Metal Interconnects for Solid Oxide Fuel Cell Power Systems

SECA Core Technology Program
Ceramatec, Inc.

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DOE-NETL

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Technical Issues Addressed

- **Technical Requirements for Metal Interconnects**
  - CTE match
  - No gas permeation
  - High temperature corrosion resistance
  - Scale conductivity
  - Scale adhesion
  - Stability in atmosphere (physical, chemical, microstructure, conductivity)
  - Stability against electrode/bond layer (poisoning effect)
  - Electrical contact with cells
  - Thermal cycle capability
Risks and Challenges

• Chromia formers preferred to provide a conductive scale
  - Continued scale growth during operation
    - Increased electrical resistance
    - Loss of adhesion
    - Porosity at interface
  - Chromium vaporization
    - Electrode Poisoning
  - Electrode compatibility
    - High resistance phase formation with electrode cations (spinel)
R&D Objectives

• **Controlled growth of conductive scale to achieve**
  - Electronic conductivity in scale
  - Low cation (metal) and anion (oxygen) diffusivity
  - Good adhesion (‘native’ scale)

• **Application of conductive layers**
  - Application techniques
    - Thermal spraying (INEEL FWP)
Approach

- **Alloy Selection** *(Fe-Cr based ferritic SS)*
  - CTE Match, Conductive scale (chromia former)
  - Choice of minor alloying elements

- **Surface Treatment & Oxidation**
  - Growth of selective oxide scale
    - Control P, T, X_i and t
  - Scale characterization
Assessment Criteria

- Weight gain with time at temperature
- Scale thickness, morphology, composition
- Electrical resistance with thermal cycles
- Exposure to relevant atmospheres
Untreated Metal - 500 hrs at 750°C

Intensity (arbitrary units)

<table>
<thead>
<tr>
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<th>Angle Two Theta</th>
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<tbody>
<tr>
<td>untreated metal</td>
<td></td>
</tr>
<tr>
<td>wet air (wet/wet air)</td>
<td></td>
</tr>
<tr>
<td>dry air (dry/wet air)</td>
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</tbody>
</table>

untreated metal
wet air (wet/wet air)
dry air (dry/wet air)
Treated Metal - 500 hrs at 750°C
Scale Resistance in H₂/H₂O
Scale Morphology

- Untreated dry air: 750°C 200 hours
- Untreated wet air: 750°C 500 hours
- Treated dry air: 750°C 500 hours
- Treated wet air: 750°C 500 hours
- Treated dry air of dual atm.: 750°C 500 hours
Phase I Summary

- **Commercial stainless steel characterized for applicability**
- **Demonstrated an appropriate treatment to achieve**
  - Low resistance interface
  - Stable morphology
  - Proper scale composition
  - Thermal cycle capability (up to five) demonstrated
- **Selected optimization parameters for additional improvements in properties**
Phase II Tasks

- **Treatment process optimization**
- **Scale growth kinetic parameter evaluation (TGA)**
- **Contact layer**
  - Application process
  - Chemical interaction
- **SOFC relevant atmosphere**
  - Effects carbon species, dual atmosphere, sulfur
- **Cr evaporation**
Stack Evaluation (SBIR Project)

- A treatment process with low resistance in coupon tests evaluated (screen printed contact layer)
- Post-test: Sr-Cr rich phase on La(Sr)CoO$_3$ cathode
Phase II Evaluations

• **Approaches to surface treatment optimization**
  - Modify intrinsic scale
    - surface treatment and thermal process
    - Objective: Limit scale growth
  - Apply extrinsic layer
    - low Cr activity composition (~LaCrO$_3$)
    - Objective: Limit Cr evaporation
  - Combine the two layers
    - graded composition
# Experimental Arrangement

<table>
<thead>
<tr>
<th>TGA</th>
<th>Coupon Couples</th>
<th>Dual atm. couples</th>
<th>Stack</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single atmosphere</td>
<td>Single atmosphere</td>
<td>Dual atmosphere</td>
<td>Dual atmosphere</td>
</tr>
<tr>
<td>No contact / electrode layers</td>
<td>Contact layer</td>
<td>Continuous current</td>
<td>Continuous current</td>
</tr>
<tr>
<td>No current</td>
<td>Intermittent current</td>
<td>Contact layer</td>
<td>Contact/electrode layers</td>
</tr>
<tr>
<td>Isothermal</td>
<td>Isothermal</td>
<td>Isothermal</td>
<td>In-plane thermal, current density gradients</td>
</tr>
</tbody>
</table>

![Diagram of coupon couples and stack](image)
Dual atmosphere couples

- Dual atmosphere
- Contact layer
- Continuous load (constant current)
- Dual atmosphere
- No contact layer
- No current
- Air atmosphere
- No contact layer
- No current

1x1 cm coupon on a larger (3.5x3.5 cm) blank
Identical treatment on mating surfaces
Contact layer: cobaltite
Extrinsic Layer - Dual atmosphere

Extrinsic Layer
Air/H$_2$(H$_2$O) at 750°C
Load ~ 200mA/cm$^2$

Time (hrs)
Resistance, milliohm.cm$^2$
Rig Validation: Graded Coating in air

Graded coating 750°C

Coupon couple in air (includes two thermal cycles)

Coupon couple in air using 'dual atm' set-up

Resistance, milliohm.cm²

Time (hrs)
Graded Coating: Dual atmosphere

Graded coating
Air/H$_2$(H$_2$O) at 750C
Load=200mA/cm$^2$

Resistance, milliohm.cm$^2$

Time (hrs)

0 50 100 150 200 250 300 350 400 450
Extrinsic layer

- Thick scale
- Poor adhesion
- Thin scale
- Scale loss?
- Thick scale under contact layer
- Sr-Cr rich interface

200 mA/cm², ~350 hrs
140 milliohm.cm²

air

fuel
Graded Coating

200 mA/cm², ~300 hrs
7 milliohm.cm²

- Thin scale (1 µm) in both regions
- No Sr-Cr rich phase
Graded coating - dual atm.

- 6 µm scale
- Influence of dual atm. away from the region?
- Thin scale
- Flakey?
- Thin scale under contact layer
- No Sr-Cr phase at the scale

200 mA/cm², ~400 hrs
15 milliohm.cm²
LSGM Short Stack Test (SBIR)

- **Changes Made**
  - Modified surface treatment of interconnect
    (low resistance in air and not optimal in dual atm)
  - Lower Sr-containing contact layer
  - Coated fuel and air feed tubes; metal manifolds
    - Environmental Barrier Coating from DOE-SBIR
  - Under layer coating
  - Functional oxide coating
  - Cr alloy
3-Cell LSGM Stack

- 500 hour test planned

3LSGM422: 800°C
Fuel Utilization ~ 54%
0.7 V/cell

<table>
<thead>
<tr>
<th>Time, Hours</th>
<th>Power, Watts</th>
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<tbody>
<tr>
<td>0</td>
<td>25</td>
</tr>
<tr>
<td>20</td>
<td>~28.5</td>
</tr>
<tr>
<td>40</td>
<td>~27</td>
</tr>
<tr>
<td>60</td>
<td>~26.5</td>
</tr>
<tr>
<td>80</td>
<td>~26</td>
</tr>
<tr>
<td>100</td>
<td>~25.5</td>
</tr>
<tr>
<td>120</td>
<td>~25</td>
</tr>
<tr>
<td>140</td>
<td>~24.5</td>
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Summary

- **New test arrangement**
  - Allows resistance measurement in dual atm. exposure
  - Allows continuous load

- **Graded coating provides low resistance and thinner oxide scale in initial tests**

- **Additional work planned**
  - Effect of coating variations
  - Effect of current density

- **Stack test validation in parallel programs**
Applicability to Industrial Teams

- **Present approaches by the SECA industrial teams**
  - Electrolyte supported co-fired planar
  - Anode supported planar
  - Cathode supported re-designed tubular
  - Anode supported thin cylinder

- **Technical applicability**
  - Metal interconnects
  - High temperature current collectors, bus bars

- **Commercial applicability**
  - Low cost materials and processes
  - Process flexibility to suit materials chemistry of mating surfaces
Activities for the next 12 Months

- **Additional improvements to pre-treatment process**
  - Graded coating

- **Determine scale growth kinetics**
  - TGA - a new unit with controlled atmosphere capability installed this month

- **Process development for conductive coating**
  - FWP at INEEL

- **Scale - coating interaction study**

- **Effect of SOFC relevant atmospheres**
  - Hydrocarbon fuel (simulated reformed methane)
  - S-bearing fuel (up to 5 ppm H$_2$S)
Acknowledgement

- Ceramatec team
- DOE-SECA project managers
- CTP teams
- SECA industrial teams