the ENERGY lab
PROJECT FACTS
Fuel Cells

Low Cost Modular SOFC Development—Pacific Northwest National Laboratory

Background

The U.S. Department of Energy (DOE) National Energy Technology Laboratory (NETL) has a mission to advance energy options to fuel our economy, strengthen our security, and improve our environment. With the Solid Oxide Fuel Cells (SOFCs) program and systems coordination from the Solid State Energy Conversion Alliance (SECA), DOE/NETL is leading the research, development, and demonstration of SOFCs for coal-fueled central generation power systems that enable low cost, high efficiency, near-zero emissions and water usage, and carbon dioxide (CO_2) capture.

Pacific Northwest National Laboratory's (PNNL) project was selected to acquire the fundamental knowledge and understanding that will facilitate research and development to enhance electrochemical performance under a diversified portfolio for anode-electrolyte-cathode development. This project is providing vital support that addresses applied technological issues common to multiple SECA Industry Teams developing SOFC systems.

PNNL has provided multi-year project support to develop advanced SOFC component materials and computational tools. In particular, interconnection components for both atmospheric and pressurized operation are being developed. Emphasis is placed on evaluation and development of materials and manufacturing processes for various SOFC components such as electrolytes, anodes, cathodes, cell-to-cell interconnections, and seals.

Project Description

This project will accelerate the development of reliable, low-cost SOFC power generation systems capable of operation on coal derived fuels by developing advanced SOFC component materials and computational tools. Interconnection components for both atmospheric and pressurized operation will be developed. Important to this work is the evaluation and development of materials and manufacturing processes for various SOFC components (electrolyte, anode, cathode, cell-to-cell interconnections, and seals) to support higher performance and robustness, less degradation, and lower cost. In addition, this project will address the effects of coal contaminants on SOFCs and how to mitigate any deleterious effects through upstream clean-up and modification of SOFC components.

This project will also identify, develop, test, validate, and optimize cost-effective cell and stack components, materials, and fabrication techniques; develop advanced computational tools and capabilities for cell and stack design analysis and optimization; test cells exposed to coal contaminants and complete analysis of the results, both post-test and using thermodynamic software; and develop and evaluate new interconnect compositions appropriate for pressurized SOFCs.

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PARTNERS

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PROJECT DURATION

Start Date	End Date
10/01/1999	09/30/2013

(annual continuations)

COST

Total Project Value
$52,889,667

DOE/Non-DOE Share
$52,889,667 / $0

AWARD NUMBER

FWP40552

U.S. DEPARTMENT OF ENERGY

Goals and Objectives

The primary goal of this project is to develop, manufacture, and evaluate advanced SOFC component materials and computational tools.

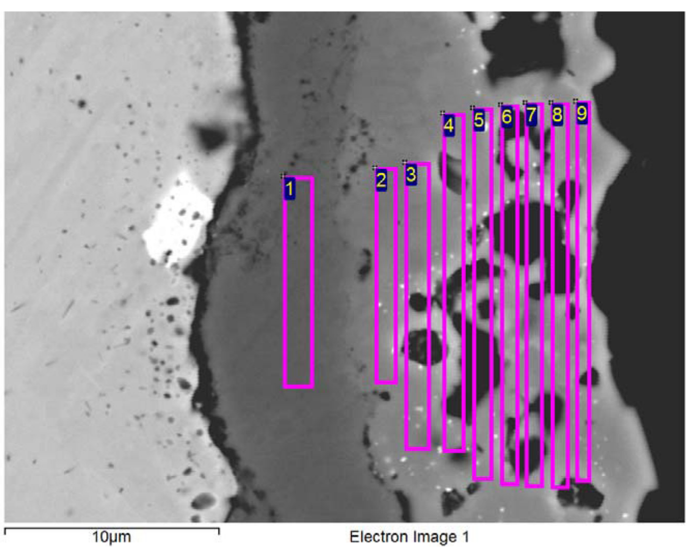
Objectives are as follows:

- Develop, test, and optimize high-performance, reliable cell and stack component materials and fabrication techniques for low-cost, reliable SOFC stacks. Develop a SOFC stack test fixture for use by PNNL and other participants.
- Develop and utilize computational techniques to assist in the development and optimization of modular SOFC systems. The electrochemical aspect of the modeling capability will be enhanced by the incorporation of instrumented stack data provided by an Industry Team. Model predictions of thermal profiles will be compared with other independent models and used to guide stack development. The American Society of Mechanical Engineers (ASME) SOFC design basis document will be updated as needed.
- Collaborate with Oak Ridge National Laboratory (ORNL) to obtain necessary material properties to support SOFC modeling. Develop materials and fabrication techniques for low-cost modular SOFC stacks. This task focuses on the testing and analyses of various materials under development (viscous and refractory seals, interconnect alloys and coatings, and cathode contact aids) in a stack environment. PNNL and ORNL will collaborate to design, experimentally evaluate, and develop seals based on viscous glass.
- Research and development on fuel related technical topics of importance to operating SOFCs on gasified coal, such as the effects of impurities in gasified coal on SOFC anodes.
- Identify, prioritize, and coordinate SECA technical activities. The management of the program involves interactions with universities, national laboratories, and SECA Industry Team members to identify, prioritize, and coordinate technical activities and to disseminate results.

Accomplishments

- Commissioned second generation stack test fixture.
- Successfully assembled and tested 3-cell stack fixture for long-term (approximately 6,000 hours) evaluation/validation of manganese-cobalt (M-C) spinel and alumina coated interconnects.
- Demonstrated excellent electrical performance of M-C spinel-coated AISI 441 for over two years (20,000 hours).
- Evaluated performance of coatings with reduced (or zero) cobalt content.
- Optimized ultrasonic spray process for interconnect coatings. An ultrasonic spray process was evaluated and successfully developed for the application of cerium oxide-modified M-C coatings aluminization on metallic interconnects.
- Conducted a microscopic study which included evaluation of interconnect coatings using composition and phase analysis. It was determined after one year that the oxide scale under the coating consisted of a continuous matrix of titanium-doped chromium oxide with dispersed grains of manganese-chromium oxide. No evidence of impending degradation was found using high-resolution transmission electron microscopy provided by Carnegie Mellon.
- Completed the ASME SOFC design basis document. This living document, modeled after those published by the ASME, acts as a repository for the current best practices in the design of structurally-sound SOFC stacks. The baseline version of the design guide covers all major aspects of stack design and is supported by five appendices of technical reference material. The document will be reviewed every six months and updated on an as-needed basis to continually reflect the state-of-the-art in SOFC stack design.
- Demonstrated that sodium volatility from soda lime glass and potassium volatility from potassium silicate had no discernible effect on performance of lanthanum strontium manganite/yttria-stabilized zirconia (LSM/YSZ) composite cathodes and lanthanum strontium cobalt ferite (LSCF) cathodes.
- Identified new coal gas contaminant absorber materials. It was found that an absorber bed containing potassium carbonate completely captures phosphorus at temperatures of 350 degrees Celsius (°C) and above, and completely captures arsenic at temperatures of 600 °C and above. This represents a possible inexpensive option for removing these contaminants from coal gas.
- Investigated the role of cell voltage on contaminant attack. A parametric study of cell voltage and coal contaminants phosphorus, arsenic, sulfur, selenium, and hydrogen chloride on anode degradation was completed. No changes in the cell degradation rate at different operating voltages were observed in the presence of phosphorus, arsenic, and hydrogen chloride; however, distinctly different responses to sulfur and selenium were observed at different voltages.
- Transferred stack test fixture capability to NETL. Single-cell stack fixtures were delivered to and commissioned at NETL. This establishes the capability at NETL for independent parallel evaluation of materials and concepts for cells and stacks.
- Devitrifying “refractory” sealing glasses were optimized in terms of softening temperature, wetting behavior, and coefficient of thermal expansion.
- Evaluated a new sealing approach incorporating a glass which is compliant at SOFC operating temperatures through single and dual atmosphere testing. Compliant seals improve mechanical robustness of SOFC stacks by reducing residual stresses during stack operation and thermal cycling. Process optimization studies were performed to control pore size and volume fraction porosity. Additions of zirconia fillers to form a glass/zirconia composite can improve dimensional stability at operating temperature. Collaborative development efforts at PNNL and ORNL involving glass only and glass/zirconia particle or fiber composites were continued.

- Continued a long-term (>20,000 hours) study evaluating the effects of applying surface modifications to ferritic stainless steel interconnect materials prior to application of protective spinel coatings. Substantial improvement in scale adhesion and spallation resistance (compared to unmodified surfaces) can be achieved, particularly through surface blasting or grinding.
- Provided topical report to SECA industry teams summarizing benefits of select surface modifications on scale adhesion / spallation resistance of steel-based interconnects.
- Effects of humidity in air on LSM- and LSCF-based cathodes were quantified as a function of operating temperature and time.
- Performed high temp XRD analyses on working LSCF cathodes as a function of operating time, voltage, and temperature. Changes in the perovskite structure and overall phase assemblage were quantified.
- Developed and implemented a continuum damage-healing modeling framework to simulate the thermal-viscoelastic behavior of compliant glass seals in SOFC stacks using finite element analysis. Sensitivity studies on the seal design parameters were performed to evaluate the effects of material properties and operating conditions on seal mechanical behavior for the seal design engineering effort.



Spectrum	O	Si	Ti	Cr	Mn	Fe	Co	Ce
1	63.90	0.27	0.27	**35.38**		0.17		
2	61.16		0.27	**13.42**	11.65	1.90	11.59	
3	60.33	0.30	0.43	**7.72**	15.94	2.78	12.22	0.28
4	59.42		0.50	**5.52**	18.42	2.99	13.15	
5	57.68	0.60	0.51	**5.25**	19.30	3.09	13.28	0.29
6	58.89	0.68	0.44	**5.07**	18.84	2.81	12.78	0.48
7	57.05	0.75	0.48	**5.28**	19.79	2.87	13.21	0.56
8	57.30	0.41	0.49	**5.37**	19.59	2.96	13.53	0.36
9	61.84	0.42	0.43	**4.61**	17.30	2.67	12.50	0.24

SEM image and results from cross-sectional EDS analysis of M-C spinel-coated surface-blasted AISI 441 after oxidation at 800 °C for 20,000 hours (element concentrations in atomic %). Note excellent scale adhesion and limited transport of Cr into the coating (right side of image).

- Developed a modeling framework that automatically creates reduced order models (ROMs) for SOFC stacks. The framework interface guides the user through the analysis procedure, samples and interrogates the multi-parameter operating space using the detailed stack model, performs regression to generate the response surfaces, and implements the response surfaces into a ROM submodel for general use within system modeling software.
- Developed a user-friendly interface for the 2D SOFC-MP software to perform pre-processing and post-processing for SOFC stack evaluations. The pre-processing capability enables users to enter and modify SOFC model geometry, stack operating conditions, and simulation control parameters, while the post-processing capability helps users to visualize the solution results for various physical quantities across the stack model domain.
- Benchmarked the 2D SOFC-MP modeling tool against experimental data. Model simulations for ten cases with different fuel compositions and temperature boundary conditions were successfully verified with experimental thermocouple data from a state-of-the-art stack for H_2 and reforming fuel compositions.
- Developed models to investigate humidity effects on cathode performance. Micro-scale modeling is used to understand competition of water with LSM activity and kinetics of possible reactions, while meso-scale modeling is used to quantify the corresponding electrochemical degradation within the electrode microstructure.
- Modified the interconnect lifetime prediction methdology to be informed from interfacial indentation data of oxide scale-substrate interfaces with different surface modifications.

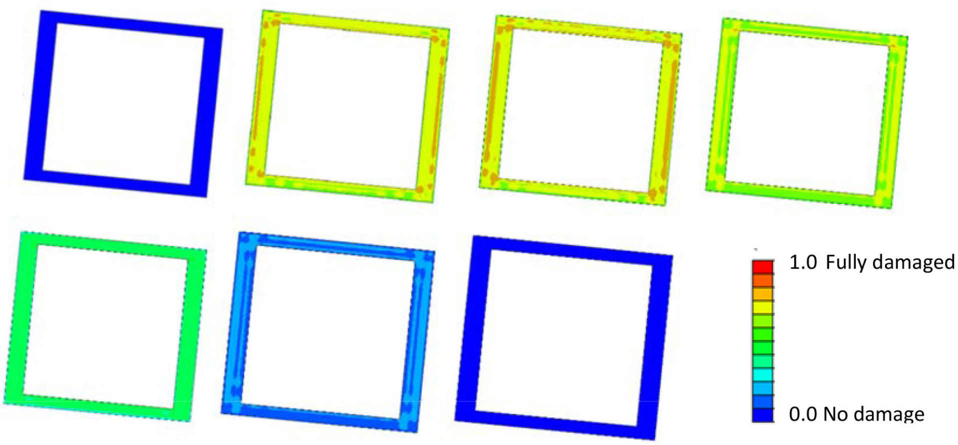
Post-test image of SCN-1 compliant glass sealed between aluminized AISI441 and an anode/electrolyte bilayer . No evidence of damage or leakage was observed after 12 thermal cycles between RT and 800 °C.

Benefits

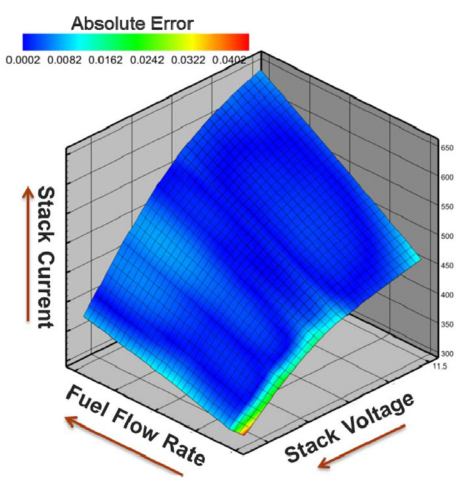
The PNNL project assists the SOFCs program in meeting its cost and performance targets by ensuring that SOFCs can achieve reliable operation over an extended operating life. The program will ultimately enable fuel cell-based near-zero emission coal and natural gas power plants with greatly reduced water requirements and the capability of capturing 97 percent or greater of carbon at costs not exceeding the typical cost of electricity available today. Achieving this goal will significantly impact the nation given the size of the market, the expected growth in energy demand, and the age of the existing power plant fleet. It will also provide the technology base to enable grid-independent distributed generation applications. Federal funding support of this research is appropriate given the game-changing nature of the technology accompanied by risks higher than the private sector initially can accept.

Interconnect research and development will improve low-cost metallic interconnects, coatings, and interfacial materials operating in the temperature range of 650 to 850 °C. This will minimize leading contributors to lifetime degradation and will address oxide growth, spallation, good electrical contact, and impact of volatile species on electrode performance. Electrode studies will improve the understanding of effects of humidity and contaminants on electrode microstructure, polarization, and resistance to support efforts to improve electrode stability and reliability during long term operation. Development of seals based on compliant glass will improve robustness of SOFC stacks towards thermal cycling and thermal gradients during operation.

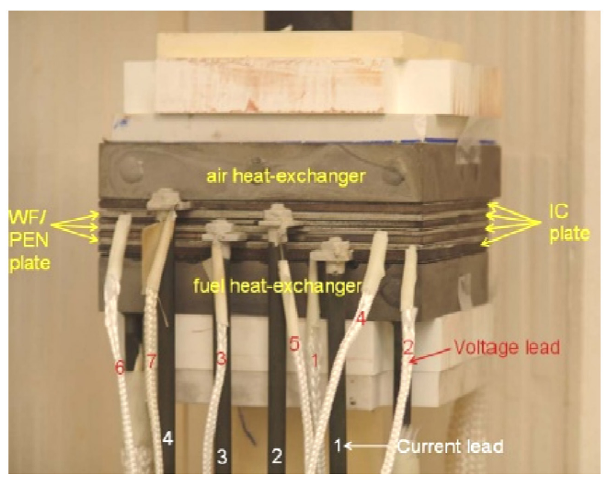
Modeling and Simulation R&D will create multi-physics models for fuel cell, stack, and system performance in order to define the optimal operating condition space, guide manufacturing, and lower costs. This will address fuel cell integrity over the required lifetime under all possible non-steady state conditions. Providing this necessary level of assurance experimentally is not cost-effective. Modeling of electrode, seal, and interconnect behavior will improve mechanistic understanding of causes of performance degradation and predict component lifetime.



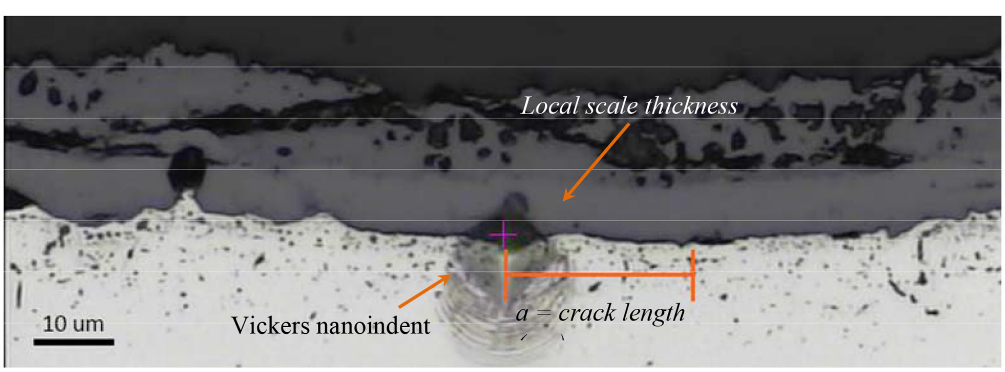
Progression of damage and healing of a viscous glass sealant during thermal-mechanical loading of an SOFC stack.



Response surface prediction and associated error for use in reduced order models (ROMs) of SOFC stack performance.



Stack test fixture used for evaluation/validation of new cell and stack materials under realistic stack environment conditions.



Use of pyramidal micro-indentation tests to characterize interfacial toughness for the interconnect scale.