Reformate Cleanup: The Case for Microchannel Architecture

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Overview

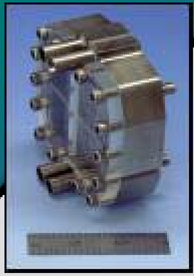
- Reformate Cleanup
  - Water Gas Shift (WGS)
  - Preferential Oxidation (PROX)
  - Desulfurization
- Balance of Plant
  - Water Management - Partial Condenser / Phase Separator

Objectives and Relevance

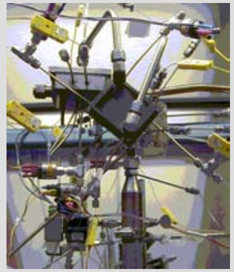
- Overall Objective
  - Apply microchannel architectures where appropriate in fuel processing for transportation, stationary, and portable applications to reduce size and weight, improve fuel efficiency, and enhance operation.

- Technical Challenges – Approaches
  - 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 Timeline



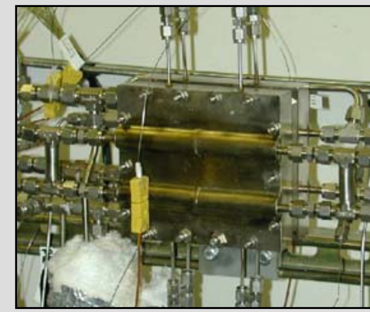
FY 1998
Full-size gasoline vaporizer/combustor
R&D100 Award



FY 1999
Fast SR kinetics observed in a microchannel reactor

FY 2000
10 kWe SR system
Invented phase separator concept

FY2001
10 kWe reactor testing
First "low dP" vaporizers
Demonstrated partial condenser



FY 2003
2 kWe WGS and PROX reactors.
Improve SR kinetics and sulfur tolerance.

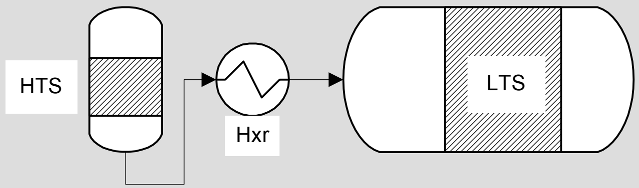
FY 2002
WGS/PROX catalyst studies
Differential temperature reactor concept
SR/WGS/PROX initial integration
Air-cooled partial condenser

- Engineered catalyst, reactor development
- Demonstrate rapid start capability
- Sulfur management
- Integrated reformer/fuel cell demonstration at ~2 kWe
- Collaborate with industrial partner(s) on manufacturing, field testing, lifetime, controls

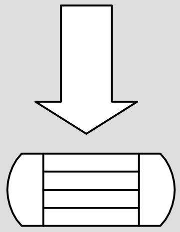
2002 — 2003 — 2004 — 2005 — 2006 — 2007 — 2008

Differential Temperature Water Gas Shift Reactor
Objective/Approach

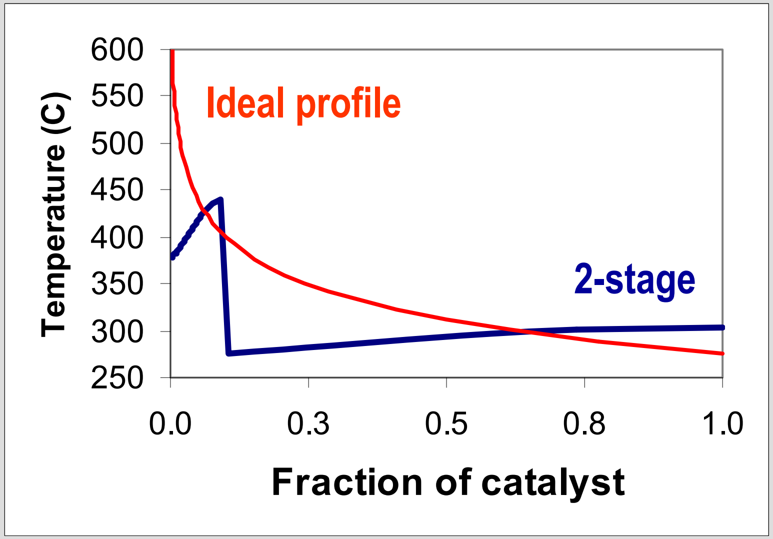
- **Objective:**
  - 90% Conversion single-stage WGS reactor < 3 liters at full-scale
- **Approach:**
  - Precious metal catalyst for high activity
  - Integrate microchannel heat exchange for temperature control
  - Optimize thermal profile
  - Reduce catalyst loading by up to 1/2
- **Relevance**
 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



Conventional 2-stage Adiabatic



Ideal profile



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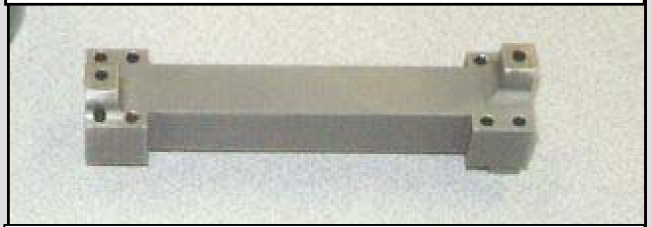
Differential Temperature Water-Gas Shift

Progress: Multi-channel Reactor Testing

Prototype 7-channel Reactor

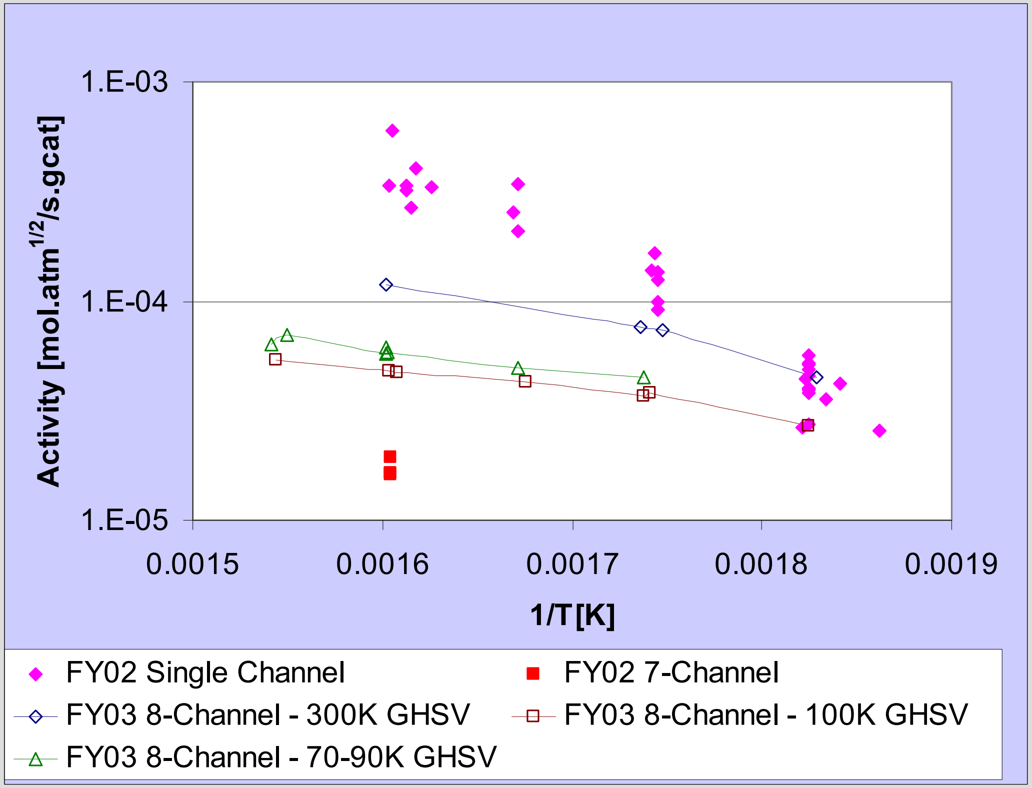


Reactor can be operated isothermally or with a temperature gradient

- PROGRESS:
  - Catalytic activity improved in multi-channel prototype reactor
  - Up to 79% CO conversion (12.4% to 2.6% dry CO) in single stage at 100,000 GHSV and 0.65 steam:gas.
- ISSUES:
  - Activity diminishes at lower GHSV
  - Loss of temperature dependence

Activity coefficients from isothermal reactor FEA model:

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



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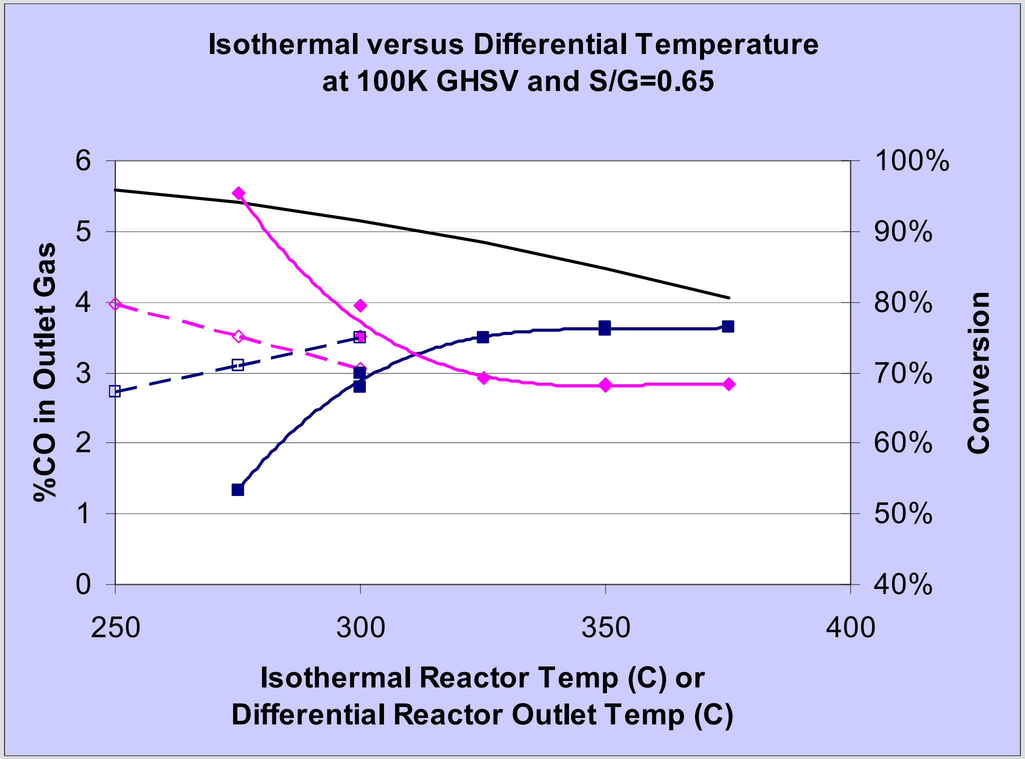
Differential Temperature Water-Gas Shift

Progress: Differential Mode Testing



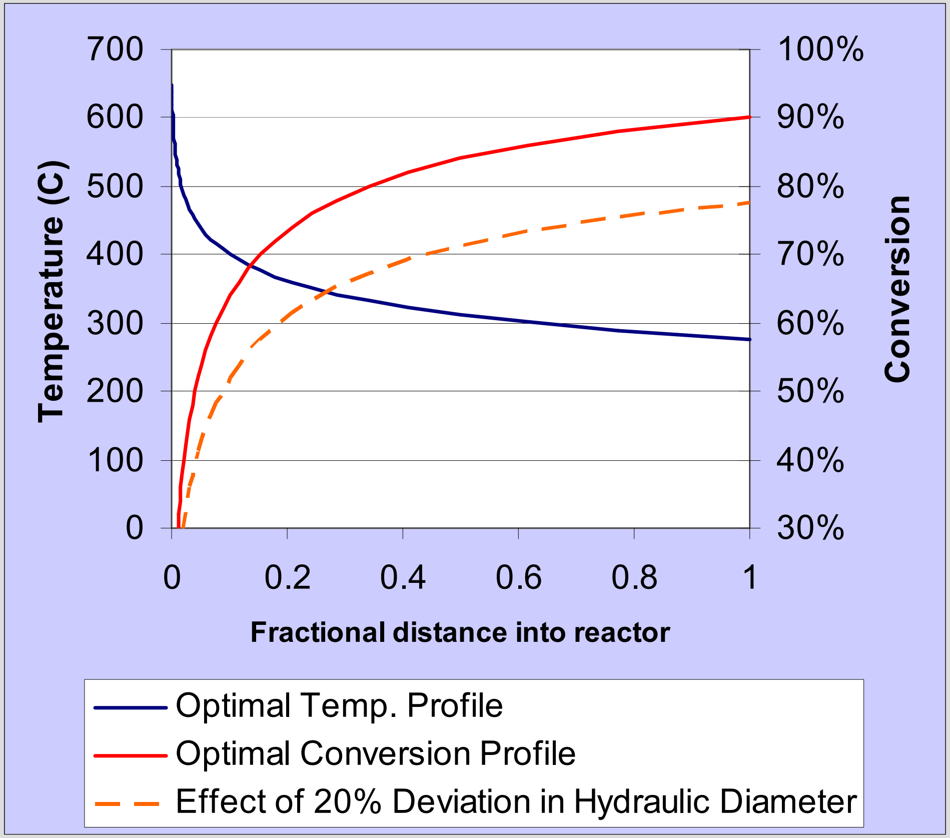
- Isothermal Operation - Outlet Gas CO%
- Differential Temperature Operation - Outlet Gas CO%
- Isothermal Operation – CO Conversion
- Differential Temperature – CO Conversion
- Equilibrium Conversion

- Conditions
  - Steam Reformate Feed
    - 12.4% dry CO
    - 14.3% dry CO_2
    - 0.65 Steam:Gas
  - 100K GHSV
  - Isothermal operation at high heat exchange flow
  - Differential temperature operation
    - Feed at 350C
    - 2 SLPM air coolant fed at outlet temperature

Differential Temperature Water-Gas Shift

Issue: Flow Maldistribution

- Flow maldistribution between channels is an important issue
  - Flow rate α d_h^3
  - Compounded by increased heat and mass transfer resistance
- Example
  - Impact of 20% distribution in hydraulic diameter on optimal conversion profile
  - Does not account for heat and mass transfer effects
  - Does not account for catalyst loading maldistribution



Differential Temperature Water-Gas Shift

Future Plans

- Continue catalyst development
  - Long-term testing
  - Start-up/shutdown and temperature cycling
  - Evaluate new formulations
    - Improved activity
    - Better activity maintenance
  - Sulfur sensitivity
  - Optimization of geometry and loading
- Multi-channel Improvements
  - Apply improved design and fabrication techniques to improve channel consistency
  - Demonstrate >90% conversion in single stage with integral heat exchange
- Develop WGS Reactor at 2 kWe-scale
  - Optimize catalyst productivity
  - Thermally integrate with 2 kWe-scale reformer system

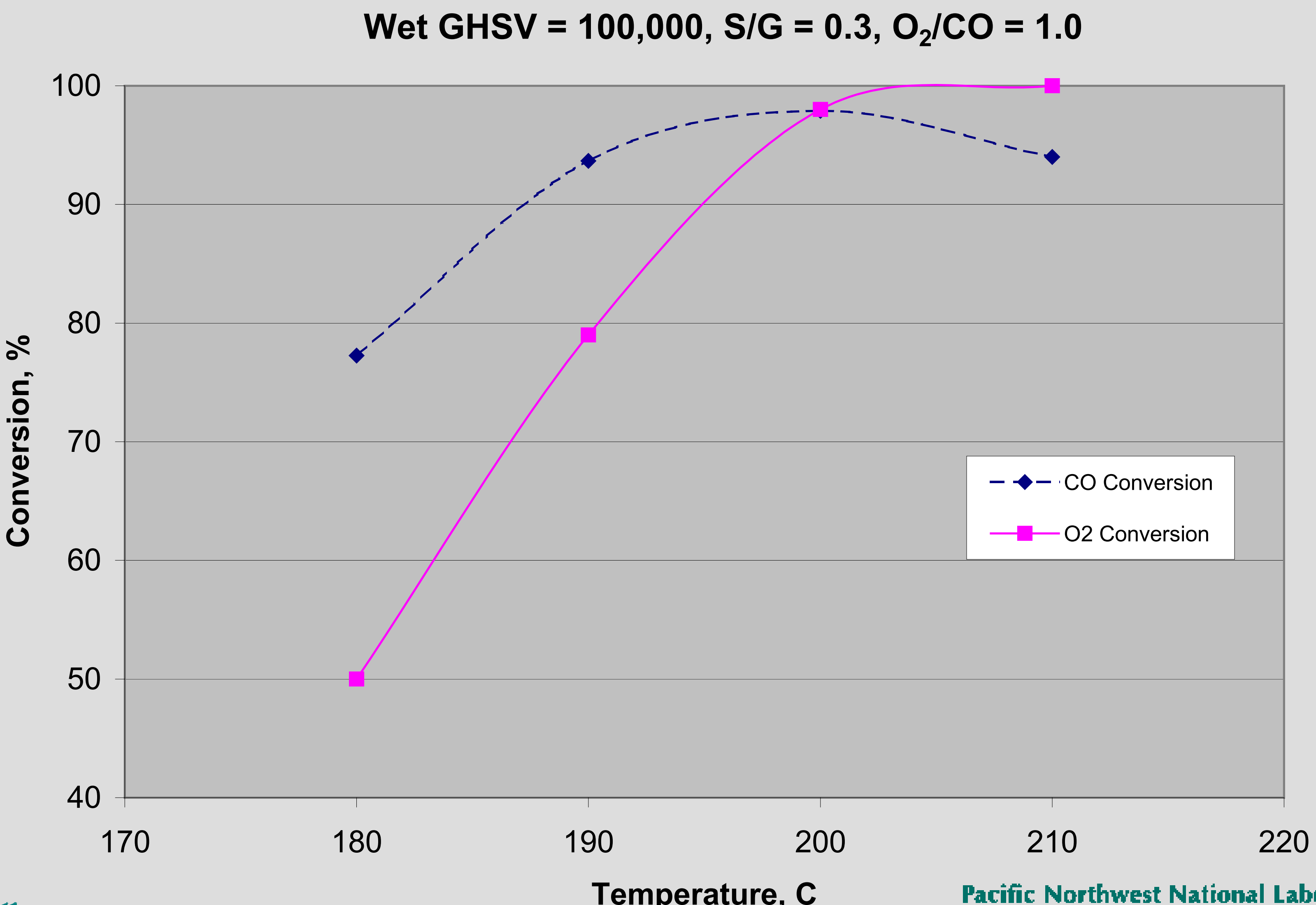
Microchannel PROX Reactor Investigations

- Objective: Determine whether microchannel architecture provides opportunities for size, weight reduction for PROX reactor.
- Approach:
  - Single-channel catalyst tests – evaluate industrial PROX catalysts for fast kinetics
  - Design & test 2 kWe PROX reactor unit; confirm favorable operational characteristics
  - Investigate weight reductions through use of low-density alloys (e.g., alloys of aluminum and titanium)
  - Investigate transient and startup characteristics
- Progress:
  - 1st Stage PROX microchannel reactor demonstrated at 2 kWe scale – exhibits high productivity due to internal microchannel heat exchangers providing temperature control

Preliminary Single Channel Catalyst Tests Used to Down-select for 2kW PROX Unit

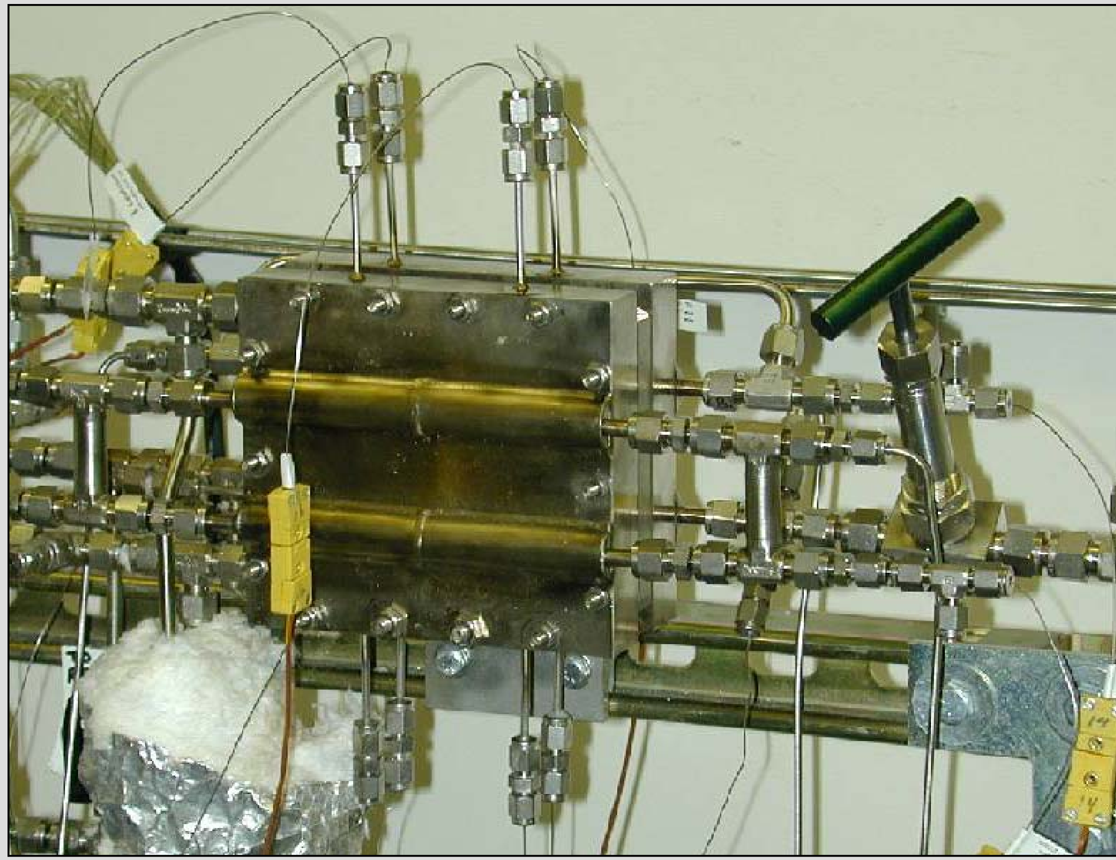
- At 1% CO in reformate, both precious metal and non-precious metal catalysts can be employed for PROX
  - Both catalysts show maximum CO conversion around 200^{o}C
  - Adiabatic operation must be avoided (excessive temperature rise)
- At 0.1% CO in reformate, precious metal catalyst are required
  - Higher CO activity at lower temperatures (~100^{o}C)
    - Lower activation energy
    - Different dependence on CO concentration (analogous to wgs)
  - Adiabatic operation may be possible
- Non-precious metal catalyst was selected for 2kW PROX test in microchannel hardware
  - CO ~1%, input from wgs reactor
  - Lower catalyst cost
  - Higher CO oxidation selectivity (lower H_2 consumption and O_2 requirements)

Selective CO Oxidation (PROX) With Non-Precious Metal Catalyst

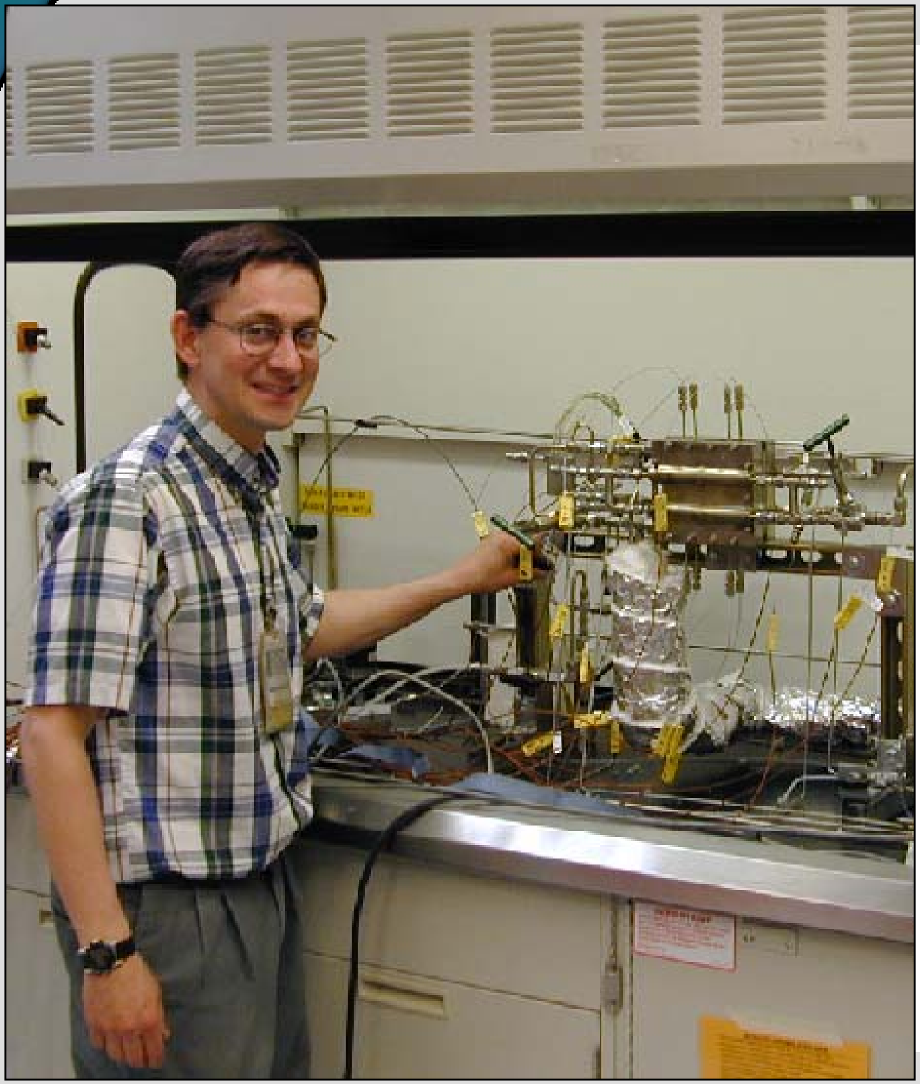


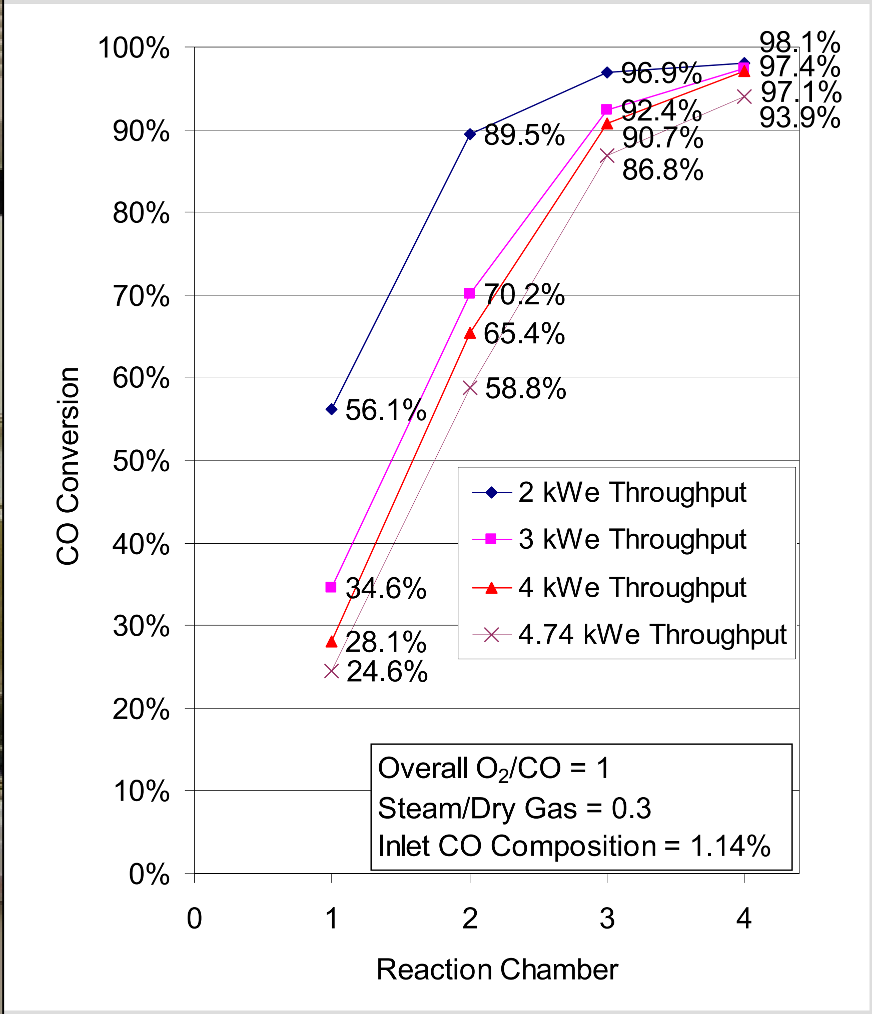
Demonstration of Microchannel PROX Unit

- Designed as 1st Stage PROX for 2 kWe operation
- Operates as a quasi-isothermal reactor
  - Incorporates microchannel heat exchange
- Employs non-precious metal catalyst
- Allow multi-compartment bleed-in of air



Operation of First Stage Microchannel PROX





Future PROX Work

- Continue to evaluate industrial PROX catalysts for use in microchannel reactors
- Evaluate aluminum and titanium alloys for microchannel PROX reactor
  - Potential weight reduction
- Integration of 1st and 2nd Stages
  - Investigate differential-temperature operation
- Test as part of integrated fuel processor with steam reformer, water-gas-shift reactors and heat exchangers
  - Evaluate fast startup, rapid transient potential
  - Evaluate thermal recuperation of PROX heat

Reformate Desulfurization Using Microchannel Devices

- Concept: microchannel absorbers can provide compact units for H_2S removal from reformate
  - ZnO absorption:
    - Fast uptake kinetics at T≥400°C but relatively high equilibrium H_2S concentration in high steam environments
    - Slower kinetics at T≤300°C but lower equilibrium H_2S concentration
  - Differential temperature microchannel absorber can provide more effective H_2S removal by operating over temperature range 300-400°C
- Challenge: absorber bed capacity in microchannel configuration
- Approach and status
  - Carry out initial studies (underway) to quantify ZnO uptake kinetics vs. T
  - Calculate optimal temperature profile for differential temperature absorber
  - Construct microchannel unit and obtain experimental performance data under differential temperature conditions
  - Compare results with isothermal operation

Balance-of-Plant

Partial Condenser / Phase Separator

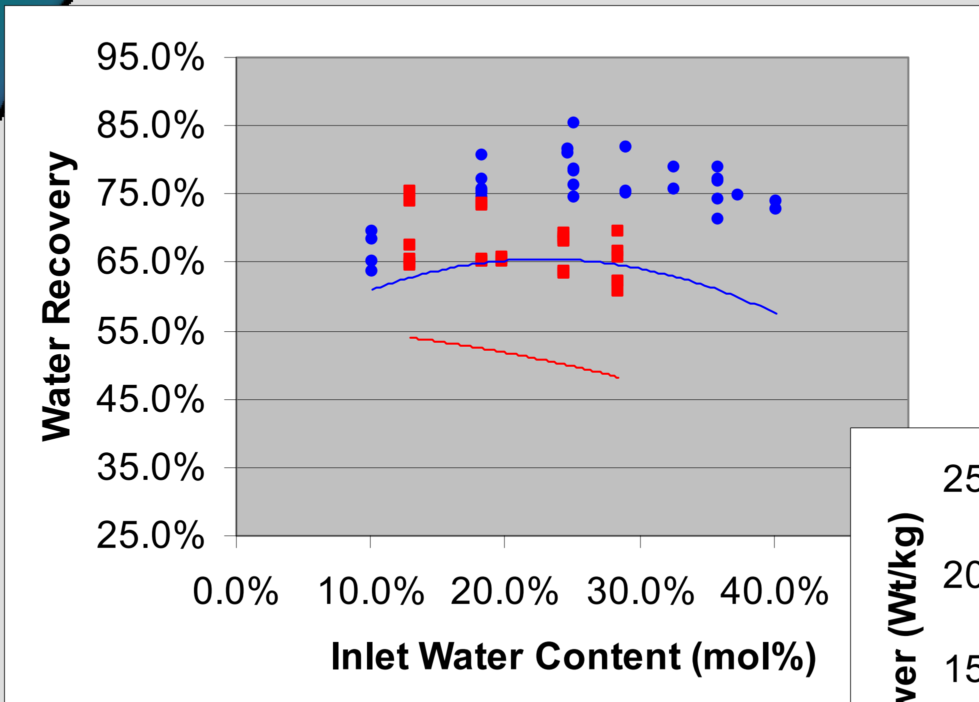
- Objective: Develop compact microchannel heat exchange technology for the recovery and recycle of water in fuel processor / fuel cell systems.

- Designed for water recovery from PEM fuel cell cathode effluent
- Condenses and separates water
- Aluminum for light weight
- Air-cooled
- Cross-flow for low coolant-air pressure drop
- 1.5-kWe-scale cathode condenser
  - Air coolant - 30 °C
  - Air pressure drop - 2.2 in. H2O
  - Weight - 260 g
  - Volume - 170 mL
  - Est. Fan Power - 14 W full-scale

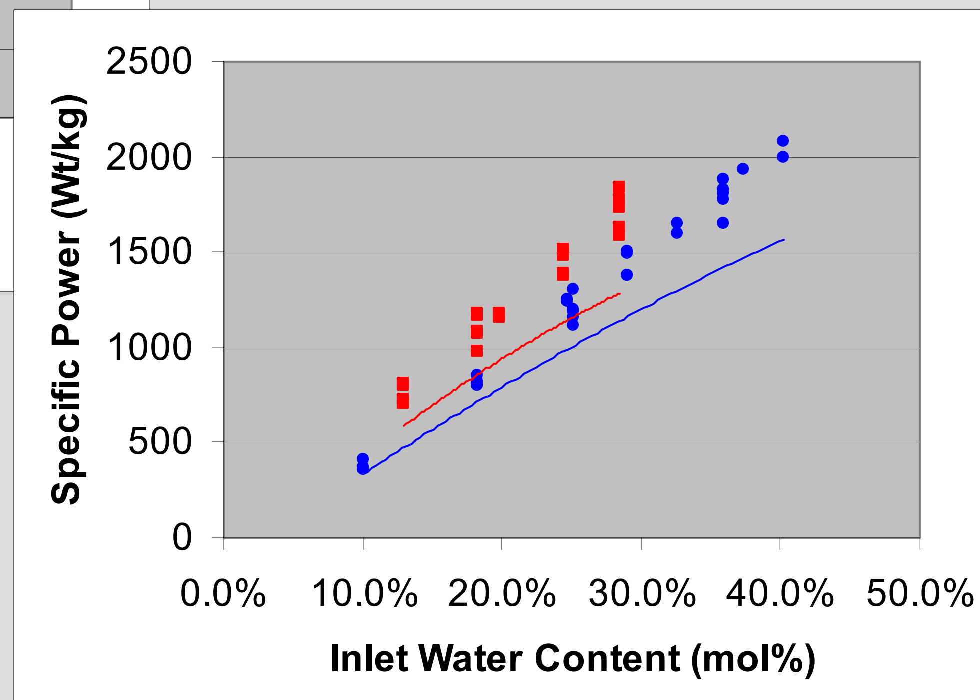
3 condensing channels (left to right)
4 cooling channels with fins

Vapor-Liquid Separator

Measured versus Predicted Performance



32 std Liter/min air flow
48 std Liter/min air flow
— Average model prediction



CONCLUSIONS

- Actual performance exceeds predicted performance.
- 2000 W_t/kg specific power at 74% water recovery.
- Water separation efficiency 90-100%.
- Coolant side pressure drop about 2X design ~ 4 in. H_2O.

Interactions

- Catalyst Development
  - Sud-Chemie, Inc. – WGS, PROX, Desulfurization
  - Engelhard - PROX
- Technology Transfer
  - Interest expressed by SOFCo for a 50-kWe-scale Differential Temperature Water Gas Shift reactor.
- Technology Spin-off
  - Microchannel Partial Condenser / Phase Separator successfully tested in zero gravity onboard NASA's KC-135 aircraft.
  - Microchannel Phase Separator is a candidate technology for NASA's Space Plane for water recovery from cathode of PEM FC power system.

Responses to Comments

- *"WGS durability may be a significant challenge…" and "Lack of WGS durability data"*
  - Catalyst deactivation has been a challenge, particularly with thermal cycles and restarts, and is under investigation.
  - A single catalyst piece has been on stream for 100 hours with nominal initial deactivation; ~ 10-20%.

- *"Little of proposed work is related to advancing fundamental understanding, which should be of high importance"*
  - WGS work is pursuing a fundamental understanding, including:
    - Kinetic model development in an engineered form
    - Finite element heat and mass transfer models of reactors for analysis and design
    - Scale-up from single channel to multi-channel reactors to understand the issues associated with scale-up, such as flow maldistribution