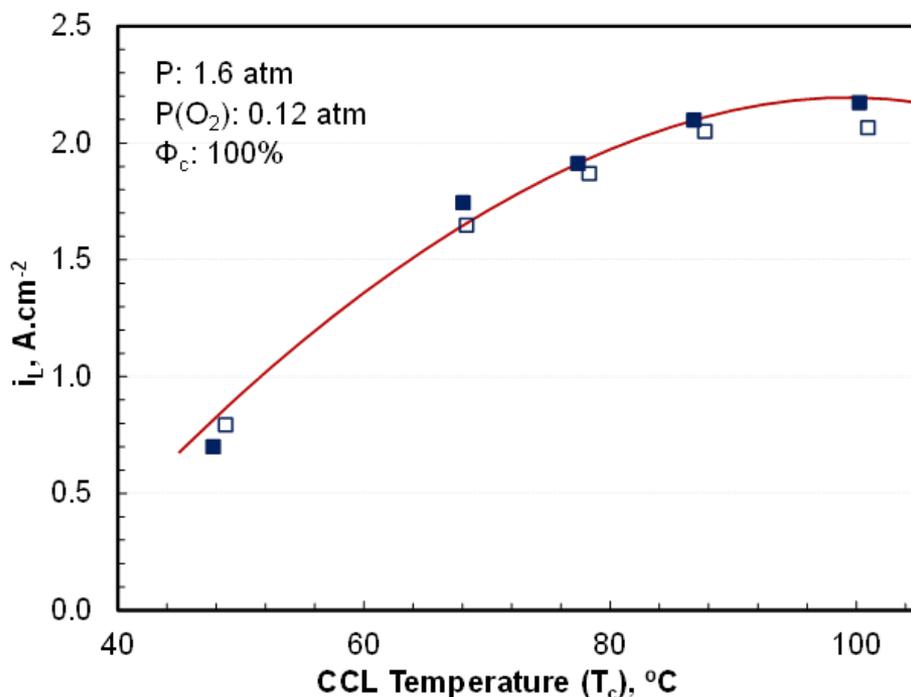
Limiting Current Density: Effect of Temperature and RH

Dependence of i_L on $T_c \gg T^{3/2}$, confirming that processes other than Fickian diffusion are rate controlling.

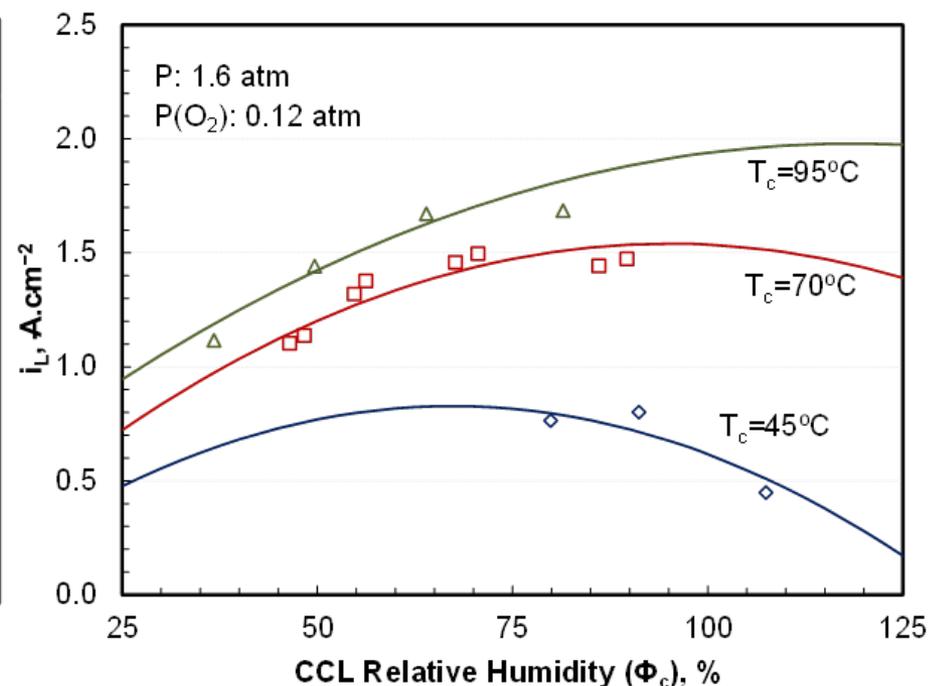
- O_2 permeability through the ionomer film on the catalyst particles

For given T_c , i_L is highest at an intermediate RH in CCL (ϕ_c^*).

- CCL flooding for $\phi_c > \phi_c^*$
- ϕ_c^* Increases at higher T_c



The plot includes all the experimental data for i_L at different T , $P=1.6$ atm, $X(O_2)=10\%$, and $\Phi=100\%$. For comparison, the model was used to rescale the i_L data to $P(O_2)=0.12$ atm, and $\phi_c=100\%$.



The plot includes all the experimental data for i_L at different Φ , $T=45, 70$ and 95°C , $P=1.6$ atm and $X(O_2)=10\%$. For comparison, the model was used to rescale the i_L data to $P(O_2)=0.12$ atm and different $T_c=45, 70$ and 95°C .

Performance of Automotive FCS with SOA d-PtCo/C Cathode Catalyst

Modeled optimal beginning of life (BOL) performance of automotive FCS subject to $Q/\Delta T=1.45$ kW/°C constraint: 0.125 mg/cm² total Pt loading; 850 EW, 14- μ m chemically-stabilized, reinforced membrane

FY2018 model based on differential cell data supports FY2017 landmark result

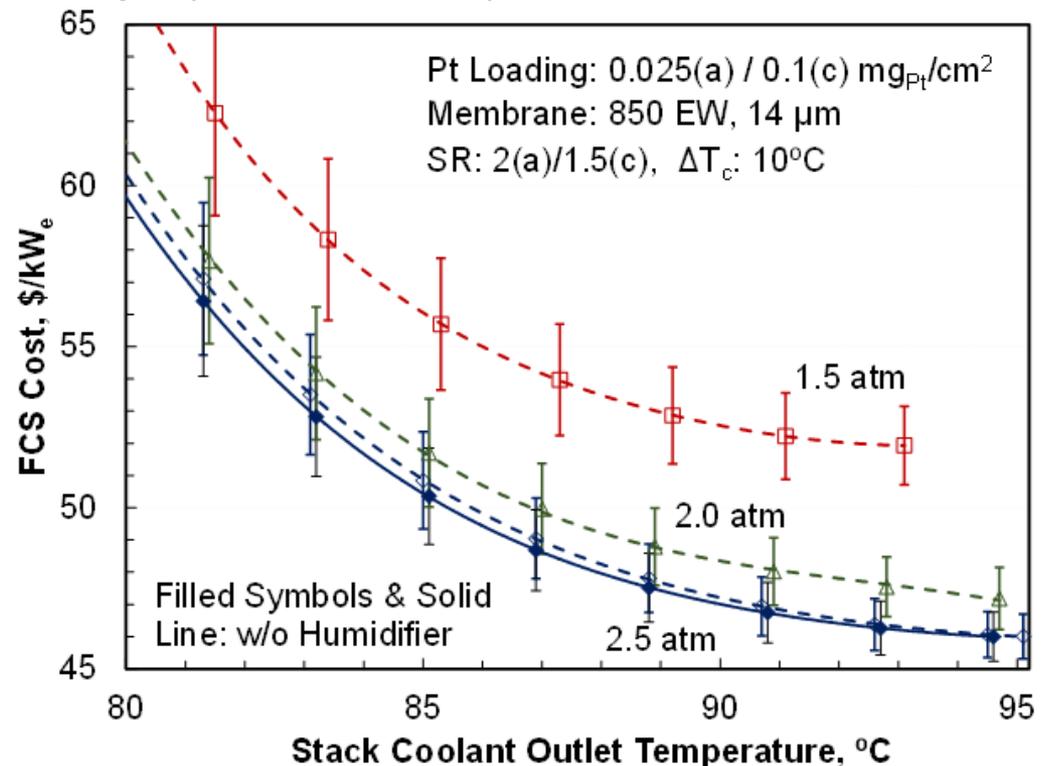
- 46.0 ± 0.7 \$/kW_e projected cost* at 2.5 atm stack inlet pressure and 95°C stack coolant outlet temperature for high volume manufacturing
- Removing membrane humidifier slightly lowers the system cost at 2.5 atm stack inlet pressure

Results are preliminary pending model validation using data from 50-cm² integral cell

- Error bars reflect variance of kinetic data in random and controlled tests and include degradation between the two series of tests

Effect of manufacturing volume on projected cost (SA)

- 500,000 units/year: 46 \$/kW_e
- 100,000 units/year: 51 \$/kW_e
- 10,000 units/year: 88 \$/kW_e



*Using 2018 cost correlations from Strategic Analysis (SA), 500,000 units/year, no H₂ blower. Includes \$2.01 cost increase in 2018 for manufacturing bipolar plates and MEAs, added controls, and CEMM price inflation.

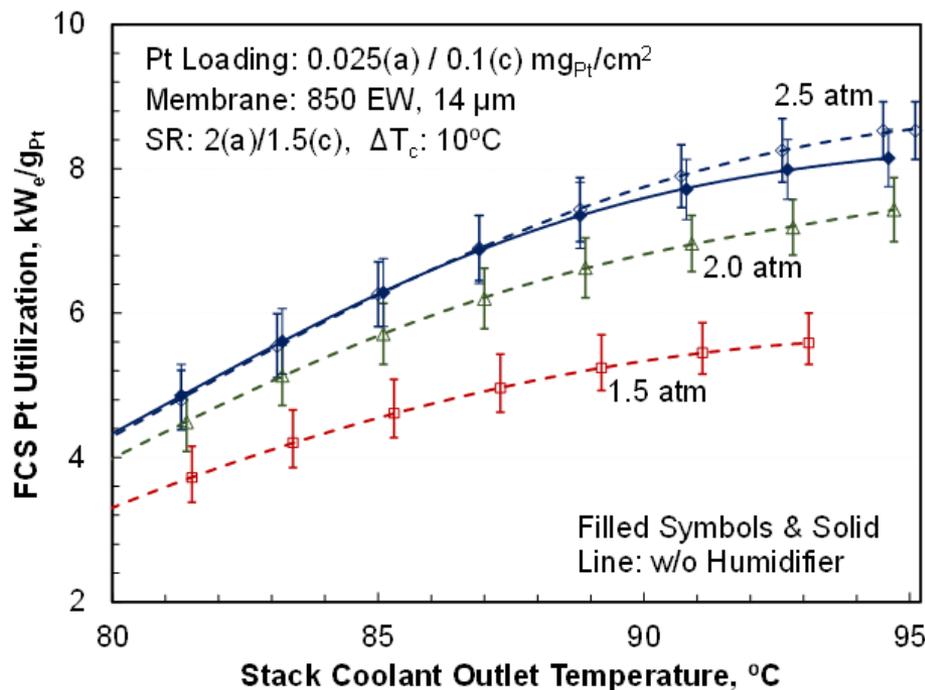
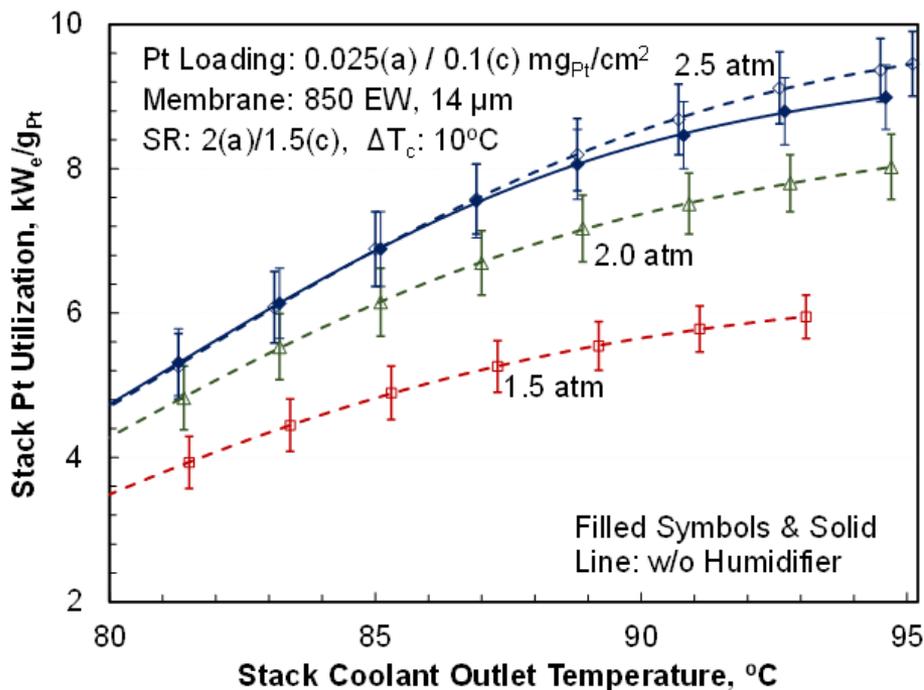


FCS with SOA d-PtCo/C Cathode Catalyst: Pt Utilization

Modeled optimal beginning of life (BOL) performance of automotive FCS subject to $Q/\Delta T = 1.45 \text{ kW}/^\circ\text{C}$ constraint: $0.125 \text{ mg}/\text{cm}^2$ total Pt loading; 850 EW, 14- μm chemically-stabilized, reinforced membrane

FY2018 model based on differential cell data supports FY2017 landmark result

- Modeled stack Pt utilization ($9.5 \pm 0.5 \text{ kW}_e/\text{g}_{\text{Pt}}$) exceeds the target ($8.0 \text{ kW}_e/\text{g}_{\text{Pt}}$)
- Modeled FCS Pt utilization ($8.5 \pm 0.4 \text{ kW}_e/\text{g}_{\text{Pt}}$) also exceeds the target
- Stack inlet pressure $\geq 2.0 \text{ atm}$ needed to meet the Pt utilization target

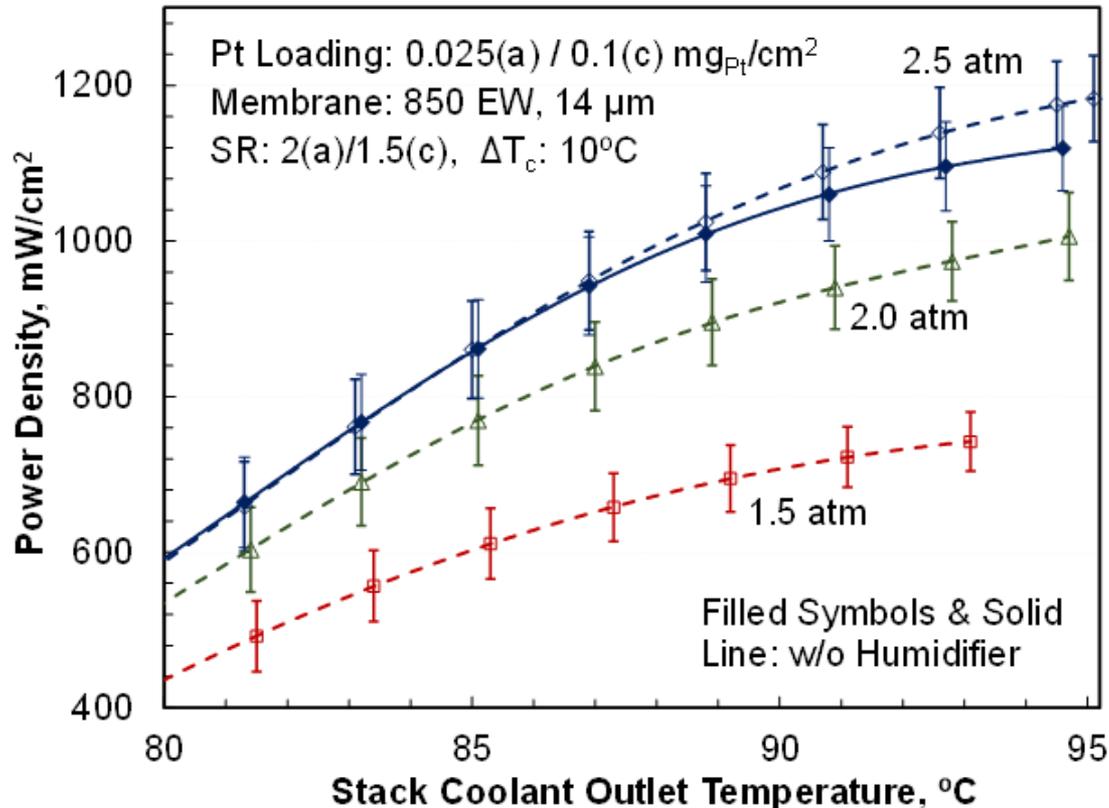


FCS with SOA d-PtCo/C Cathode Catalyst: Power Density

Modeled optimal beginning of life (BOL) performance of automotive FCS subject to $Q/\Delta T=1.45$ kW/°C constraint: 0.125 mg/cm² total Pt loading; 850 EW, 14- μ m chemically-stabilized, reinforced membrane

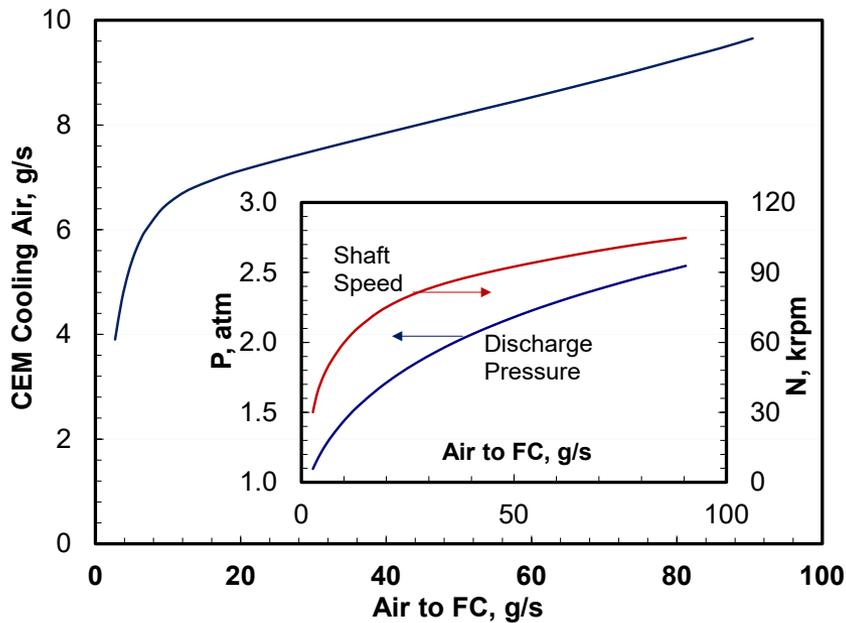
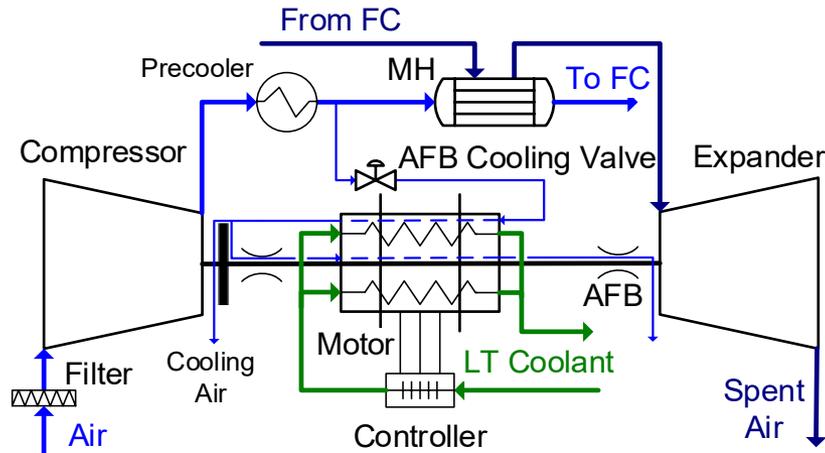
FY2018 model based on differential cell data supports FY2017 landmark result

- Modeled gross stack power density (1180 ± 55 mW/cm²) exceeds the target (1000 mW/cm²) at low Pt loading (0.125 mg-Pt/cm² total)
- Stack inlet pressure ≥ 2.0 atm needed to meet the power density target



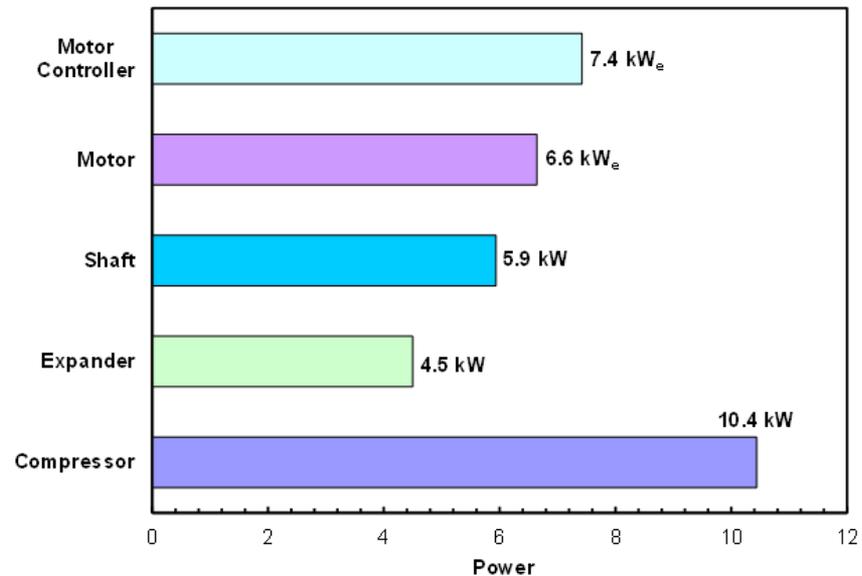
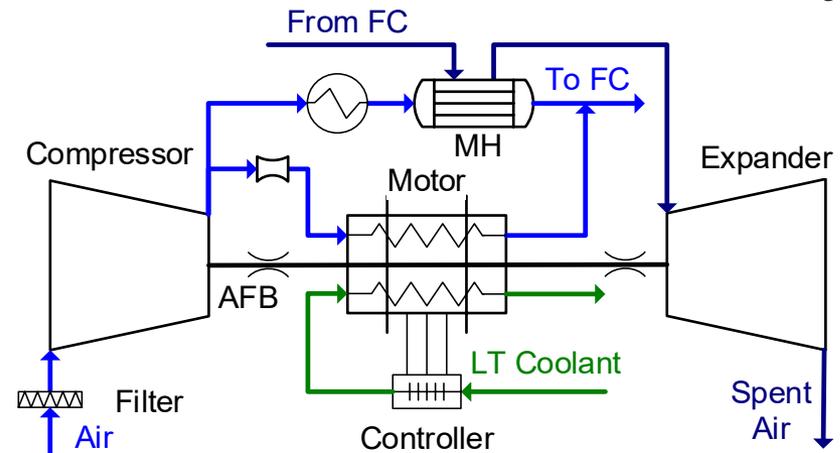
Air Management System

CEMM w/o Bleed Air Recovery



Projected cooling air loss using Honeywell data for CEMM

Modeled CEMM with Bleed Air Recovery



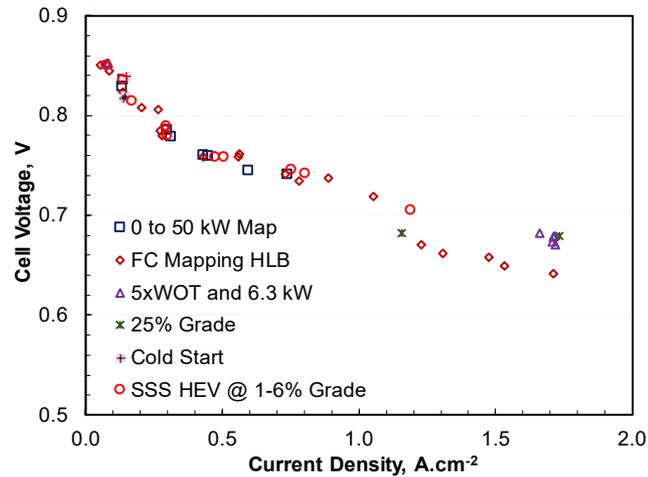
Simulation results: 73 g/s air flow rate, and 2.5 atm compressor discharge P; 2.3 atm expander inlet P, 85°C inlet T, fully saturated; 40°C ambient T



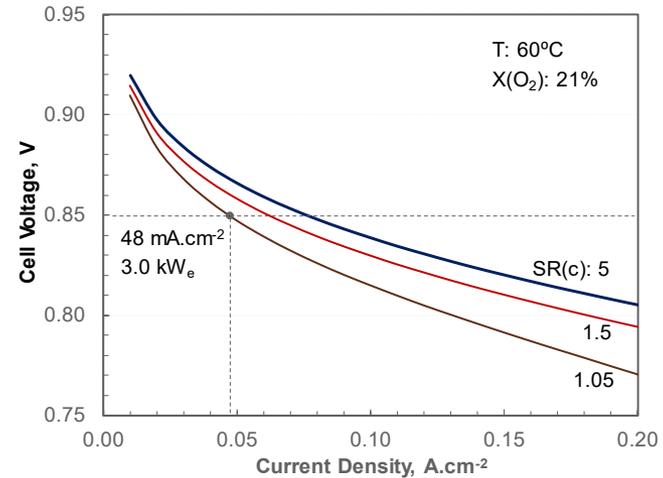
*AFB: Air foil bearing

Mitigation of High Cell Voltages

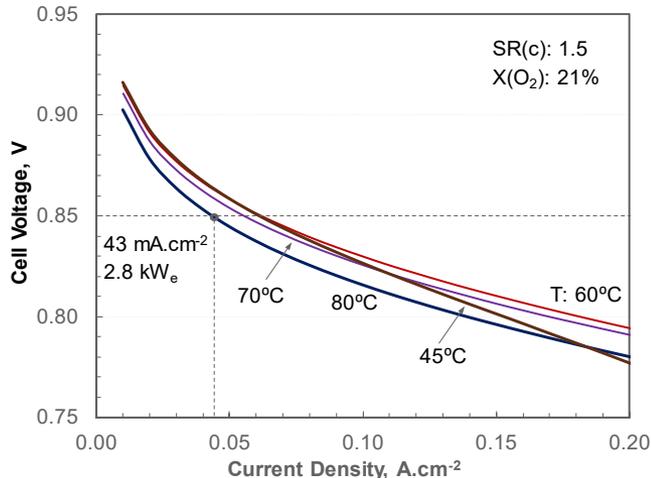
SOA stack limits cell voltage to 0.85 V during idle



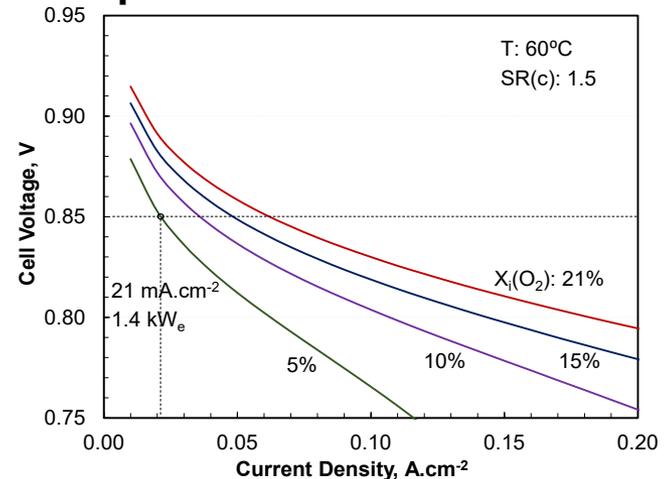
Reducing cathode stoichiometry may not provide adequate control of cell voltage



Lowering stack temperature may also be inadequate and has thermal inertia



Reducing O₂ concentration by recycling spent air provides most control



M. Sato, "Fuel cell System and method of Controlling the Same," US 2017/0047602 A1, Feb. 16, 2017.

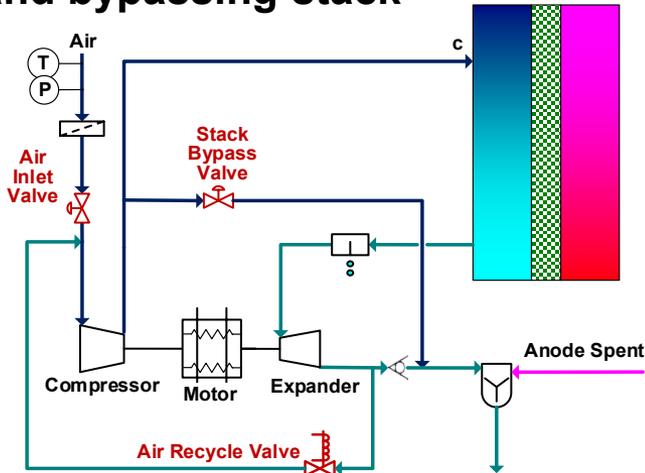
S. Kawahara, K. Suematsu, M. Toida, and R. Akaboshi, "Fuel Cell System and Control Method Thereof,"

US 9,531,022 B2, Dec. 27, 2016.

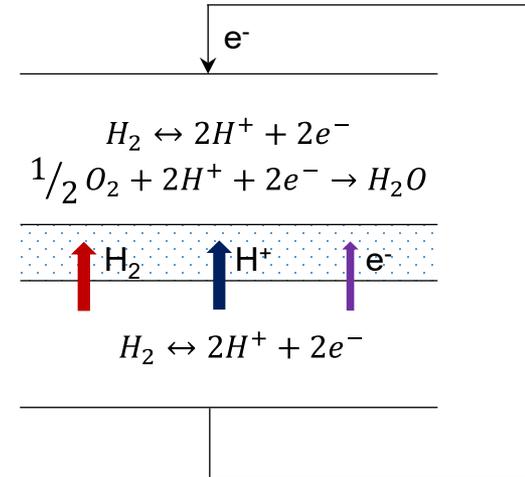
Controlling O₂ Concentration at Stack Inlet

Controlling O₂ concentration at stack inlet by recycling cathode spent air and bypassing stack*

Model for mixed cathode potential at low cathode stoichiometry

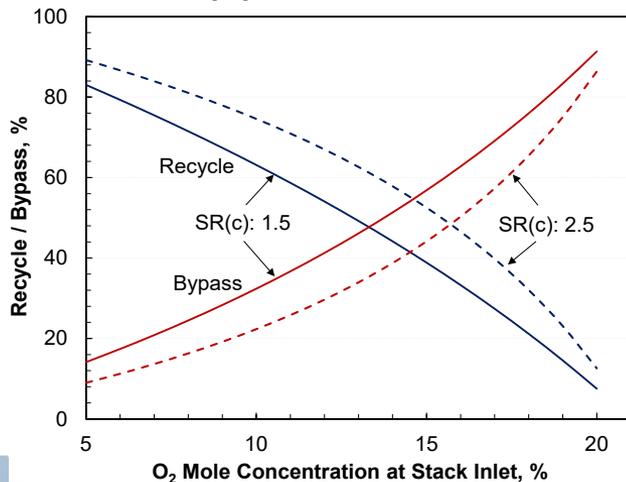


Air inlet and recycle valves not included in Argonne 2018 FCS configuration



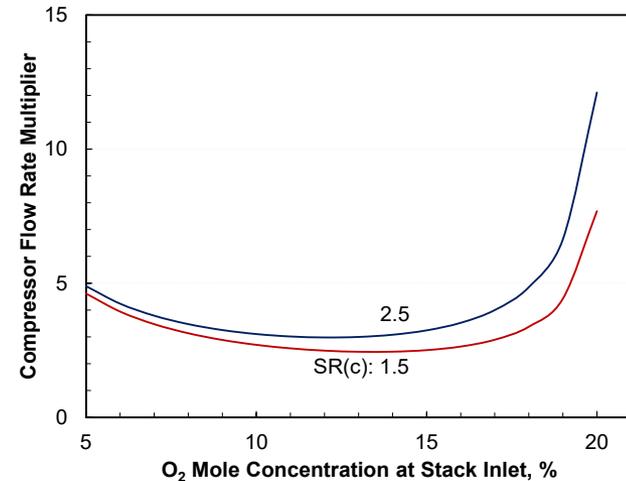
Recycle and bypass depend on desired reduction in O₂ concentration at stack inlet and SR(c)

Increase in compressor flow rate to reduce inlet O₂ concentration below 21%



$$R = \dot{m}_R / \dot{m}_C$$

$$B = \dot{m}_B / \dot{m}_C$$

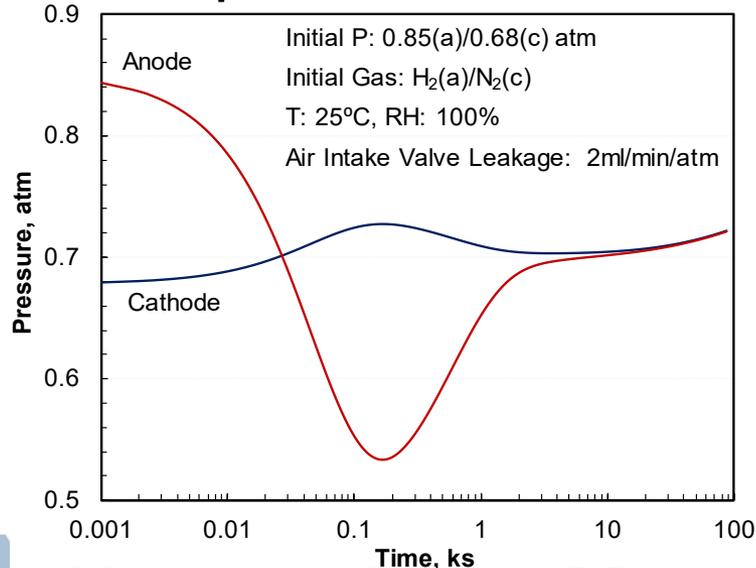


Protected Shutdown

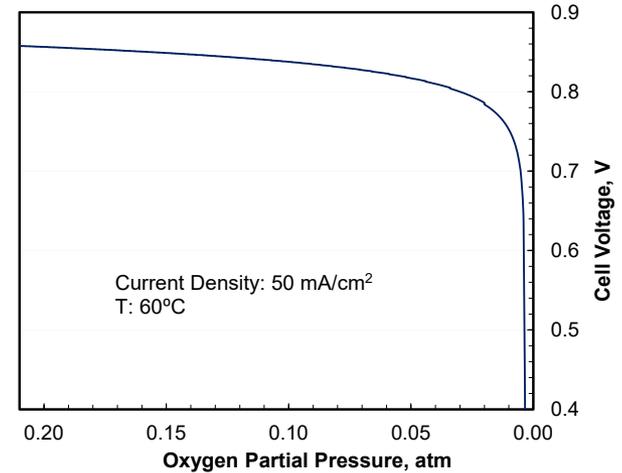
Protected shutdown algorithm

- Step 1: Isolate the cathode circuit by closing the air intake valve and opening the air recycle valve
- Step 2: Deplete oxygen by applying load while supplying hydrogen
- Step 3: Shut the compressor (if on) and isolate H₂ circuit by closing the H₂ main valve
- Step 4: Allow H₂ and N₂ in anode and cathode circuits to equilibrate

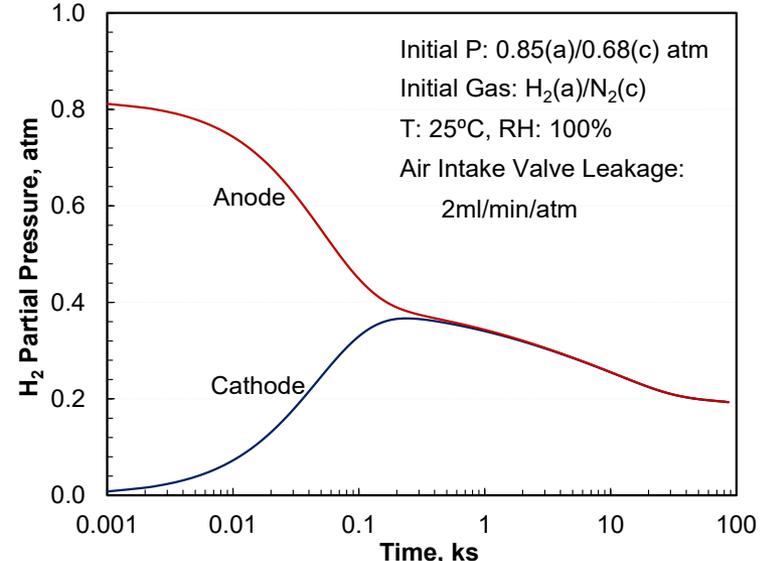
Step 4: Equilibration of anode and cathode pressures



Step 2: Depletion of oxygen with cathode air recycled by compressor



Step 4: Equilibration of H₂ partial pressures in anode and cathode circuits



Collaborations

Air Management	Honeywell: Cost and Performance Enhancements for a PEM Fuel Cell Turbocompressor (FC27)
	Eaton: Roots Air Management System with Integrated Expander (FC103)
Stack	3M: High Performance, Durable, Low Cost Membrane Electrode Assemblies for Transportation (FC104)
	Ballard/Eaton: Roots Air Management System with Integrated Expander (FC103)
	JMFC and UTRC: Rationally Designed Catalyst Layers for PEMFC Performance Optimization (FC106)
	FC-PAD: Fuel Cell Performance and Durability Consortium (FC135, FC136, FC137, FC138, FC139)
	GM: Highly-Accessible Catalysts for Durable High-Power Performance (FC144)
	GM: Durable High-Power Membrane Electrode Assemblies with Low Pt Loadings (FC156)
Water Management	Gore, Ford, dPoint: Materials and Modules for Low-Cost, High-Performance Fuel Cell Humidifiers (FC067)
Thermal Management	3M, Honeywell Thermal Systems
Fuel Management	3M, University of Delaware (Sonijector)
Fuel Economy	ANL-Autonomie (SA044), Aalto University (Fuel Cell Buses)
H ₂ Impurities	3M
System Cost	SA: Manufacturing Cost Analysis of Fuel Cell Systems and Transportation Fuel Cell System Cost Assessment (FC163)
Dissemination	IEA Annex 34, Transport Modeling Working Group, Durability Working Group, Catalysis Working Group

- Argonne develops the fuel cell system configuration, determines performance, identifies and sizes components, and provides this information to SA for high-volume manufacturing cost estimation

Proposed Future Work

1. Continue to support DOE development effort at system, component, and phenomenological levels
2. Continue to support SA in high-volume manufacturing cost projections, collaborate in life-cycle cost studies
 - Optimize system parameters considering costs at low-volume manufacturing
 - Life cycle cost study for medium and heavy duty vehicles (Ballard, Eaton, SA)
3. Alternate MEAs with advanced alloy catalysts
 - State-of-the-art low PGM Pt and Pt alloys (FC-PAD collaboration)
 - Alternate electrode structures (FC-PAD FOA projects collaboration)
 - Durability models (FC-PAD and GM collaboration)
4. System architecture and balance-of-plant components
 - Air management system with centrifugal and Roots compressors and expanders (Honeywell/Eaton collaboration)
 - Fuel and water management systems: anode gas recirculation, internal/external humidification
 - Bipolar plates and flow fields for low pressure drops and uniform air/fuel distribution, cell to stack performance differentials
 - Strategies and controls for stack idling, startup and shutdown, subfreezing temperatures
5. Incorporate durability considerations in system analysis
 - System optimization for cost, performance, and durability on drive cycles (Advanced alloy catalyst systems)

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



Project Summary

Relevance:	Independent analysis to assess design-point, part-load and dynamic performance of automotive and stationary FCS
Approach:	Develop and validate versatile system design and analysis tools Apply models to issues of current interest Collaborate with other organizations to obtain data and apply models
Progress:	Projected 46.0 ± 0.7 $\$/kW_e$ FCS cost at high volume manufacturing and 8.5 ± 0.4 kW_e/g_{Pt} FCS Pt utilization with SOA d-PtCo/C cathode catalyst, reinforced 14- μm 850 EW membrane, and $Q/\Delta T = 1.45$ $kW/^\circ C$ constraint Verified that the stack can achieve 1180 ± 55 mW/cm^2 power density exceeding the target at low Pt loading (0.125 $mg-Pt/cm^2$) Projected <5% penalty in power density if the cathode humidifier is removed, and ~15% penalty if stack inlet pressure is lowered to 2 atm from 2.5 atm Showed that parasitic power approaches 25 kW_e if the compressor discharge pressure is raised to 4 atm Modified the reference system configuration to include valves and controls for protected shutdown, safe startup from sub-freezing temperatures, and limiting cell voltage to 0.85 V during idle
Collaborations:	3M, Eaton, GM, Gore, JMFC, SA, UTRC, UDEL/Sonijector
Future Work:	Fuel cell systems with emerging high activity catalysts Alternate balance-of-plant components System analysis with durability considerations on drive cycles

Reviewers' Comments

Sample comments and feedback

- ANL employs sound approach in forecasting performance but need more validation
- Impressive range of accomplishments, excellent study of catalyst systems, but needs more focus on Ni loss and other degradation mechanisms
- Excellent collaborations, but needs better definition of engagement with FC-PAD.
- Closer collaboration with OEMs to obtain stack and system data
- The project addresses multiple barriers including cost, performance, thermal/water/air subsystems, but more validation needed.
- Future work appears to be appropriate. More emphasis on durability in future.

Work scope consistent with above recommendations

- ✓ On-going work on differential cell data for PtCo/C dispersed catalysts in collaboration with FC-PAD and an industrial partner. Initial results on performance and durability are included in FC-PAD presentations. The PI is FC-PAD coordinator for modeling and validation thrust area.
- ✓ Examining durability during idling, startup/shutdown and sub-freeze start. Added valves and controls to mitigate catalyst degradation.
- ✓ Analyzing system simplification (compressor air bleed), component elimination (humidifier, H₂ recirculation blower) and alternative operating conditions (higher pressures)
- ✓ Expanded collaboration with an OEM on model validation using 10-50 cm² cells. Analyzing data from a SOA automotive stack and fuel cell system.
- ✓ ANL is a subcontractor to SA on FC-163 project, responsible for supplying performance and design data. Plans and recent results are discussed in bi-weekly calls.



Technical Back-Up Slides

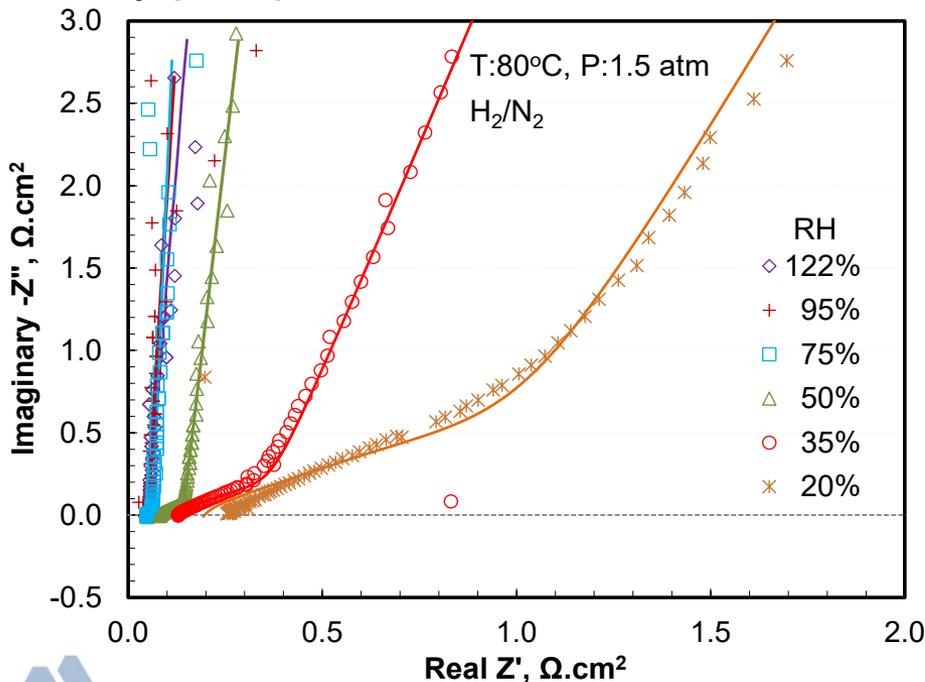


Electrode Resistance

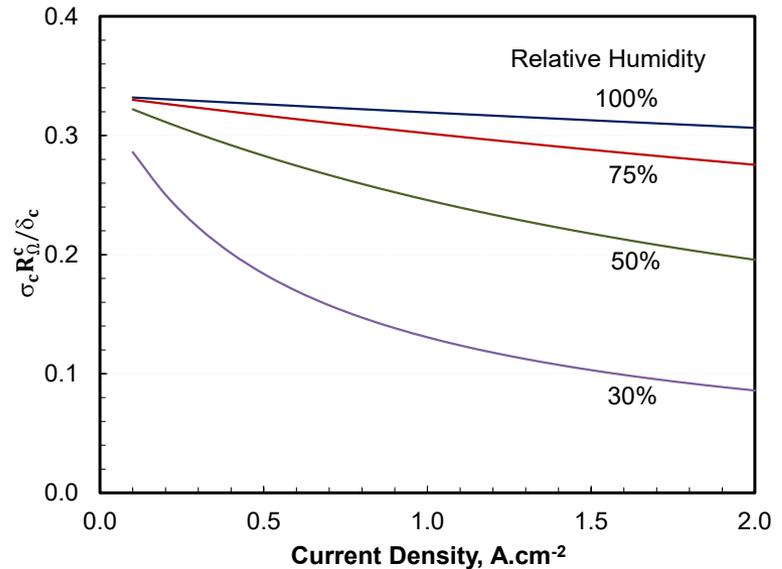
Electrode (σ_c) and membrane conductivities (σ_m) from Galvanostatic impedance data in H_2/N_2 at 0.4 V with 5 mV perturbation

- ZVIEW transmission line model (100 repeat units)

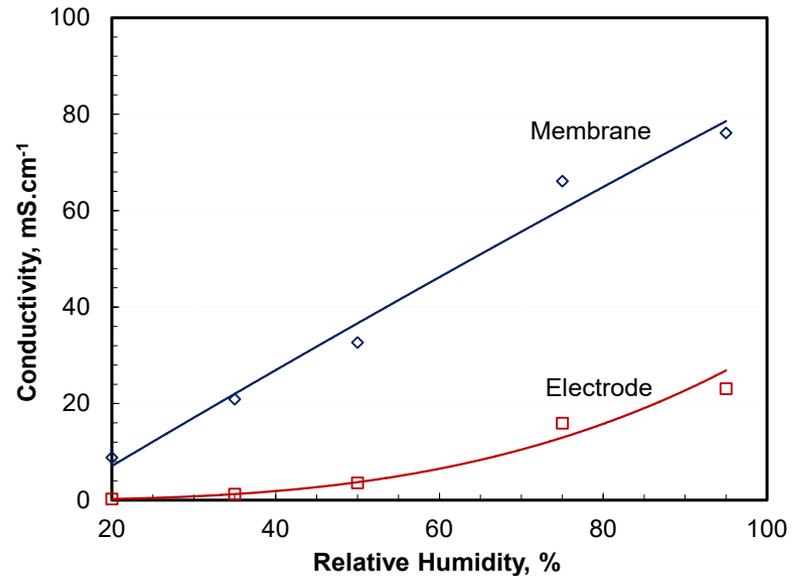
Nyquist plot consistent with RC circuit



Effective electrode resistance



Electrode and membrane conductivities



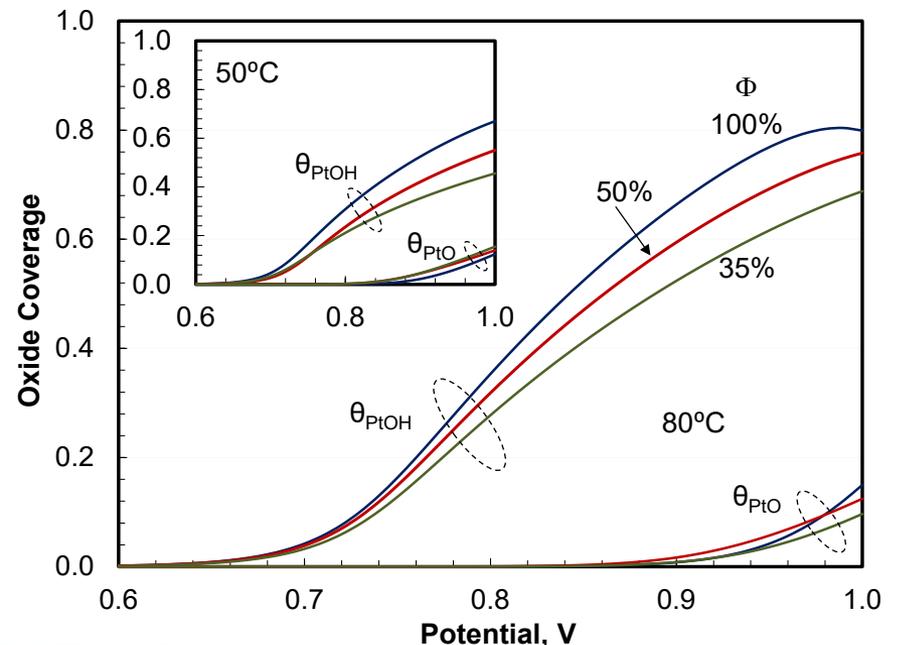
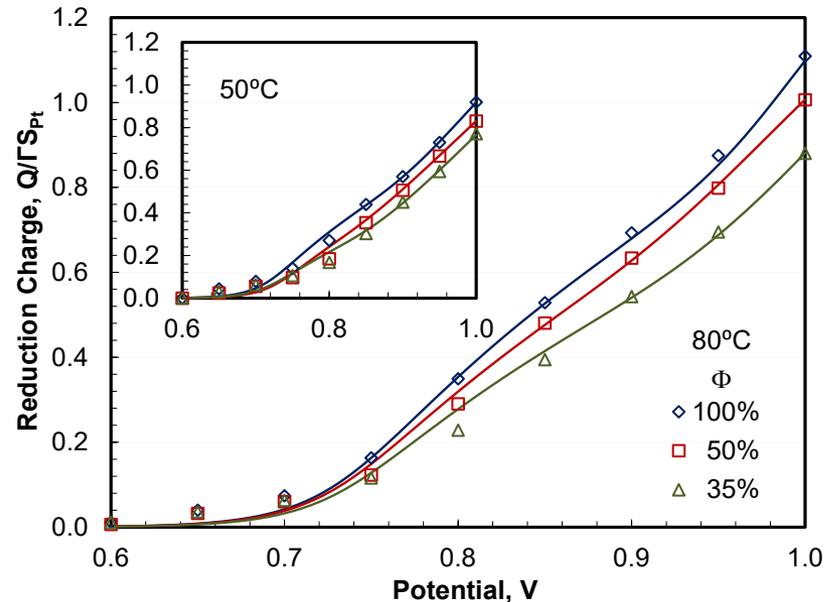
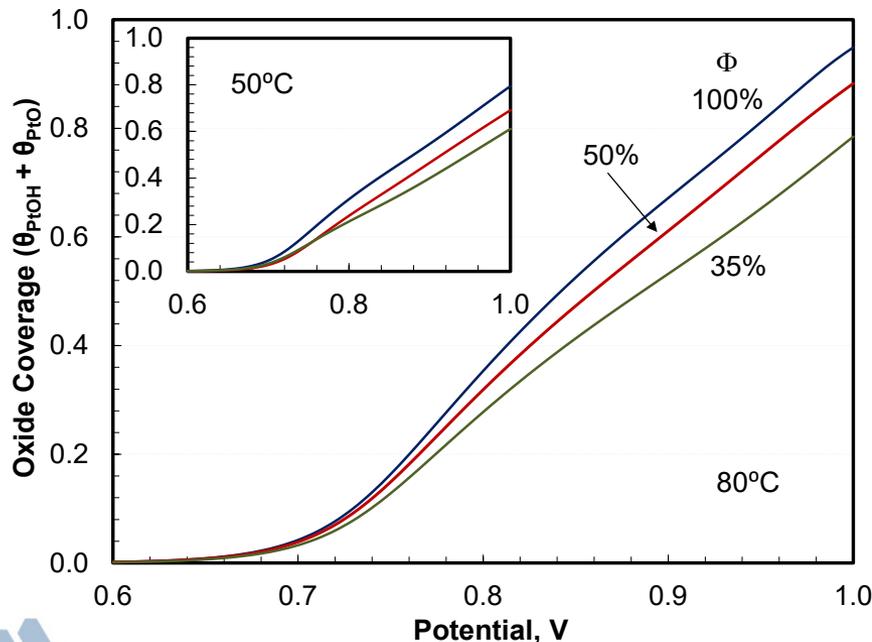
Oxide Formation on d-PtCo/C Catalyst

- Solid solution model for PtO_x formation developed from measured CV reduction charge after 45-min hold at constant potential¹



$$\theta = \theta_{PtOH} + \theta_{PtO}$$

$$K_i = c_{H^+} \left(\frac{\theta_i}{\theta_{i+1}} \right) e^{-\frac{F}{RT}[E - E_{oi}]} = K_{i0} e^{-\frac{\omega_i \theta_i^{x_i}}{RT}}$$



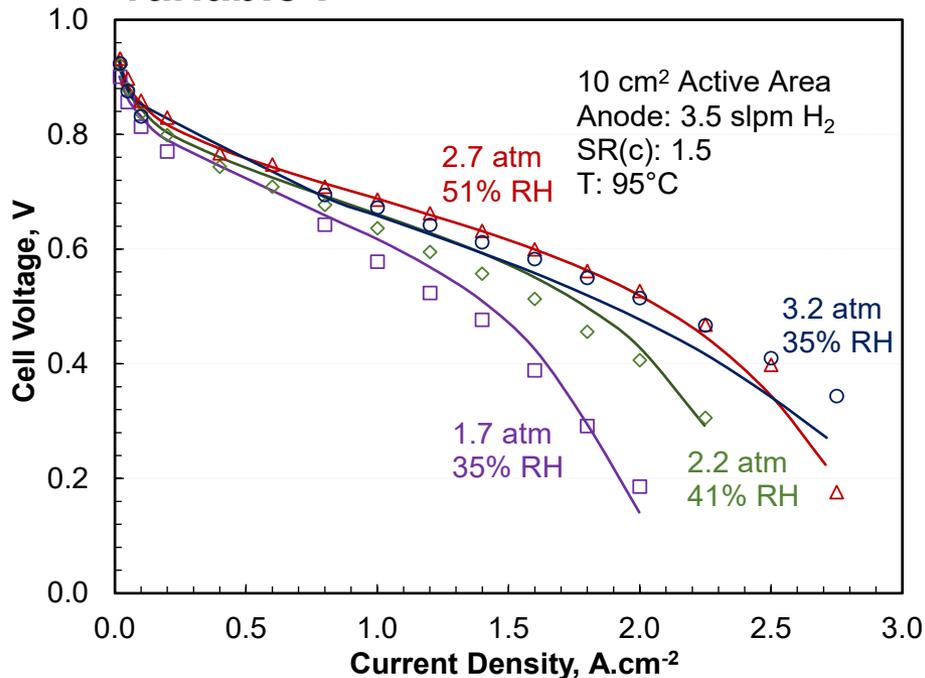
Work in Progress

- Validation tests on 50-cm² integral cell with controlled SR(c) and SR(a)
- Differential cell tests with 0.05 and 0.2 mg/cm² Pt loadings in cathode
- Extracting resistances for O₂ transport in GDL, CCL pores and ionomer

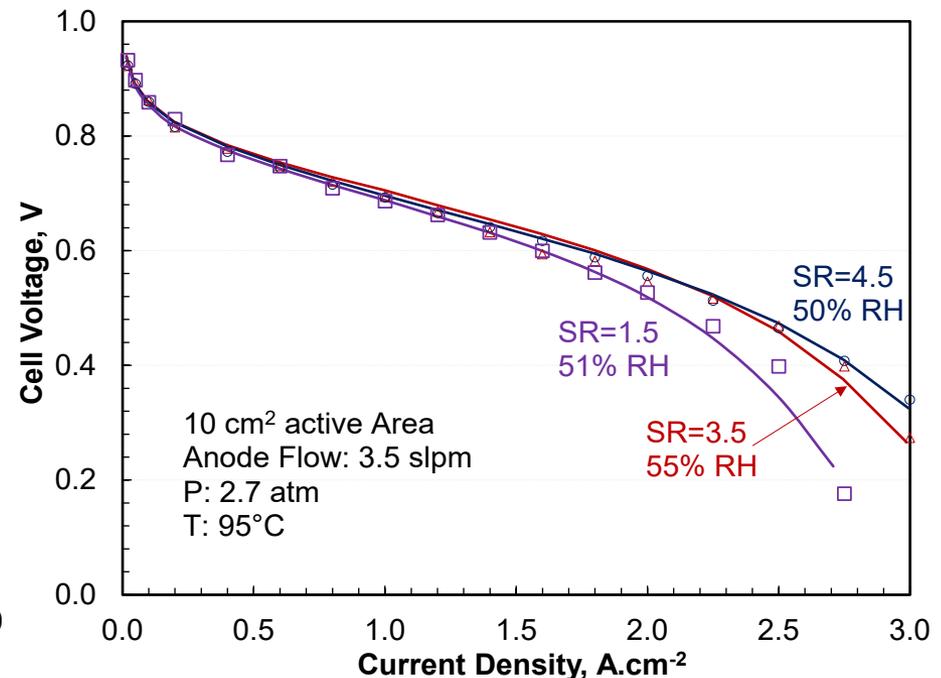
Future Work

- Catalyst accelerated cell tests to model durability

Differential cell operated in integral mode: Constant SR(c), variable P



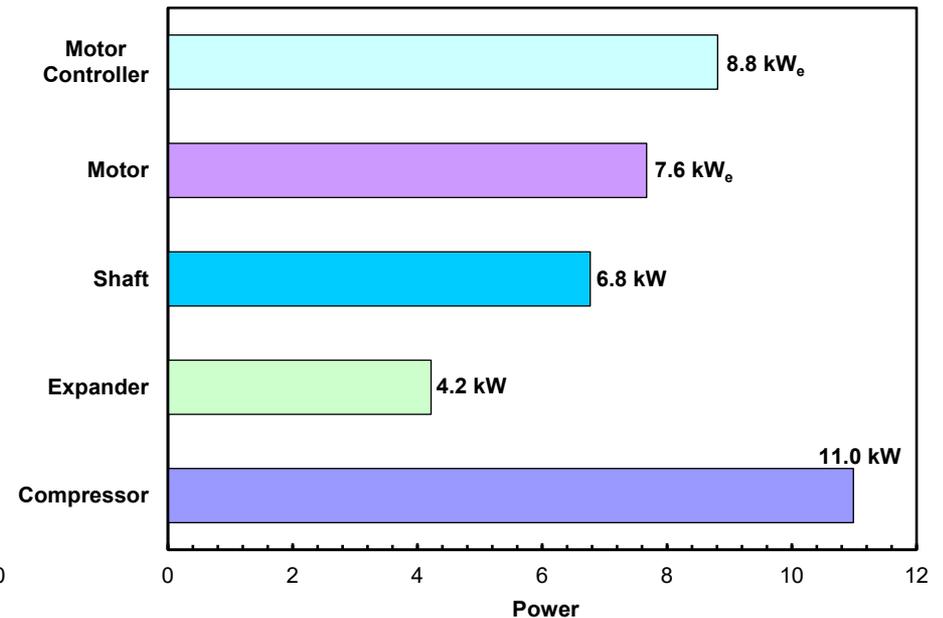
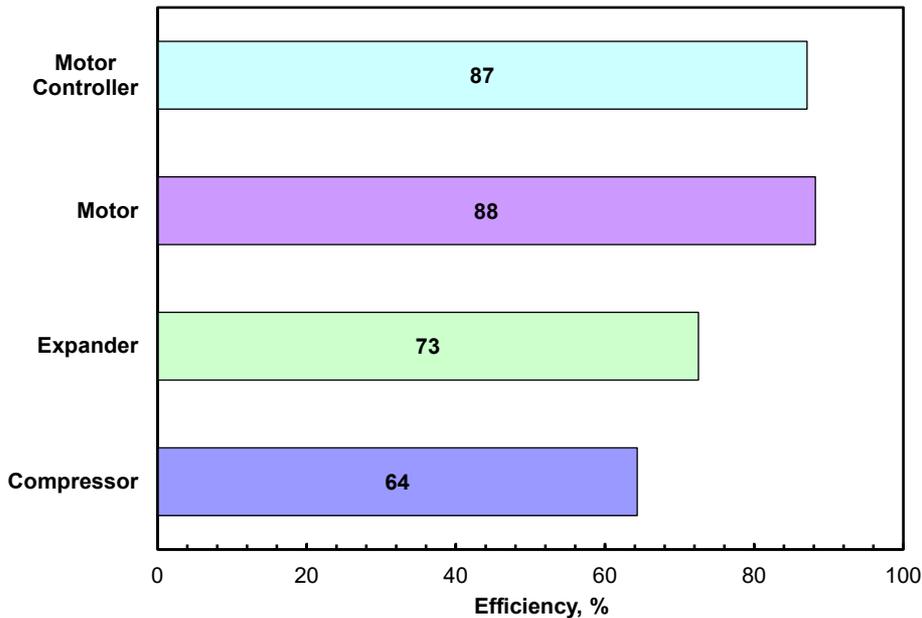
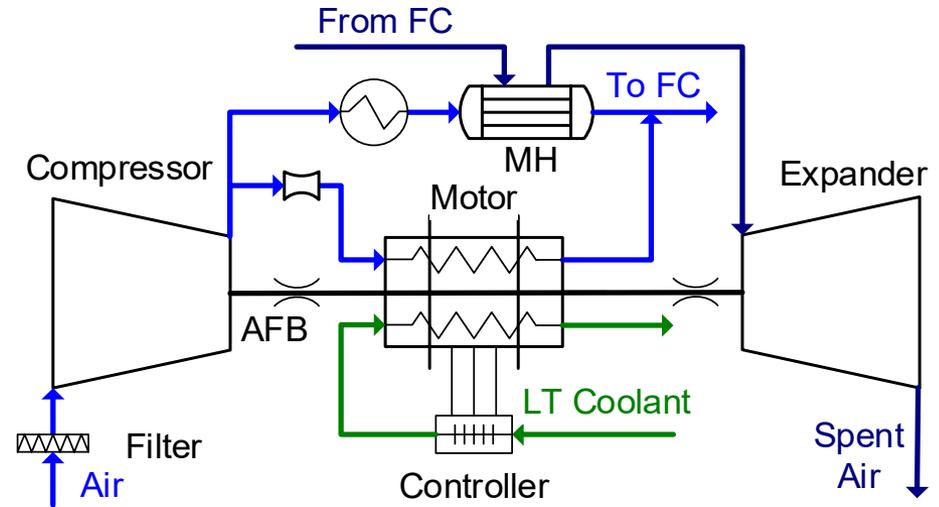
Differential cell operated in integral mode: Variable SR(c), constant P



Projected Performance of CEMM with Bleed Air Recovery

Assumption: CEMM cooling air can be recovered and combined with humidified air upstream of the PEFC stack

- **Data are for as-built components. Efficiencies and performance can be improved by resizing the components to match the actual operating conditions**

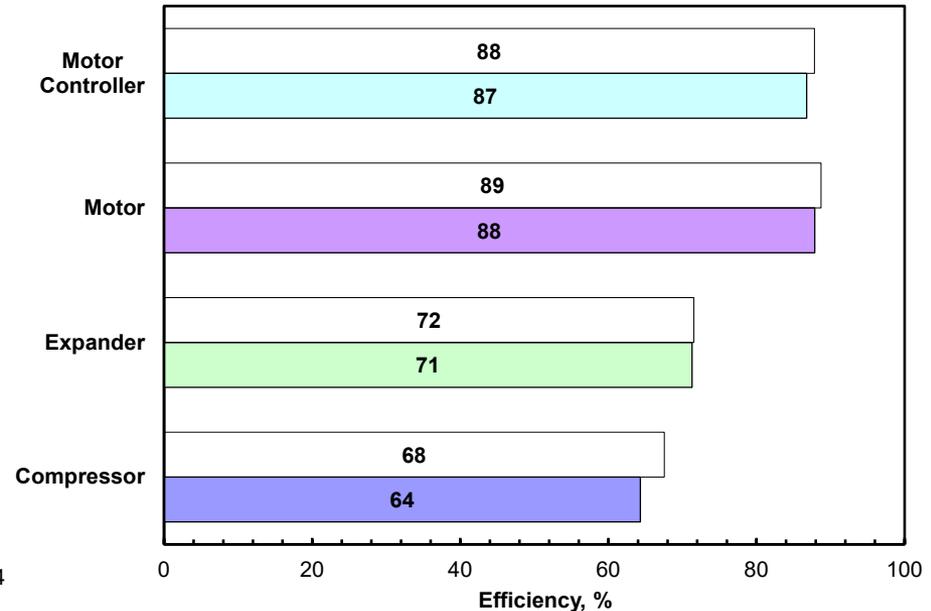
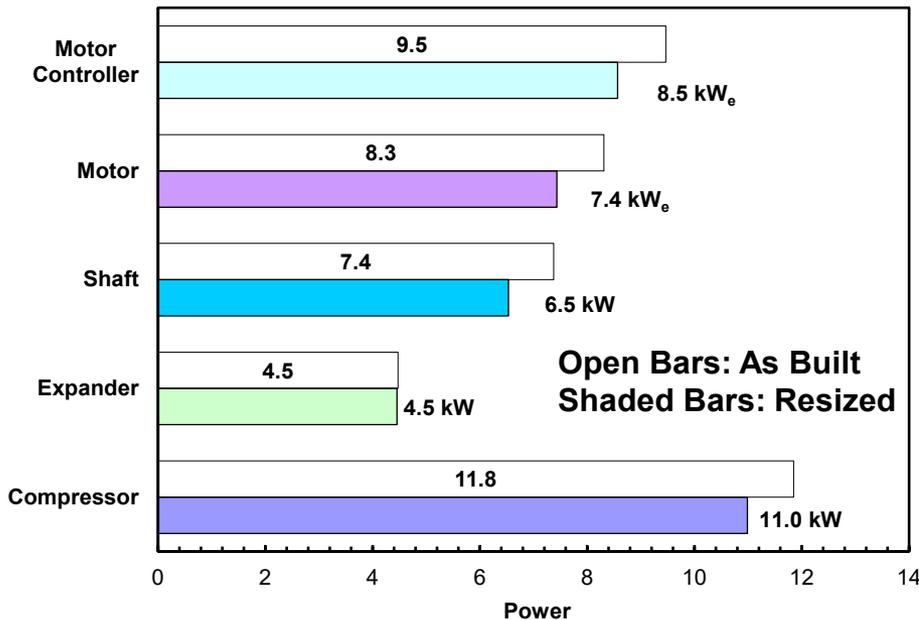
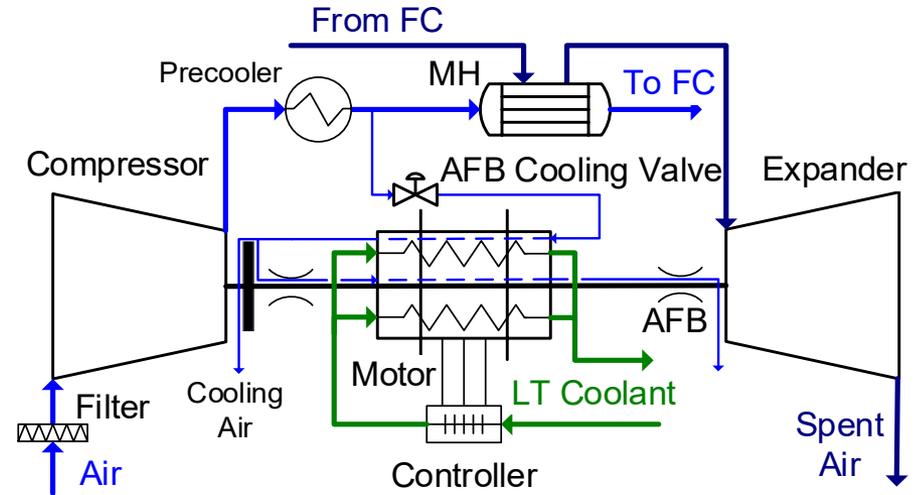


Simulation results for 73 g/s air flow rate and 2.5 atm compressor discharge P; 2.3 atm expander inlet P, 85°C inlet T, fully saturated; 40°C ambient T

Comparative Performance of CEMM w/o Bleed Air Recovery

A control valve used to split pre-determined amount of air downstream of the pre-cooler to cool AFB and motor; this cooling air is lost irrecoverably.

- Data are for as-built components. With resizing, the parasitic power can be reduced to 7.0 kW_e with bleed air recovery and 8.1 kW_e w/o bleed air recovery.



Simulation results for 73 g/s air flow rate and 2.5 atm compressor discharge P; 2.3 atm expander inlet P, 85°C inlet T, fully saturated; 40°C ambient T

