

# Fuel Cells Systems Analysis

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# Overview

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## Timeline

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

## Budget

- FY10 funding: \$650K (+\$300K)  
DOE share: 100%
- FY09 funding: \$600K (+\$300K)

## Barriers

- B. Cost
- C. Performance
- E. System Thermal and Water Management
- F. Air Management
- J. Startup and Shut-down Time, Energy/Transient Operation

## Partners/Interactions

- Honeywell CEM+TWM projects
- DTI, TIAX
- 3M, Emprise, PermaPure
- ISO-TC192 WG12, HNEI, JARI, LANL
- IEA Annexes 17 and 20
- FreedomCAR fuel cell tech team

- 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



# Objectives

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Develop a validated system model and use it to assess design-point, part-load and dynamic performance of automotive and stationary fuel cell systems.

- 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



# Approach

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

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

Validate the models against data obtained in laboratory and at Argonne's Fuel Cell Test Facility.

- Collaborate with external organizations

Apply models to issues of current interest.

- Work with FreedomCAR Technical Teams
- Work with DOE contractors as requested by DOE



# Collaborations

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Air Management	Honeywell Turbo Technologies
Stack	3M, Nuvera
Water Management	Honeywell Aerospace, Emprise, PermaPure
Thermal Management	Honeywell Thermal Systems
Fuel Economy	ANL (PSAT)
H <sub>2</sub> Impurities	HNEI, JARI, LANL, ISO-TC-192 WG
System Cost	DTI, TIAX
Dissemination	IEA Annex 22 and 25

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



# Technical Accomplishments

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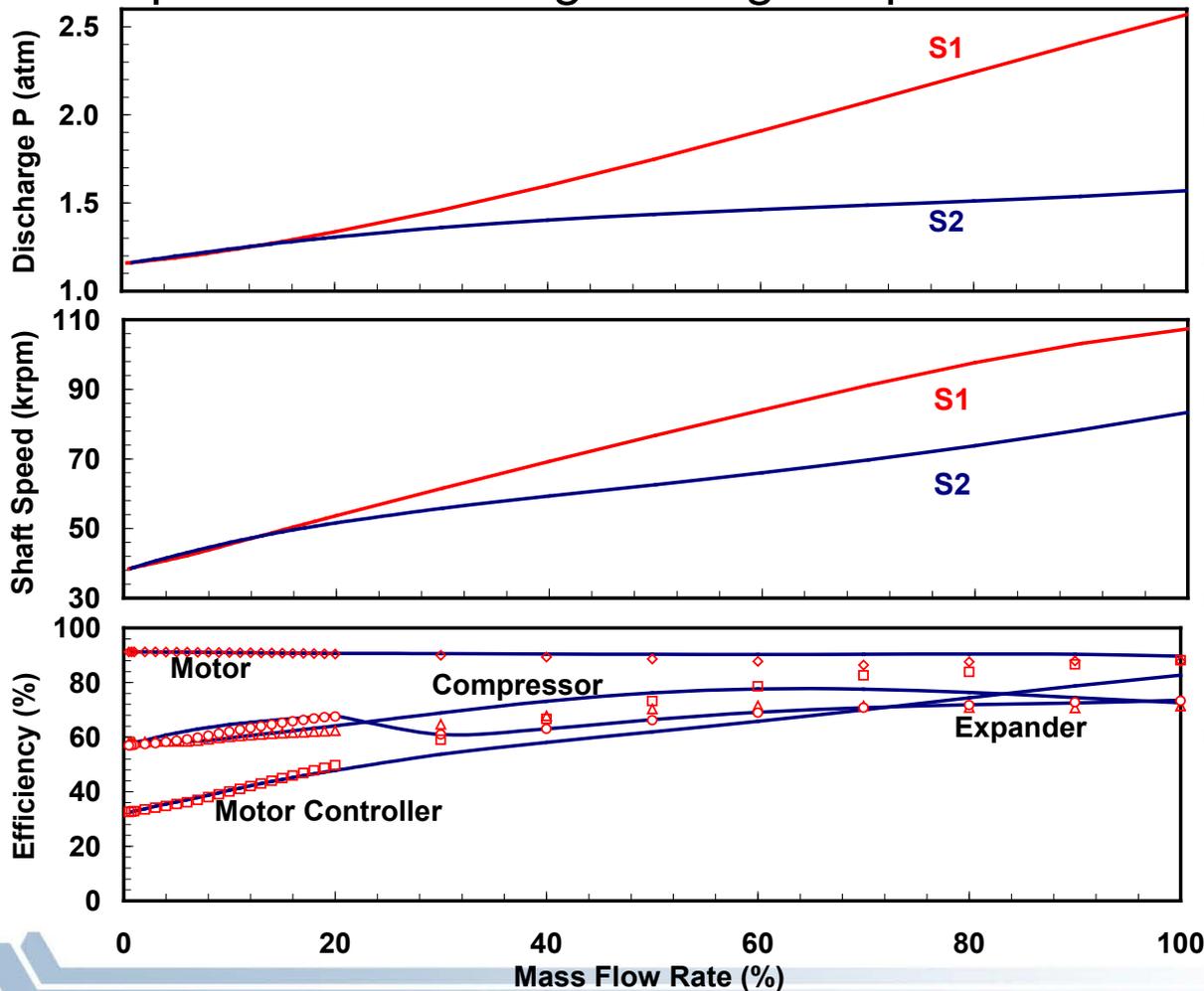
1. System analysis to update the status of technology
  - **Stack:** Determined the performance of NSTFC stacks with 0.15 mg/cm<sup>2</sup> Pt loading and 20- $\mu$ m membrane
  - **Air Management:** Evaluated the performance of Honeywell's compressor-expander-motor module for 1.5-atm operation
  - **Fuel Management:** Evaluated the performance of parallel ejector-pump hybrids
  - **Water Management:** Constructed performance maps for planar membrane humidifiers
  - **Thermal Management:** Collaborated with Honeywell to evaluate performance of microchannel automotive radiators
  - **Drive Cycle Simulations:** GCTool-PSAT simulations for fuel economy of hybrid FCEVs
  - **Cost:** Assisting TIAX in projecting cost of Argonne FCS-2010 at high volume manufacturing
2. Hydrogen impurity effects (Backup Slides)
  - Conducting dynamic simulations to projected combined effect of H<sub>2</sub> impurities at ISO specs
  - Providing modeling support to ISO-TC192 WG-12 efforts





# Air Management System

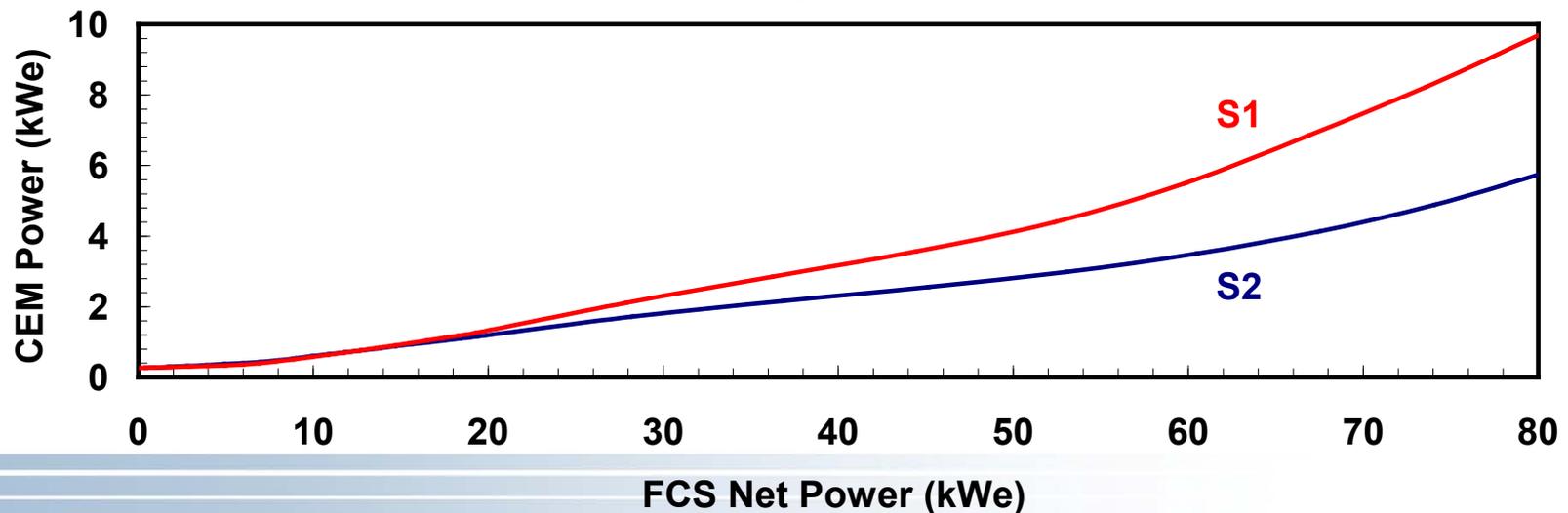
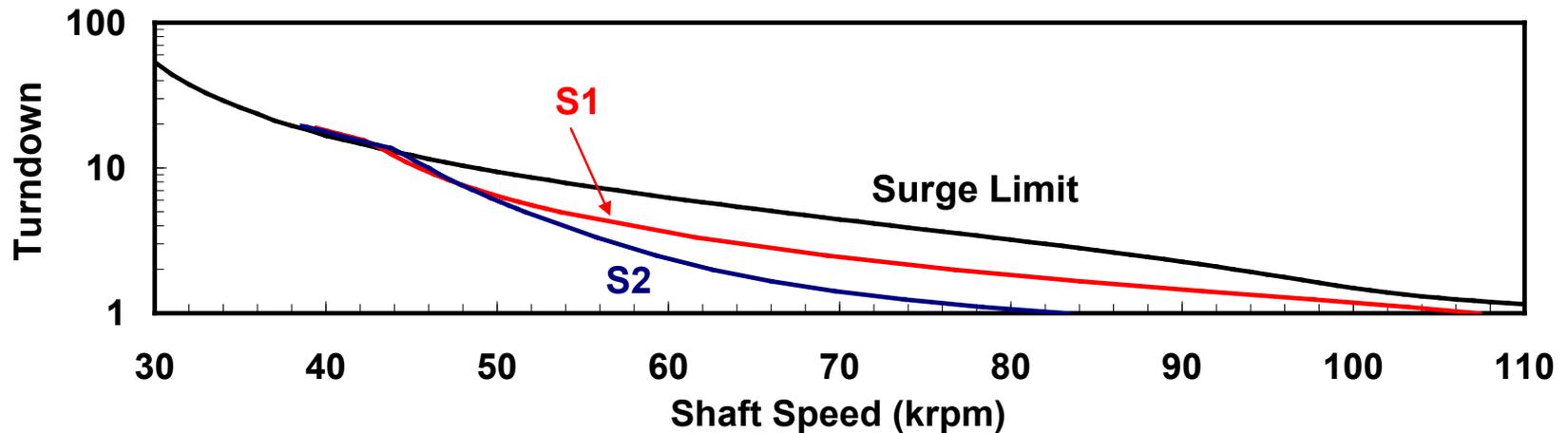
- Determined performance of Honeywell's compressor-expander-motor module (CEMM) originally designed for 2.5-atm peak P (S1)
- Comparable component efficiencies for S1 and S2 which may improve with redesign for higher rpm



- Mixed axial flow compressor
- Variable nozzle turbine (VNT)
- 3-phase brushless DC motor, liquid and air cooled
- Motor controller, liquid cooled
- Air foil bearing (AFB)

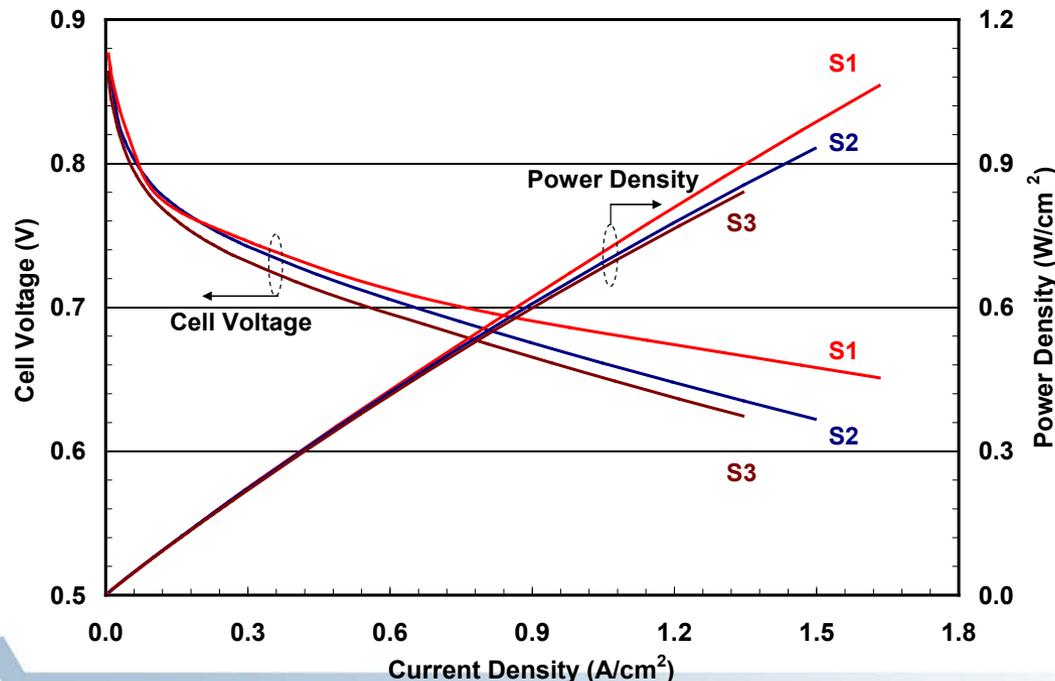
# Performance of Integrated CEM Module

- Maximum turndown may be limited by compressor surge for shaft speeds less than 45 krpm
- At rated power, the CEMM consumes  $\sim 10 \text{ kW}_e$  in S1,  $< 6 \text{ kW}_e$  in S2
- CEMM min. power between  $270\text{-}400 \text{ W}_e$ , determined by AFB durability



# Reference Stack with 3M's NSTF Catalysts

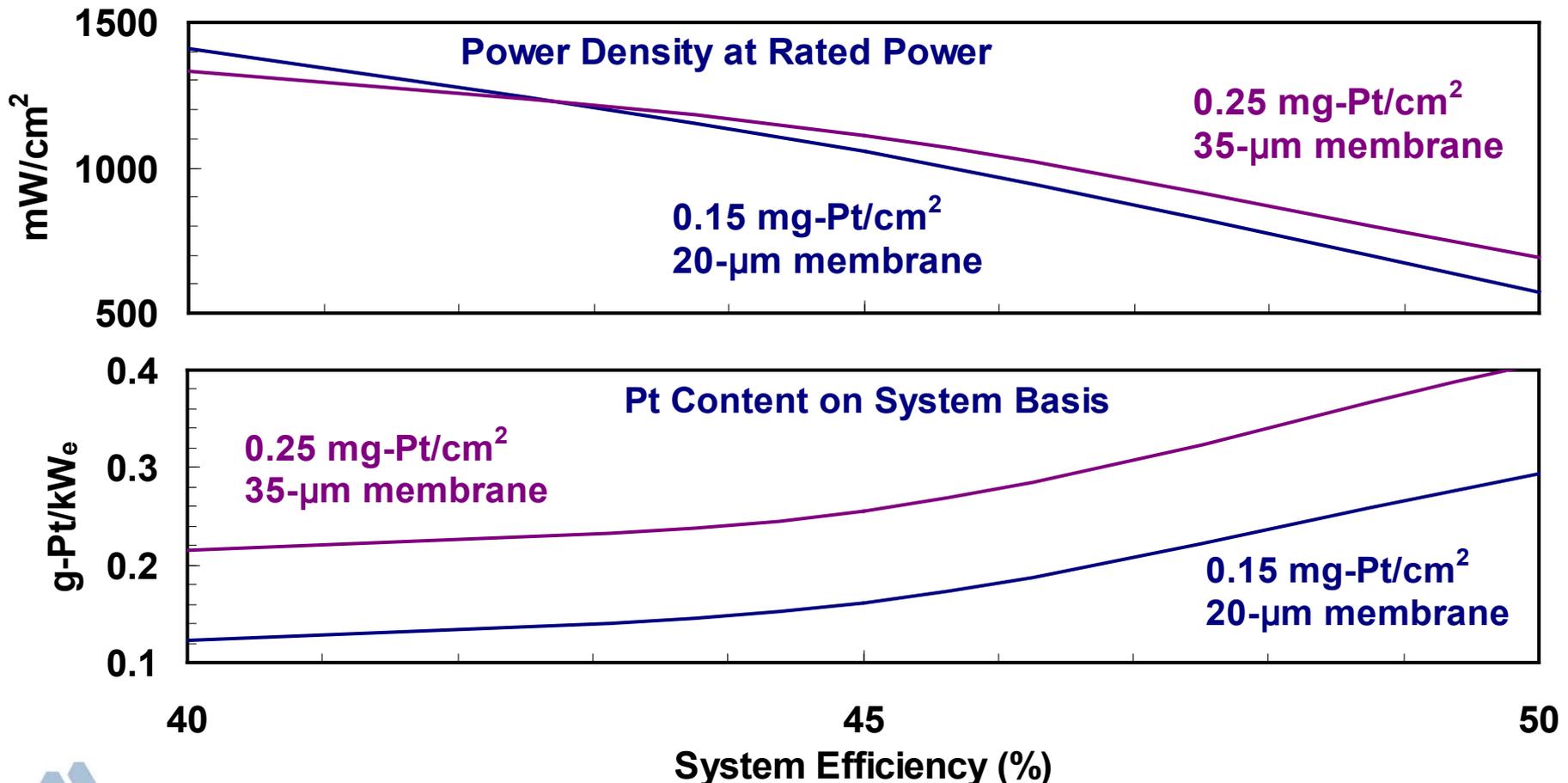
- 3M's single cell data with 0.1(a)/0.15(c) mg/cm<sup>2</sup> Pt, ternary PtCoMn catalyst, and 35- $\mu$ m 850 EW membrane
- 3M's single cell data with 0.05(a)/0.1(c) mg/cm<sup>2</sup> Pt, ternary PtCoMn catalyst, and 20- $\mu$ m 850 EW membrane
- ECSA, specific activity, short and crossover currents and HFR data from CV, EIS and H<sub>2</sub>/air cells at 0.9 V, 70-120°C, 20-100% RH
- Determined optimum combination of stack temperature and inlet RH as a function of pressure, stoichiometry and MEA parameters



FCS	S1	S2	S3
Stack P, atm	2.5	1.5	1.5
Stack T, °C	85	75	65
Dew Point T (c), °C	64	61	22
Dew Point T (a), °C	59	53	22

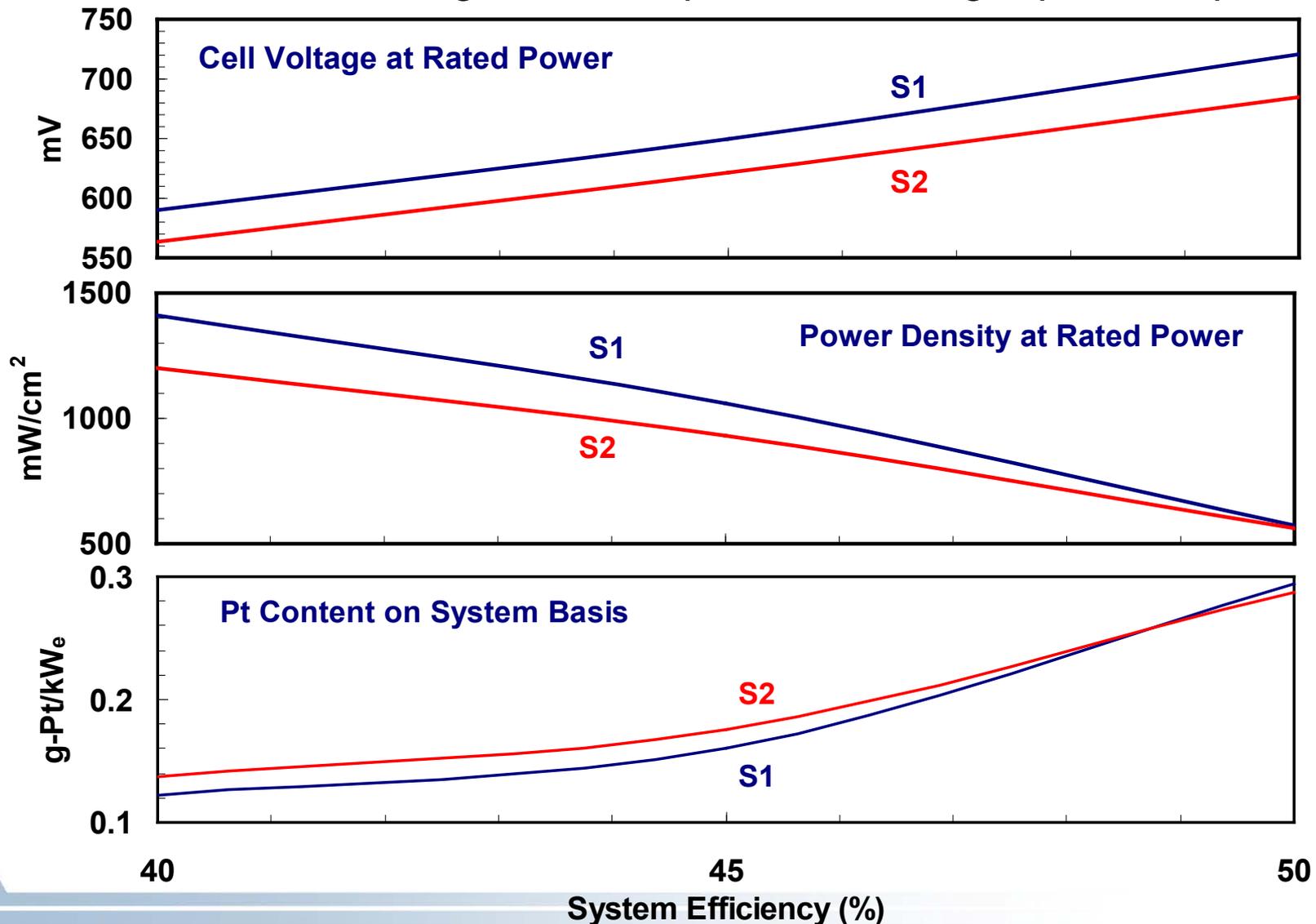
# Stack Performance – Effect of Pt Loading

- Reference 2009 S1 system: 0.25 mg-Pt/cm<sup>2</sup>, 35- $\mu$ m membrane
- Reference 2010 S1 system: 0.15 mg-Pt/cm<sup>2</sup>, 20- $\mu$ m membrane
- 30-45% projected reduction in Pt content because of lower Pt loading and thinner membrane, 0.12-0.30 g-Pt/kW<sub>e</sub>(net)



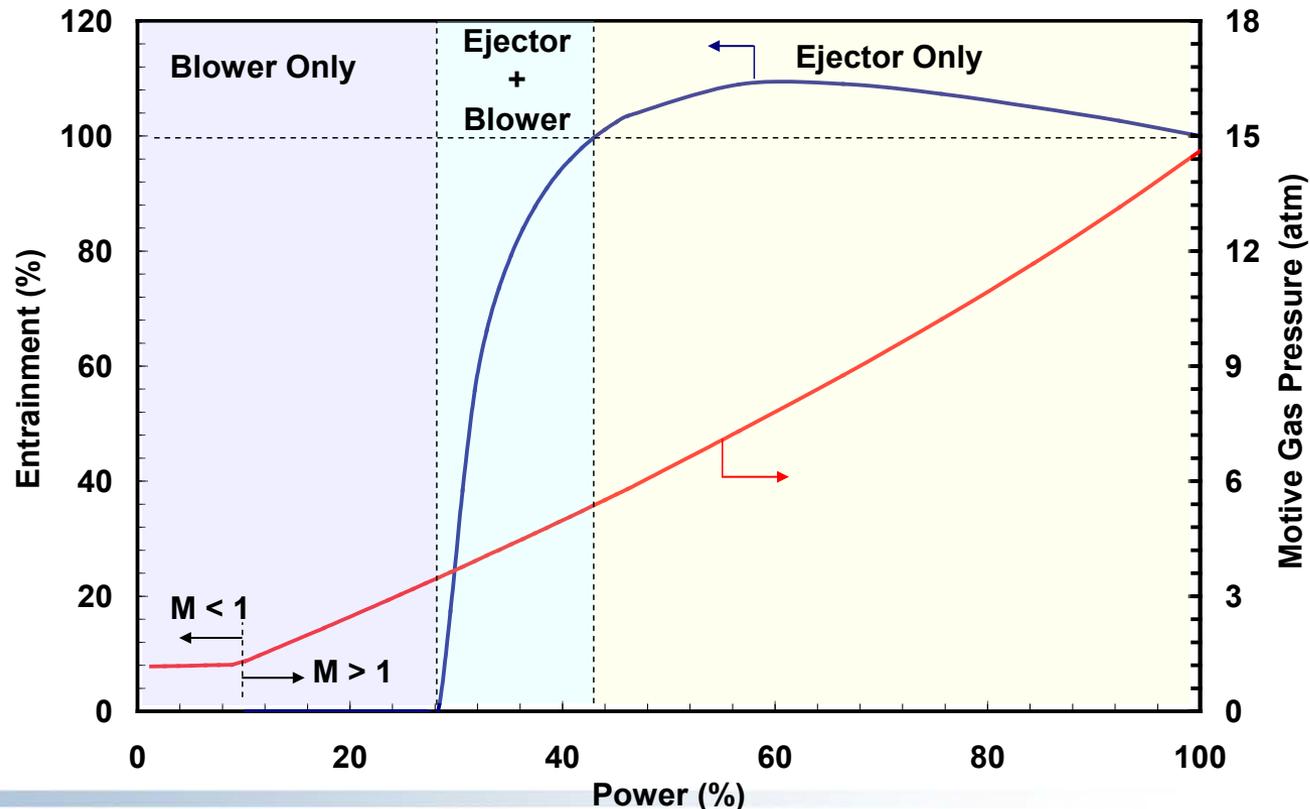
# Stack Performance – Effect of Operating Pressure

- S1 has higher power density and lower Pt content even though cell V has to be 25-35 mV higher to compensate for larger parasitic power



# Fuel Management System

- Parallel ejector-pump hybrid
- Ejector performance
  - Motive (p) gas:  $\text{cH}_2$ , 15-atm maximum P, MW=2
  - Suction (s) gas:  $\text{H}_2$  with water, 1-1.5 atm, 75°C, MW=3-7
  - Lift pressure: <3 psi; Recycle ratio:  $m_s/m_p = 2-5$
  - Blower flow rate(%): 100-Entrainment(%)

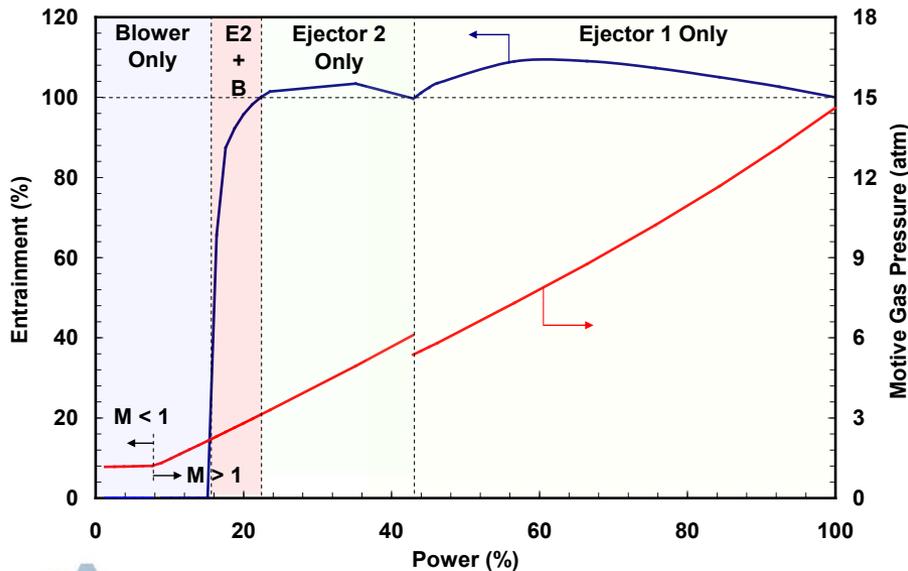


# Alternate Arrangements

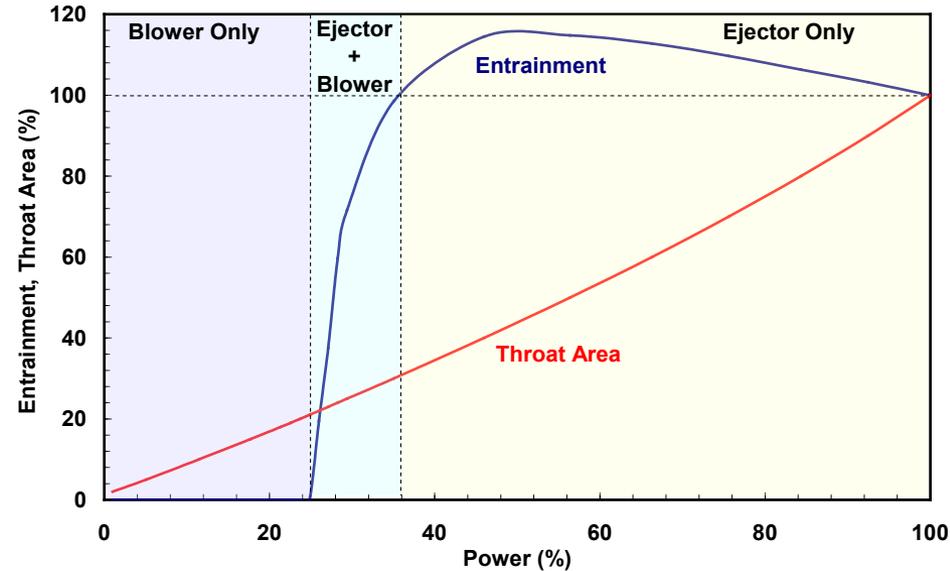
- A blower is always needed if the FCS has a turndown >3 with single ejector, >4 with variable geometry ejector, and > 5 with dual ejectors

		Ejector Arrangement			
Blower		None	Single	Variable Geometry	Dual
Flow Rate	L/s	20	5.6	4.5	3.1
Pressure Head	psi	3	0.8	0.65	0.4
Power	W	400	35	25	10

## Dual Ejectors + Blower

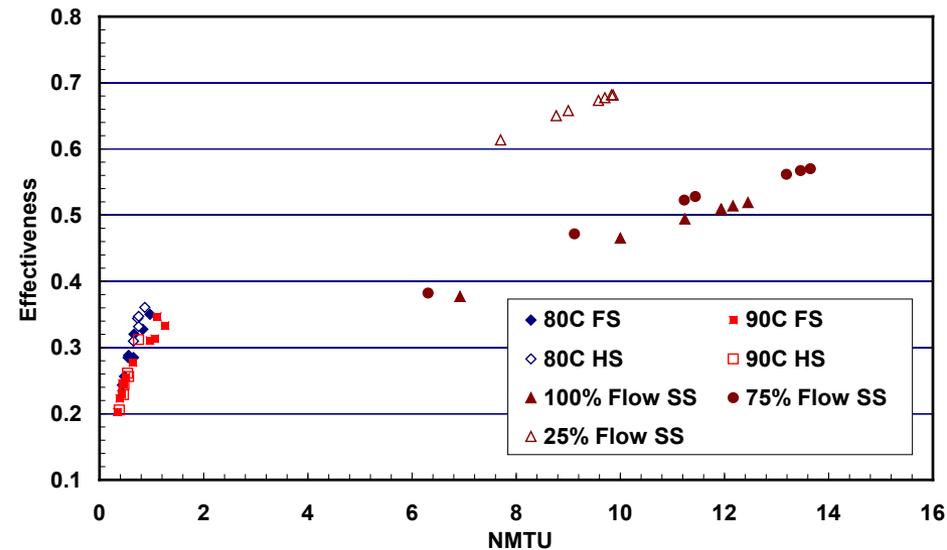
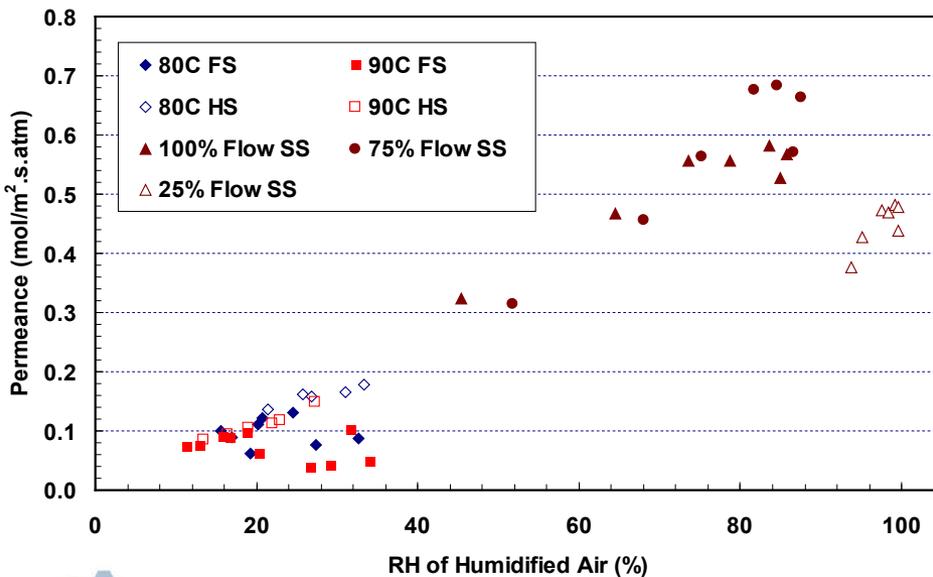


## Variable Geometry Ejector + Blower



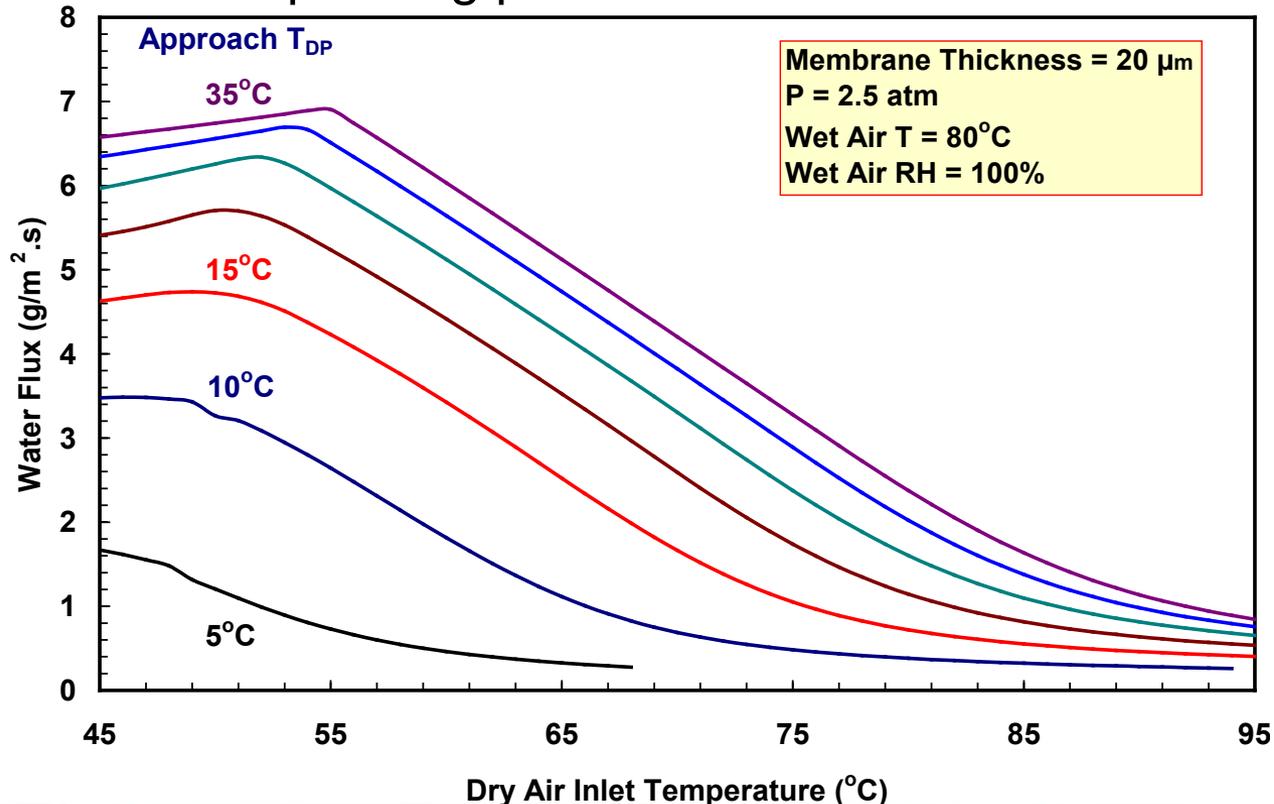
# Water Management System

- Analyzed Honeywell and PermaPure data for full-scale (FS), half-scale (HS) and 1/10<sup>th</sup> sub-scale (SS) membrane humidifiers
- Permeance ( $\kappa_m$ ) for the units (not local values) can be represented in terms of the dry-air outlet RH.
  - $\kappa_m$  defined similarly as LMTD for heat transfer
  - $\kappa_m$  also depends on temperatures
- Effectiveness ( $\varepsilon_m$ ) can be represented in terms of NMTU (number of mass transfer units)



# Water Mass Transfer Flux

- Model results for mass transfer from saturated wet stream
- Optimum dry-air inlet temperature ( $T_m$ ) for maximum flux
  - Flux decreases for  $T_d > T_m$  because of low uptake (too dry)
  - Flux decreases for  $T_d < T_m$  as humidified air approaches saturation
- Flux is also a strong function of the membrane thickness, temperature of the wet air and operating pressures



# Thermal Management System

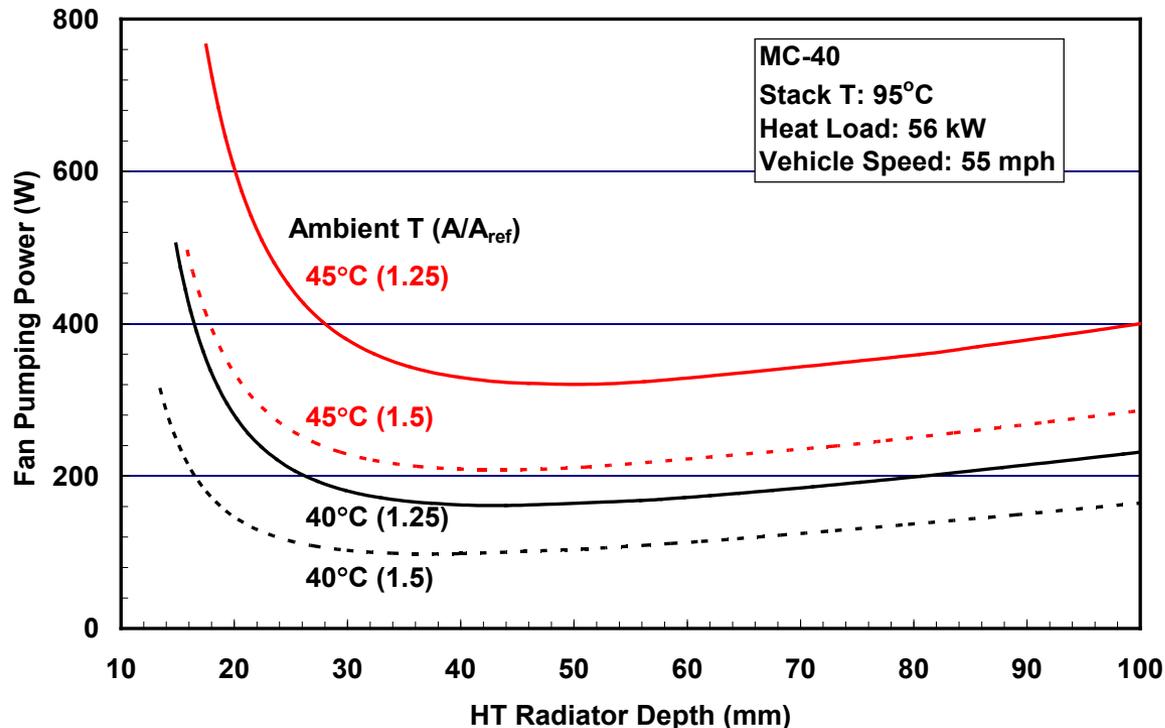
- Heat rejection more difficult in FCVs than ICEVs
  - Higher efficiency, but similar or higher heat loads but smaller  $\Delta T$
  - Additional FCS and electric drivetrain components requiring cooling to lower temperatures: stacked A/C condenser, LTR and HTR
  - Heat rejection in FC powertrains is most challenging while driving at 55 mph on 6.5% grade with 600 kg payload
  - Allow stack temperature to rise when heat rejection is difficult
  - Expander needed even in S2 and S3

		S1			S2			S3		
Rated Power	kWe	80	80	80	80	80	80	80	80	80
System Efficiency	%	50	45	40	50	45	40	50	45	40
Stack Pressure	atm	2.5	2.5	2.5	1.5	1.5	1.5	1.5	1.5	1.5
Stack Temperature	°C	85	85	85	75	75	75	65	65	65
Heat Rejection										
Demand Power	kWe	61.5	61.5	61.5	61.5	61.5	61.5	61.5	61.5	61.5
Cell Voltage	mV	741	681	634	732	682	649	734	688	657
Stack Pressure	atm	2.4	2.4	2.3	2.1	2.1	2	2.1	2.1	2
Stack Temperature	°C	95	95	92	95	95	92	80	80	75
HT Radiator Heat Load	kW	49.5	59.5	68.3	49.3	57.6	63.5	49.4	56.7	62.2
LT Radiator Heat Load	kW	16.2	16.5	16.2	13.6	13.8	13.5	13.3	13.5	13.3



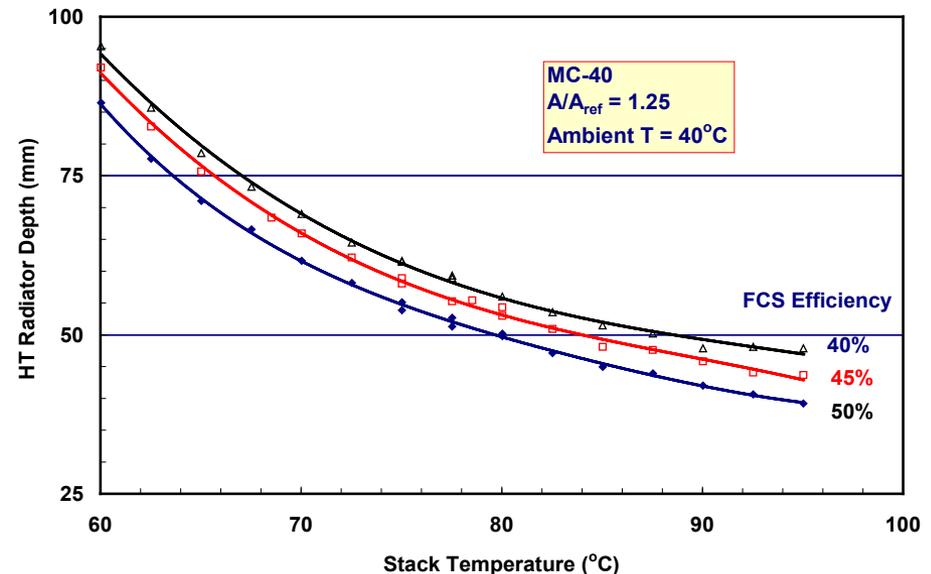
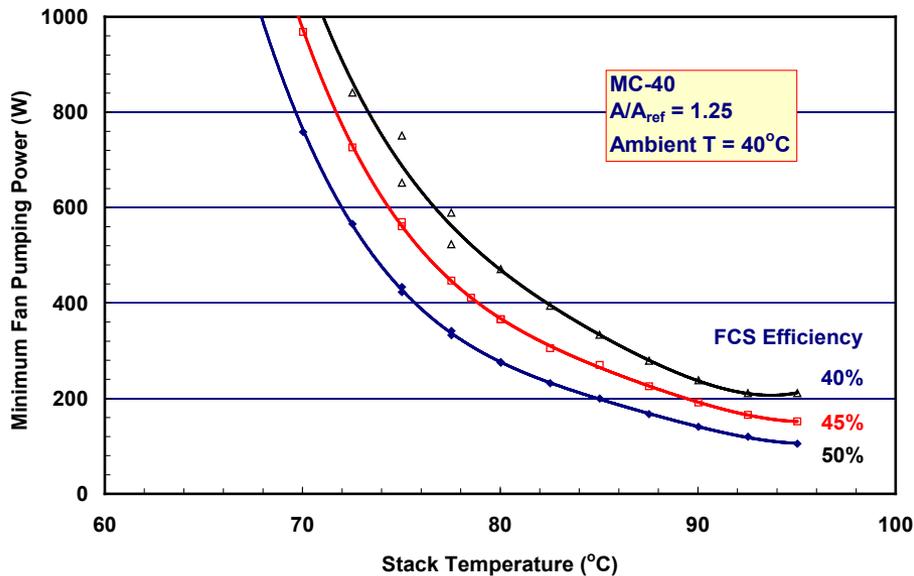
# Microchannel Radiator

- Derived  $f$  and  $j$  factors from Honeywell data for 9"x9"x1.3" subscale and 27.6"x17.7"x1.3" full-scale radiators
  - 18 and 24 fpi louver and 40 and 50 fpi microchannel fins
- FCS radiator can be more compact with 40-fpi microchannel than with standard automotive 18-fpi louver fins
- FC powertrains will likely be derated at  $T_a > 40^\circ\text{C}$  since the fan power doubles for every  $5^\circ\text{C}$  increase in ambient temperature ( $T_a$ )



# Heat Rejection vs. Stack Efficiency at Rated Power

- At low stack temperatures, the minimum fan power increases nonlinearly with heat load (i.e., FCS efficiency at rated power)
- For given fan power, 40% FCS (S2) must be capable of operating at 5-10°C higher stack temperature than 50% FCS
- From the standpoint of heat rejection, 40% FCS may be acceptable if it can be operated at 95°C (S3 not OK)

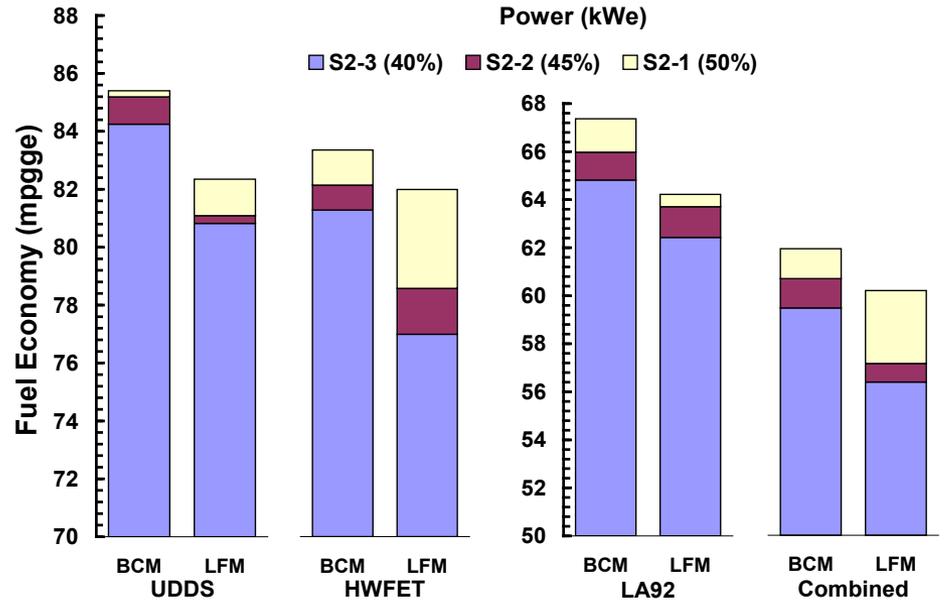
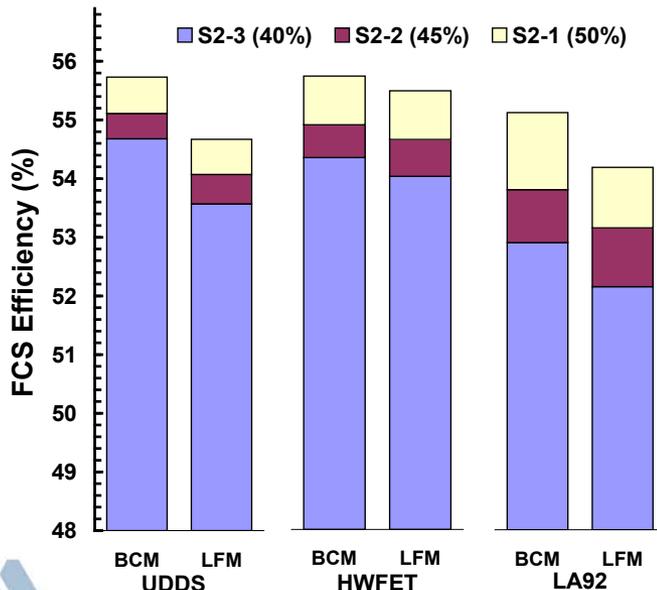
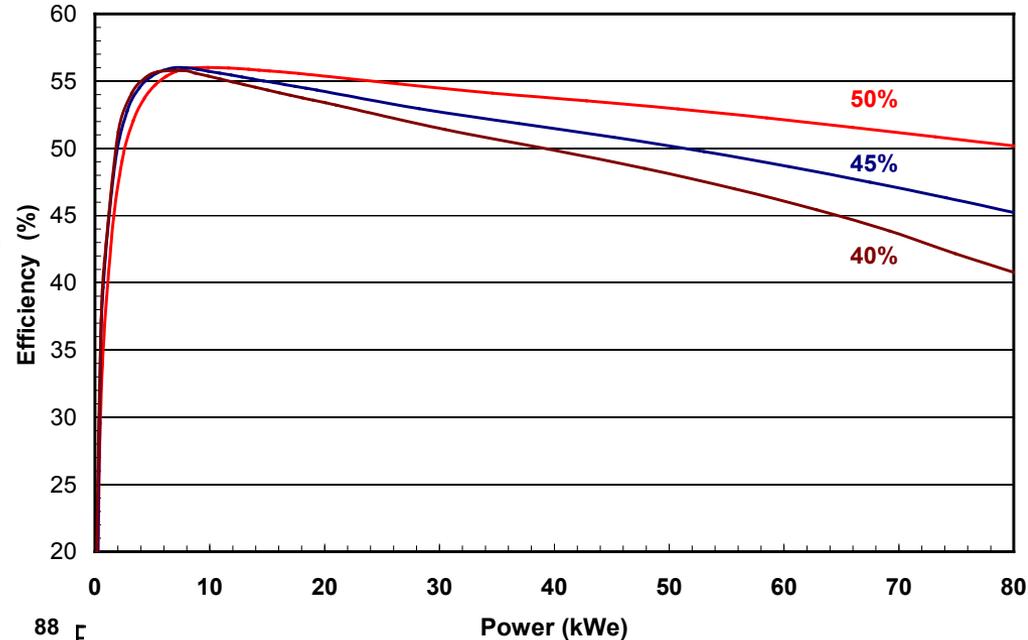


# System Performance - Summary

Lowering rated-power efficiency from 50% to 40% decreases

- peak efficiency by <1%
- fuel economy by 4% in battery-charging mode (BCM) & 7% in load-following mode (LFM)
- and system Pt content by 50%

No. of start-stops on UDDS: 58 in BCM, 4 in LFM2



PSAT drive-cycle simulation results for a mid-size hybrid vehicle

# Future Work

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## 1. Systems Analysis

- Support DOE/FreedomCAR development effort at system, component, and phenomenological levels
- Collaborate with 3M to develop durability models for NSTFC electrode structures
- Continue cooperation with Honeywell and others to validate air, fuel, thermal, and water management models
- System optimization for cost, performance, and durability
- Drive cycle simulations for durability enhancement
- Alternate membrane, catalyst structures, and system configurations
- Support DTI and TIAX in high-volume manufacturing cost projections

## 2. Hydrogen Quality

- Validate impurity models against U.S. and JARI data
- Project effects of proposed standards on stack performance
- Provide modeling support to ISO-TC192 WG-12 and the Codes and Standards Technical Team

# Appendix



# Dynamic Behavior of Fuel Hydrogen Impurities in Polymer Electrolyte Fuel Cells

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1. SMR-PSA may be the pathway for producing  $H_2$  in the early stages of hydrogen economy
  - CO and  $N_2$  levels determine the  $H_2$  recovery from PSA and influence the cost of producing  $H_2$
  - Of the two impurities, CO degrades the cell performance more but accumulation of  $N_2$  accumulation can significantly dilute the  $H_2$  concentration
  - Our simulations show that  $N_2$  crossover from air is the main source of  $N_2$  buildup in the anode recycle stream and determines the purge schedule
2.  $H_2S$  and  $NH_3$  are extremely harmful to PEFC efficiency but are easily removed in PSA beds
  - Dynamic simulations show that  $NH_3$  cannot accumulate in the anode recycle stream

# Hydrogen Purification Drivers (PSA)

Species	Adsorption Force	ISO TC 197 WG 12 (14687) Draft Spec	ATR Mol %	Purification Ratio for ATR	SMR Mol %	Purification Ratio for SMR	OVERALL EFFECT
Helium (He)	Zero	100 ppm (total inert)	500 ppm	5	500 ppm	5	<b>NOT POSSIBLE</b>
Hydrogen (H2)	Weak	99.99%	40-45%		75-80%		Impacts PSA recovery & Capital Cost
Oxygen (O2)		5 ppm	50 ppm	10	-	-	Impacts PSA recovery & Capital Cost
Argon (Ar)		100 ppm (total inert)	500 ppm	5	500 ppm	5	Impacts PSA recovery & Capital Cost
Nitrogen (N2)		100 ppm (total inert)	34-38%	3800	1000 ppm	10	Impacts PSA recovery & Capital Cost
Carbon Monoxide (CO)		0.2 ppm	0.1 -1 %	50000	0.1-4%	200000	Impacts PSA recovery & Capital Cost
Methane (CH4)		2 ppm (incl THC)	0.5 – 2%	10000	0.5 – 3%	15000	Impacts PSA recovery & Capital Cost
Carbon Dioxide (CO2)		2 ppm	15-17%	85000	15 -18%	90000	Relatively easier to remove
Total HC's			2 ppm (incl CH4)	0.1 %	500	0.5%	2500
Ammonia	Strong	0.1 ppm	Low ppm		Low ppm		Relatively easier to remove
Total Sulfur	Strong	0.004 ppm					Relatively easier to remove
Halogenates	Strong	0.05 ppm					Relatively easier to remove
Water (H2O)	Strong	5 ppm	Dew Point		Dew Point		Relatively easier to remove

DOE H2QWG Draft Roadmap. Courtesy, Bhaskar Balasubramanian (Chevron)

# CO/CO<sub>2</sub> Poisoning Model

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## ■ Hydrogen Oxidation Reaction

- $\text{H}_2 + 2\text{M} \leftrightarrow 2\text{M-H}$  (Dissociative Adsorption)
- $\text{M-H} \rightarrow \text{M} + \text{H}^+ + \text{e}^-$  (Electrochemical Oxidation)

## ■ CO Poisoning of Pt

- $\text{CO} + \text{M} \leftrightarrow \text{M-CO}$  (Associative Adsorption on Linear Sites)
- $\text{CO}_2 + 2\text{M-H} \rightarrow \text{M} + \text{M-CO} + \text{H}_2\text{O}$  (Reverse Water-Gas Shift)
- $\text{M-CO} + \text{H}_2\text{O} \rightarrow \text{M} + \text{CO}_2 + 2\text{H}^+ + 2\text{e}^-$  (Electrochemical Oxidation)

## ■ Reactions with Oxygen

- $\text{M-CO} + \frac{1}{2} \text{O}_2 \rightarrow \text{M} + \text{CO}_2$  (CO Oxidation)
- $2\text{M-H} + \frac{1}{2} \text{O}_2 \rightarrow 2\text{M} + \text{H}_2\text{O}$  (H<sub>2</sub> Oxidation)

# H<sub>2</sub>S Poisoning Model

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- Sequential Sorption of H<sub>2</sub>S on Pt
  1.  $nM + H_2S \leftrightarrow M_n-H_2S$  (Associative chemisorption)
  2.  $M_n-H_2S \leftrightarrow M_nS + 2H^+ + 2e^-$  (Electrochemical reaction)
  3.  $M_nS + 3H_2O \rightarrow nM + SO_3 + 6H^+ + 6e^-$  (Electrochemical oxidation)
- Multi-site sorption of H<sub>2</sub>S, n is a function of total sulfur coverage ( $\theta_S$ )
  - $n \rightarrow 1$  as  $\theta_S \rightarrow 1$ ,  $n \rightarrow N$  as  $\theta_S \rightarrow 0$
- Near OC, M<sub>n</sub>S can re-convert to M<sub>n</sub>-H<sub>2</sub>S ( $E_2 = 0.14$  V), and H<sub>2</sub>S can desorb for partial recovery
- At a high anode overpotential ( $E_3 = 0.89$  V), M<sub>n</sub>S can oxidize to SO<sub>3</sub>, SO<sub>3</sub> assumed completely soluble in water and removed from the system

# NH<sub>3</sub> Poisoning Model

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## Experimental Observations

- Reversible at low concentrations but may be irreversible at high concentrations (Uribe 2002)
- CV traces suggest that NH<sub>3</sub> is not significantly adsorbed on the electrocatalyst (Uribe 2002 & Soto 2003).
- AC impedance spectroscopy data indicate that increased membrane resistance cannot account for the observed overpotentials due to NH<sub>3</sub> impurity (Soto 2003).

## Model

- Transient stack model with steady-state option
- NH<sub>3</sub> uptake in ionomer modeled as a reversible absorption-desorption reaction
- Same approach for NH<sub>3</sub> uptake in membrane except it is exposed to both anode and cathode streams
- Effect of NH<sub>4</sub><sup>+</sup> on conductivity from experimental data
- Postulated effect of NH<sub>4</sub><sup>+</sup> on ORR kinetics

# HCHO/HCOOH/C<sub>7</sub>H<sub>8</sub> Poisoning Model

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## ■ Hydrogen Oxidation Reaction

- $\text{H}_2 + 2\text{M} \leftrightarrow 2\text{M-H}$  (Dissociative Adsorption)
- $\text{M-H} \rightarrow \text{M} + \text{H}^+ + \text{e}^-$  (Electrochemical Oxidation)

## ■ Poisoning of Pt

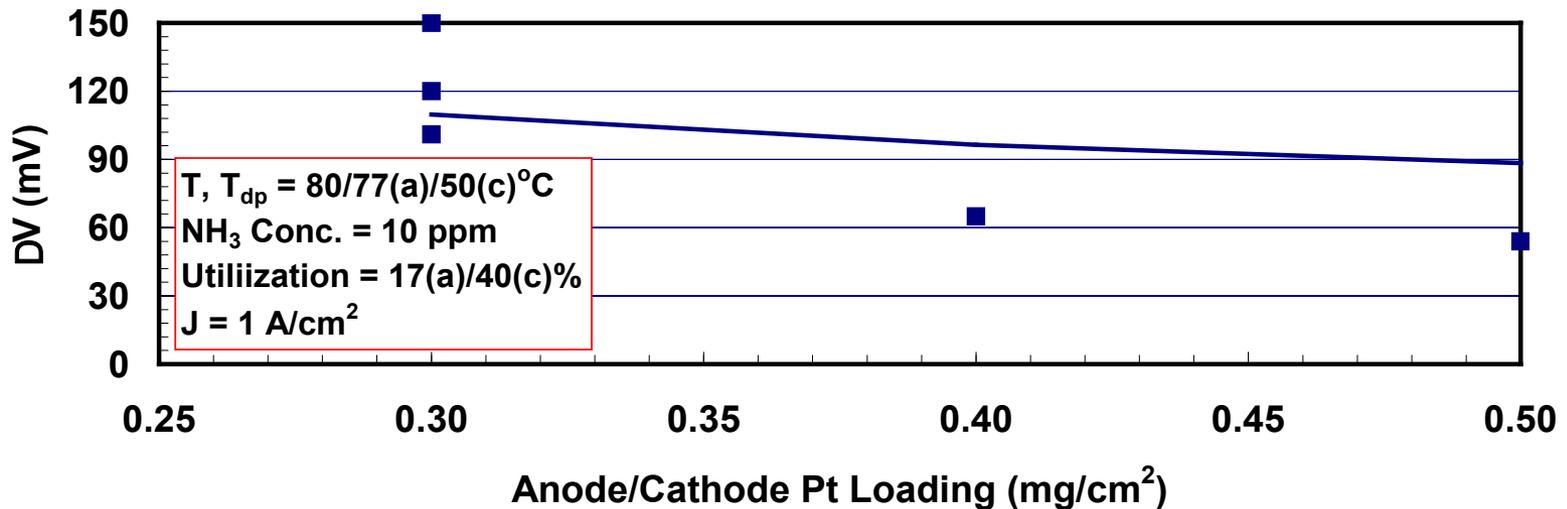
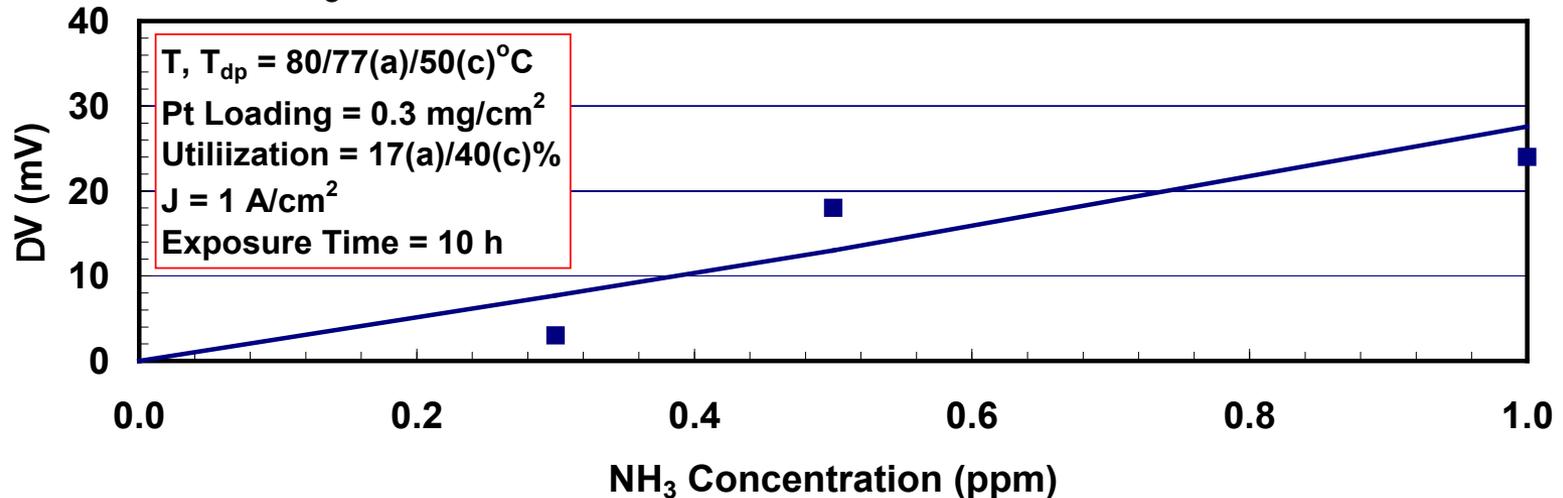
- $\text{HCHO} + \text{M} \rightarrow \text{M-CO} + 2\text{H}^+ + 2\text{e}^-$  (Dissociative Adsorption)
- $\text{HCOOH} + \text{M} \rightarrow \text{M-CO} + \text{H}_2\text{O}$  (Dissociative Adsorption)
- $\text{C}_7\text{H}_8 + \text{M} \leftrightarrow \text{M-C}_7\text{H}_8$  (Associative Adsorption)

## ■ Reaction with Hydrogen

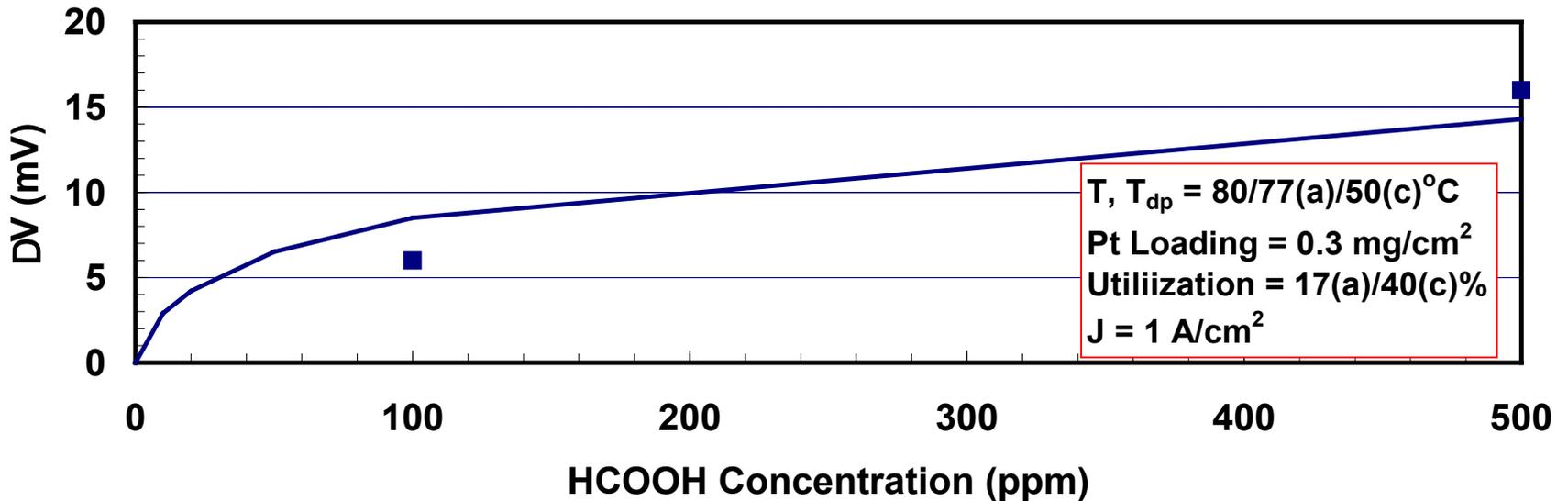
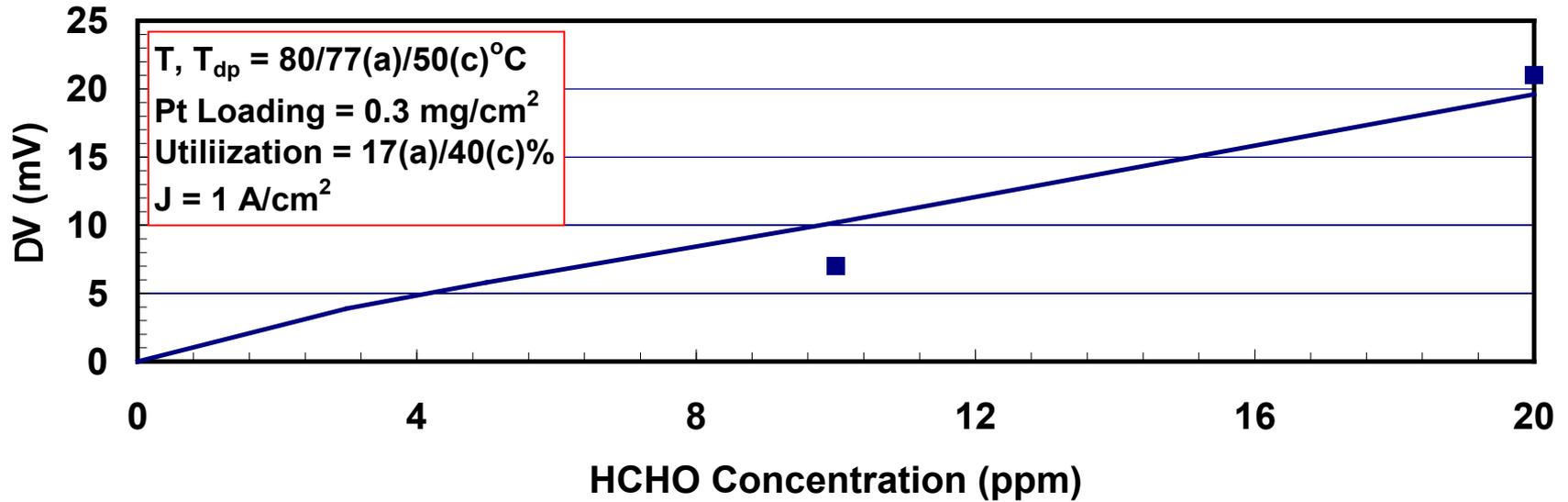
- $\text{C}_7\text{H}_8 + 6\text{M-H} \rightarrow 6\text{M} + \text{C}_7\text{H}_{14}$  (Hydrogenation to MCH)

# NH<sub>3</sub> Impurity Model Development (JARI Data)

- Modeled NH<sub>3</sub> crossover is a function of uptake and current density

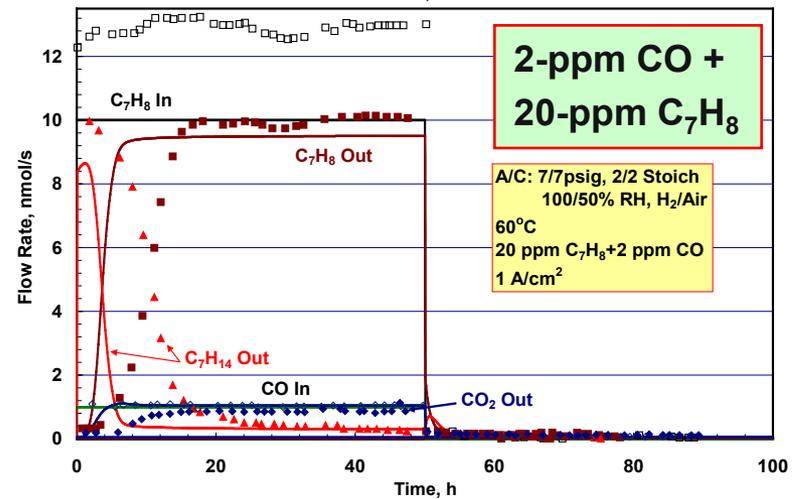
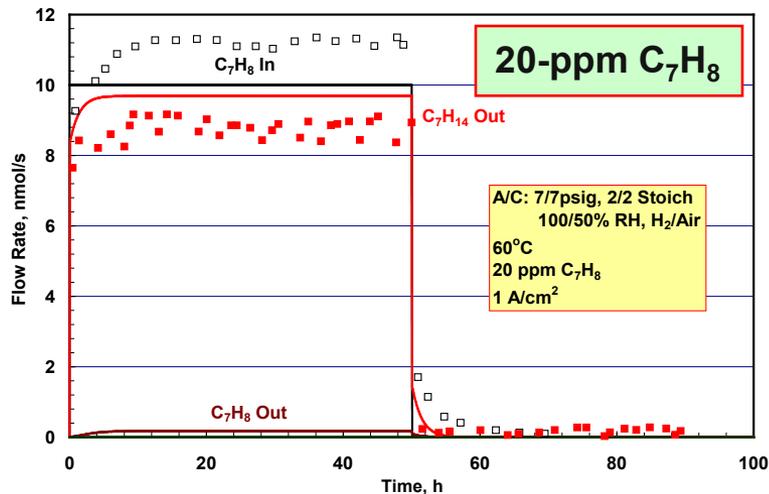
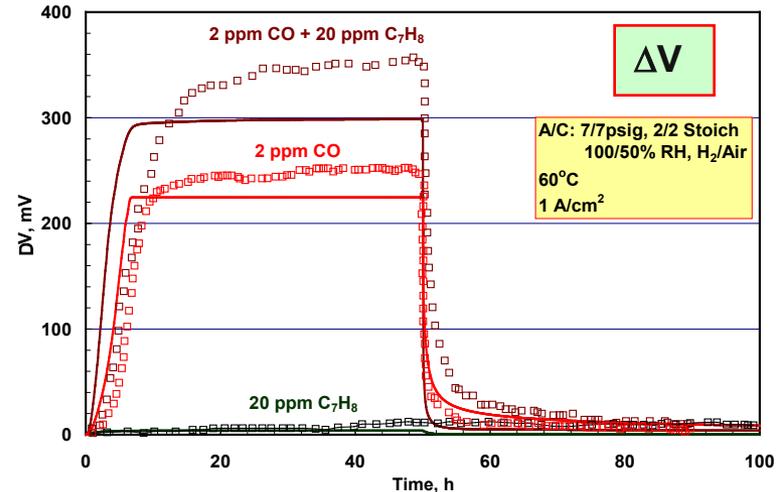
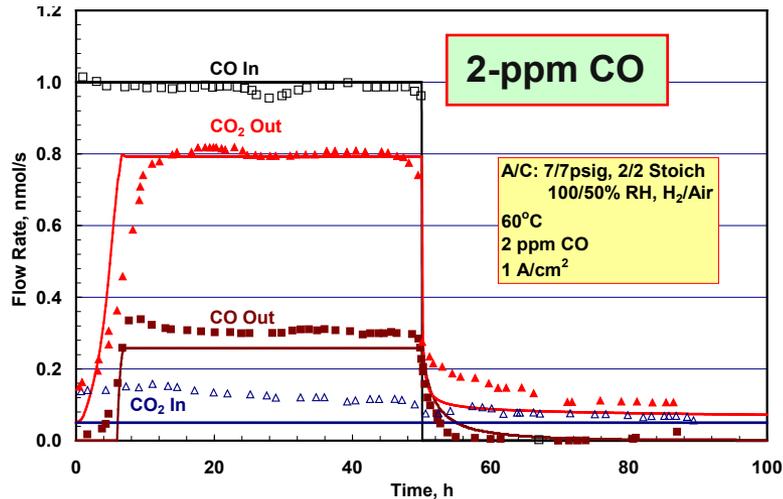


# HCHO/HCOOH Impurity Model Development: JARI Data



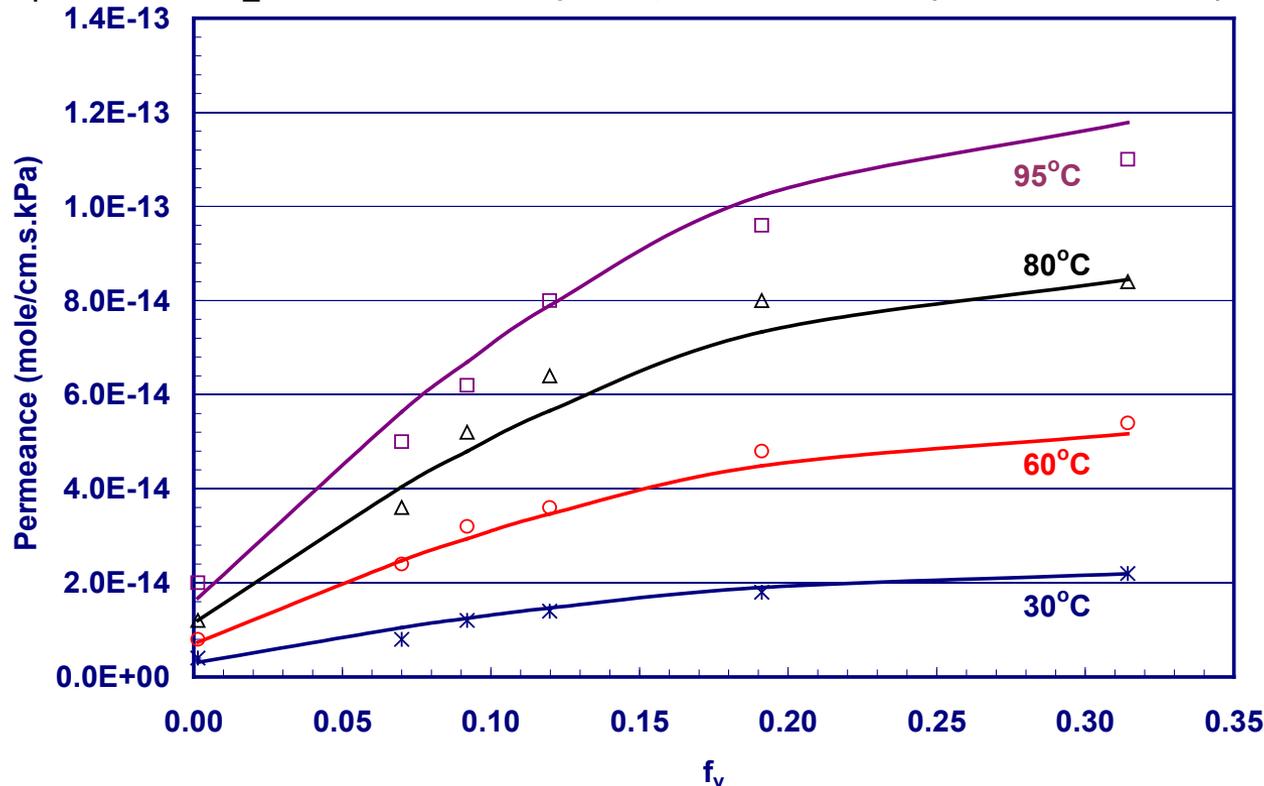
# Analysis of HNEI Data: Toluene

- Toluene alone: Small  $\Delta V$ , significant conversion to methyl-cyclohexane
- Toluene + CO: Larger  $\Delta V$ , insignificant hydrogenation



# Effect of Inerts (N<sub>2</sub>, He, CH<sub>4</sub>)

- Giner data for N<sub>2</sub> permeance as a function of T and membrane water content (f<sub>v</sub>)
  - N<sub>2</sub> crossover from cathode air may potentially exceed N<sub>2</sub> in H<sub>2</sub>
- He permeance from JARI data: 2.4x10<sup>-13</sup> mol/cm.s.kPa at 80°C, 77°C dew point, 1.6 higher than H<sub>2</sub> permeance
- CH<sub>4</sub> and CO<sub>2</sub> are relatively impermeable (HNEI, JARI)



# Reference Conditions, Assumptions, Definitions

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## 1. Pressurized stack

- 87 kWe at 1 A/cm<sup>2</sup> with pure H<sub>2</sub>
- 50% O<sub>2</sub> & 70% per-pass H<sub>2</sub> utilization
- N111 membrane, 0.2(c)/0.1(a) mg/cm<sup>2</sup> Pt loading
- Stack pressure: 2.5/1.9/1.6 atm at 100/25/5% power
- Inlet RH: 60/80/90% at 100/25/5% power

## 2. Purge criteria

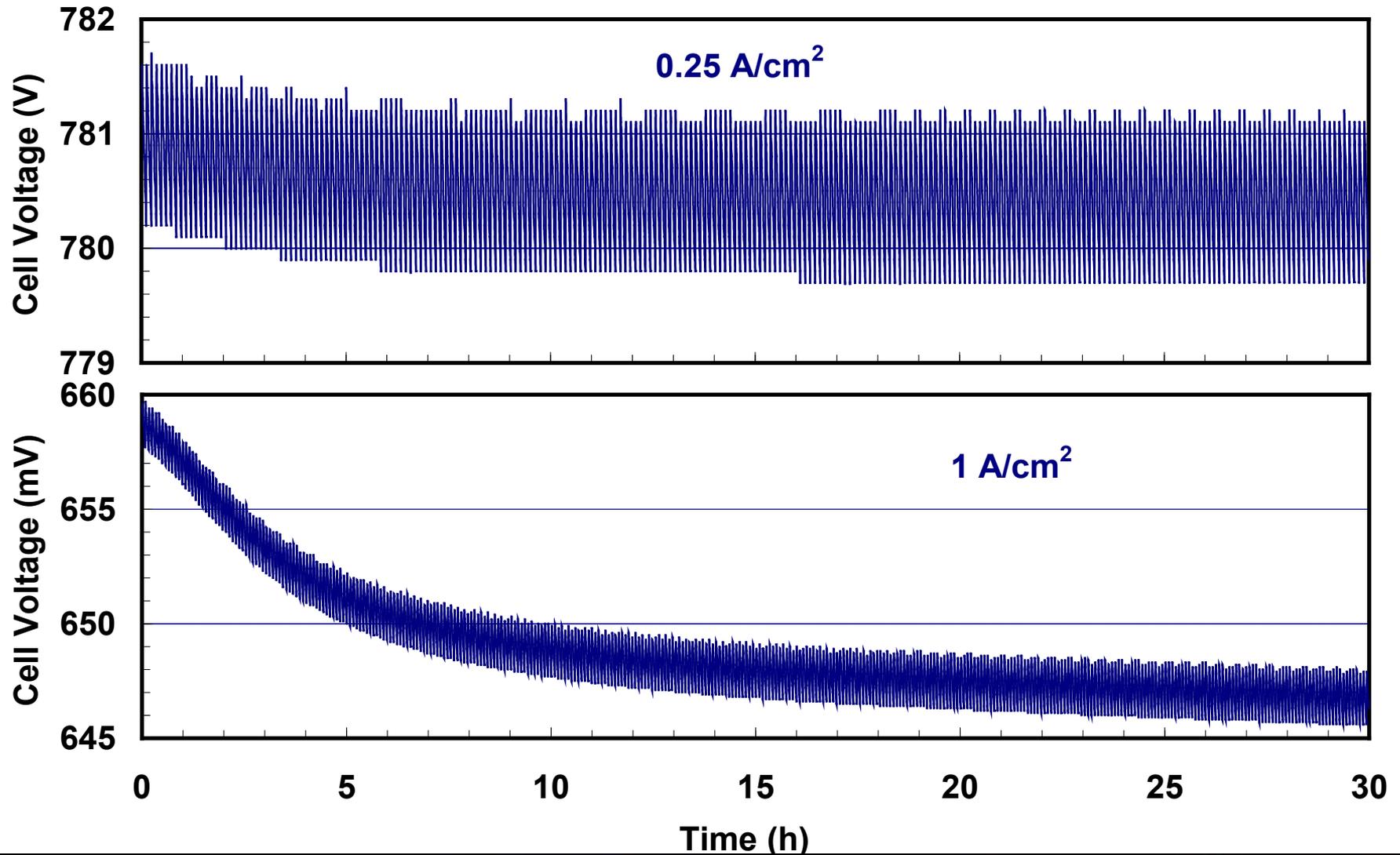
- Single purge (14 L) equivalent to 2 volumes of anode subsystem restores gas in anode channels to the H<sub>2</sub> fuel specs

## 3. Definitions

- Concentrations generally reported as max. values after 30-h exposure
- Stack efficiency defined as ratio of DC power produced to the LHV of H<sub>2</sub> consumed (chemically and electrochemically) and purged

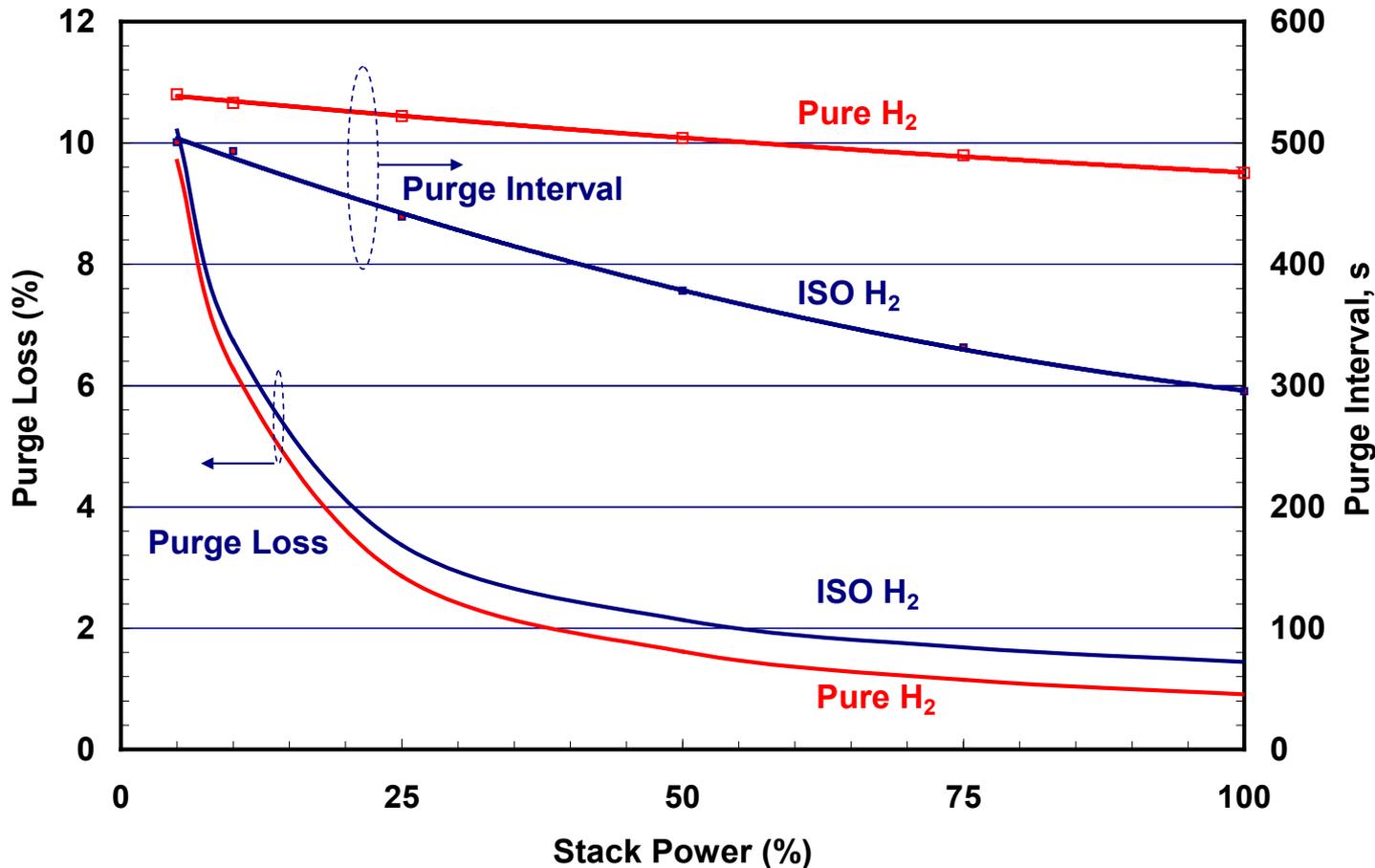
# Dynamic simulations of Impurity Effects

- Purge occurs whenever impurity concentration reaches 15%



# Purge Loss and Schedule

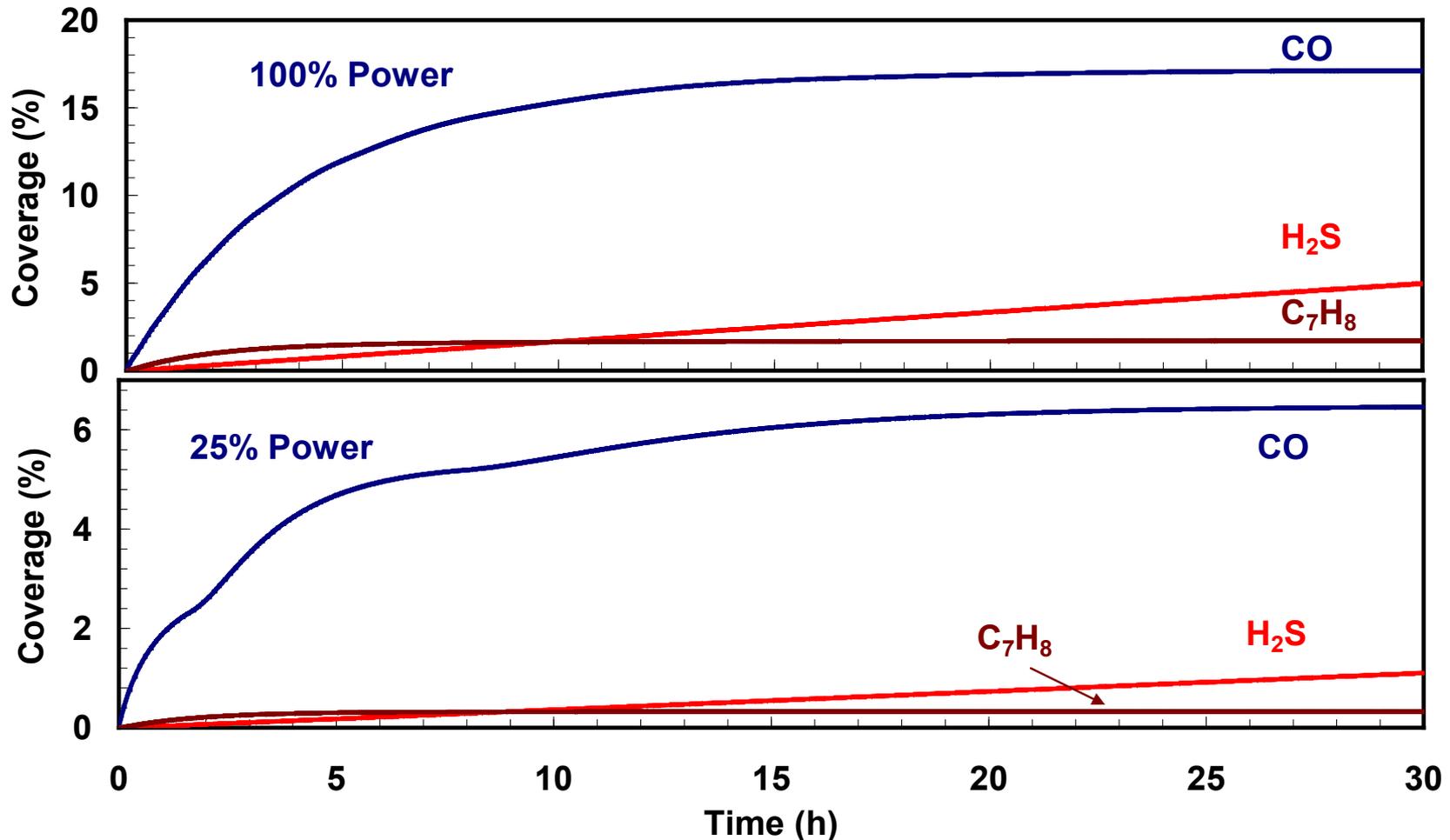
- At low power, purge interval determined primarily by N<sub>2</sub> crossover
- At high power, buildup of fuel impurities also affects purge interval



CO:	0.2 ppm
CO <sub>2</sub> :	2 ppm
N <sub>2</sub> :	100 ppm
He:	200 ppm
NH <sub>3</sub> :	0.1 ppm
HCHO:	10 ppb
HCOOH:	0.2 ppm
C <sub>7</sub> H <sub>8</sub> :	2 ppm
H <sub>2</sub> S:	4 ppb

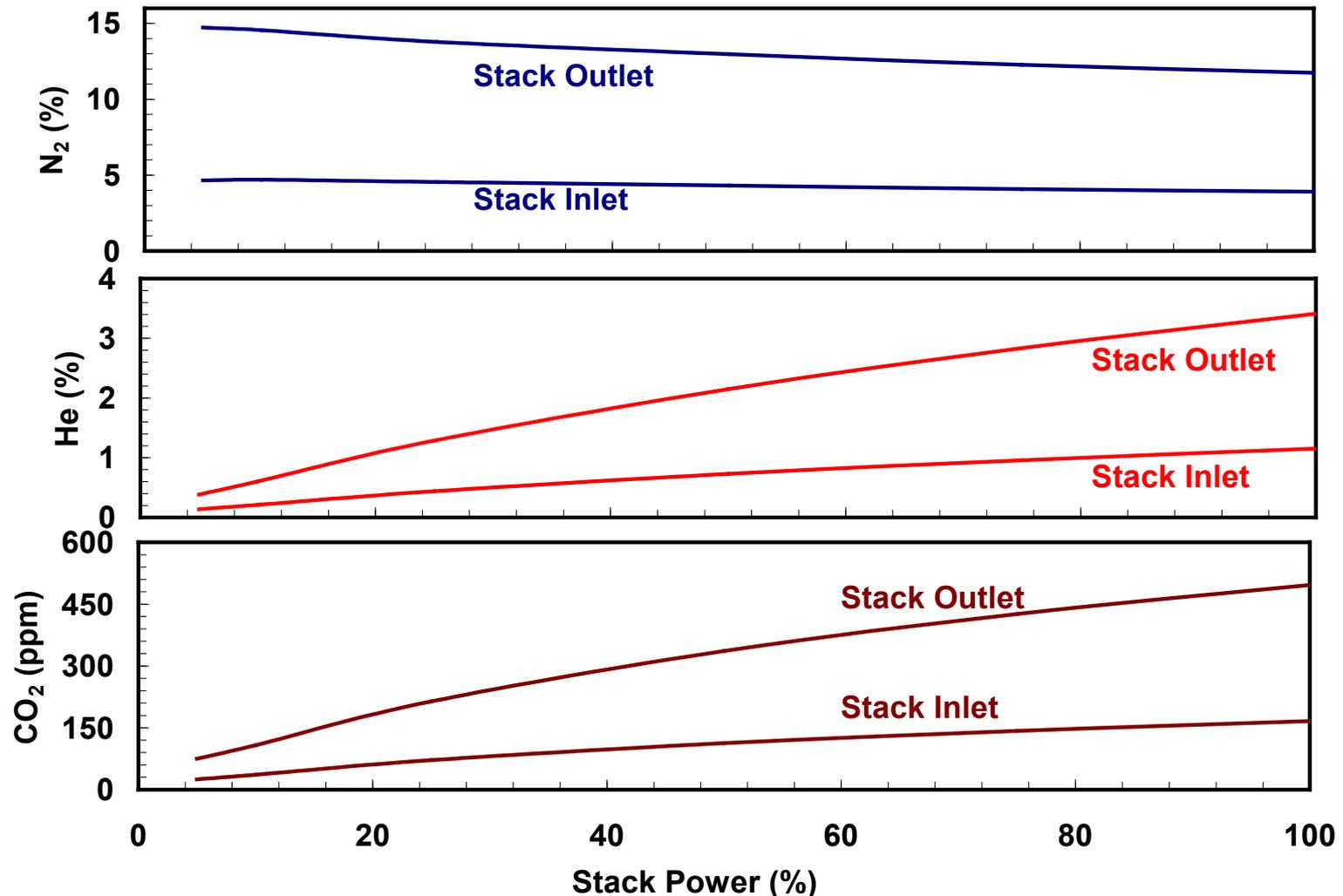
# Dynamic Pt Coverage

- Steady-state coverage of CO & C<sub>7</sub>H<sub>8</sub> functions of P & overpotentials
- No SS coverage for H<sub>2</sub>S which competes with CO & C<sub>7</sub>H<sub>8</sub> for vacant sites



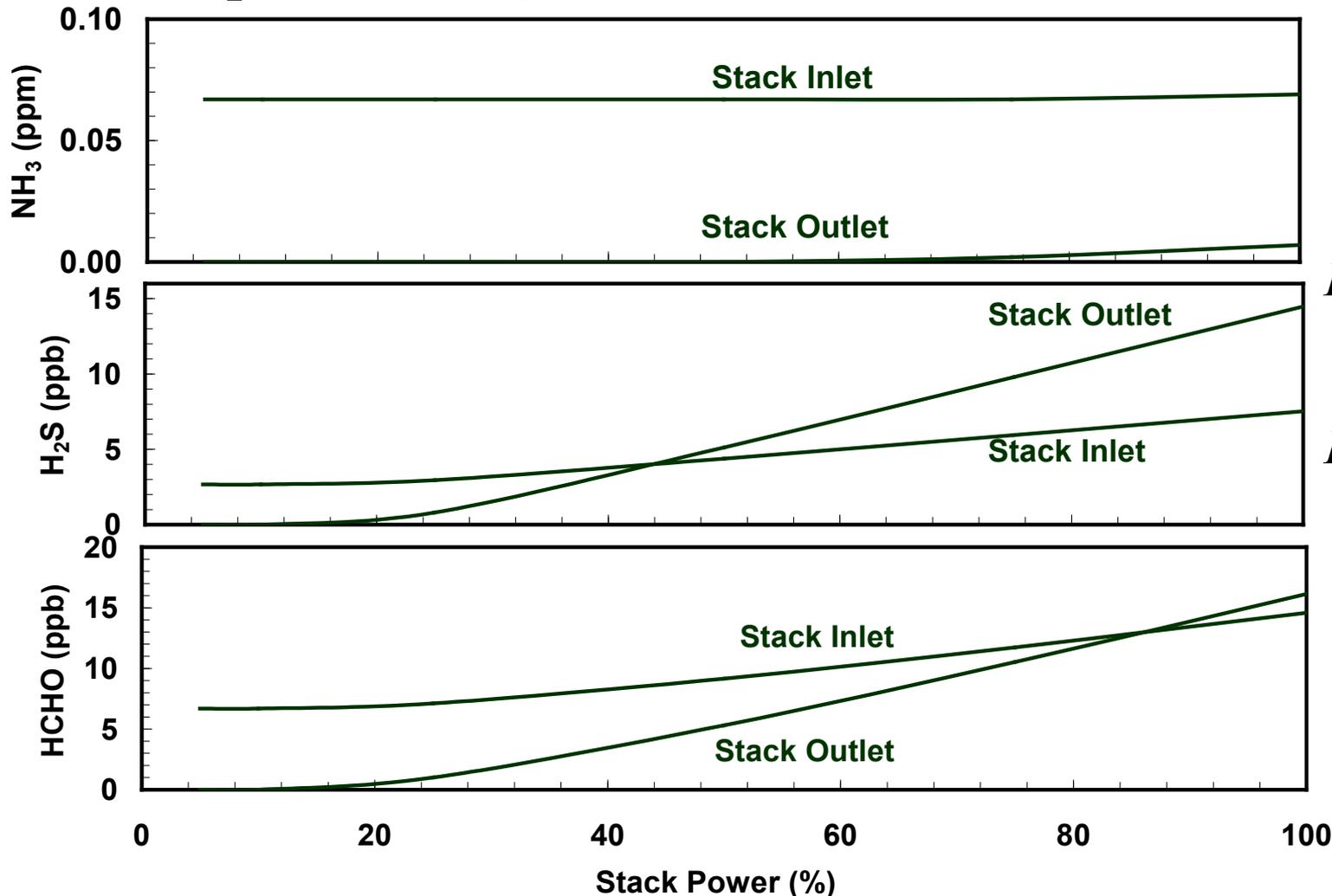
# Buildup of Impurities with Dynamic Purge: N<sub>2</sub>, He, CO, CO<sub>2</sub>

- N<sub>2</sub> buildup almost entirely due to N<sub>2</sub> crossover from cathode
- No buildup of CO as it is quantitatively converted to CO<sub>2</sub>



# Dynamic Buildup of NH<sub>3</sub>, H<sub>2</sub>S and HCHO

- No buildup of NH<sub>3</sub>, NH<sub>3</sub> crossover increases with current density
- H<sub>2</sub>S breakthrough at more than 20% power, >30 h exposure



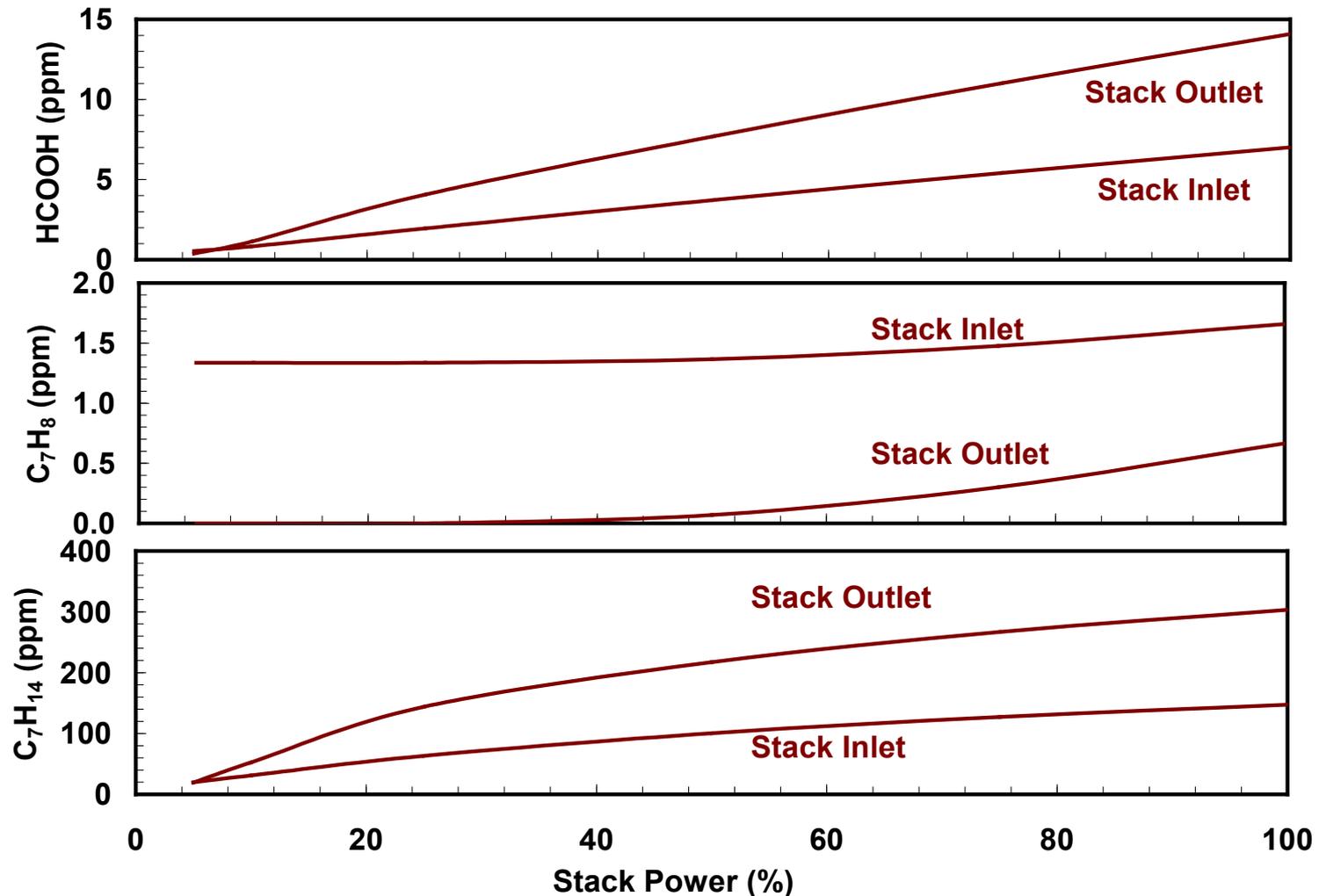
Enrichment

$$E_i = \frac{C_i^{out}}{C_i^{in}}$$

$$E_i = \frac{(1 - \Phi_i)}{(1 - \Phi_{H_2})}$$

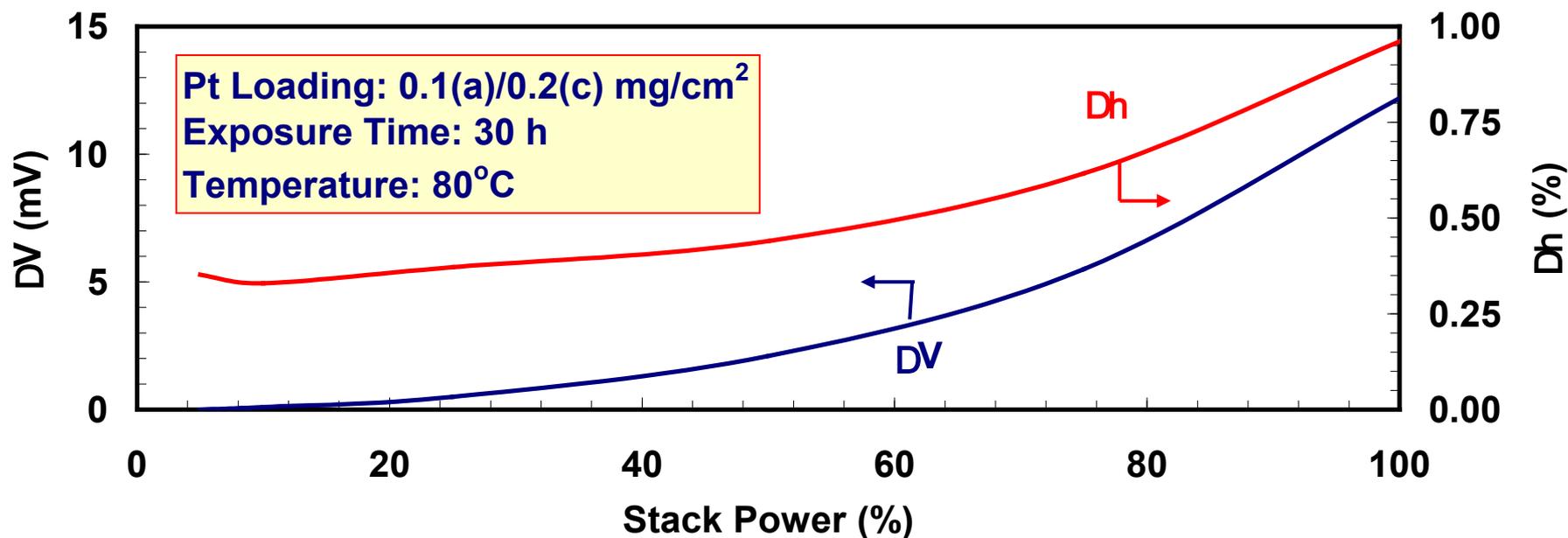
# Dynamic Buildup of HCOOH and C<sub>7</sub>H<sub>8</sub>

- Slower dissociative adsorption of HCOOH than HCHO
- Conversion of toluene to methyl-cyclohexane slows at higher power



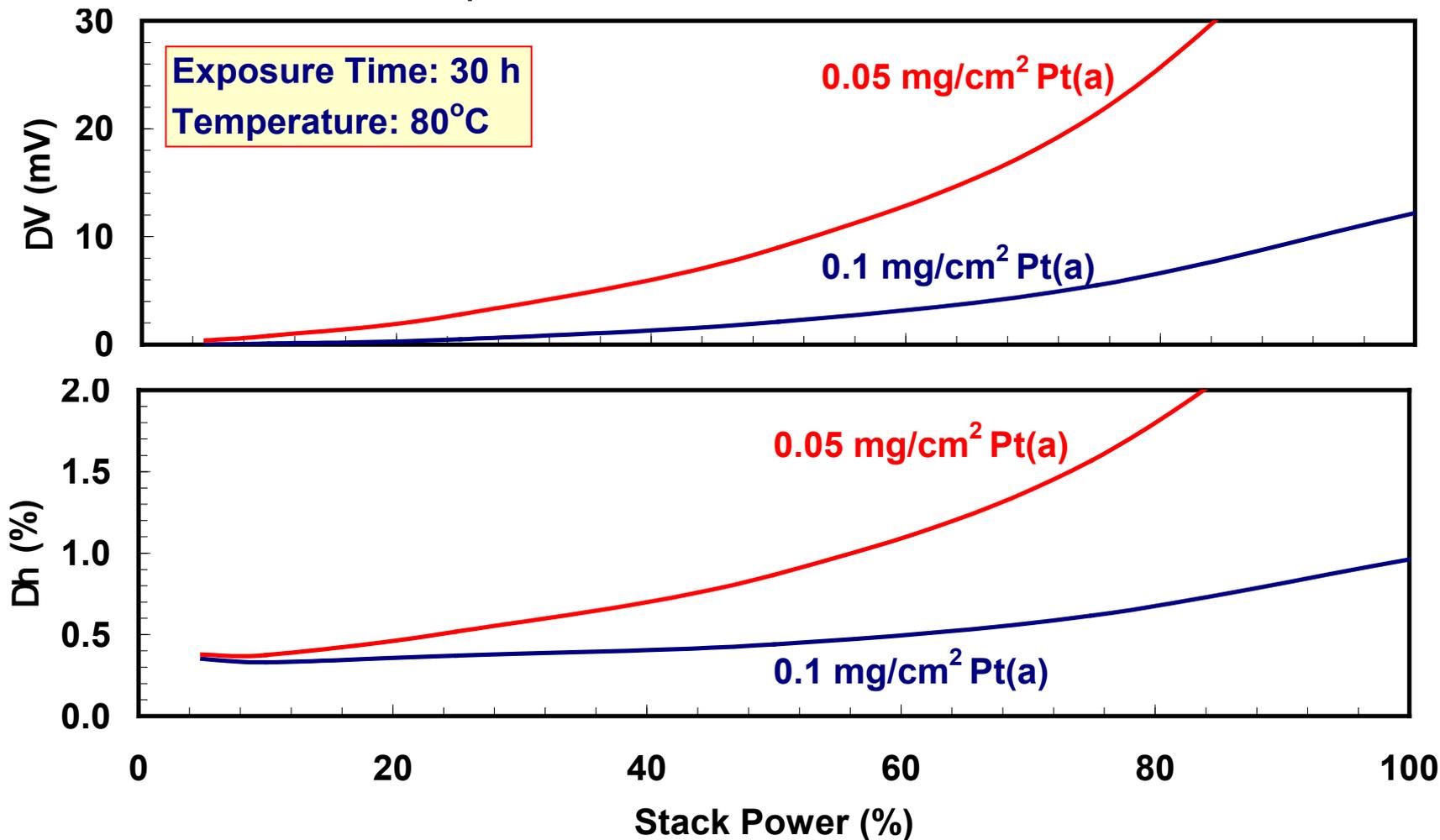
# Simultaneous Effect of Impurities

- Losses defined with respect to cell voltage and stack efficiency for pure fuel hydrogen accounting for  $N_2$  crossover from cathode
- At <30% power,  $\Delta V$  approaches zero but 0.35 percentage point reduction in stack efficiency due to buildup of impurities
- Slow increase in  $\Delta V$  expected at longer exposure because of  $H_2S$  uptake



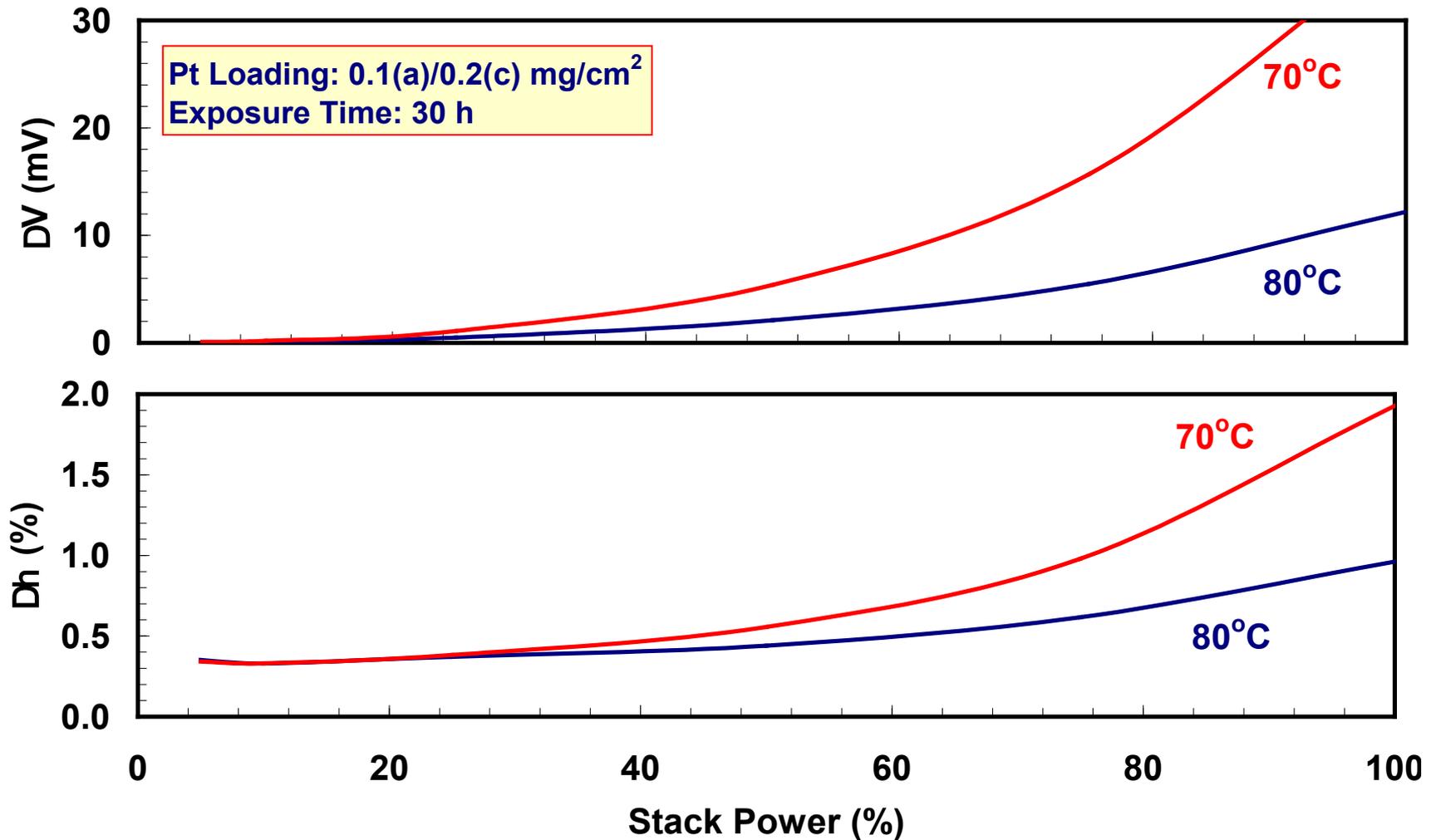
# Effect of Anode Pt Loading

- Substantially higher  $\Delta V$  and  $\Delta \eta$  with 0.05 mg/cm<sup>2</sup> Pt (a) for >50% stack power



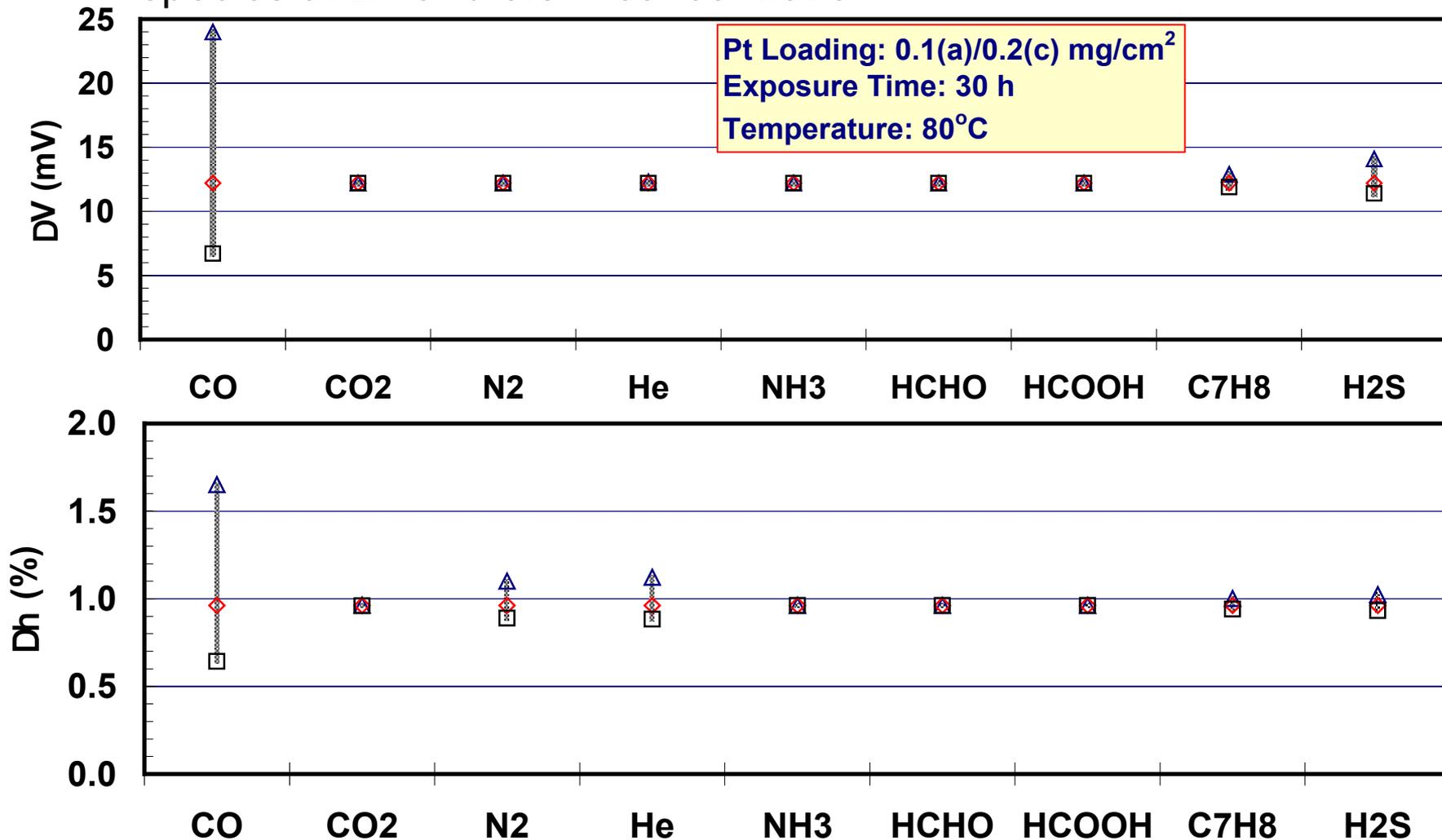
# Effect of Stack Temperature

- 70°C stack temperature, 1.5 bar inlet pressure at rated power



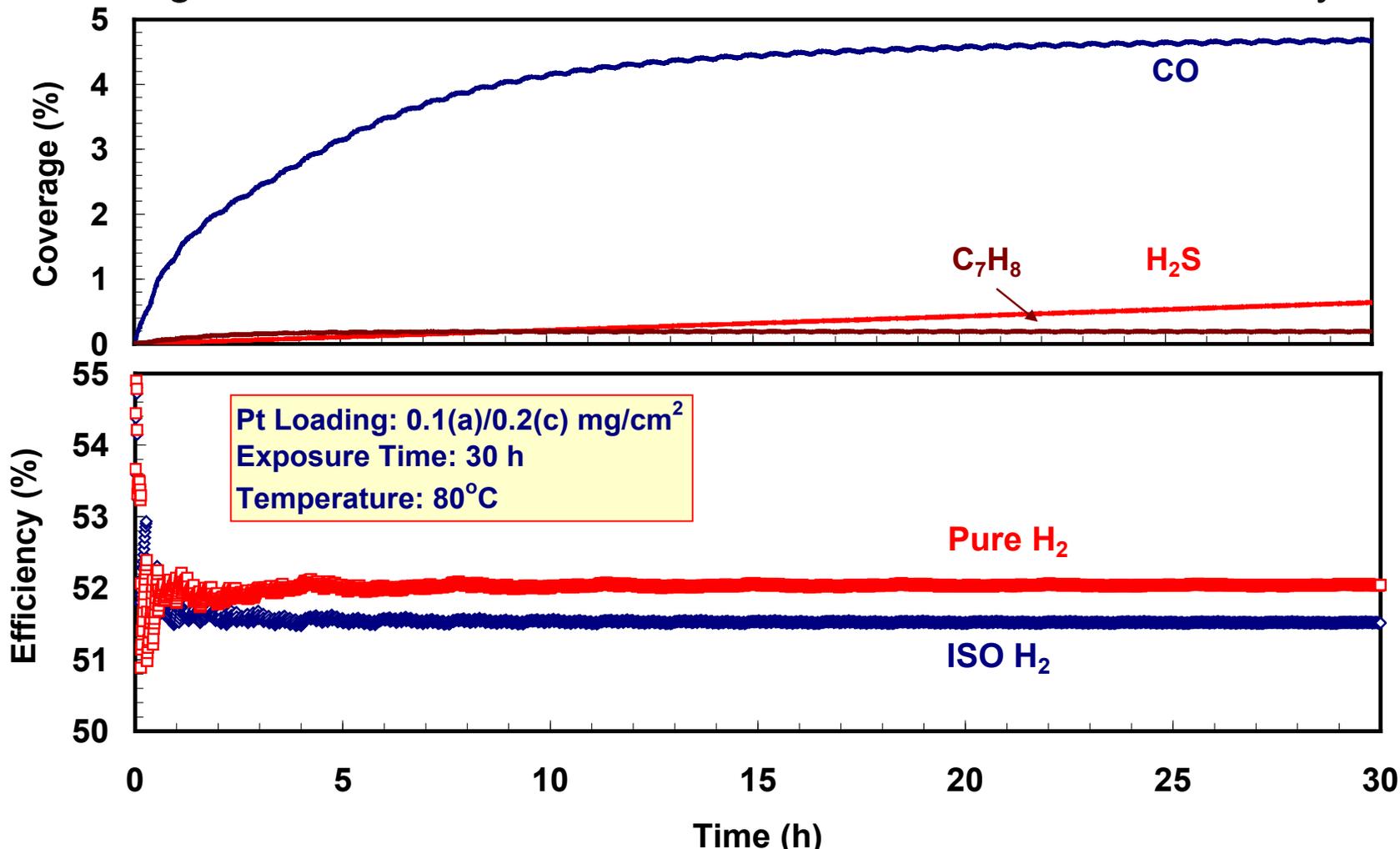
# Sensitivity Analysis

- Results for 100% stack power, ISO max impurity specs with one species at 2X and 0.5 X concentration



# Drive Cycle Simulations

- Multiple repeats of warm FUDS (1372 s) and FHDS (740 s) for 30 h
- Insignificant differences in VI curve for the last FUDS-FHDS cycle



# Summary and Conclusions

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- $\text{N}_2$  crossover from air is the main source of  $\text{N}_2$  buildup in the anode recycle stream and determines the purge schedule
- Dynamic simulations show that  $\text{NH}_3$  cannot accumulate in the anode recycle stream
  - Significant crossover of  $\text{NH}_3$  to cathode air
  - Cumulative degradation in performance due to  $\text{H}_2\text{S}$
- Cyclic buildup of formaldehyde, formic acid and methyl-cyclohexane
- Critical data needs
  - $\text{CO}/\text{CO}_2$ :  $\text{H}_2$  pump data for independent verification of chemical vs. electrochemical  $\text{CO}$  oxidation
  - $\text{NH}_3$  crossover as function of current density,  $\text{NH}_3$  isotherms as functions of RH and T
  - $\text{H}_2\text{S}$ : Isolation of anode vs. cathode overpotentials
  - Effect of T on behavior of  $\text{HCHO}$ ,  $\text{HCOOH}$  and  $\text{C}_7\text{H}_8$