Fuel Processors for PEM Fuel Cells


University of Michigan
College of Engineering
May 25, 2004

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Project Objectives

• Develop high performance, low-cost materials
  - High capacity sulfur adsorbents for liquid fuels
  - High activity and durable Autothermal Reforming (ATR), Water Gas Shift (WGS) and Preferential Oxidation (PrOx) catalysts
• Design and demonstrate microreactors employing high performance catalysts
• Design and demonstrate microvaporizer/combustor
• Design and demonstrate thermally integrated microsystem-based fuel processors
• Evaluate system cost

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Total Budget (as of March, 2004)

Year 4
$1,418,201

Year 1
$975,000

Year 2
$975,000

Year 3
$1,950,000

<table>
<thead>
<tr>
<th></th>
<th>DoE</th>
<th>Cost-Share</th>
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<tbody>
<tr>
<td>Received</td>
<td>1,250k</td>
<td>517k</td>
</tr>
<tr>
<td>Due</td>
<td>1,750k</td>
<td>383k</td>
</tr>
<tr>
<td></td>
<td>41%</td>
<td>22%</td>
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Fuel Processor (Fuel Cell) Technical Barriers

- Fuel Processor Startup/Transient Operation
  - Improved catalysts, sorbents and reactors
  - Thermal integration
  - Decreased unit operations

- Durability
  - Improved impurity tolerance
  - Improved resistance to coking and sintering

- Emissions and Environmental Issues

- Hydrogen Purification/CO Cleanup
  - Improved catalysts, sorbents and reactors

- Fuel Processor System Integration and Efficiency

- Cost
  - Improved catalysts, sorbents and reactors
  - Integration and decreased unit operations

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## Fuel Processor (Fuel Cell) Technical Targets

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Units</th>
<th>Current Status (2003)</th>
<th>Target for Year:</th>
<th></th>
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<tbody>
<tr>
<td>Energy efficiency</td>
<td>%</td>
<td>78</td>
<td>78</td>
<td>80</td>
</tr>
<tr>
<td>Power density</td>
<td>W/L</td>
<td>700</td>
<td>700</td>
<td>800</td>
</tr>
<tr>
<td>Specific power</td>
<td>W/kg</td>
<td>600</td>
<td>700</td>
<td>800</td>
</tr>
<tr>
<td>Cost</td>
<td>$/kWe</td>
<td>65</td>
<td>25</td>
<td>10</td>
</tr>
<tr>
<td>Cold startup time to max power</td>
<td>min</td>
<td>TBD</td>
<td>2.0</td>
<td>1.0</td>
</tr>
<tr>
<td>@ -20 °C ambient temperature</td>
<td>min</td>
<td>&lt;10</td>
<td>&lt;1</td>
<td>&lt;0.5</td>
</tr>
<tr>
<td>@ +20 °C ambient temperature</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Transient response (10% to 90% power)</td>
<td>sec</td>
<td>15</td>
<td>5</td>
<td>1</td>
</tr>
<tr>
<td>Emissions</td>
<td></td>
<td>&lt;Tier 2 Bin 5</td>
<td>&lt;Tier 2 Bin 5</td>
<td>&lt;Tier 2 Bin 5</td>
</tr>
<tr>
<td>Durability</td>
<td>hours</td>
<td>2000</td>
<td>4000</td>
<td>5000</td>
</tr>
<tr>
<td>Survivability</td>
<td>°C</td>
<td>TBD</td>
<td>-30</td>
<td>-40</td>
</tr>
<tr>
<td>CO content in product stream</td>
<td>ppm</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Steady state</td>
<td>ppm</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>Transient</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>H₂S content in product stream</td>
<td>ppb</td>
<td>&lt;200</td>
<td>&lt;50</td>
<td>&lt;10</td>
</tr>
<tr>
<td>NH₃ content in product stream</td>
<td>ppm</td>
<td>&lt;10</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
</tr>
</tbody>
</table>

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High Performance Materials + High Degree of Integration = Microsystems

Project Director: Levi Thompson (ltt@umich.edu)
Co-PIs: Gulari, Savage, Schwank & Yang (ChE);
Assanis, Im, Ni & Wooldridge (ME);
Dahm & Powell (Aero)
Subcontractors: Ricardo, Inc. (MI); Osram Sylvania;
IMM (Germany); MesoFuel (NM)

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Project Safety

• Preliminary Identification of Safety Vulnerabilities (e.g. FMEA, HAZOP)
• System Safety Assessment
• Risk Mitigation Plan
• Safety Performance Assessment
• Communications Plan
Project Timeline

Phase I: Components
- Design and Modeling
  - Desulfurizer Demonstration
  - Microreactor Demonstrations
  - Microcombustor/microvaporizer Demonstration
  - Microchannel System Development

Phase II: 1 kW Processor
- System Design and Modeling
  - System Fabrication

Phase III: ≤10 kW Processor
- System Evaluation

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Project Timeline

Phase I: Components

11/01-10-02 11/-2-10/-3 11/03-10/04 11/04-10/05

Phase II: 1 kW Processor

Phase III: ≤10 kW Processor

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**π-Complexation Mechanism:**

- Cu ions occupy faujasite 6-ring windows sites. Thiophene approaches site.
- σ-donation of thiophene π-electrons to the 4s orbital of Cu(I) or Ni(II)
- d-π* backdonation of electrons from 3d orbitals of Cu(I) or Ni(II) to π* orbitals of thiophene

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Sulfur Adsorber Prototype

- Three Sorbent Layers
  - Activated Carbon (12.4 wt%)
  - Activated Alumina (23 wt%)
  - Ni(II)-Y (64.6 wt%)
- Gasoline Rate: 50 mL/hr
- Equivalent H₂ Output: 2.8 moles/hr (100 W)
- Effluent Concentration: ~ 0.3 ppmw sulfur
- Operation Cycle: 9-10 hrs

Yang et al., U.S. and foreign patents applied.

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Microreactors

- Materials of Construction
  - Silicon Microfabrication
  - Micromachined Metals
  - Low Temperature Co-Fired Ceramics (LTCC)

- Metal Microreactors
  - 1\textsuperscript{st} Generation (GEN1) Micro-reactor
    - Design and Fabrication
  - 2\textsuperscript{nd} Generation (GEN2) Micro-reactor
    - Design Overview and Achievements

- Semi-solid Forming (SSF) Process

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GEN2 Prototype Design

• Flexible design
• Assembled reactor module is 77 x 64 x 54 mm (25 stacks)

Fabricated Parts

Assembled module

Heater
Gasket retainer
Foam
Gasket
Separation wall

Core Layers

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Breadboard System

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ATR Prototype Results
(100 \text{ We})

Experimental Conditions: \( \text{H}_2\text{O}/\text{C} = 2.0, \text{O}/\text{C} = 1.0 \)
Reactor Skin Temperature: 590 °C; Reactor Exit Temperature: 385 °C
1.5 SLPM air, 0.6 mL/min Iso-octane, 1.1 mL/min H\text{2}O

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Minimal Coke Deposition

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WGS Prototype Results

- Temperature: 240°C
- Flow rate: 40 ccm (1 Wₑ)
- GHSV: 53,333 h⁻¹
- Feed composition

<table>
<thead>
<tr>
<th>Component</th>
<th>Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO</td>
<td>10%</td>
</tr>
<tr>
<td>H₂O</td>
<td>31%</td>
</tr>
<tr>
<td>CO₂</td>
<td>6%</td>
</tr>
<tr>
<td>H₂</td>
<td>39%</td>
</tr>
<tr>
<td>N₂</td>
<td>15%</td>
</tr>
</tbody>
</table>

Conversion (%) vs. Time (hr):

- Packed Bed (120 mg)
- Single Foam (300 mg)

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WGS Prototype Results (100 We)

3.75 l/min

Conversion (%)

Time (min)

Dual: 340 & 290 °C
Single: 330-390 °C

CO = 6.1%

20 Channels

340 °C

CO ~ 1.1%

15 Channels

290 °C

CO < 0.6%

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PrOx Prototype Results

- 4 % Pt-Al₂O₃ sol-slurry hybrid washcoat
- WHSV = 50 lit hr⁻¹ g-cat⁻¹
- Increased catalyst loading of ~250 mg/foam
- Inlet stream compositions (simulated WGS exhaust):
  - CO : 0.79 – 0.81 %
  - O₂ : 0.81 – 1.19 %
  - CO₂ : 14.91 – 15.28 %
  - H₂ : 30.58 – 31.32 %
  - H₂O : 15.54 %
  - N₂ : 36.23 – 36.99 %
PrOx Prototype Results

Performance of assembled PrOx module

Conversion/Selectivity (%)

Temperature (°C)

02: CO ratio = 1.0

220

02: CO ratio = 1.5

200

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Catalytic Tailgas Combustor Prototype

Burner Characteristics:

- 100 W nominal capacity mesoscale burner
- 80 ppi Pt-coated FeCrAlloy metal foam
- 8.0 L/min tailgas low-H₂ surrogate flow rate

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Catalytic Tailgas Burner and Heat Exchanger Prototype

- Performance tests conducted for 1.5% - 8% H₂ concentrations
- Current test results show single-sided efficiencies of 35-45%
- Double-sided efficiencies anticipated in 65-80% range

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# GEN2 100 W<sub>e</sub> Prototype Design

## Table

<table>
<thead>
<tr>
<th></th>
<th>Vap/Com</th>
<th>ATR</th>
<th>WGS</th>
<th>PrOx</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Temperature (°C)</strong></td>
<td>450</td>
<td>600</td>
<td>340</td>
<td>290</td>
</tr>
<tr>
<td><strong>Modules</strong></td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td><strong>Catalyst Type</strong></td>
<td>Ni/CeZrO&lt;sub&gt;2&lt;/sub&gt;</td>
<td>Au/CeO&lt;sub&gt;2&lt;/sub&gt;</td>
<td>Au/CeO&lt;sub&gt;2&lt;/sub&gt;</td>
<td>Pt/Al&lt;sub&gt;2&lt;/sub&gt;O&lt;sub&gt;3&lt;/sub&gt;</td>
</tr>
<tr>
<td><strong>Catalyst Weight (g)</strong></td>
<td>1.5</td>
<td>6</td>
<td>4.5</td>
<td>2.4</td>
</tr>
<tr>
<td><strong>No. of Foam cores</strong></td>
<td>10</td>
<td>20</td>
<td>15</td>
<td>30</td>
</tr>
<tr>
<td><strong>Foam Volume (cc)</strong></td>
<td>4</td>
<td>8</td>
<td>6</td>
<td>12</td>
</tr>
<tr>
<td><strong>Power Density (W/L)</strong>*</td>
<td><strong>Based on Foam</strong></td>
<td>5,500</td>
<td>25,000</td>
<td>7,142</td>
</tr>
<tr>
<td></td>
<td><strong>Target</strong></td>
<td>5,882</td>
<td>10,417</td>
<td>2,525</td>
</tr>
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Interactions and Collaborations

- **Osram Sylvania (some IP transfer):** Joel Christian - scale up of catalysts
- **Ricardo:** Marc Wiseman - system optimization and cost analysis
- **Mesofuel:** Doyle Miller - heat exchanger design and fabrication
- **IMM:** Volker Hessel - reactor design optimization

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Responses to Previous Year Reviewers’ Comments

- Capacity of Cu(I) zeolite too low
- Coking of Ni-based ATR catalysts
- Verify performance of WGS catalysts
- Bottoms up approach
- Slow progress in developing microreactors
- Minimal involvement by companies
- Microprocessor work appears to be similar to PNNL
- Recommendations: Sulfur-tolerant ATR and hot gas sulfur sorbent

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Future Work

• Remainder of FY03
  - Increase module power densities
    • Increase catalyst loading and utilization
    • Decrease parasitic weight (reactor and foam)
  - Assemble 100 W breadboard fuel processor
  - Evaluate cost and final size
  - Estimate start-up time

• FY04 (through end of 2004)
  - Demonstrate integrated module
  - Assemble 1 kW breadboard fuel processor

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Stack Level Integration

Flow Path

<table>
<thead>
<tr>
<th>Flow Path</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>ATR</td>
<td>650</td>
</tr>
<tr>
<td>By-Pass</td>
<td>400</td>
</tr>
<tr>
<td>HtEx</td>
<td>650</td>
</tr>
<tr>
<td>200</td>
<td>HtEx</td>
</tr>
<tr>
<td>HTS</td>
<td>450</td>
</tr>
<tr>
<td>HtEx</td>
<td>100</td>
</tr>
<tr>
<td>LTS</td>
<td>300</td>
</tr>
<tr>
<td>100</td>
<td>HtEx</td>
</tr>
<tr>
<td>150</td>
<td>PrOx</td>
</tr>
<tr>
<td>HtEx</td>
<td>RT</td>
</tr>
</tbody>
</table>

Ceramic Insulating Layer

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Thank You

High Performance Materials + High Degree of Integration + Microsystems

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