Progress on Performance, Durability, and Reliability of LGFCS SOFC Technology
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Outline

● LGFCS Program Overview
  ● Manufacturing Automation
  ● Integrated String Test
● Performance and Durability Testing
  ● Block Scale Results
  ● Subscale Results
● Cell technology status and optimization for commercial product
  ● Primary Interconnect (PIC)
  ● Anode Development
  ● Cathode Development
  ● Candidate materials for future cell technology
● Structural Reliability
LG Production Research Institute developing automated strip assembly equipment

- Eliminate manual operations of:
  - Edge glassing
  - SIC wire application
  - Subassembly (manifolds to printed substrates)
LGFCS Integrated String Test Program

- Test of a 220 kW system demonstrator incorporating all key subsystems
  - Fuel Processing
  - Pressurized Generator Module including turbogenerator
  - Power Electronics
  - Pipeline natural gas and grid connection
  - 3Q-4Q 2014 commissioning/4Q testing

- SECA program supports further improvements in cell/stack lifetime up through Block-scale (19 kW) testing
Outdoor Test Pad under construction for receipt of major IST components.
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● Structural Reliability
Block Test replicates Plant Configuration

- Block metric testing matches full system cycle, components (less TG and recuperator), operation and boundary conditions

**LGFCs NG “Dry Cycle” Configuration**

- Thermally self-sustaining insulation system
- Anode and cathode ejectors
- Reformers and heat exchangers
- Off-gas burners
- System control methodology
Phase 1 Test – Performance and Durability

- Initial ASR = 0.34 ohm-cm²
- Fuel Utilization System equivalent ~78.8%
- Anode Loop Efficiency (system equivalent) ~61.1%
- Current density = 380 mA/cm² (NOC)
- Test ended prematurely due to secondary interconnect failure.

860°C Nominal Temperature, 6.4 Bara Pressure
Power Degradation = 0.5%/1000 hrs
Phase II Test met SECA Target

Power Degradation = 1.1%/1000 hours
(Rate higher than Phase I due to wider temperature range)

1. Air heater TC fault. Repaired and test restarted after 30 h (A)
2. Main fuel mass flow controller unstable at high flow rate. Ran flow controller at lower stable conditions. Repaired and test restarted after 145 h (B)
3. Current limiter set to 20.0 A on HMI. Limited current to 93.5% of design point. Correct during end-of-year facility maintenance shutdown at 335 h (C)
4. Blockage in gas analysis line at stack inlet after approx. 500 h. Unable to determine CH₄ conversion in reformer and stack inlet composition. Assumed equilibrium CH₄ conversion thereafter for ASR calculations
5. IV/OPV deviation alarm on HMI set too close to rig operating point. Resulted in trip and controlled shutdown at 1245 h (D).
Block Performance as Expected

(Current, fuel flow, and temperature drop forced to match data)

Data at 343 hours
Phase II Block Testing Summary

- Initial performance within expectations
  - 18.8 kW output at design point current
  - ASR ~ 0.345 ohm-cm² for strips in design temperature range
- Power degradation 1.1%/1000 hours
  - Achieves SECA target of 1.5%/1000 hrs at 3000 hours
- Degradation higher than subscale and Phase I Test
  - Wider temperature range than for Phase I test. Parts of the block are outside of design temperature range.
  - Degradation observed due to printing defect which has since been addressed and correction validated
  - Degradation of strips near design average temperature was similar to Phase I test
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Bundle Testing Shows Consistent Performance

- Agrees well with model from pentacell data
Testing on desulfurized natural gas to confirm performance on real world fuel

- Fueled from small scale SCSO* system using pipeline natural gas
- Initial performance and degradation are good

*SCSO = Selective Catalytic Sulfur Oxidation
Subscale Durability Map Demonstrates Trends and Guides Cell Development

- Performance mapped over operating envelope
- Detailed performance separation achieved
- Durability performance confirmed at larger scales

ASR @ 16K hrs = 0.465 ohm-cm²

Projected Points

Shaded area represents performance envelope which meets product requirements
Consistent performance across scales validates durability testing approach

- 860°C Nominal Temperature
- 6.4 Bara except for PBT5
- Wider temperature range brings out print defect - since corrected and validated

Differences between block and subscale:
1. Cathode air and temperature distribution
2. Bundle-to-bundle fuel distribution
3. BOP chrome, insulation contaminants
4. Manufacturing variability

T1314 Data is excluding 1/4-strips outside of Design Temperature Range
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● Structural Reliability
Two-Year Life was Demonstrated for Primary Interconnect (PIC) Design and Materials

- Cathode: 8.5 - 11.3% O₂, 1.2%, H₂O, bal N₂
- Anode: reformate fuel
- Elapsed Time, hours: 0 - 16000
- PIC ASR change from start to the end is negligible

Graph shows the comparison of PIC ASR for PCT63B: 860°C and PCT89B: 800°C. The graph also indicates the presence of anode, cathode, anode current collector, and cathode current collector.
Modified PIC Shows Improved Performance

- Further mitigate degradation mechanism for 3-5 year life
- Higher conductivity PIC materials and design modification
- PIC ASR is as low as 0.03 ohm-cm² and stable up to 4300 hrs
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Baseline Anode Microstructural Change (ODOD*)

- Baseline anode (Ni-YSZ) tested at 925°C for 4000 hrs
- Significant microstructure change
  - Porosity increases and metal phase depletion at anode/ACC interface

* This work is supported by Ohio Department of Development
Single Layer Anode for Improved Durability

- 1980hrs in ambient + 3006 hrs in simulated system conditions

![Graph showing cell ASR over elapsed time](image)

- **PCT150B1: SL Anode (thick), 4 bara**
- **SCT6-101B: SL Anode, 1bar**
- **SCT6-101B, SL Anode, 4bar**

Test rig related issue

Bundle outlet fuel, 12%O2-1.2%H2O, 925°C,
Detailed Microstructural Analysis of Single Layer Anode by 3D Reconstruction (ODOD*)

- Metal phase generally is uniform across the anode

![Graph showing volume fraction of pores, ceramic, and metal across distance from interfaces.](image)

- SCT6-101B
- 925°C, 5000 hrs, bundle outlet

- ≈ 1 µm from Electrolyte/Anode interface
- ≈ 1 µm from Anode/substrate interface

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Lower Anode ASR Demonstrated for Future Cell Technology

- High thermal expansion substrate allows use of higher conductivity anode current collecting material
- Thinner substrate reduces fuel diffusion resistance

![Graph showing ASR reduction and temperature over time](image)
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Cathode Microstructural Change after 16,000 hrs

- Accumulated free MnOx at electrolyte interface at 800°C
- At 860°C also see densification

PCT89B: 800C/16,000 hrs

PCT63B: 860C/16,000 hrs
Approaches for LSM-Based Cathode Optimization

- Evaluation of different cathode compositions – LSM and ionic phase
  - Thermodynamic consideration
  - Second phase/impurities
- Doped LM/LSM for microstructural stability at high temperatures
Cathode Optimization to Eliminate Free MnOx

- Free MnOx was identified only in baseline cathode pellet

Baseline

Candidate A

Candidate B

Candidate C

As-fired cathode pellets

Baseline

Candidate A

Candidate B

Candidate C

Cathode pellets aged for 1000 hours at high temperature
Accelerated Testing Method Developed for Cathode Screening

- Allows screening of cathodes in 500 hrs
- Accelerated testing indicates next generation cathode is more stable
- Results repeated for both baseline and next generation cathodes

500 hrs at accelerated testing conditions
Next Generation Cathodes Show Improved Durability (800°C)

- For baseline degradation in 2nd year attributed to cathode (process at ~100 Hz)
- The change in the AC impedance peak not clearly linked to electrode microstructure change
Next Generation Cathodes Show Improved Durability (900°C)

- Tests at higher temperature show potentially better performance for next generation cathodes compared to baseline

**900°C: Cell ASR vs Time**

- Bundle outlet fuel, 12%O2-1.2%H2O, 4.0 bara
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Bundle ASR Improvement by using High Conductivity Via Material

- Average bundle ASR is 0.25 ohm-cm² vs. 0.28 ohm-cm² for baseline bundle without increasing the cost.
Expected Performance for Next Generation Anode, Cathode, and PIC Technology

- Average repeat unit (RU) ASR of 0.25 ohm-cm² at 1 bara
- Projected to 0.22 ohm-cm² at 4 bara and ≤0.20 ohm-cm² using higher conductive ACC & thinner substrate
Need for Alternate Cathode Driven by Desire for Lower Operating Temperatures

- Focusing on nickelate cathodes due to its CTE, lower ASR and activation energy
Modified Nickelate Cathode Shows Significant Interface Improvement

- Fine microstructure/more triple phase boundaries
- Stronger interface may improve both durability and reliability
- The key challenge of phase stability needs further effort

Button cells tested for 150 hrs at 790°C
Nickelate Cathode Shows Promising Short Term Durability

- Nickelates also show low-temperature steam effect
- Lower degradation after stabilization for optimized nickelate cathode
Improved ASR for Future Cell Technologies Gives Operating Flexibility

- Block operating temperature: 810-910°C for baseline
- Allow fuel cell system operation at lower temperature and/or improved efficiency

Anode: bundle inlet, cathode: dry air, 1 bara
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Structural Reliability Considerations

- LGFCS utilizes all ceramic strip
- Weibull probabilistic analysis required for all components
- Evaluating reliability against fast-fracture/infant mortality and time-dependent mechanisms:
  - Initial emphasis on understanding slow-crack growth of porous substrate
  - Now adding focus on properties of dense ceramics and glass-ceramic based joints

Bundle assembly (~350W): Serial fuel and current flow

~17"H x 12"W x 16"D

Block assembly (~20kW): 5 strips of 12 fuel-parallel bundles
LGFCS Substrate Exhibiting High $K_{th}$ for Slow Crack Growth

Typical Slow Crack Growth Curve for Ceramics Showing Three Regions
($I =$ threshold, $II =$ linear, $III =$ instability)

Time to failure can be predicted:

$$t = \frac{2(K_{IC}^{2-n} - K_{th}^{2-n})}{(2-n)A\sigma^2Y^2}$$

Porous MMA substrate
900°C
50% H$_2$O, 48.3% N$_2$, 1.7% H$_2$

$K_{th}$ is $\sim 70\%$ of $K_{IC}$

$K_{th} = 1.6$ MPa$\sqrt{m}$
Summary

- Stack power degradation rate met SECA Phase 2 target
- Accelerated testing technique developed under this program proving to be a good tool for cathode material screening for long term stability
- Next generation/optimized electrodes being identified under long-term durability testing to advance to 5-year service life
- Improved ASR for future cell technologies gives operating flexibility and allow fuel cell system operation at lower temperature and/or improved efficiency
- Porous MMA substrate material showing promising properties to benefit long-term structural reliability
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