Carbon Composite Bipolar Plate for PEM Fuel Cells

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Hydrogen and Fuel Cells Merit Review Meeting
Berkeley, California May 19-22, 2003
Objective

To develop a slurry-molded, carbon fiber material with a carbon chemical vapor infiltrated (CVI) sealed surface as a bipolar plate that would meet cost and property goals.

<table>
<thead>
<tr>
<th>Property</th>
<th>Specification</th>
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</thead>
<tbody>
<tr>
<td>Bulk Conductivity</td>
<td>&gt; 100 S/cm</td>
</tr>
<tr>
<td>H₂ permeability</td>
<td>&lt;2 x 10⁻⁶ cm³/cm²-sec</td>
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<tr>
<td>Corrosion rate</td>
<td>&lt;16 µA/cm²</td>
</tr>
<tr>
<td>Cost</td>
<td>&lt;$10/kW</td>
</tr>
</tbody>
</table>
Approach

• Bipolar plate utilize carbon/carbon concept
• Preform is slurry-molded carbon fibers
  – similar to paper or felt production
  – fibers ~100 μm plus filler particles
  – features stamped/embossed into preform
• CVI with carbon
  – seals and makes hermetic high-density surfaces
  – provides continuous, high-conductivity material
Advantages of Approach

• Preforms prepared from slurry-molded carbon fibers
  – net shape process/press-in features
  – process can be continuous (i.e., papermaking)
  – low-cost materials
• Appropriate surfaces sealed via deposition of carbon
  – high-conductivity (graphitic) carbon coating all surfaces
  – infiltration makes component fully integral
  – potential for continuous or semi-batch processing
• Negligible impurities/poisons with minimal corrosion
• Strength and toughness of carbon/carbon
• Very light weight (about half that of other approaches)
• Potential for integral diffuser/catalyst support, therefore, lower ohmic losses
## Timeline of Project Accomplishments

<table>
<thead>
<tr>
<th>Date</th>
<th>Event Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>March 1998</td>
<td>Project Initiated</td>
</tr>
<tr>
<td>July 1998</td>
<td>3-cm disks with machined flowfield</td>
</tr>
<tr>
<td>March 1998</td>
<td>High conductivity meas.</td>
</tr>
<tr>
<td>June 1999</td>
<td>100-cm² plates with machined flowfield</td>
</tr>
<tr>
<td>June 1999</td>
<td>Good cell performance</td>
</tr>
<tr>
<td>June 2000</td>
<td>Light weight demonstrated</td>
</tr>
<tr>
<td>June 2000</td>
<td>High strength/toughness</td>
</tr>
<tr>
<td>June 2000</td>
<td>Initial samples to industry</td>
</tr>
<tr>
<td>June 2000</td>
<td>Pressed preforms</td>
</tr>
<tr>
<td>June 2000</td>
<td>Freeze/thaw testing</td>
</tr>
<tr>
<td>June 2000</td>
<td>Low corrosion meas.</td>
</tr>
<tr>
<td>June 2000</td>
<td>Multiple samples to industry</td>
</tr>
<tr>
<td>April 2001</td>
<td>License signed with Povair Fuel Cell Technologies</td>
</tr>
<tr>
<td>June 2001</td>
<td>Two-sided 100-cm² plates with pressed flowfield evaluated</td>
</tr>
<tr>
<td>June 2001</td>
<td>Thermal conductivity meas.</td>
</tr>
<tr>
<td>June 2001</td>
<td>Resistivity/polarization meas.</td>
</tr>
<tr>
<td>June 2001</td>
<td>Measured mechanical properties of plate material</td>
</tr>
<tr>
<td>June 2001</td>
<td>Significantly improved wetting of bipolar plate surface</td>
</tr>
<tr>
<td>June 2001</td>
<td>Determined surface roughness of finished plate</td>
</tr>
<tr>
<td>June 2002</td>
<td>Developed a series of bipolar plate materials/specimens that meet specific fuel cell manufacturer's design needs</td>
</tr>
<tr>
<td>June 2002</td>
<td>With licensee, developed manufacturing capability for specific bipolar plate designs through sample preparation, process modeling, and consultation</td>
</tr>
<tr>
<td>Sept. 2002</td>
<td>Developed carbon composite with graphite particulate filler</td>
</tr>
<tr>
<td>Sept. 2003</td>
<td>UT-BATTENELLE</td>
</tr>
</tbody>
</table>
Current Accomplishments

• With licensee, have reduced thickness of plates from 2.5 mm to 1.5 mm
• Determined influence of CVI temperature on depth of infiltration
• Further characterized and measured mechanical properties of carbon composite plate material
• Determined electronic properties, including effect of surface roughness on resistance
• Developed initial model of chemical vapor infiltration process that is scalable to production – model demonstrated value through guidance on flow control improving uniformity
Carbon Composite Plate Fabrication

**Slurry Molding of Preforms**

- 100 µm Milled Fiber
- Phenolic Durez® Resin
- 400 mesh Sieve Screen

**Vacuum Molding Station**

- Molded Preform

**Press or Stamp Features**

**CVI Carbon**

- EXHAUST GASES TO VACUUM SYSTEM
- COMPONENTS BEING INFILTRATED
- REACTANT GASES IN

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Oak Ridge National Laboratory
U.S. Department of Energy
Porvair Preform Material Used in Study of Infiltration Depth as a Function of Reactor Conditions and Position

CVI Conditions:

Temperature
High Rate - 1500°C
Low Rate - 1350°C

Flow
1 sccm methane
2.5 sccm argon

Pressure
4 kPa

CVI Reactor Chamber Showing Position of Samples
Effect of CVI Conditions/Material on Density Profile

Image analysis reveals density gradients through plate thickness more pronounced for:
- ORNL material, which has no filler particles and thus higher green state porosity
- More rapid infiltration (higher infiltration temperature) in ORNL material

Mosaic of Optical Images of Plate Cross-Section (Porvair Material)

Through Thickness Density

Reactor Position/Conditions
- Upstream-Left, 4 hours at 1500°C
- Downstream-Right, 4 hours at 1500°C
- Upstream-Left, 24 hours at 1350°C
- Downstream-Right, 24 hours at 1350°C
- Uninfiltrated
- ORNL material

SEM Image of a Fracture Surface of an Infiltrated Bipolar Plate Produced From Porvair Preform Material Indicating That Carbon Deposition Is Limited Largely to the Exposed Surfaces
2-D Computer Model for CVI Reactor

- Commercial fluid dynamics code (CFD-ACE from CFD Research, Huntsville, AL) Cross-section reactor/preform geometry
- Kinetics and transport parameters from literature and from experiments at ORNL
- Finite-volume method with 136,000 element structured grid
- Boundary conditions match experiment
- Steady-state solution for gas flow

**Gas Channels**
- 3.0 mm above preform
- 3.5 mm below preform

**Inlet**
- 1500 sccm CH$_4$
- 2500 sccm Ar

**Preform**
- 2.0 mm thick
- 140 mm with flow
- 121 mm cross flow

**Outlet**
- 8000 Pa

1753 K
1773 K
Original Inlet Design Yielded Non-Uniform Flow

- Single recirculation cell near inlet
- Parabolic, laminar flow above/below preform
- Higher velocity above preform
Installation of a Nozzle Yielded Uniform Flow

- Nozzle turns and centers inlet flow
- Top/bottom recirculation cells
- Uniform velocity above/below preform
Electronic Properties Are Weakly Related to Surface Roughness

- 2-probe measurements are sums of all contact resistances
- Surface roughness does not correlate closely with contact resistance
- Sample no. 4 was not infiltrated and thus has fewer “interfaces” to offer resistance
- Sample no. 6 was polished and indicates a significantly lower contact resistance than that of unpolished samples (all others)
- Bulk conductivity as measured by a 4-probe technique varies little with sample preparation

<table>
<thead>
<tr>
<th>Porvair Material</th>
<th>Infiltration Treatment</th>
<th>Surface Roughness Rz µm</th>
<th>Resistance 2 Probe DC Ω</th>
<th>Conductivity 4 Probe V/I S/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. 1 - V162C-L2 A</td>
<td>4 hours at 1500°C</td>
<td>8.83</td>
<td>1.86</td>
<td>365</td>
</tr>
<tr>
<td>No. 2 - V1631-R1-C</td>
<td>24 hours at 1350°C</td>
<td>12.9</td>
<td>1.37</td>
<td>417</td>
</tr>
<tr>
<td>No. 3 - V1631-R1-P</td>
<td>Uninfiltrated</td>
<td>13.5</td>
<td>0.86</td>
<td>369</td>
</tr>
<tr>
<td>No. 4 - V162C-L2 D-1</td>
<td>4 hours at 1500°C</td>
<td>10.2</td>
<td>2.16</td>
<td>386</td>
</tr>
<tr>
<td>No. 5 - V163AR1A-1</td>
<td>24 hours at 1350°C</td>
<td>14.3</td>
<td>1.57</td>
<td>383</td>
</tr>
<tr>
<td>No. 6 - V163AR1-B</td>
<td>24 hours at 1350°C</td>
<td>2.9</td>
<td>0.84</td>
<td>408</td>
</tr>
</tbody>
</table>
Porvair Material Has Reasonable Strength and Apparent Notch Insensitivity

- The resistance of materials to crack propagation can be evaluated by the notch sensitivity test
- Test specimens with holes of various sizes are subjected to monotonic tensile loading
- Ultimate strength (un-notched) is at a minimum the zero intercept of the strength vs. 2a/w

![Image]

Loading rate: 5 \( \mu \)m/s

Uninfiltrated Material
Industrial Interactions

- Supporting licensee Porvair Fuel Cell Technology in scaling up technology
- Porvair has been awarded a $6.1 M DOE program for scaling and applying the carbon composite bipolar plate technology
- Other companies evaluating plates, with some under development
Continuing Development and Technology Transfer of Carbon Composite Bipolar Plates

- Milestone FY 03 - Develop a carbon composite bipolar plate component with reduced thickness and optimized mechanical properties. (Sept. 2003)
- Develop CVI process model that is scalable to pilot and production facilities
- Determine electronic properties as a function of surface roughness
- Milestone FY 04 - Support continued scale-up of the licensee’s operation through modeling of fabrication processes. (Sept. 2004)
Some Advisory Panel Comments From Last Year and Responses

• “Demonstrate durability of materials manufactured by industrial partner in larger hardware”
  – Mechanical property measurements were made to better characterize the material which showed notch insensitivity (good toughness)
  – Partner has produced significant numbers of plates that have performed in customer hardware

• “Providing different hydrophobicity could be interesting area to explore”
  – Licensee does not give this a high priority

• “Test method extensively”
  – Developing mechanical and electrical property test results