V.D.4 Affordable, High Performance, Intermediate Temperature Solid Oxide Fuel Cells

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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 ~600°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 ~1 kW_e stack for IT operation under combined heat and power (CHP) conditions with natural gas and minimal external reforming.

Fiscal Year (FY) 2016 Objectives

- Demonstration of bilayer electrolyte cell with OCP performance of ≥0.9 V at both button cell and larger 10 cm x 10 cm size for operation at ≤600°C.
- Demonstration of 10 cm x 10 cm cell operating at ≤600°C with a cell area specific resistance ≤0.2 Ωcm^2 and power density ≥1 W/cm^2.
- Demonstration of 10 cm x 10 cm bilayer cell stack in CHP conditions with natural gas.

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 (≤600°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 kWe). Lower operating temperature SOFCs 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 stationary fuel cell technical targets.

- Equipment Cost: $\$1{,}700/kW_{avg}$
- Transient Response (10–90% rated power): 2 min
- Degradation with Cycling: 0.3%/1,000 h
- Electrical Efficiency: >45%
- CHP Efficiency: 90%

FY 2016 Accomplishments

Accomplishments during the current project period include:

- Demonstration of a 10 cm x 10 cm bilayer cell with an OCP of 0.91 V at ~600°C.
- Successful scale-up of a porous anode-supported cell architecture to the 10 cm x 10 cm size as a step toward matching the 1.25 W/cm^2 seen at the button cell level at 600°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 [GDC] electrolyte) cells to a larger 10 cm x 10 cm production size and achieve similar performance as button cells. Furthermore, we have scaled up production of the erbia-stabilized bismuth oxide (ESB) material which 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 and 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 are necessary to take full advantage of SOFC fuel flexibility. Achieving these advancements while maintaining high power density at intermediate temperatures (~600°C) will help us deploy this technology for distributed generation and CHP.

APPROACH

Redox's approach involves the synergistic use of two electrolyte materials in a bilayer structure to result in 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 at 600°C, but due to the electronic leakage that occurs in reducing environments (i.e., fuel conditions), can result in decreased cell efficiency. The second material is bismuth oxide-based such as ESB, which has 60 times the conductivity of yttrium stabilized zirconia 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 manganate [LSM]–ESB) with exceptionally low area specific resistance and are leveraging past work on infiltration of electrodes that can 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 such as 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 ~600°C, an advanced, custom multi-physics model, which takes into account the unique thermochemical and physical properties of the Redox materials, is used. This is critical due to variations in conductivity and chemical activity of GDC as a function of temperature and effective oxygen partial pressure PO_2, which varies significantly down the channel in SOFC anodes with increasing fuel utilization [3].

RESULTS

During FY 2016, Redox worked to scale up our results from the button cell level to a cell size of 10 cm x 10 cm for a porous anode-supported bilayer electrolyte SOFC with high OCP and high power density at 600°C. In FY 2015, we adjusted the relative and total GDC–ESB bilayer thickness to improve the cell OCP. Figure 1A shows the typical

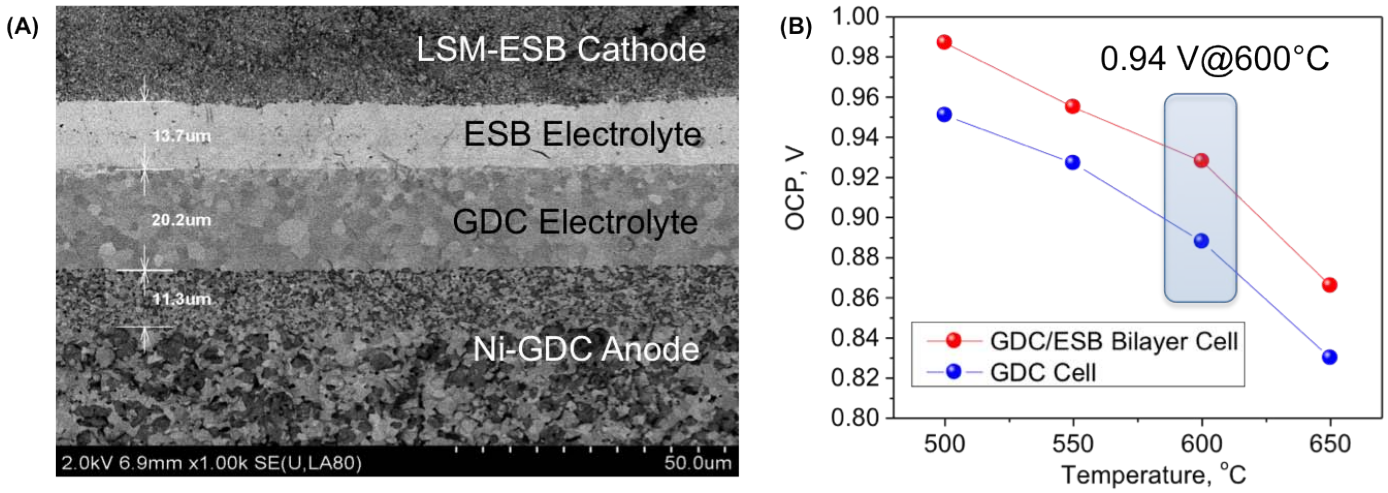

FIGURE 1. Button cell bilayer electrolyte cell results with (A) a scanning electron microscopy cross-section displaying the typical microstructure, and (B) OCP results for a bilayer electrolyte cell versus a GDC-only electrolyte cell from 500°C to 650°C

microstructure for a bilayer cell. As previously reported the GDC thickness was varied between ~12 µm and ~40 µm, while the ESB thickness was kept between 4 µm and 20 µm. The cells were tested in a standard button cell reactor using humidified hydrogen and air. Figure 1B shows the OCP at different temperatures (500–650°C) for a bilayer electrolyte cell and a GDC-only electrolyte cell. In these cases the GDC layer was ~20 µm for both cells, while the bilayer cell additionally had an ESB layer that was ~4 µm thick. There was a ~40 mV increase in OCP at each temperature, which can translate to a gain in cell efficiency for operating temperatures ≤600°C.

As the scale-up efforts on the porous anode support progressed during FY 2016, we used standard Redox production cells to scale up the bilayer electrolyte configuration to the 10 cm x 10 cm size. As shown in Figure 2, we demonstrated a 10 cm x 10 cm ESB–GDC bilayer electrolyte cell exhibiting an OCP of 0.91 V at ~600°C. This demonstration was achieved by the integration and scaling of earlier project efforts and deliverables. The cell was a standard Redox production cell with a ~20 µm GDC layer, a ~12 µm ESB layer, and a ~20 µm LSM–ESB cathode. While this was higher than the target OCP (≥0.9 V), the value was lower than what was achieved for the button cells. Our multi-physics model predicted that this was due in part to an undesirable porosity in the ESB layer for the 10 cm x 10 cm cell, which could have caused a decreased conductivity and/or reduced triple phase boundary, which, due to the mixed electronic and ionic conductivity in the GDC layer, can result in a lower OCP. Figure 3A shows the microstructure of the ESB layer for the button cell which was sintered at temperature T1. Figure 3B and 3C show the microstructure for a 10 cm x 10 cm cell sintered at T1 and T2, respectively, where T1 > T2. Due to furnace temperature gradients, the 10 cm x 10 cm cell whose results are shown in Figure 2 was actually fired at T2 even though the furnace setpoint was the same as in the button cell fabrication. When the furnace was adjusted so the actual temperature was T1, the larger size cell still was denser but still more porous than the button cell. Additional optimization of the ESB layer is needed to match the density achieved for the button cells.

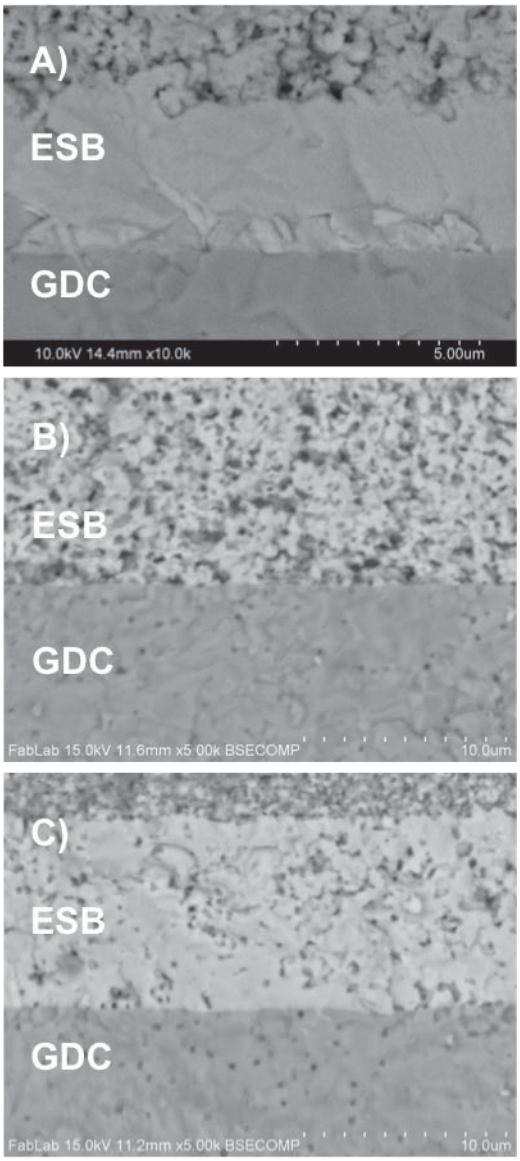

FIGURE 3. Scanning electron microscopy cross-sections comparing ESB microstructure for (A) a button cell sintered at T1, (B) a 10 cm x 10 cm cell fired at T2 < T1, and (C) a 10 cm x 10 cm cell fired at T1

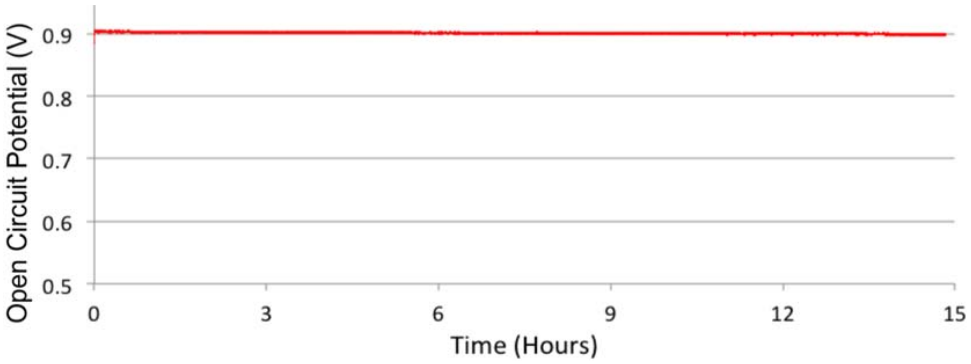

FIGURE 2. OCP for Redox 10 cm x 10 cm production cell tested at ~600°C in hydrogen

As reported previously, in FY 2015, we achieved more than 1.25 W/cm^2 at ≤600°C (cell area specific resistance <0.180 Ωcm^2) using a porous anode-supported, bilayer cell and various electrode catalyst infiltrants. Figure 4 compares results of a first attempt at a scaled up porous anode-supported 10 cm x 10 cm cell and the optimized version. The half cells utilized production tape cast layers. As seen in Figure 4A, the initial cells had a great deal of closed porosity in the anode support layer (ASL) and a dense anode functional layer (AFL). In order to gain a more interconnected pore network we used a broader particle size distribution for the pore former. We also made a porous AFL, which together with the optimized ASL resulted in a microstructure that was very similar to those of the porous anode-supported button cells used to achieve >1.25 W/cm^2 at ≤600°C.

CONCLUSIONS AND FUTURE DIRECTIONS

Conclusions from the FY 2016 work include the following:

- The bilayer electrolyte configuration was shown to have increased the OCP in excess of the target of 0.90 V using a 10 cm x 10 cm cell at ~600°C.
- The porous anode-supported cell has been successfully scaled up to the 10 cm x 10 cm size, and has similar microstructure as the button cells that displayed >1.25 W/cm^2.

Future work will include the following:

- Optimization of the ESB microstructure to further boost OCP for a 10 cm x 10 cm cell.
- Implementation and demonstration of a 10 cm x 10 cm, porous anode-supported bilayer cell with a power density ≥1 W/cm^2 at ≤600°C.
- Demonstration of a 10 cm x 10 cm bilayer cell stack in CHP conditions with natural gas.

FY 2016 PUBLICATIONS/PRESENTATIONS

1. Bryan Blackburn, "Affordable, High Performance, Intermediate Temperature Solid Oxide Fuel Cells," 2016 U.S. Department of Energy Hydrogen and Fuel Cells Program Annual Merit Review and Peer Evaluation Meeting, Washington, D.C., June 9, 2016.

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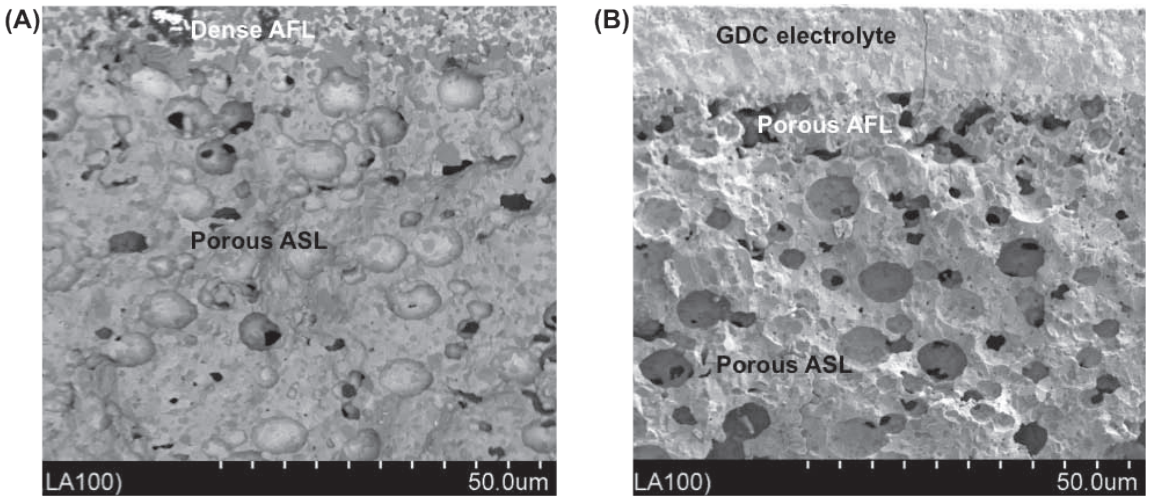

FIGURE 4. Scanning electron microscopy cross-sections comparing microstructure of a production manufactured, porous anode-supported cells using (A) original and (B) optimized scaled-up tape materials