

V.F.8 Affordable, High Performance, Intermediate Temperature Solid Oxide Fuel Cells

Bryan Blackburn (Primary Contact),
Thomas Langdo, Dong Ding, Lei Wang,
Luis Correa, and Colin Gore
Redox Power Systems
387 Technology Drive
College Park, MD 20742
Phone: (301) 314-1959
Email: bryan@redoxenergy.com

DOE Manager
David Peterson
Phone: (720) 356-1747
David.Peterson@ee.doe.gov

Contract Number: DE-EE0006735

Subcontractor
University of Maryland, College Park, MD

Project Start Date: October 1, 2014
Project End Date: March 31, 2016

- Demonstration of a bilayer electrolyte cell with OCP performance of ≥ 0.9 V in both a button cell and a larger 10 cm by 10 cm cell in operation at $\leq 600^\circ\text{C}$
- Demonstration of an advanced SOFC operating at $\leq 600^\circ\text{C}$ with a cell area specific resistance (ASR) $\leq 0.2 \Omega\text{-cm}^2$ and power density $\geq 1 \text{ W/cm}^2$

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Fuel Cell Technologies Office Multi-Year Research, Development, and Demonstration Plan:

- (A) Durability
- (B) Cost
- (C) Performance

Technical Targets

This project's goals are focused on stack improvements with increased power density for operation at intermediate temperatures ($\leq 600^\circ\text{C}$), which supports DOE technical targets. The results of this project can be applied to the design of advanced natural gas fueled power systems in the low and intermediate power range (1–100 kW_e). SOFCs with lower operating temperatures can result in simpler, more compact, and lightweight systems with longer lifetimes. Furthermore, higher power densities at lower temperatures result in reduced system and operating costs, while IT operation still maintains sufficient exhaust temperatures for efficient CHP applications. Such systems have the potential to meet or exceed the following 2020 DOE fuel cell technical targets:

- Equipment Cost: $\leq \$1,700/\text{kW}_{\text{avg}}$
- Transient Response (10–90% rated power): ≤ 2 minutes
- Degradation with cycling: $\leq 0.3\%/1,000$ h
- Electrical Efficiency: $\geq 45\%$
- CHP Efficiency: $\geq 90\%$

FY 2015 Accomplishments

- Integrated bilayer electrolyte mechanism into custom Redox multi-physics model for cell and stack, with validation of model using button cell data under different operating conditions (e.g., temperature)
- Demonstrated button cells with OCP ≥ 0.9 V at $\leq 600^\circ\text{C}$ using a gadolinium doped ceria (GDC)/erbium-stabilized bismuth oxide (ESB) bilayer

Overall Objectives

Redox Power Systems' overall objectives in this project are to improve the performance and durability of Redox solid oxide fuel cell (SOFC) technology through the:

- Development of an optimized bilayer electrolyte with increased open circuit potential (OCP) and thus greater fuel efficiency for natural gas fueled, intermediate temperature (IT) operation of $\sim 600^\circ\text{C}$.
- Optimization of compositions and microstructures for the cathode to increase power density, and the anode to improve carbon- and sulfur-tolerance in hydrocarbon fuels for IT operation.
- Use of a custom multiphysics model and advanced materials to optimize the performance of bilayer stack designs for IT operation.
- Creation of bilayer cell performance maps and demonstration of a $\sim 1 \text{ kW}_e$ stack for IT operation under combined heat and power (CHP) conditions with natural gas and minimal external reforming.

Fiscal Year (FY) 2015 Objectives

- Validation of multi-physics bilayer cell and stack model to experiment

- Demonstrated button cells with power densities exceeding 1.25 W/cm^2 at $\leq 600^\circ\text{C}$



INTRODUCTION

We have previously demonstrated a high power density SOFC technology using advanced materials in a novel bilayer electrolyte design with graded electrode structures at the button cell level. Redox has also been able to scale our base (single layer gadolinia doped ceria, or GDC, electrolyte) cells to a larger 10 cm by 10 cm production size and achieve a degree of performance similar to that of the button cells. Furthermore, we have scaled-up production of the ESB material that together with GDC comprises the bilayer electrolyte. However in addition to integrating the ESB/GDC bilayer into the larger format cells, significant improvements in cell performance/efficiency can still be achieved to make systems based on our technology more commercially viable with lower costs, higher efficiency, and superior durability. Moreover, integrated development of the anode for operation on readily available hydrocarbon fuels with a maximum degree of internal reforming is necessary to take full advantage of SOFC fuel flexibility. Achieving these advancements while maintaining high power density at intermediate temperatures ($\sim 600^\circ\text{C}$) will help us deploy this technology for distributed generation and CHP applications.

APPROACH

Redox's approach involves the synergistic use of two electrolyte materials in a bilayer structure to achieve superior performance (i.e., higher conductivity) at lower temperatures [1]. The first material is cerium oxide based, such as GDC, which has more than five times the ionic conductivity of conventional yttrium-stabilized zirconia (YSZ) at 600°C , but which, due to the electronic leakage that occurs in reducing environments like those in the fuel cell, can result in decreased cell efficiency. The second material is bismuth oxide based, such as ESB, which has 60 times the conductivity of YSZ at 600°C but is unstable in reducing environments. The bilayer electrolyte combines the cerium oxide and bismuth oxide layers (i.e., GDC/ESB) with the latter being situated on the cathode air side of the cell. In this case, the GDC protects the ESB from decomposing while the ESB blocks the electronic leakage in the GDC, thus boosting cell power and efficiency at lower operating temperatures. We have also developed new, bilayer-compatible cathode materials (e.g., composite lanthanum strontium manganite [LSM]-ESB) with exceptionally low ASR and are leveraging past work on infiltration of electrodes to enhance the performance of the cathode and anode. For the anode, catalyst infiltration can significantly increase power density and stability in the presence of hydrocarbon fuels like natural

gas, even in the presence of sulfur compounds like H_2S [2]. To assist in the development of cells and stacks for operation at $\sim 600^\circ\text{C}$, Redox makes use of our advanced multi-physics model that takes into account the unique thermochemical and physical properties of the Redox materials. This model is critical due to variations in conductivity and chemical activity of GDC as a function of temperature and effective oxygen partial pressure P_{O_2} , which varies significantly down the channel in SOFC anodes with increasing fuel utilization [3].

RESULTS

During FY 2015, Redox integrated the physics of the GDC/ESB bilayer into our custom single-channel SOFC multiphysics model. The bilayer model employs SOFC layers consisting of Ni-GDC and/or Ni-YSZ anodes, GDC and ESB bilayer electrolytes, and an LSM-ESB cathode. We used the model to simulate both button cells and stack components with outputs consisting of polarization curves, three-dimensional temperature profiles, and composition distributions, based on temperature dependent input kinetic parameters and electrical properties associated with the electrolyte and electrode materials. The bilayer model parameters were fit to the button cell data at 600°C , and validated by correctly predicting performance at 650°C (Figure 1). The only change made to the model input during validation was the operating temperature, which demonstrates that the charge-transfer reaction rate expressions and proposed kinetic parameters are adequate to predict the GDC/ESB bilayer cell performance under different conditions. This validation has also been confirmed with different data generated at higher temperatures and

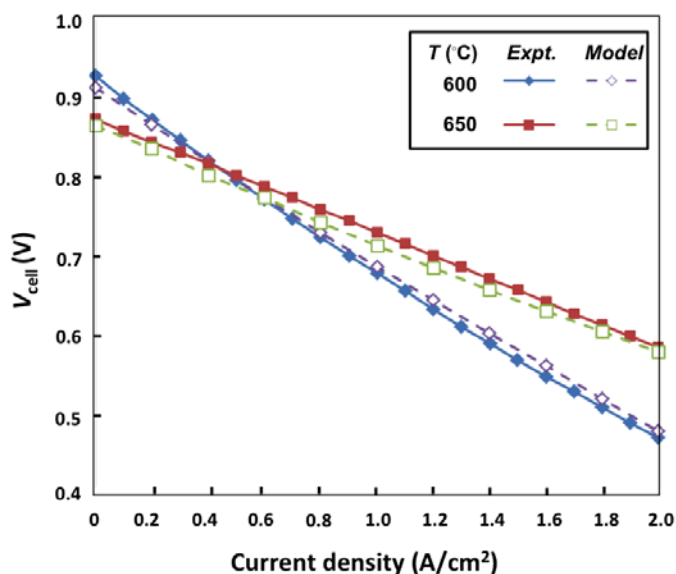


FIGURE 1. Comparison of predicted electrochemical performance of the stack model with experimental I-V curves at 600°C and 650°C

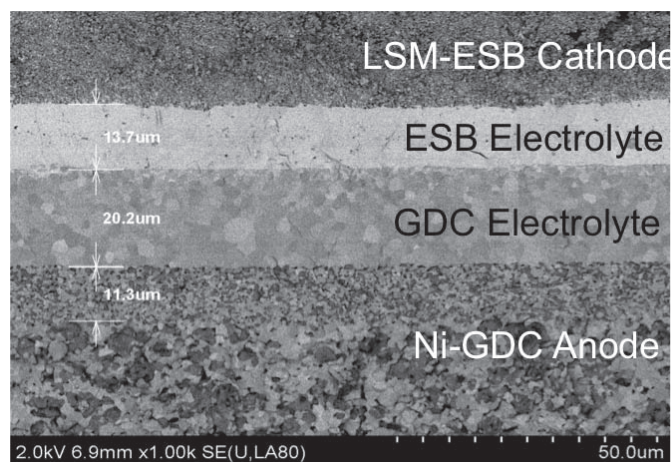


FIGURE 2. Example of SEM micrograph showing bilayer microstructure and geometry for cells

by predicting performance at lower temperatures, as well as with isothermal conditions for variations in the ESB and GDC thickness. The model also captures the kinetics of electrochemical and heterogeneous internal reforming reactions in the anode, and therefore now represents a complete multi-physics tool for cell/stack investigations.

In the first six months of this project, we adjusted the relative and total GDC/ESB bilayer thickness to improve OCP. Figure 2 shows an example scanning electrode microscopy (SEM) micrograph of a GDC/ESB bilayer cell used in the thickness optimization trials. Thicknesses were on the order of 20 μm for the GDC and $\sim 5\text{--}20\ \mu\text{m}$ for the ESB. Using these cells we successfully demonstrated a bilayer button cell operating on humidified hydrogen fuel and air (1 atm) with an OCP as high as 0.94 V at 600°C. As shown in Figure 3, we also demonstrated a button cell with an OCP of 0.93 V at 600°C and a maximum power density of 1.27 W/cm². The cell also had an ASR of 0.171 $\Omega\text{-cm}^2$. These very high power densities at intermediate operating temperatures were accomplished through additional bilayer electrolyte optimization as well as through the introduction of catalyst infiltrants into porous electrodes using methods that are scalable in manufacturing.

CONCLUSIONS AND FUTURE DIRECTIONS

Conclusions from the FY 2015 work include:

- Redox's bilayer electrolyte configuration can effectively boost the open circuit voltage at intermediate operating temperatures ($\sim 600^\circ\text{C}$) and therefore the efficiency of SOFCs when compared to cells consisting of a single cerium oxide electrolyte.
- When the bilayer electrolyte is combined with infiltration of catalysts in the cathode and/or anode,

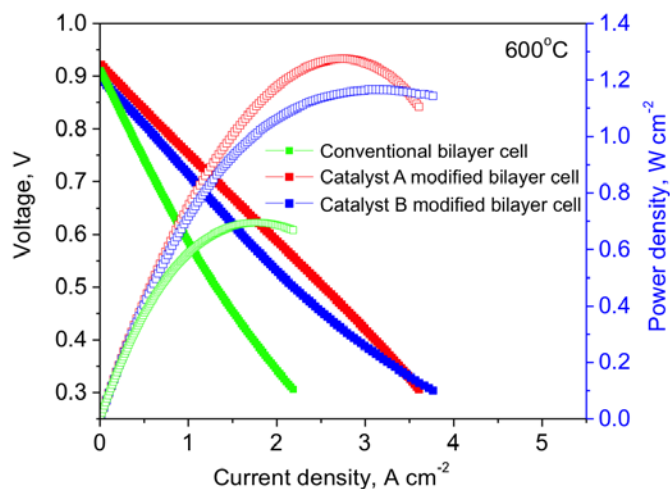


FIGURE 3. Initial results for bilayer button cell showing an OCP ≥ 0.9 V and a power density of 1.25 W/cm² at 600°C in humidified hydrogen

performance can be enhanced further with cells having power densities > 1.25 W/cm² at 600°C.

- The addition of the bilayer electrolyte to the custom Redox multi-physics model makes it a valuable tool for optimizing designs at both the cell and stack levels.

Future work will include the following:

- The implementation and demonstration of the bilayer electrolyte at the 10 cm by 10 cm cell size with an OCP ≥ 0.9 V at $\leq 600^\circ\text{C}$
- The demonstration of the bilayer 10 cm by 10 cm cells in a ~ 1 kW_e stack under CHP conditions using natural gas

FY 2015 PUBLICATIONS/PRESENTATIONS

1. Bryan Blackburn, Affordable, High Performance, Intermediate Temperature Solid Oxide Fuel Cells, U.S. Department of Energy's 2015 Annual Merit Review and Peer Evaluation Meetings (AMR) for the Hydrogen and Fuel Cells Program, Arlington, VA, June 11, 2015.

REFERENCES

1. Eric D. Wachsman and Kang Taek Lee, *Science*, **334**, 935–939 (2011).
2. Zhangbo Liu, Beibei Liu, Dong Ding, Mingfei Liu, Fanglin Chen, and Changrong Xia, Fabrication and Modification of Solid Oxide Fuel Cell Anodes Via Wet Impregnation/Infiltration Technique, *Journal of Power Sources*, **237**, 243–259 (2013).
3. Lei Wang, Greg S. Jackson, and Bryan M. Blackburn, *ECS Transactions*, **57**(1), 2583 (2013).