MICROCHANNEL FUEL PROCESSING COMPONENTS

DOE Fuel Cells for Transportation Program Annual Review

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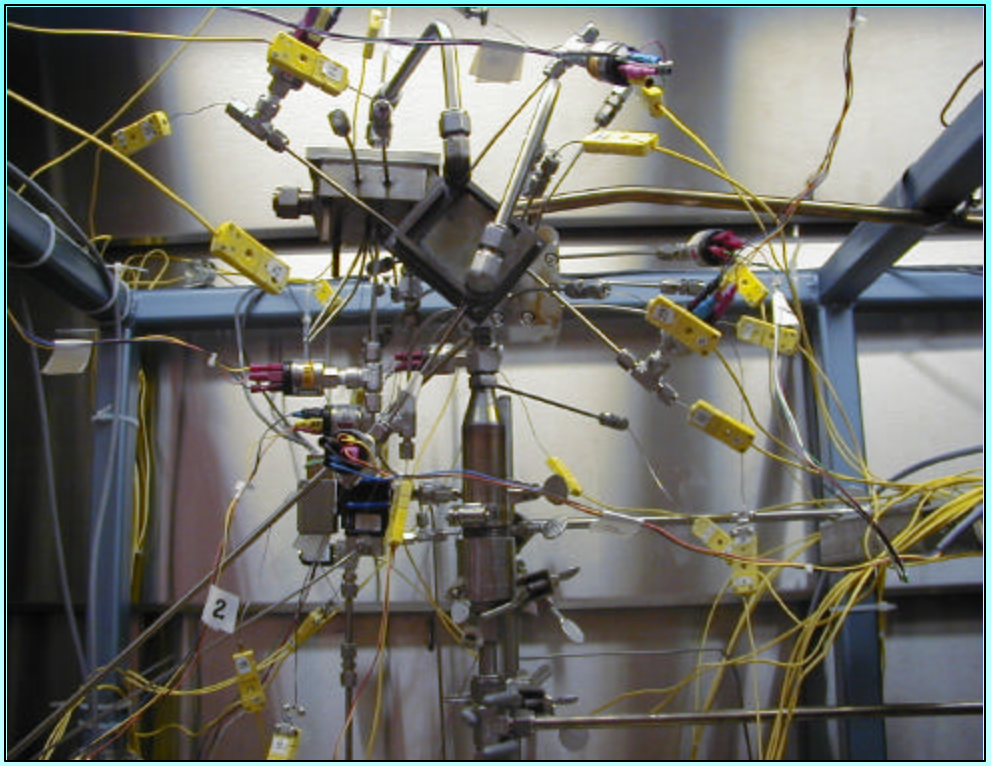
June 24, 1999

ACCOMPLISHMENTS: MICROCHANNEL GASOLINE VAPORIZER

- 50 kWe capacity: Four cells each of microchannel reactors and heat exchangers
- Volume: 0.3 liters
- Interactions with Industry:
  - Units delivered to Epyx and Hydrogen Burner Technology for testing (September 1998)
  - Met performance goals during testing at Epyx (May 1999)
- FY99 Cost Studies:
  - $66/unit based upon 500,000 units manufactured per year

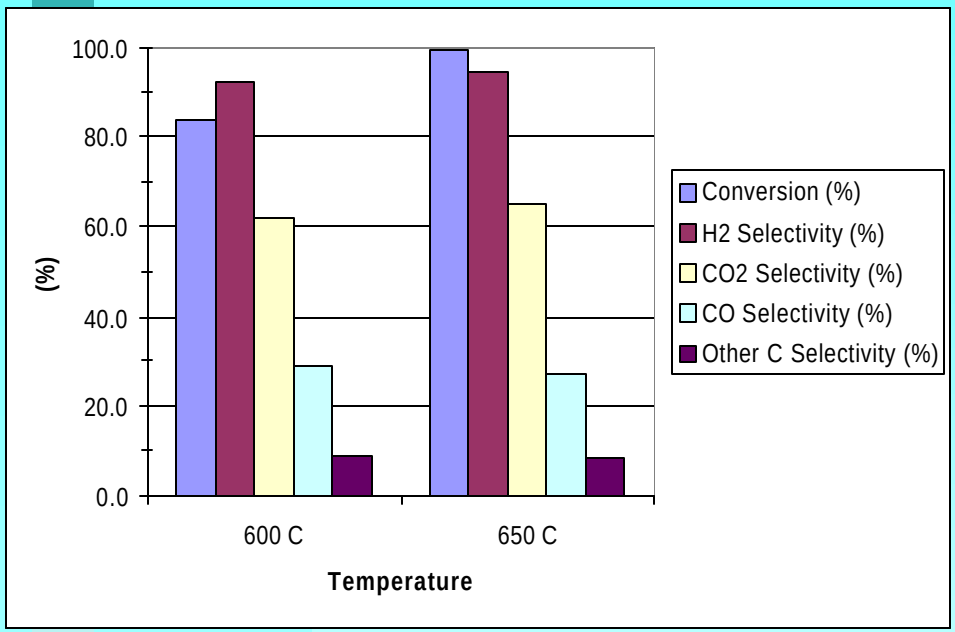
ACCOMPLISHMENTS: DEMO OF MICROCHANNEL STEAM REFORMER

- First proof-of-principle testing of a single-cell, microchannel steam reformer (June 1999)
- Conditions include:
  - 1.1 to 2.3 millisecond residence time
  - Steam : Carbon ratios of 3:1 to 6:1
  - 67-72% H_2 in product stream

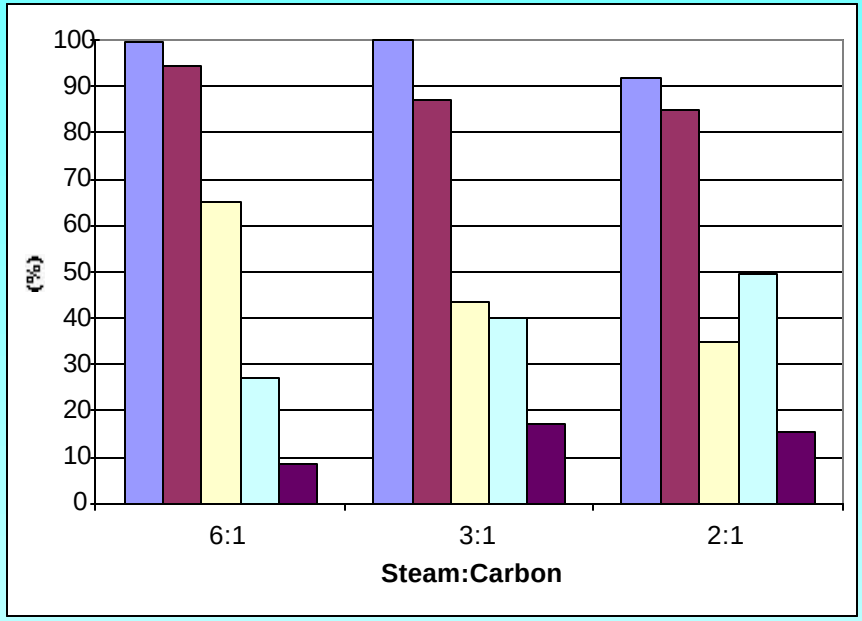


Implications: 8-10 Liters (0.30 - 0.35 ft^3) overall fuel processing system hardware volume (for 50 kW_e fuel cell)

ACCOMPLISHMENTS: PERFORMANCE OF ISOOCTANE SR ON ENGINEERED CATALYST



(6:1 steam ratio)



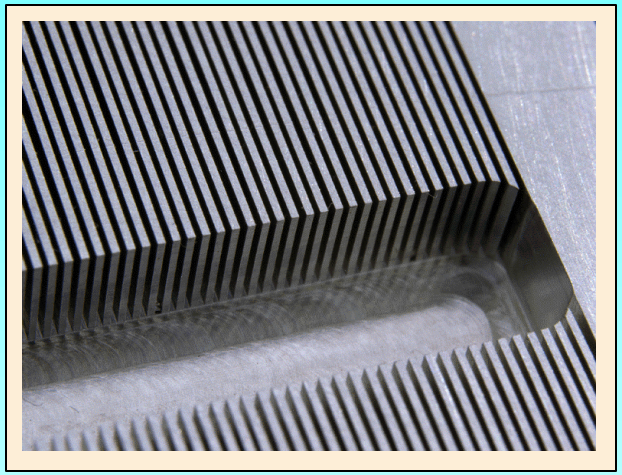
(650 C)

> 90% conversion and > 90% selectivity to H_2 with millisecond residence times

100 hour test runs successfully completed

OBJECTIVES

- Develop ultra compact reactors, separators and heat exchangers -- for the onboard, automotive production of hydrogen from liquid hydrocarbons -- *based on heat and mass transport advantages that can be realized using engineered microstructures*



- Goals:
  - Microchannel Fuel Processor System Volume: < 1.0 cubic foot
  - Cost: < $500/unit (based on 500,000 units per year)
  - Performance: 40-50% system efficiency (including fuel cell)

TECHNICAL APPROACH

- FY99 Approach
  - Microchannel gasoline vaporizer for POx and/or autothermal reformers
    - Reacts remaining hydrogen in the fuel cell anode offgas to provide gasoline heat of vaporization
    - 0.3 liter hardware volume for 50 kWe product stream
  - Integral microchannel steam reforming system
    - Includes reformers, combustors, recuperative heat exchangers, fuel vaporizers and steam generators

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- Development Report
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- Conclusions
  - Interactions with Industry, other Collaborations and Technology Transfer
  - Program Direction / Next Steps

Draft Functional Specifications

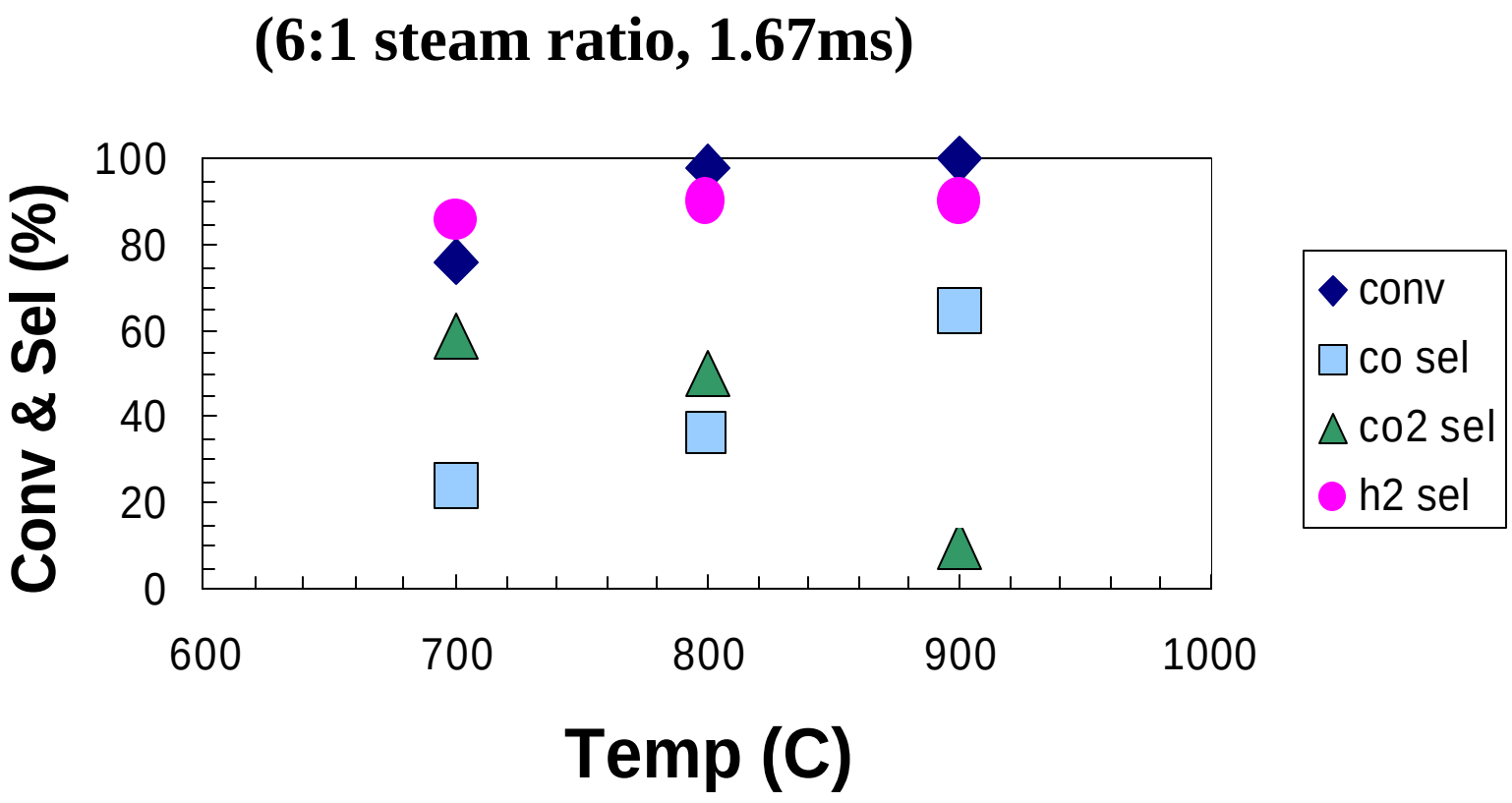
Level 3: Catalyst Requirements

- Reaction *residence* time 8 ms
  - 3 ms goal
- Steam to carbon ratio
  - 3:1 design point
  - 2:1 goal
- Coke formation: negligible
- Lifetime: >2500 hrs
- Temperature range 650-850 C
  - 700 C goal

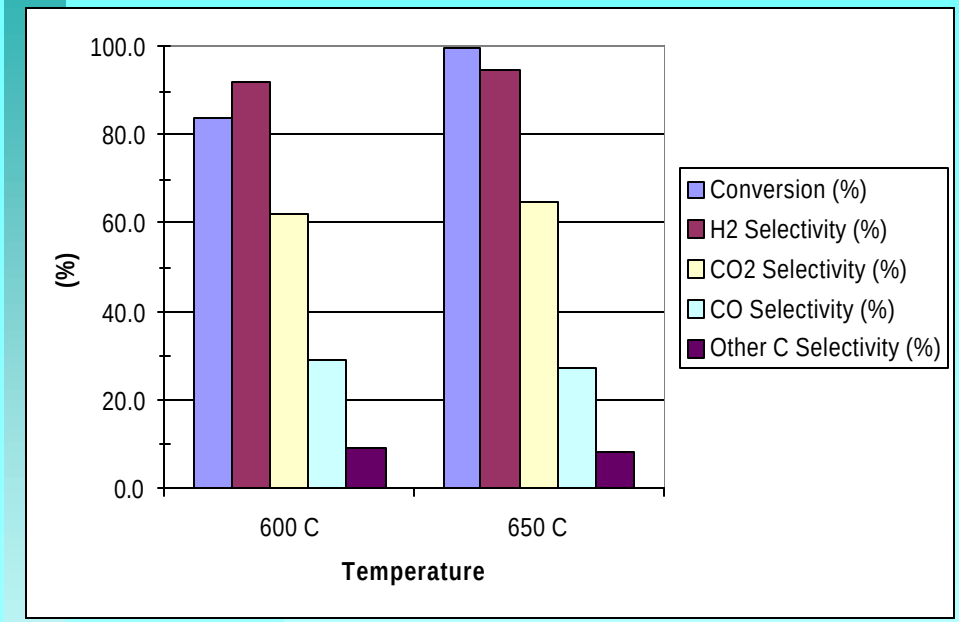
Catalyst Development:
Microchannel Reactor Catalyst Development Path

- Scientific Feasibility
  - Initial screening of powder catalyst for intrinsic kinetics
  - Desired performance
- Engineering Feasibility
  - System required performance met using engineered catalyst
  - Catalyst life issues
- Deployment
  - Microchannel reactor with integrated engineered catalyst

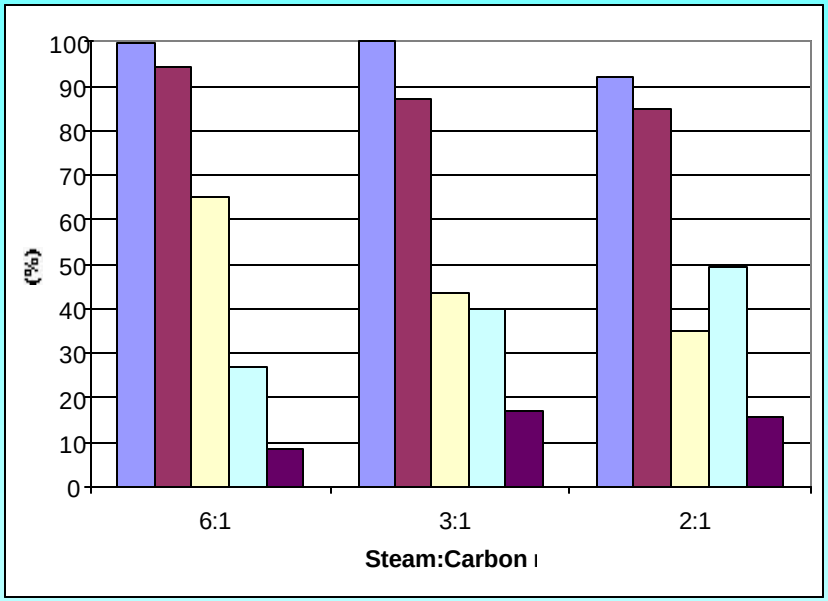
Catalyst Development:
Proprietary Iso-octane SR Powder Catalysts



Catalyst Development:
Performance of Proprietary Iso-octane SR Engineered Catalyst



Temperature effect (6/1 steam ratio)



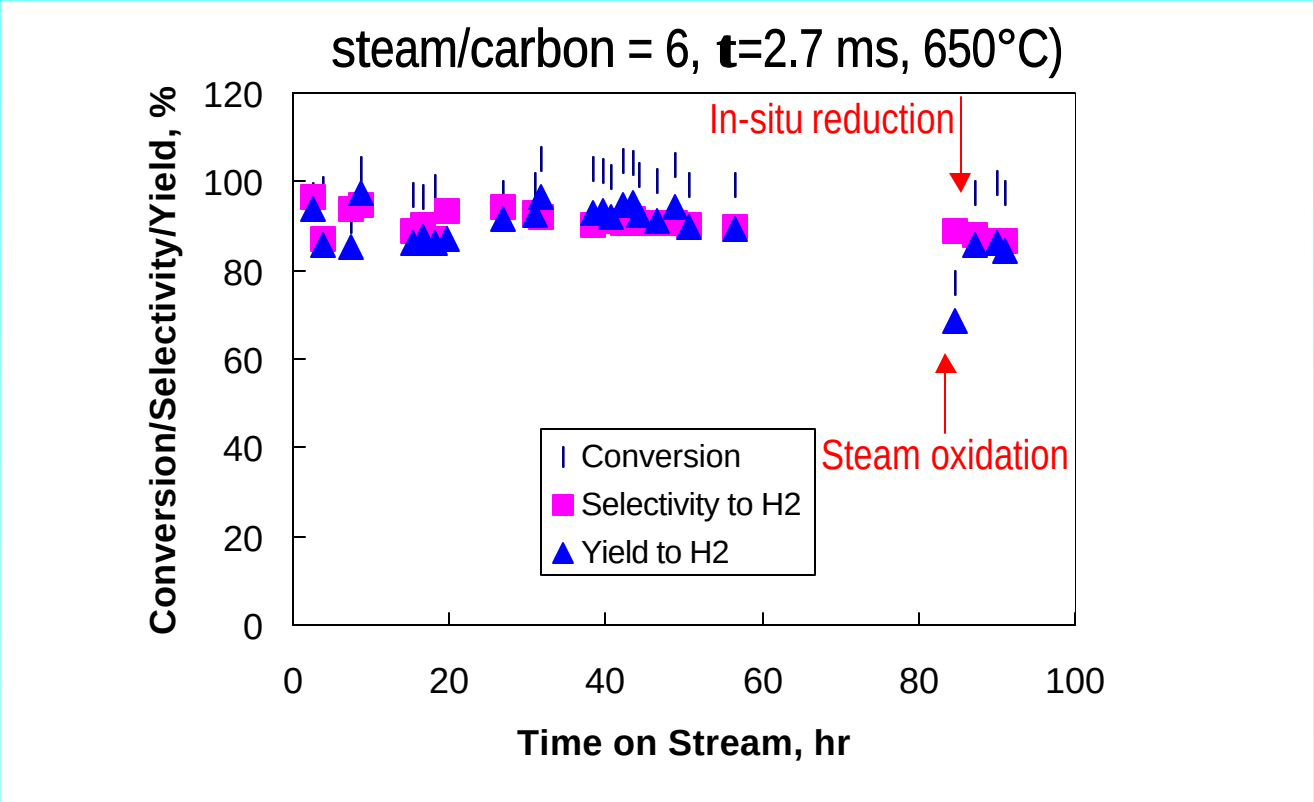
Steam ratio effect (650C)

>90% conversion and >90% selectivity to H_2 can be achieved at <700C and 0.86 ms *residence* time

<u>AND</u> potentially with 2:1 steam:carbon ratio (currently 85% conversion and 83% selectivity at 650C)

Catalyst Development:

Life Testing of Proprietary Engineered Catalyst for Iso-octane SR



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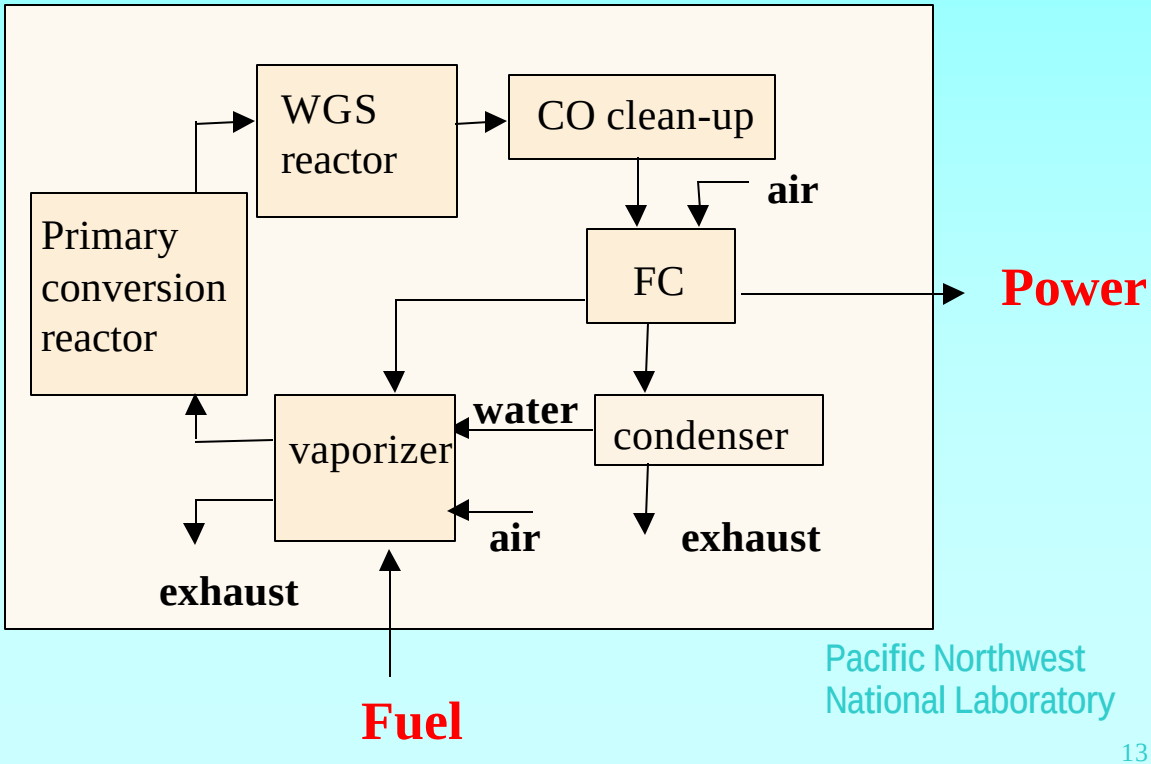
Automotive Fuel Processor

- Primary Conversion
- Water gas shift
- CO Clean-up
- Vaporizer

Fuel processor components



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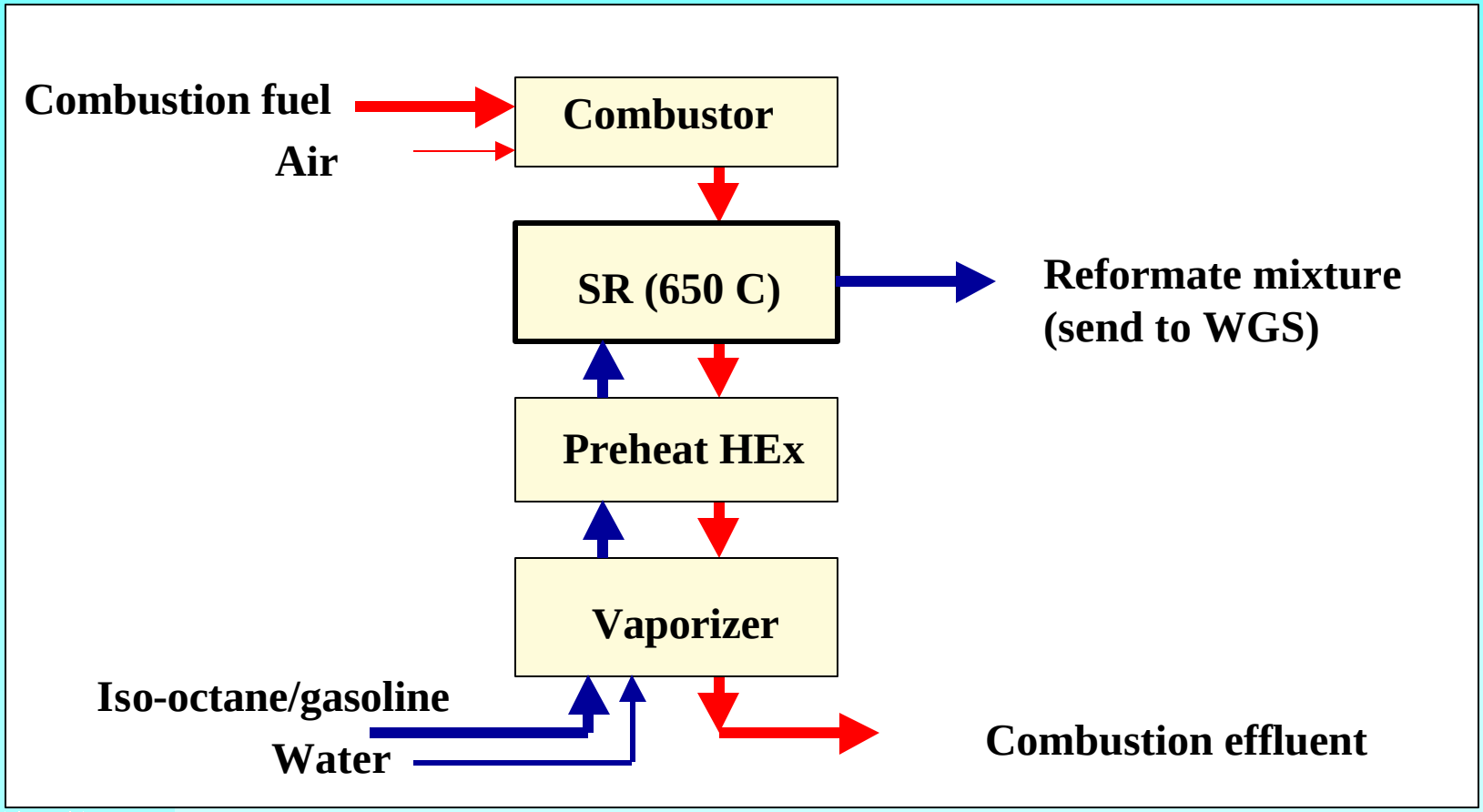
Process Chemistry:
Automotive Fuel Processor

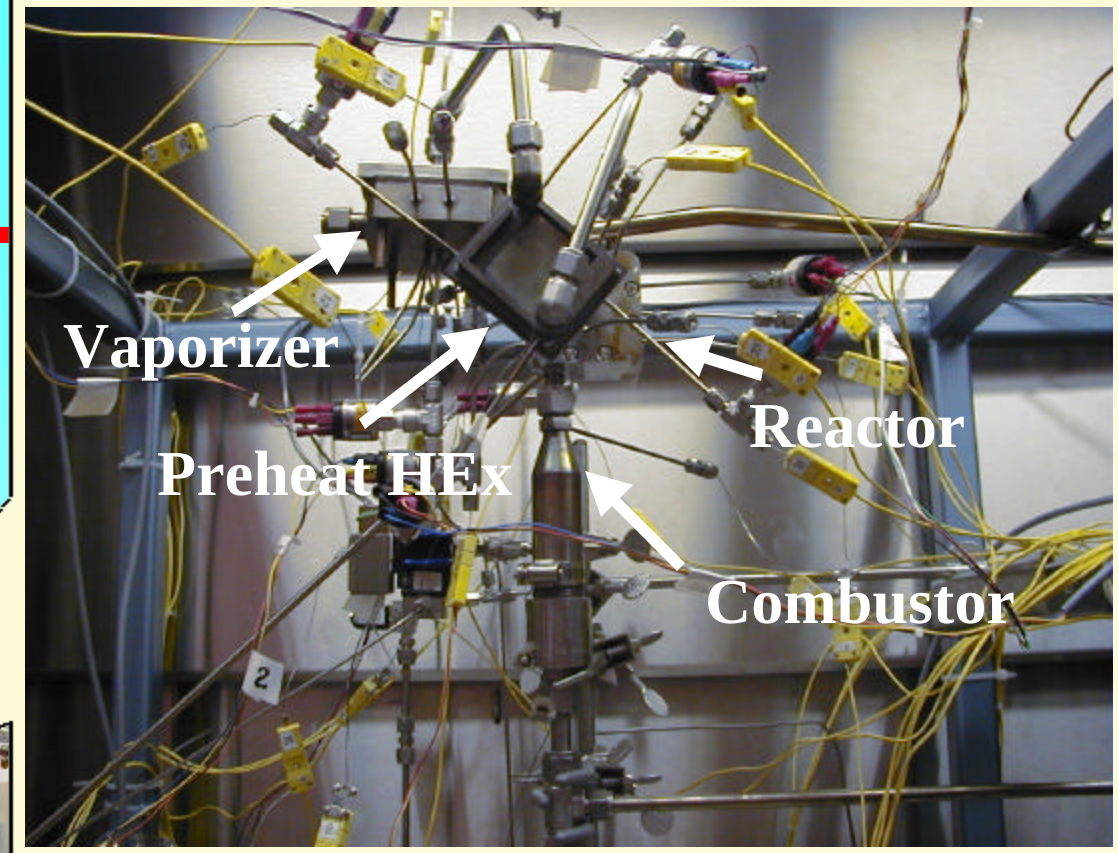
- Iso-octane steam reforming (gasoline simulant)
  - $C_8H_{18} + 8\ H_2O = 8\ CO + 17H_2$ ΔH_r = 1345 kJ/mol
  - $CO + H_2O = CO_2 + H_2$ (some high T shift)
  - Undesired side/series reactions:
    - $CO+CO = CO_2 + C(s)$
    - Cracking reactions
    - Methane formation
  - <u>Conditions (conventional hardware)</u>:
    - Temperature ~ 800C
    - Steam : Carbon ~ 6+
    - Residence time > 1 sec

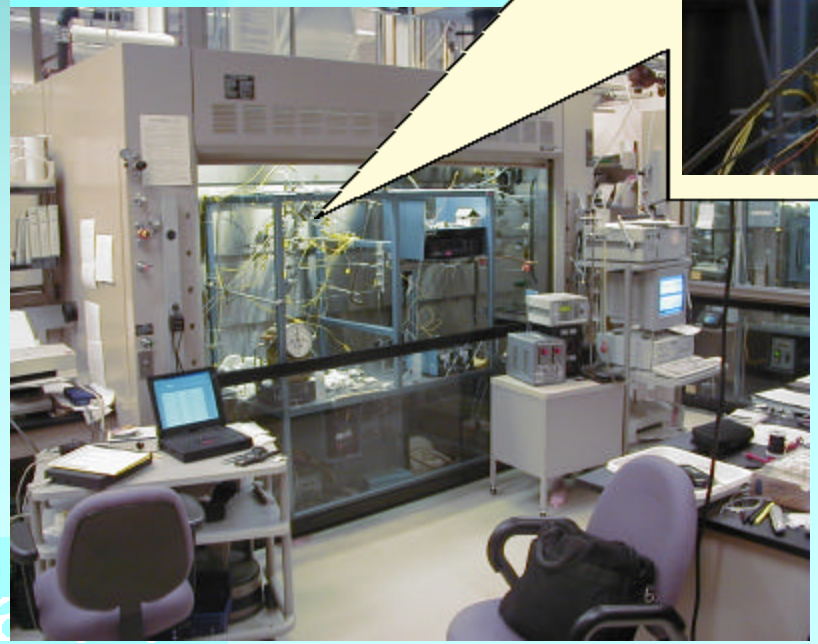
Primary conversion:
Proprietary Catalyst and Microreactor

- Steam reforming demonstrated in a microchannel reactor



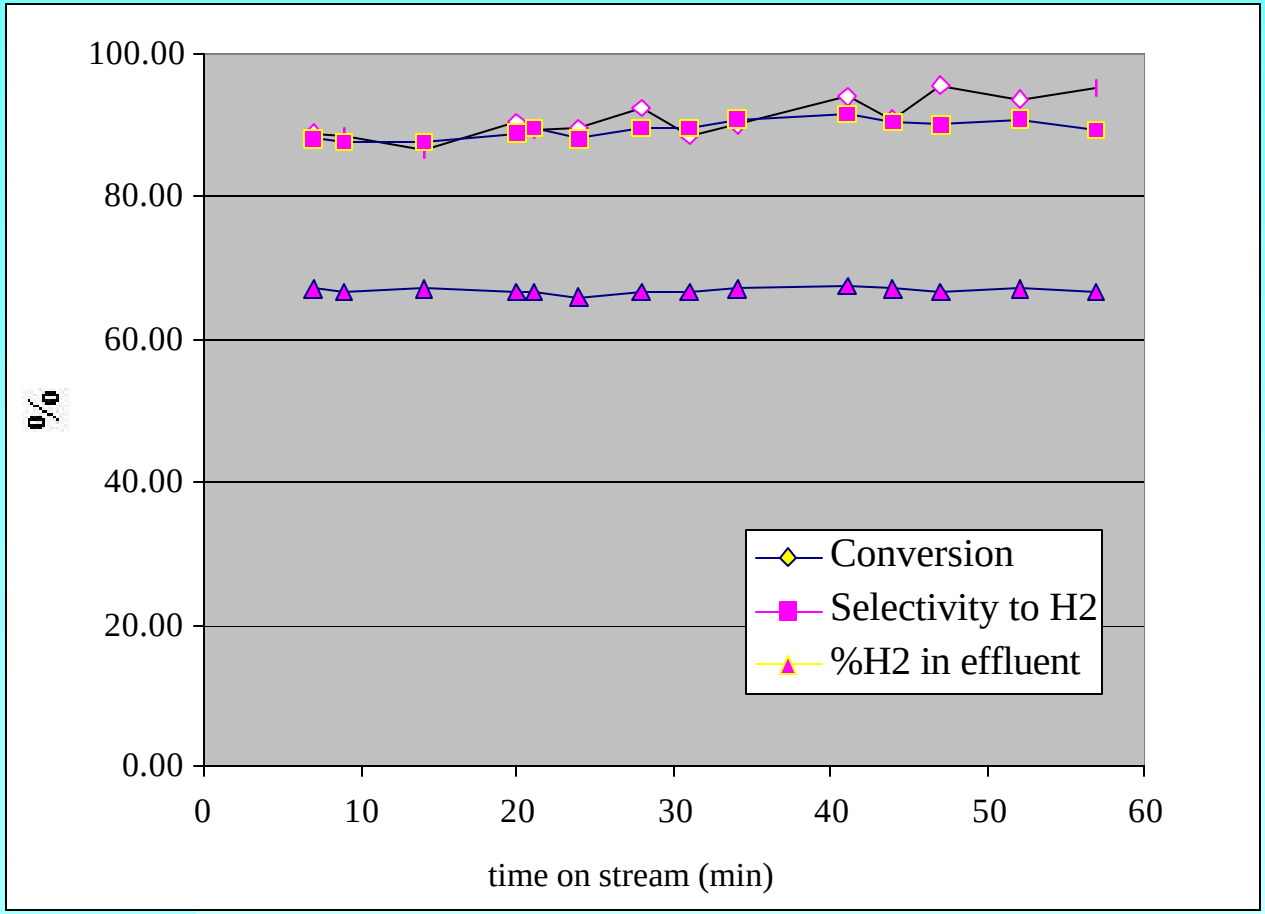
Microchannel Steam Reformer





Reactor cell volume = 18 cm^3

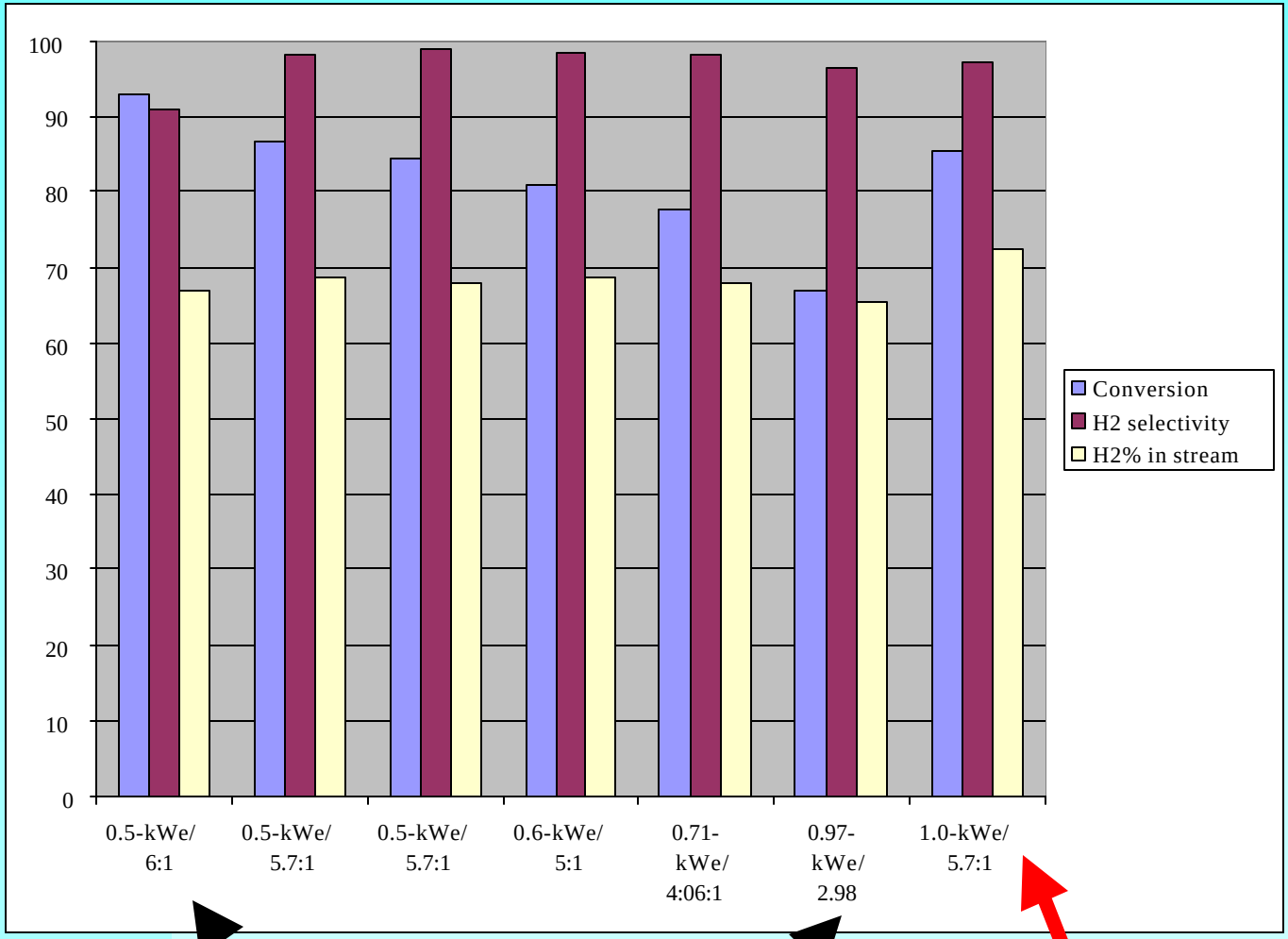
Iso-octane Steam Reforming:
First hour



τ=2.3 ms
T=650 C
S:C = 6
0.5-kW_e

SR Performance:

Function of power, S:C, residence time, heat flux



Residence time ~ 2.3 +/- 0.2 ms

t ~ 1.15 ms

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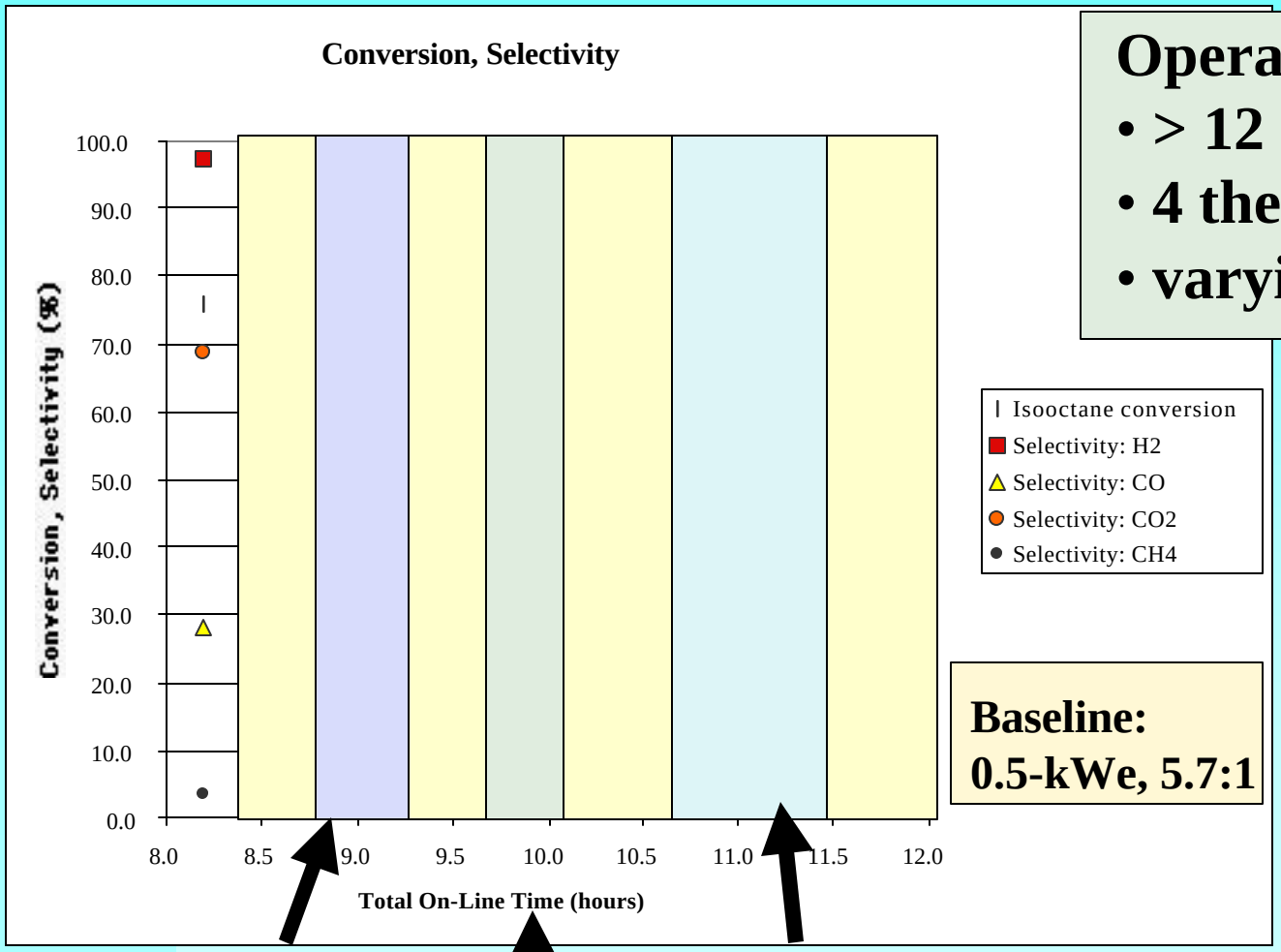
SR Performance:
function of time and load



Operated
- **> 12 hrs**
- **4 thermal cycles**
- **varying loads**

Baseline:
0.5-kWe, 5.7:1

0.6-kWe, 5:1

0.71-kWe, 4:1

0.97-kWe, 3:1

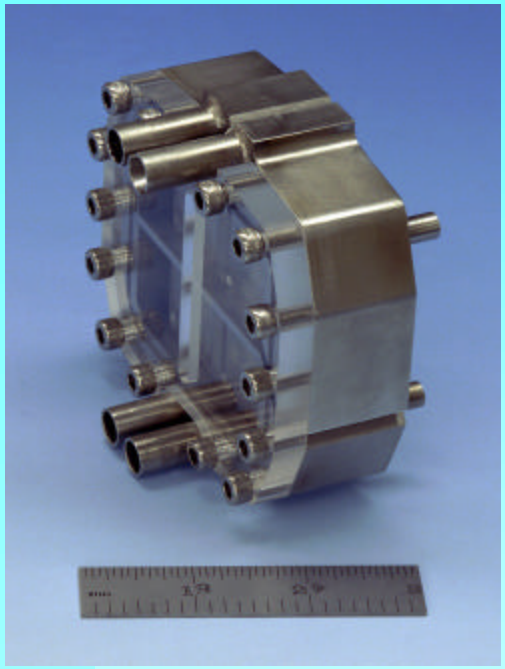
Steam Reformer Summary

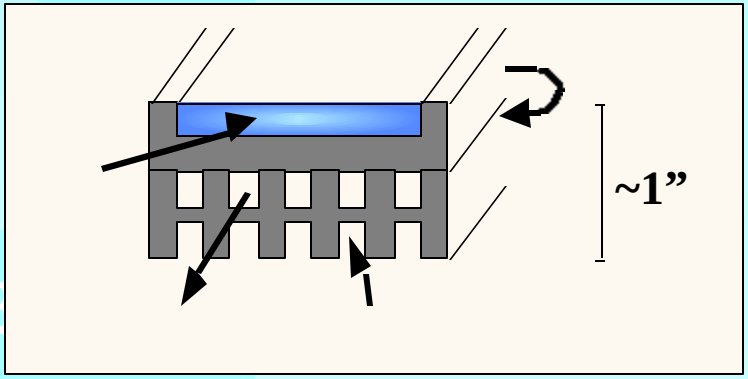
- Capacity (cell volume 18 cm^3)
  - Initial experiments: 0.5 to 1.0-kW_e at 1 atm
  - Design point: 5-kW_e at 5 atm
- Range of test conditions
  - Residence time: 1.1 to 2.3 milliseconds
  - Steam:carbon : 3:1 to 6:1
  - Temperature: 630 to 670 C
- Performance
  - Conversion = 67 to 99%
  - H_2 Selectivity = 91 to 99%
  - H_2 content = 67 to 72%
  - No degradation observed
- Implications: complete full-scale SR System (50-kW_e) ~4L

Automotive Fuel Processor

- Steam reformer
- Water gas shift
- CO Clean-up
- Vaporizer
  - Combust dilute H_2 present in anode effluent to provide heat of vaporization for fuel

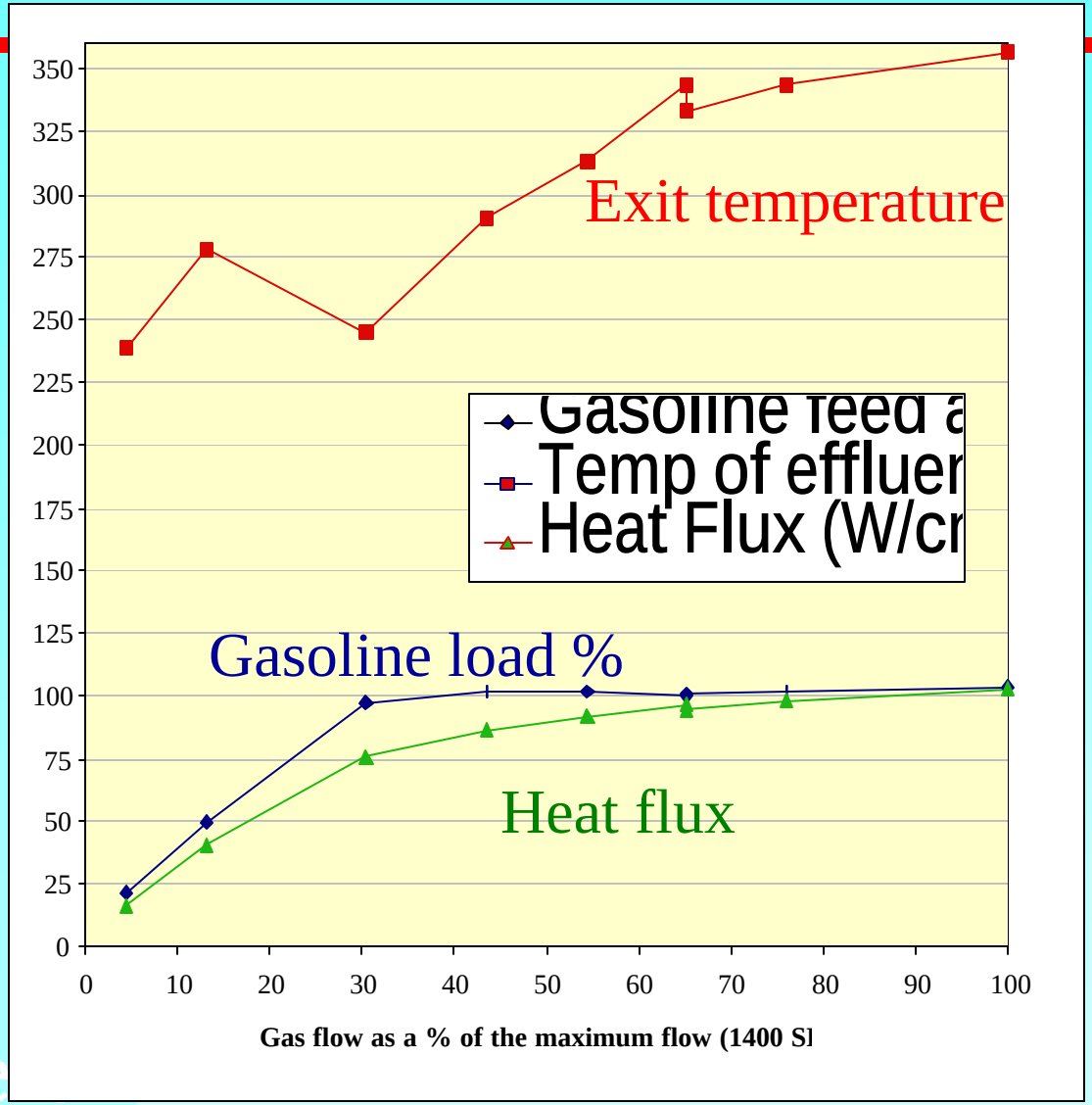
Full-Scale Microchannel Gasoline Vaporizer





- Attributes: Four parallel cells of microchannel reactors and four cells of microchannel heat exchangers
- Size: 3” by 4” by 1.5”
- Capacity: Vaporized gasoline for 50-kW_e fuel processing system
- Implications: Complete fuel processor system = 0.3 ft^3
- Fabrication: Laminate process
- Pressure drop: ΔP < 2psi through microchannels at 1400 SLPM
- Delivered : Epyx and H2 Burner

Compact Gasoline Vaporizer:
Full-Scale Performance

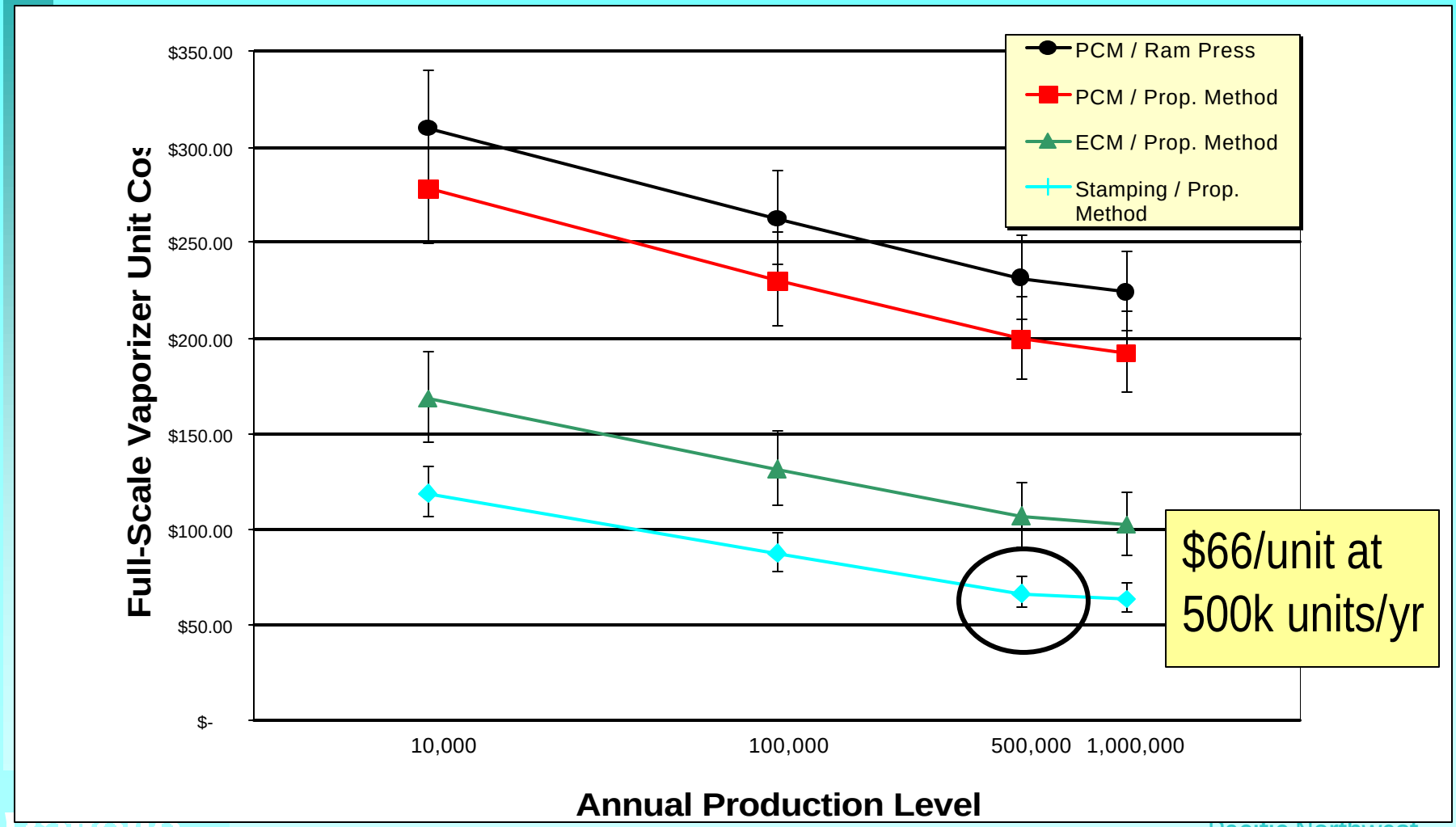


Gasoline vaporized: ~ 300 mL/min

Feed gas (anode effluent + air: ~ 1400 SLPM

Demonstrated at Epyx (5/18/99)

FULL-SCALE GASOLINE VAPORIZER
Manufacturing Cost Study Results



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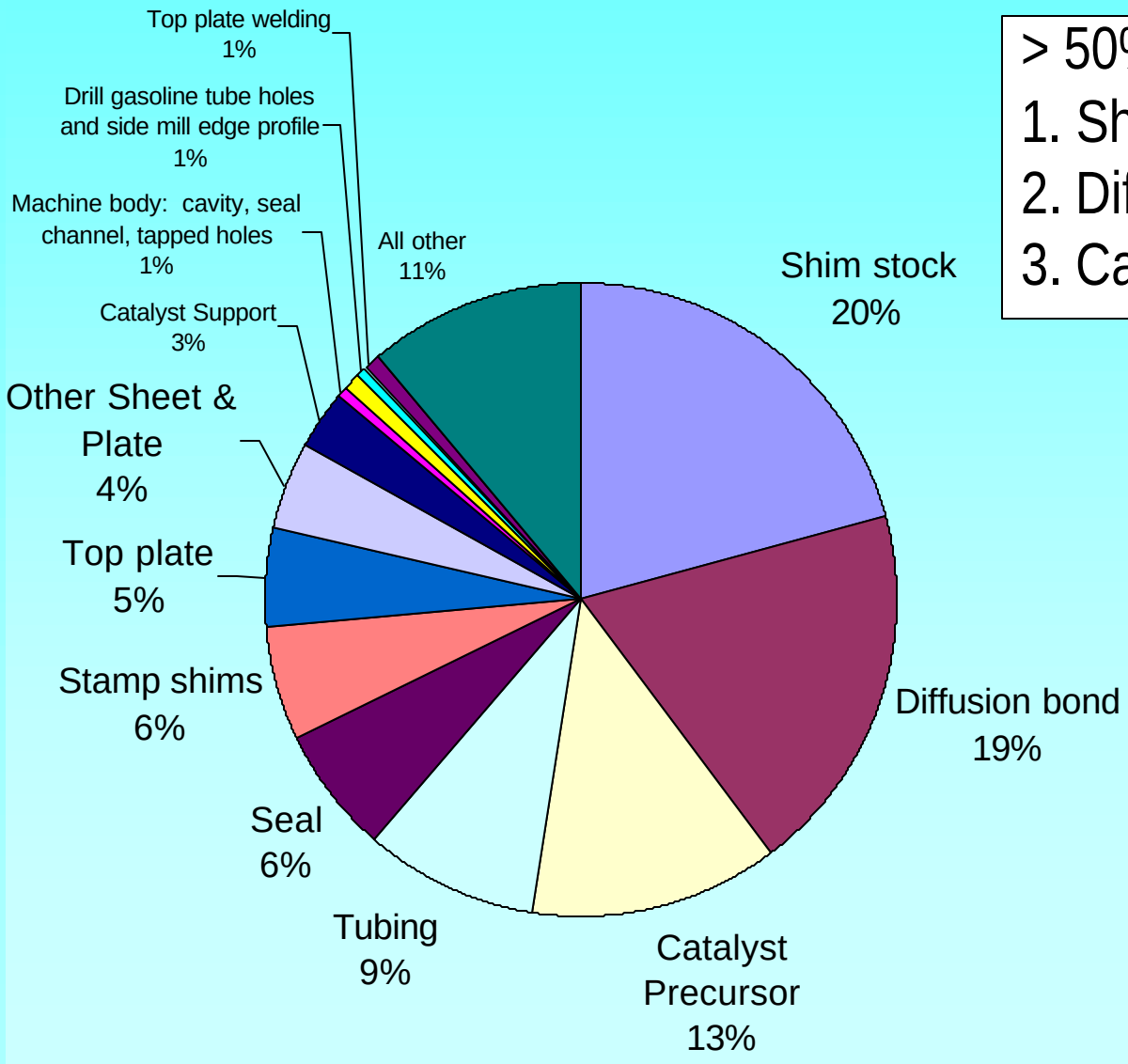
Manufacturing Cost Study Results



> 50% of Unit Cost:
1. Shim Stock
2. Diffusion Bonding
3. Catalyst Precursor

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INTERACTIONS WITH INDUSTRY, OTHER COLLABORATIONS AND TECH TRANSFER

- Microchannel gasoline vaporizer at Epyx and Hydrogen Burner for testing as part of integrated fuel processing system
- Short residence time performance of Argonne's autothermal catalyst to be evaluated
- Conferences and Papers (see "References" appendix for specific papers during FY99)
  - AICHE/DECHEMA International Conferences on Microreaction Technology
  - International Symposium of Chemical Reaction Engineering
- Nondisclosure Agreements and meetings with automobile industry and fuel cell developers

PROGRAM DIRECTION / NEXT STEPS

- TARGET: Demonstrate closed-loop microchannel fuel processing system with fuel cell operation (5 kWe) during FY01
- FY99-00 Tasks
  - Microchannel Gasoline Vaporizer:
    - Support Epyx and Hydrogen Burner testing of microchannel gasoline vaporizers
    - Publish manufacturing costs evaluation
  - Manufacturing Cost Studies
    - Demonstrate low-cost fabrication technologies

PROGRAM DIRECTION / NEXT STEPS (Continued)

- FY99-00 Tasks
  - Microchannel Steam Reformer System
    - Continue testing of catalysts and current reformer unit
    - Transition to gasoline and other transportation fuels
    - Test high efficiency recuperative microchannel heat exchanger
    - Design and test integrated reactor/combustor core
    - Develop detailed design for integral steam reformer system including reactors, combustors, recuperative heat exchangers, fuel vaporizers and steam generators
  - Microchannel Gas Conditioning
    - Investigate microchannel gas absorption and adsorption methods for removal of hydrogen sulfide, carbon monoxide and carbon dioxide

CONCLUSIONS

- Microchannel vaporizers currently at Epyx and Hydrogen Burner Technology for testing
- Engineered catalysts in development to meet system specs while operating at millisecond residence times
- First proof-of-principle testing of microchannel steam reformer
- Complete microchannel fuel processing system estimated at 8-10 Liters (0.30 - 035 cubic feet)
- Manufacturing cost evaluations suggest acceptable costs for microchannel reactors and heat exchangers
- Next steps can lead to demonstration of benchscale (5 kWe) microchannel fuel processing system with fuel cell possible as soon as FY2001

Steam Reformer Fuel Processing System

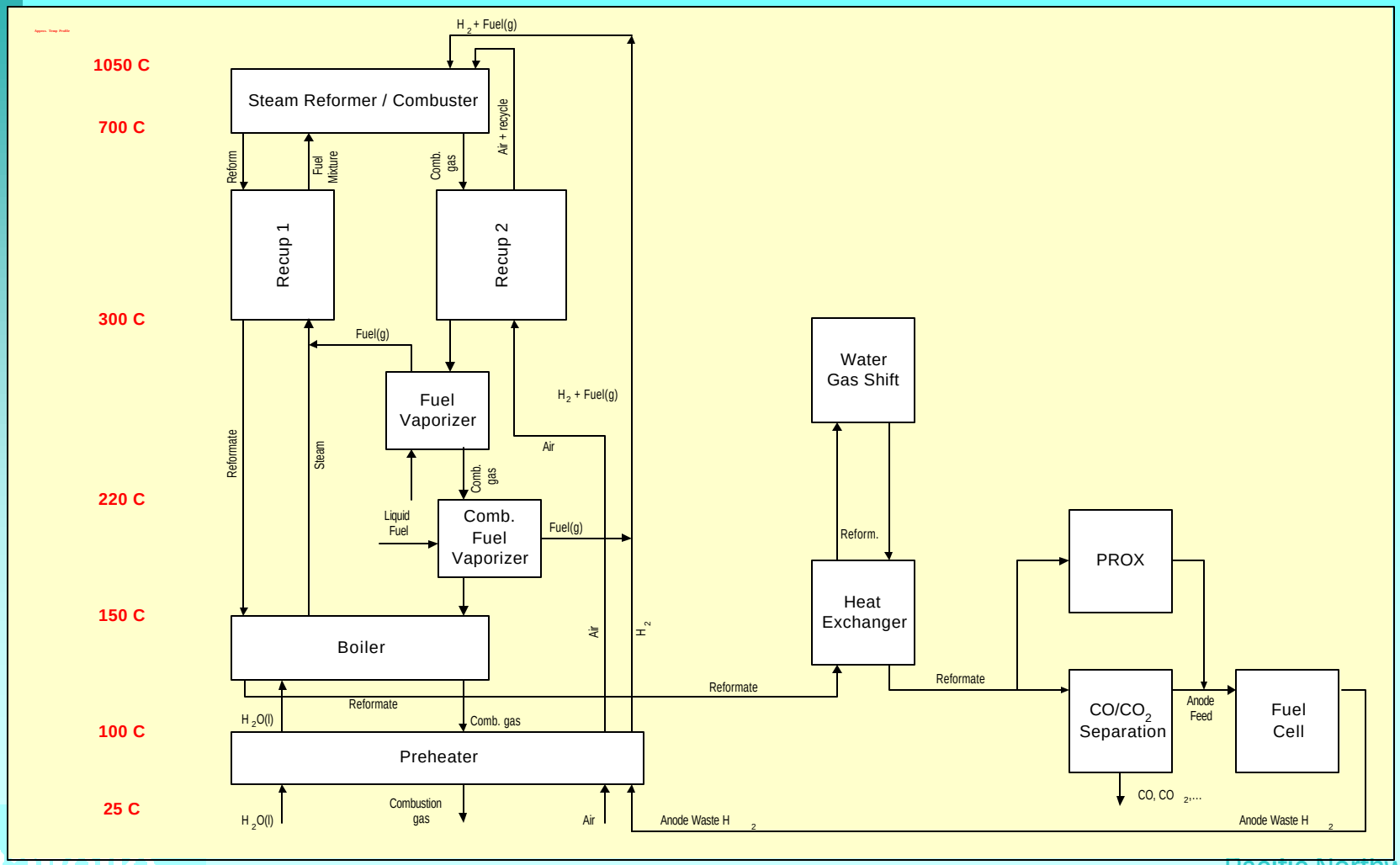
Conceptual Design -- 20 kWe size

APPENDIX I -- FY99 REFERENCES

- Madson, DW, PM Martin, DC Stewart, AY Tonkovich, GL Roberts and M White, Fabrication of Microchannel Chemical Reactors and Separation Systems, 3rd International Conference on Microreaction Technology, AICHE/DECHEMA, Frankfurt Germany, April 1999
- Tegrotenhuis, WE, RJ Cameron, VV Vishwanathan, and RS Wegeng, Solvent Extraction and Gas Absorption using Microchannel Contactors, 3rd International Conference on Microreaction Technology, Frankfurt Germany, AICHE/DECHEMA, April 1999
- Tonkovich, AY, JL Zilka, MJ Lamont, Y Wang, and RS Wegeng, Microchannel Reactors for Fuel Processing Applications, I. Water Gas Shift Reactor, presented at Bi-Annual International Symposium of Chemical Reaction Engineering, Newport Beach, CA, September 1998. Also accepted for publication in Chemical Engineering Science, 1999.
- Tonkovich, AY, JL Zilka, MJ Lamont, S Fitzgerald, DP Vanderwiel, Y Wang, and RS Wegeng, Microchannel Reactors for Fuel Processors, 3rd International Conference on Microreaction Technology, AICHE/DECHEMA, Frankfurt Germany, April 1999
- Wegeng, RS, MK Drost, and DL Brenchley, Process Intensification through Miniaturization, 3rd International Conference on Microreaction Technology, AICHE/DECHEMA, Frankfurt Germany, April 1999

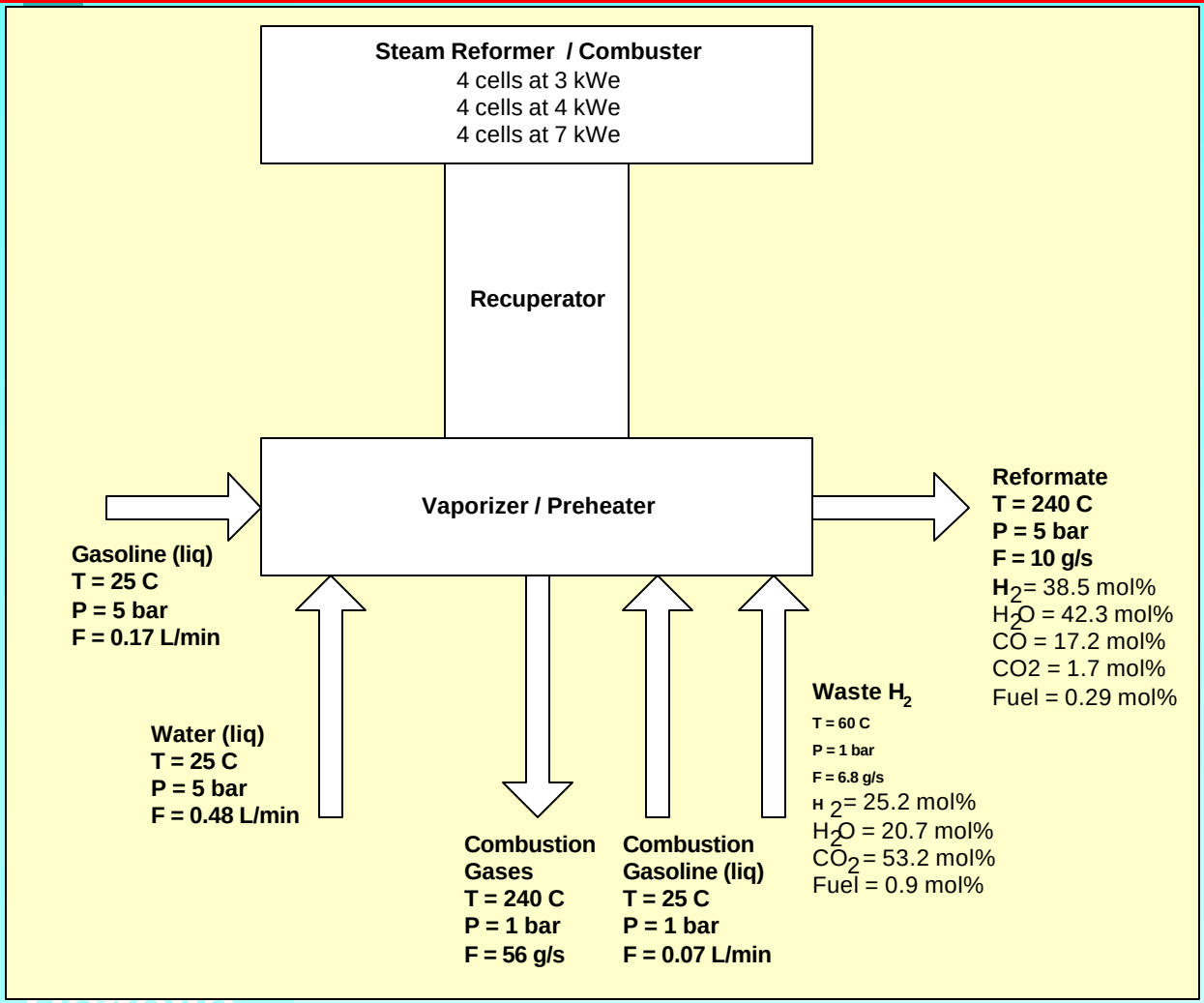
APPENDIX II -- Draft Functional Specifications

Level 0: Process Layout



APPENDIX II -- Draft Functional Specifications

Level 1: 800 C Steam Reformer Subsystem



Requirements:

- Hardware ≤ 4 L
- 50 kWe power output
- Combustion temp < 1010 C

Performance:

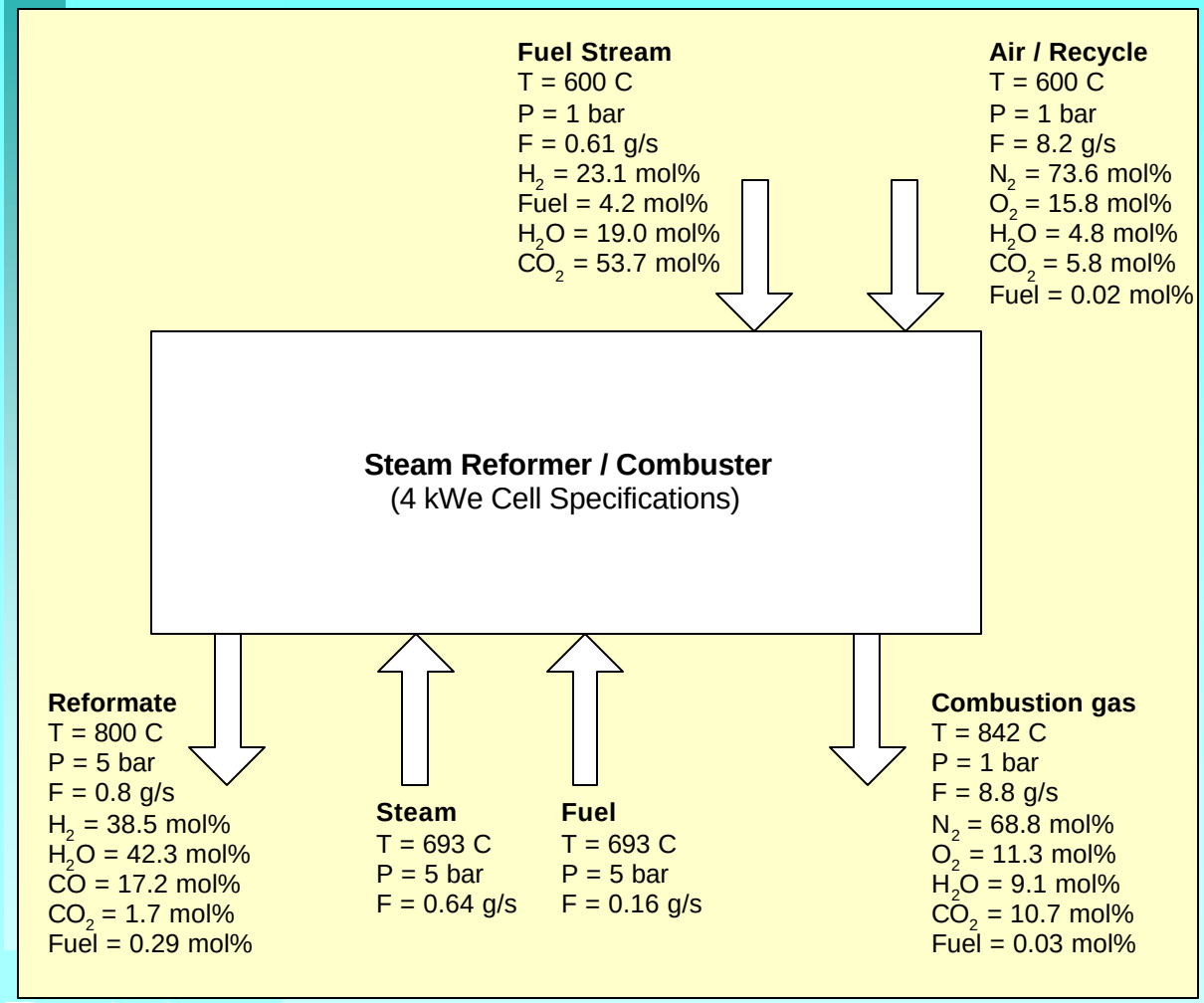
- 40.8% Fuel efficiency
- 81.5% SR fuel conversion efficiency

Assumptions:

- 0.8v Fuel Cell
- 85% H2 Utilization
- 90% WGS Conversion

APPENDIX II -- Draft Functional Specifications

Level 2: 800 C Steam Reformer / Combustor



Assumptions:

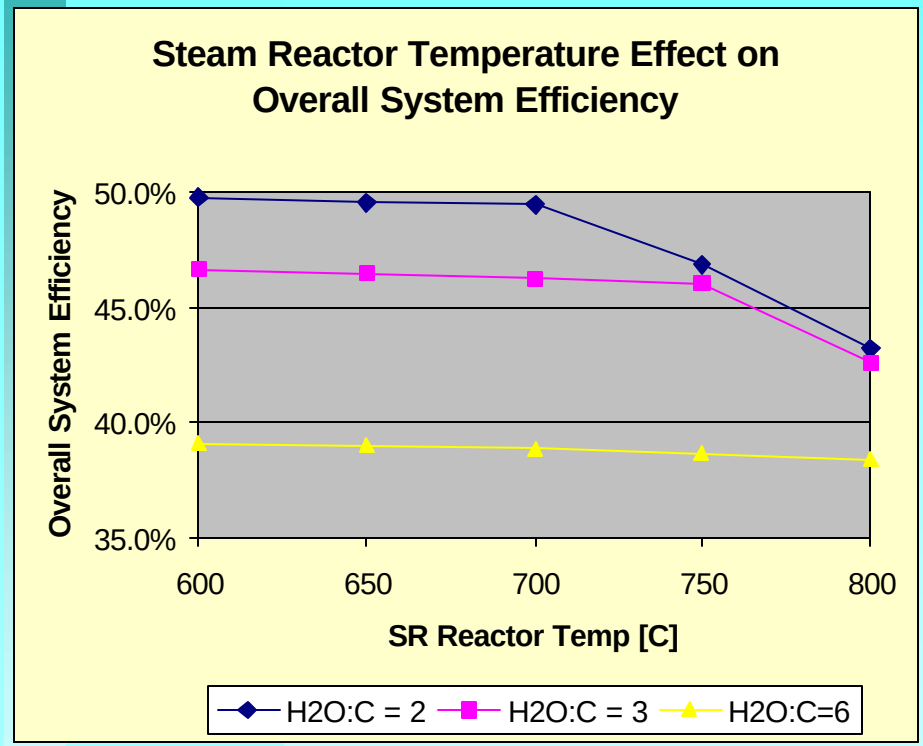
- 0.8v Fuel
- 85% H2 Utilization
- 90% WGS Conversion
- Combustion temp < 1010 C

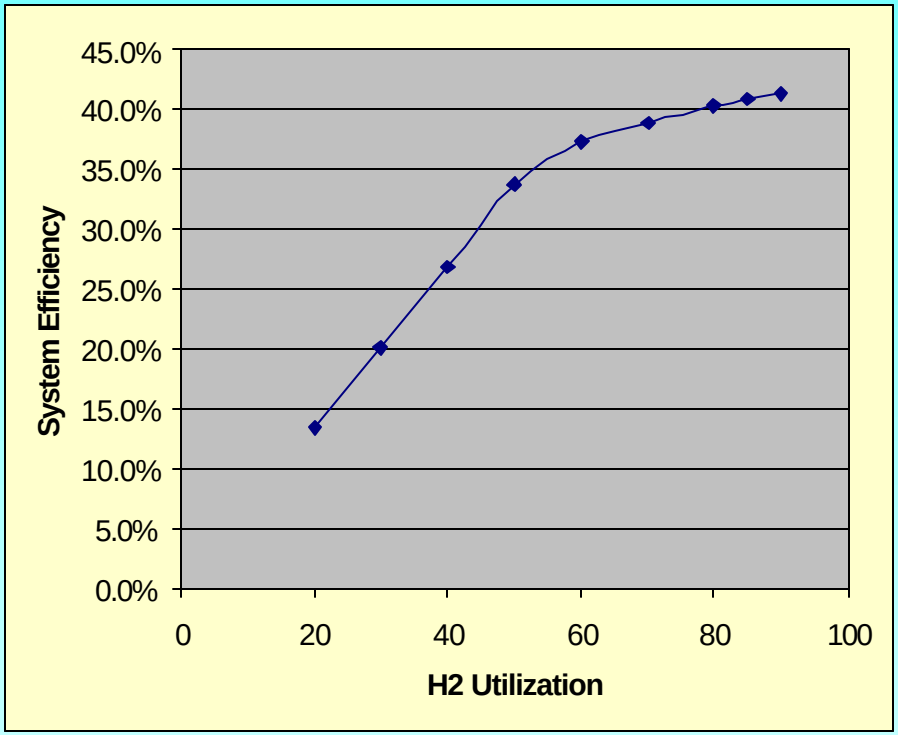
Initial design basis:

- 90% Fuel Conversion
- 3:1 H2O:C ratio
- CO/CO2 = 10 in product
- 800 C Reactor Temp.
- 90% fuel combustion
- 100% H_2 combustion
- 80% Equiv HEx Effectiveness

- Total Steam Reformer/ Combustor Hardware Volume (all cells) ≤ 1.7 L

APPENDIX II -- Reaction Temperature and Steam:Carbon Ratio Effects





Substantial advantages would be associated with lower temperature catalysts and lower steam to carbon ratios