Microchannel Reformate Cleanup:
Water Gas Shift and Preferential Oxidation

2004 DOE Hydrogen, Fuel Cells & Infrastructure Technologies Program Review

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Objectives

- Overall Objectives
  - Apply microchannel architectures where appropriate in fuel processing for transportation, stationary, and portable applications to reduce size and weight, improve fuel efficiency, and enhance operation.
  - Develop a prototype microchannel-steam-reforming fuel processor at 2 kWe scale that will meet DOE performance targets when scaled up to 50 kWe.

Performance Criteria	2004 Projected Performance	2004 Demo Target
Cold (20C) Start Time	12 sec, reformer only	<60 s to 90%
Start up Energy	<7 Mj (a)	2 MJ
Power Density	2307 W/L (a)	700 W/L
Efficiency	78%	78%
Durability	>1000 hr	2000 hr

(a) based on individual components only, excludes tube, duct, insulation etc.

- Task Objectives
  - Demonstrate **90%** CO conversion in a single-stage WGS reactor that scales to less than **3** liters at full-scale
  - Determine whether microchannel architecture provides opportunities for size and weight reduction for PROX reactor

Budget

- Continuing Project, $119K of Carryover Funding
- FY04 Funding Level $700K, allocated:
  - $300K for WGS development (this poster)
  - $400K funding for reforming activities (presentation)

Technical Barriers and Targets

- Fuel-Flexible Fuel Processor Technical Barriers Addressed
  - L. Hydrogen Purification/Carbon Monoxide Clean-up
  - M. Fuel Processor System Integration and Efficiency

- Key DOE Technical Targets Addressed:
  - CO Content in Product Stream
    - (< 10 ppm steady-state, < 100 ppm transient)
  - Power Density
    - (> 800 W/L)
  - Specific Power
    - (> 800 W/kg)

Approach

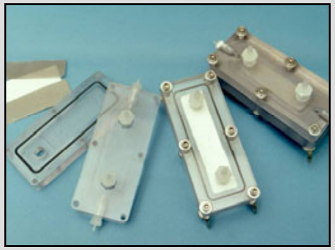
- Compact size and weight to meet packaging requirements
  - rapid heat and mass transfer for high hardware productivity
- Thermal management
  - high heat transfer effectiveness in heat exchangers and reactors for maximum heat utilization and high fuel efficiency
- Water management
  - compact and efficient air-cooled partial condensers
- Rapid start-up
  - imbedded heat transfer in reactors facilitates rapid heating
- Cost
  - improved productivity of precious metal catalysts

Project Safety

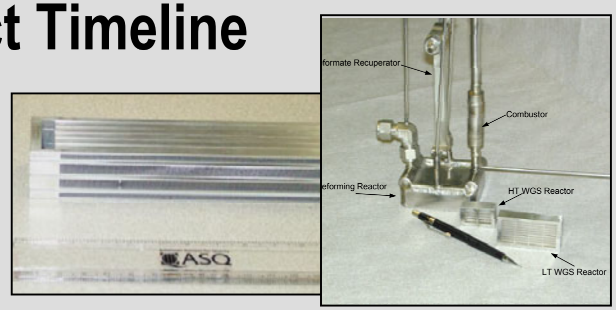
- Training of staff on laboratory-specific hazards is assured by limiting access to the lab through IOPS (Integrated Operation System)
- Standard operating procedures for all experimental systems
  - Reviewed and approved by ES&H representatives, a cognizant space manager for the lab space, and a building manager prior to work beginning.
  - Identifies hazards, required personal protective equipment, and operational constraints.
- The microchannel architecture, due to low internal volume, minimizes inventories of flammable gas.

CO Cleanup / Balance of Plant Project Timeline

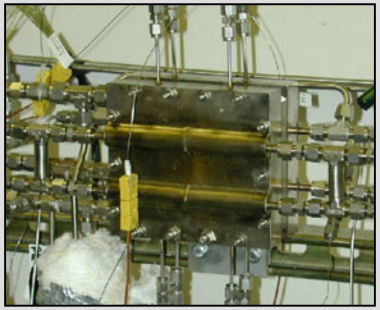
FY 2000
Invented phase separator



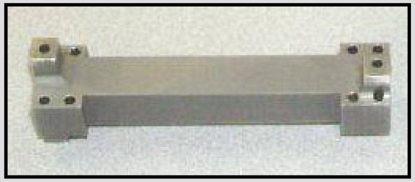
FY2001
Demonstrated partial condenser with phase separator

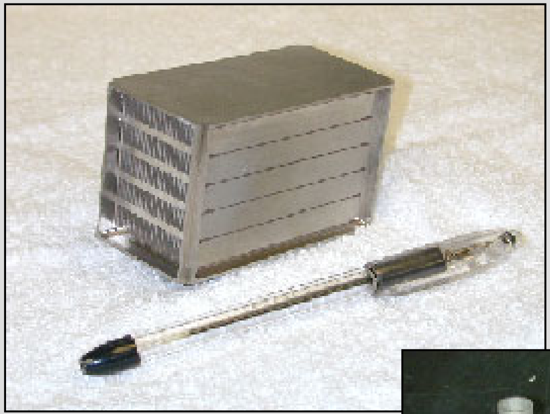


FY 2002
WGS/Prox catalyst studies
WGS reactor concept
Initial SR/WGS/Prox integration
Air-cooled condenser



FY 2003
2 kWe PROX reactor
Multi-channel WGS demo





FY2004
2 kWe differential temp. WGS reactor
5 kWe phase separator built for NASA

Engineered catalyst, multi-channel reactor development

Demonstrate WGS and PROX at 2 kWe

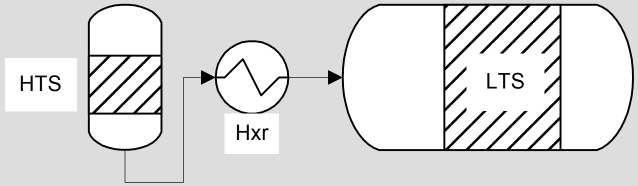
Integrated fast-start reformer / fuel cell demonstration at ~2 kWe

Collaborate with industrial partner(s) on manufacturing, field testing, lifetime, controls

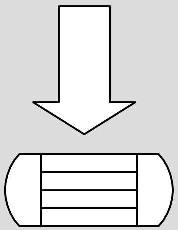
2003 2004 2005 2006 2007 2008

Differential Temperature Water Gas Shift Reactor
Approach

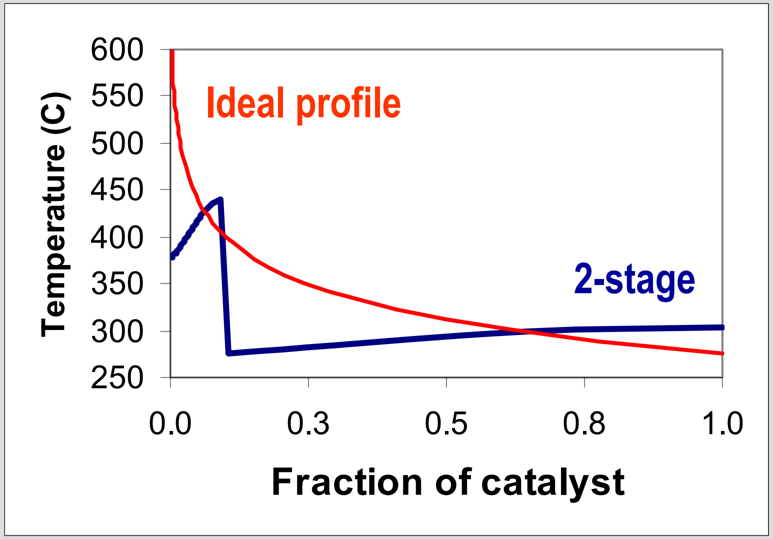
- Objective:
  - 90% Conversion single-stage WGS reactor < 3 liters at full-scale
- Approach:
  - Precious metal catalyst for high activity
  - Integrate microchannel heat exchange for temperature control
  - Optimize thermal profile
  - Reduce catalyst loading by up to 1/2
- Relevance
  - Smaller size – higher power density and specific power
  - Reduced cost
    - Improved catalyst efficiency
    - 3 devices collapsed into 1
  - Potential for higher energy efficiency



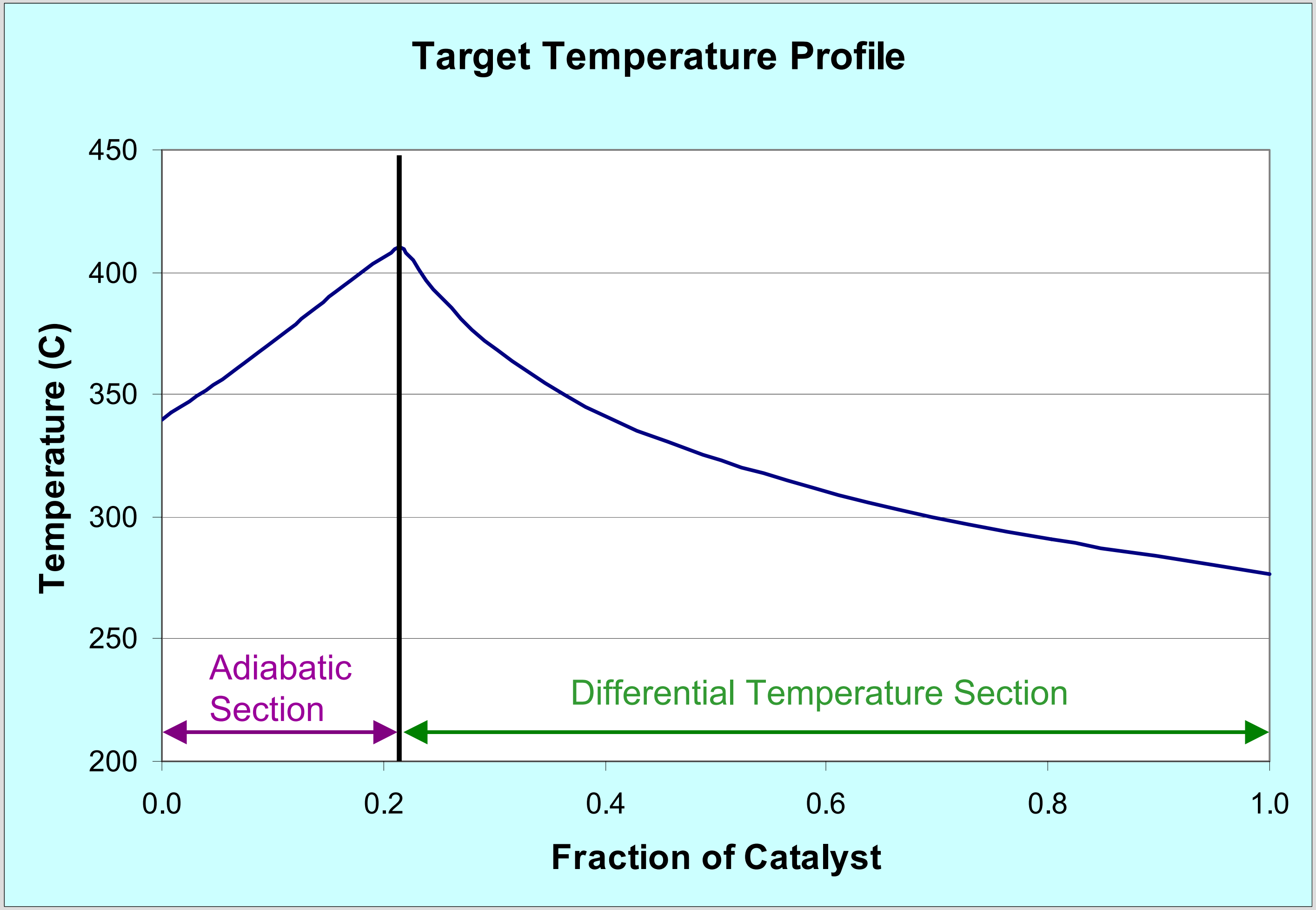
Conventional 2-stage Adiabatic



Ideal profile

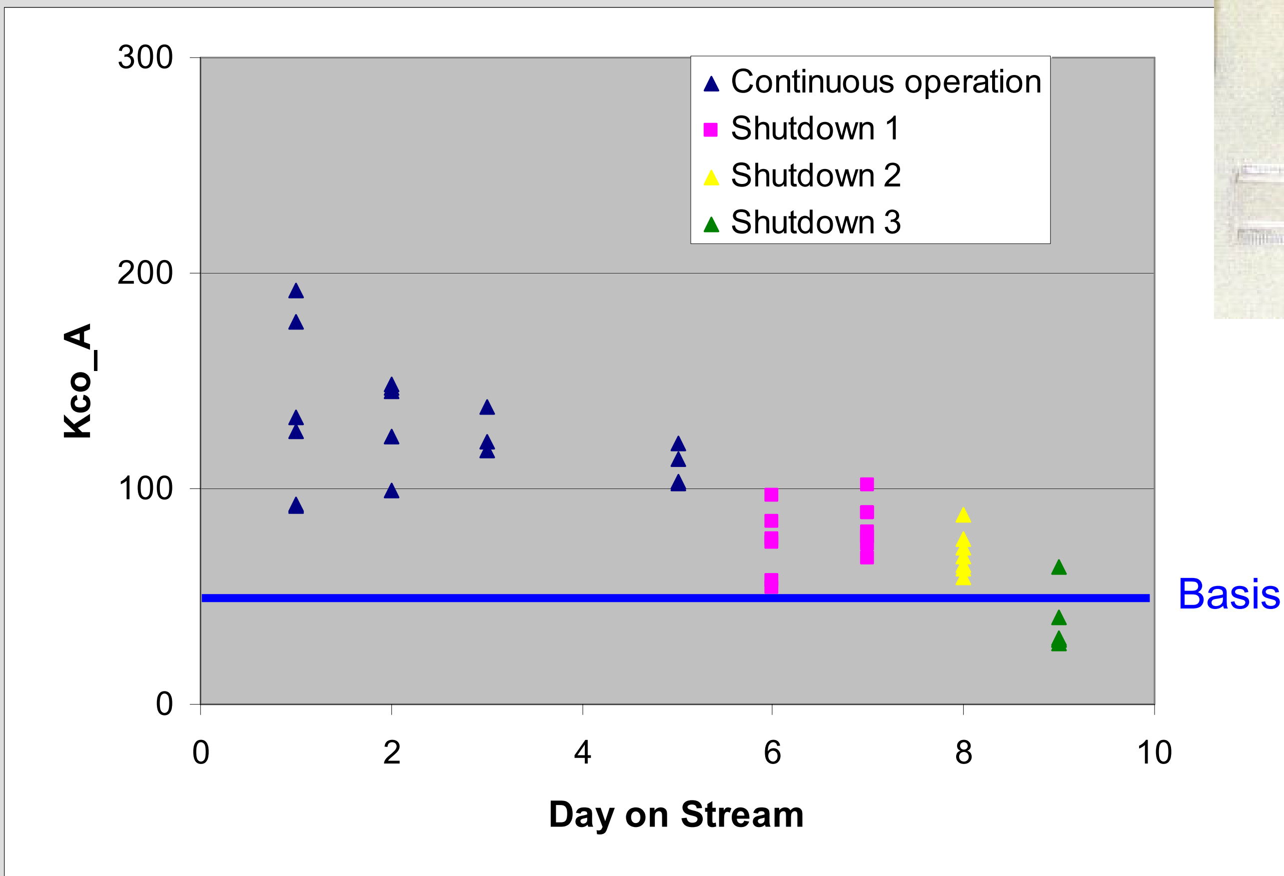


Differential Temperature Water-Gas Shift
New Reactor Concept



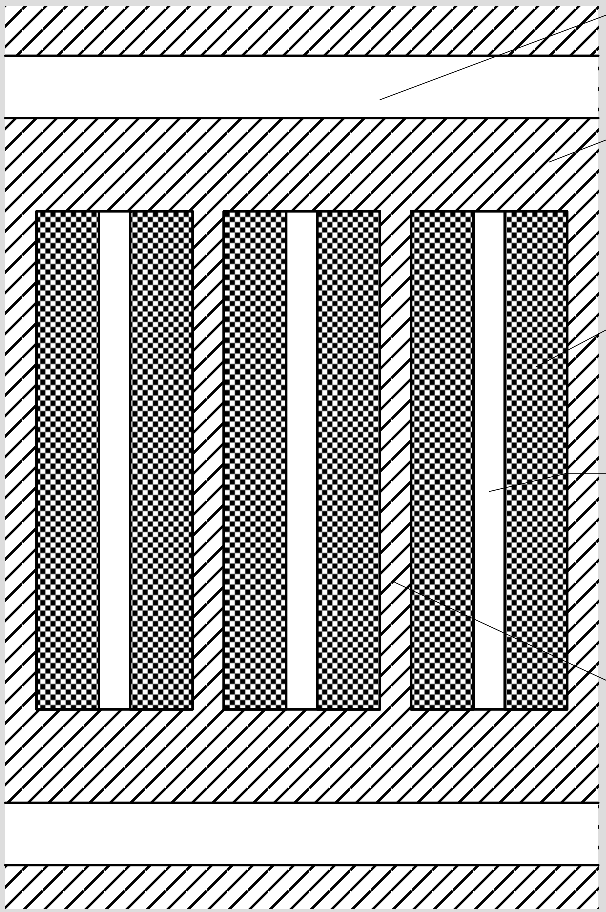
Water-Gas Shift
Extended Testing of Engineered PM Catalyst

$$r_{CO} = \rho_B \, k_{CO\,A} \exp(-k_{CO\,B}/T) \; p_{CO}^{-1} \; p_{H_2}^{-1} \; p_{CO_2}^{-1/2} \left(p_{H_2O} \, p_{CO} - \frac{p_{H_2} \, p_{CO_2}}{K_{eq}(T)} \right)$$



Basis for 2 kWe reactor design

WGS Differential Temp Reactor
Extended Area for Mass Transfer



- **Problem**
  - **WGS mildly exothermic**
  - **Interleaved approach gives too much heat transfer area**
  - **Reaction quenched prematurely**
- **Solution**
  - **Utilize concept of extended area in reaction channels**
- **Result**
  - **Heat exchange will support required heat flux**
  - **52% of reactor volume is catalyst**
  - **ΔT in fins < 0.1°C**

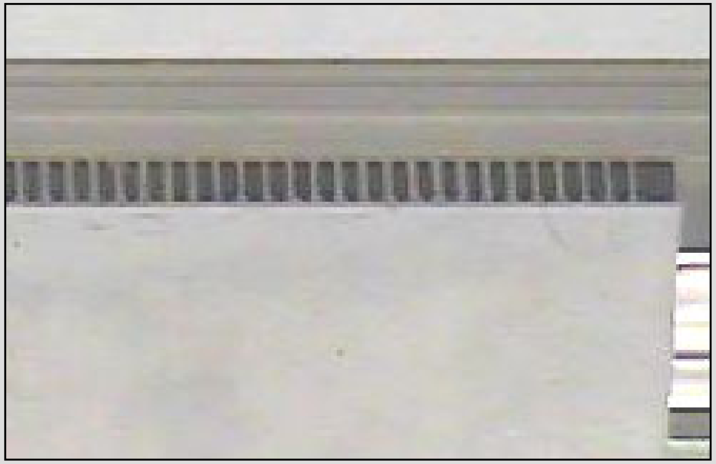
Water-Gas Shift 2 kWe Reactor
Design

Water-Gas Shift Differential Temperature Reactor
Comparison to Conventional 2-Stage Adiabatic

- Assumptions
  - 350 g/L catalyst loading onto a 600 cpi monolith
  - Same catalyst activity as used in differential temperature design
  - Negligible mass transfer resistance
  - Total volume 25% greater than catalyst volume
  - 5 W/cm^3 heat transfer power density in heat exchanger
  - Steam reformate with 3:1 original steam to carbon ratio
- 50 kWe scale
  - Differential temperature reactor volume – **4.1 Liters**
  - Conventional 2-stage adiabatic – **9.9 Liters (2.2X larger)**
    - High temperature stage – 0.8 Liter
    - Heat exchanger – 1.25 Liters
    - Low temperature stage – 7.8 Liters
    - Does not include piping between devices

Water-Gas Shift Differential Temperature Reactor
Manufacturability

- Methods for loading catalyst powder in situ are being developed under other programs
- Extrusion of microchannel arrays has been demonstrated



- Additional investments in low cost manufacturing techniques are required for adoption of the technology

Differential-Temperature WGS Summary

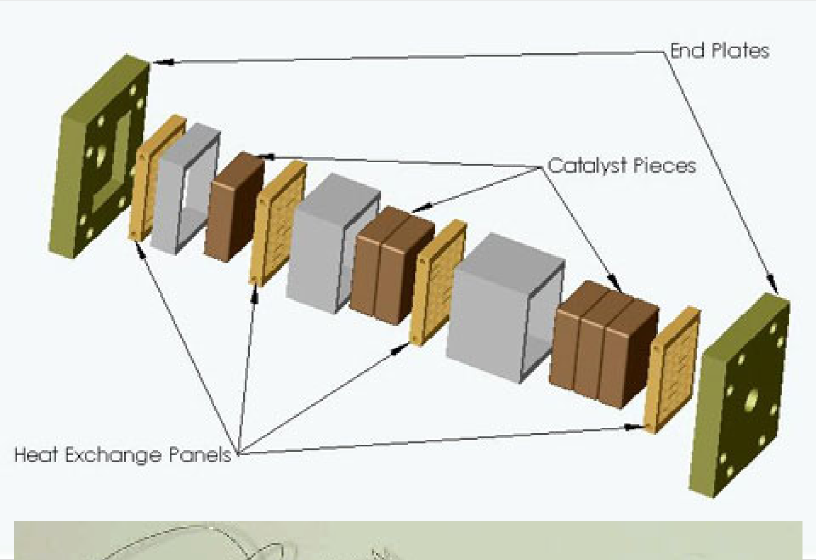
- Hybrid approach adopted to optimize size and weight
  - Adiabatic front end
  - Differential temperature back end
  - One single integrated device
  - Novel reactor concept gives high volumetric productivity
- Prototype 2 kWe reactor in development
  - Benchmark size and weight
  - To be integrated into a fuel processing system
- Full-scale 50 kWe WGS reactor projections:
  - Scaled from 2 kWe reactor design
  - 4.1 Liters (12,000 We/L)
  - 9 kg weight (5,600 We/kg)
  - 1.2 MJ to heat from ambient to operating temperature
  - Additional size reductions with increased catalyst activity

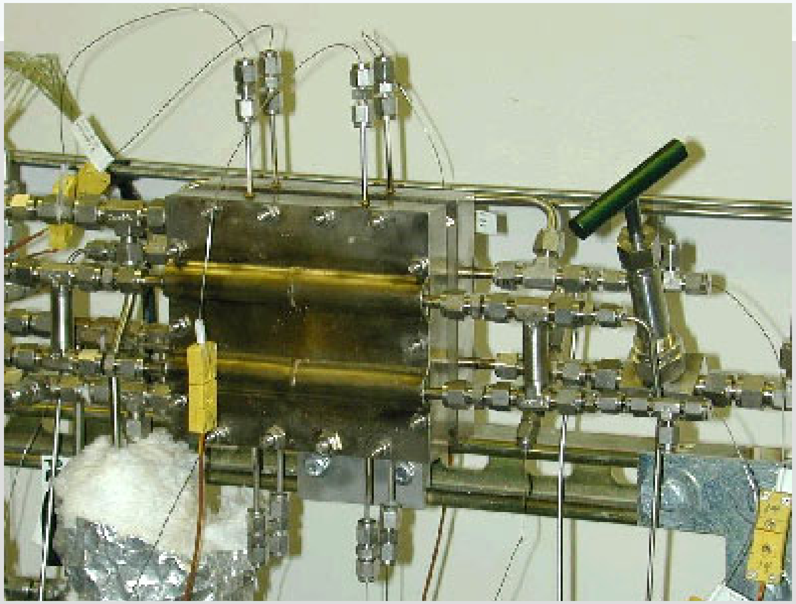
Microchannel PROX Reactor Investigations

- Objective: Determine whether microchannel architecture provides opportunities for size and weight reduction for PROX reactor.
- Approach:
  - ☑ Single-channel, isothermal catalyst tests
    - ☑ Evaluate industrial PROX catalysts
    - ☑ Characterize kinetics
    - ☑ Identify temperature and oxygen sensitivities
  - ☑ Design and test a 2 kWe PROX reactor
    - ☑ Demonstrate PROX functionality
    - ☑ Confirm favorable operational characteristics
    - ☑ Establish design basis for future prototype reactors
  - Investigate weight reduction by using low-density alloys (e.g., aluminum and titanium)
  - Design and build a prototype reactor optimized for size and weight
  - Investigate transient and startup characteristics
- Progress:
  - 1^{st} Stage PROX microchannel reactor demonstrated at 2 kWe scale – exhibits high productivity due to internal microchannel heat exchangers providing temperature control
  - Combined 1^{st} and 2^{nd} stage PROX reactor achieves 1% to 10 ppm CO reduction at a 2 kWe scale

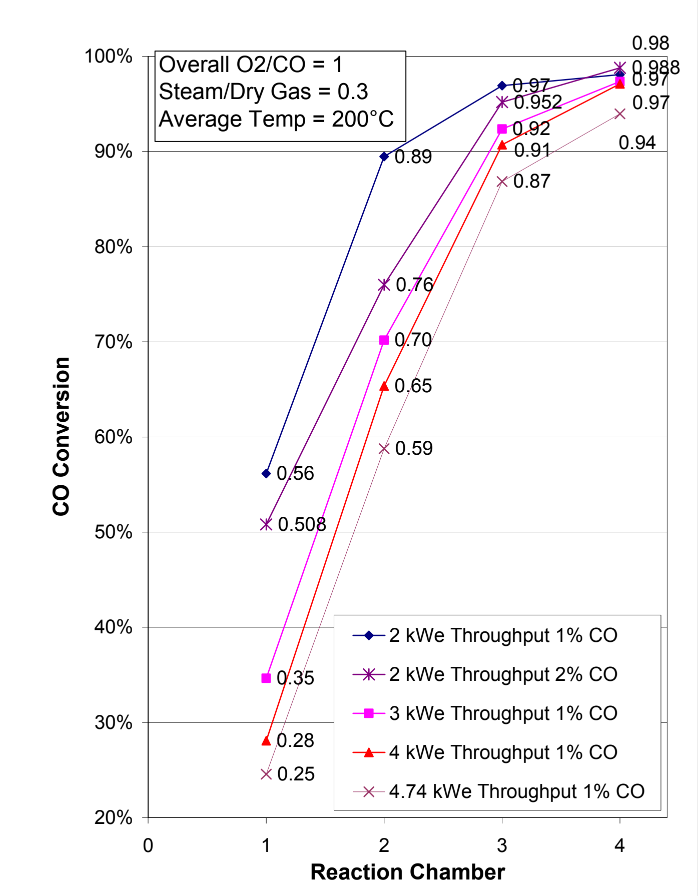
Microchannel PROX Reactor

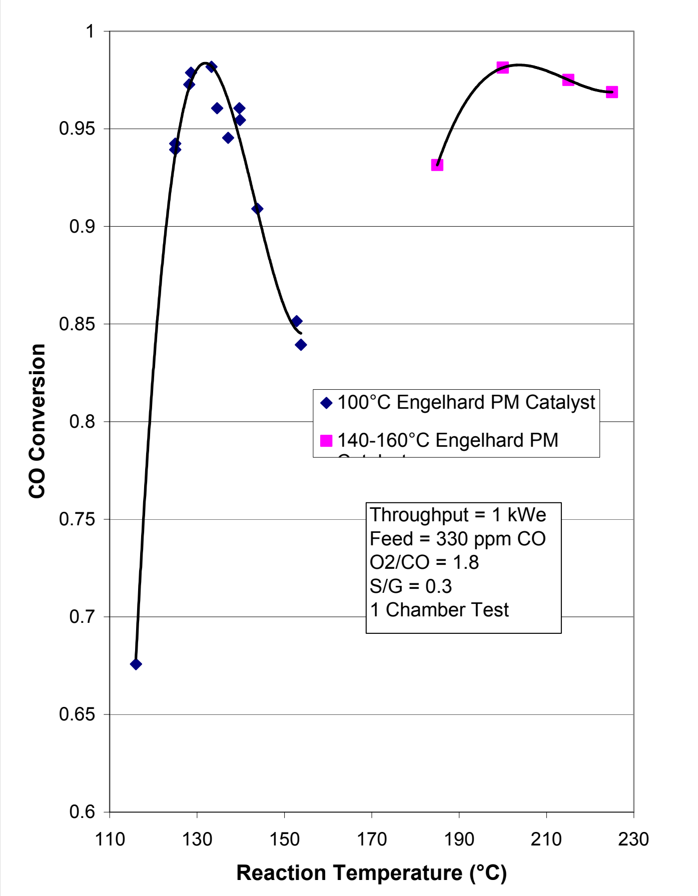
- Objectives of PROX Testing
  - Support fast kinetics
  - Minimize H_2 Oxidation
  - Maximize CO Conversion
- Approach Taken
  - Control temperature
    - Alternating catalyst pieces and microchannel heat exchangers
    - Conduction to remove heat from catalyst
  - Inject air within each chamber of the reactor
  - Adjust amount and type of catalyst in each chamber
- Reactor Design
  - Designed for 1st Stage PROX at 2 kWe throughput
  - Includes 4 Chambers with air bleed-in and sample measurement at each chamber





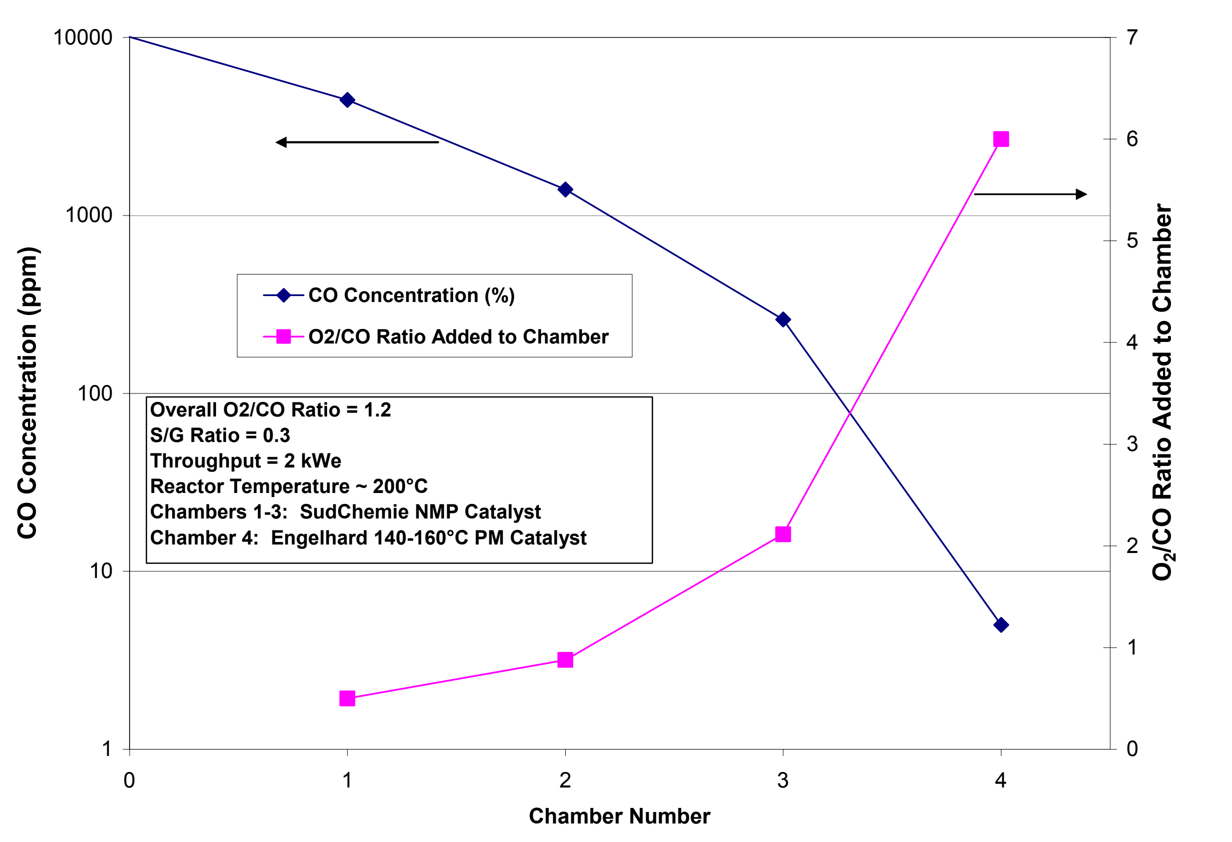
Results with 1st and 2nd Stage PROX





- First Stage (1% CO)
  - Sud Chemie NPM Catalyst Results
  - Air addition after each stage results in high conversions up to 4 kWe with 1 and 2% CO Feed
- Second Stage (330 ppm CO)
  - Engelhard PM Catalyst Results
  - 140-160°C 2nd Stage Catalyst operates at the same temperature as NPM 1st Stage Catalyst

Combined 1st and 2nd Stage PROX in Microchannel Reactor



- Results
  - Combined Sud Chemie NPM Catalyst and Engelhard PM Catalyst
  - Added higher ratios of O_2/CO in later chambers to increase conversion
  - Decreased CO from 1% to < 10 ppm at 2 kWe throughput with an overall O_2/CO ratio of 1.2 and a combined GHSV of 93,000

PROX Summary

- Future PROX work:
  - Evaluate aluminum and titanium alloys for microchannel PROX reactor
    - Potential weight reduction
  - Design a next generation 2 kWe reactor
    - Optimize size and weight
    - Facilitate heat integration within fuel processing system
    - Designed for rapid start and transient operation
  - Test as part of integrated fuel processor with steam reformer, water-gas-shift and heat exchangers
  - Couple to a PEM fuel cell

- No current scope for continuing microchannel PROX reactor development

Interactions

- Catalyst Development
  - Sud-Chemie, Inc. – WGS, PROX
  - Engelhard - PROX

- Technology Spin-off
  - R&D 100 award submitted for microchannel phase separator technologies.
  - NASA PEM Fuel Cell Interests
    - Microchannel Partial Condenser / Phase Separator successfully tested in zero gravity onboard NASA's KC-135 aircraft.
    - Microchannel phase separator built for NASA at 5-kWe-scale; tested by NASA JSC and onboard NASA's KC-135 aircraft.

Responses to Comments

- *"This technology has good potential for this application, but the specific relevance needs better definition."*
  - A preliminary 'apples-to-apples' comparison of microchannel WGS to conventional adiabatic approach is provided.
  - FY04 scope does not include continued PROX development.
- *"Durability is still missing from list of technical targets." and "Need evidence or pathway for realization of 2,000 hr target."*
  - Catalyst deactivation has been a challenge, particularly with thermal cycles and restarts, and is under investigation.
  - Efforts have been focused on concept demonstration. Other issues including durability and fast-start will be addressed in future work.

Future Work

- Complete testing of 2 kWe WGS reactor
- Integrate PROX and WGS reactors with steam reformer at 2 kWe scale
  - Demonstrate rapid start
  - Evaluate the startup time, energy, and CO content vs time for this system
- Resolve durability technical challenges
  - Identify source of WGS deactivation with shutdown/restart
  - Demonstrate 2000 hour durability