High-Temperature Viscous Sealing Glasses for Solid Oxide Fuel Cells

Cheol-Woon Kim, Cynthia L. Schwartz, Joe Szabo, Kevin Barr, and Ted E. Day
MO-SCI Corporation, Rolla, MO; ckim@mo-sci.com
Richard K. Brow and Zhongzhi Tang
Department of Materials Science and Engineering
Missouri University of Science & Engineering, Rolla, MO; brow@mst.edu

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Phase I Technical Objectives
1. Develop viscous sealing glass compositions that can be used at 650-850°C
   a. Requisite Thermal and Physical Properties
      - Long-term stability in viscosity (800-850°C)
      - Stable, slow crystallization kinetics
      - Viscosity: 10⁷ Pa·s (10⁷ Poise) - 10¹² Pa·s (10¹³ Poise)
      - CTE (parallel plate) 10-15 x 10⁻⁶/°C (YSZ-SS441)
   b. Preparation of a total of 45 compositions and measured properties (TLiq, Tg, Ts, and CTE) of all of the 45 compositions
   c. Preparations to establish sealing properties of the glass in electrolyte bilayers (Ni/YSZ - supported YSZ electrolyte bilayers)

2. Conduct hermetic sealing tests
   SOFC Materials: Aluminized SS441/YSZ supported YSZ electrolyte bilayers supplied by PNNL

3. Characterize thermochemical reactions
   a. Volatilization of glass components
   b. Interfacial reaction

Viscosity Measurements
<table>
<thead>
<tr>
<th>Glass</th>
<th>TLiq (°C)</th>
<th>Tg (°C)</th>
<th>Ts (°C)</th>
<th>CTE x 10⁶/°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glass 2</td>
<td>1150</td>
<td>1150</td>
<td>1100</td>
<td>10⁻⁶</td>
</tr>
<tr>
<td>Glass 4</td>
<td>1150</td>
<td>1150</td>
<td>1100</td>
<td>10⁻⁶</td>
</tr>
<tr>
<td>Glass 28</td>
<td>1150</td>
<td>1150</td>
<td>1100</td>
<td>10⁻⁶</td>
</tr>
</tbody>
</table>

Viscosity-cont.
- High temperature measurements (viscosity range 1-10⁸ Pa·s) by plate-plate technique
- Low temperature measurements (viscosity range 10⁷-10¹² Pa·s) by the cylinder compression (parallel plate) technique
- Viscosity-temperature curves fit using the Corning viscosity model (UC Mauro, PNAS, 2005)

Reactivity Characterization
- Pastees were made from glass powders (~45 µm) and acetone, and used to bond Ni/YSZ to lower and SS441 (substrates from PNNL)
- Sandwich seals held in air at 650°C for 500 hours
- Seals were cross-sectioned, polished, then analyzed by analytical SEM
- General observations:
  - Pastees were made from glass powders (~45 µm) and acetone, and used to bond Ni/YSZ to lower and SS441 (substrates from PNNL)
  - Sandwich seals held in air at 650°C for 500 hours
  - Seals were cross-sectioned, polished, then analyzed by analytical SEM
- Relative atomic concentrations (EDS): 68.9 ± 1, 68.9 ± 1, 68.9 ± 1

Phase I Conclusions
1. Glass compositions from the Ba-borosilicate (or Ba-borate) system have been identified that possess the properties desired for 'self-sealing' behavior:
   - Liquidus temperatures as low as possible (< 800°C)
   - High temperature measurements (viscosity range 1-10⁸ Pa·s)
   - Low temperature measurements (viscosity range 10⁷-10¹² Pa·s)
   - Viscosity-temperature curves fit using the Corning viscosity model (UC Mauro, PNAS, 2005)

2. Hermetic seals have been made between Ni/YSZ and SS441
   - Survive 75 thermal cycles between 750°C and room temperature
   - Next to increase the glass (sub-Tg) CTE for better match to other SOFC components
   - Need to redesign hermeticity testing rig to evaluate 'self-sealing' behavior

3. Provided foundation for Phase II
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