# V.I.10 Development of kW Scale Coal-Based Solid Oxide Fuel Cell Technology*

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*Congressionally directed project

## Objectives

Develop a kilowatt scale coal-based solid oxide fuel cell (SOFC) technology. The outcome of this research and development effort will form the technological basis for developing a megawatt scale coal-based SOFC technology. Objectives for 2009 included the following:

- Evaluate the anode and cathode catalyst activity and a low-cost interconnect durability.
- Investigate the factors governing the anode catalyst activity for the electrochemical oxidation of coal.
- Refine the coal injection and fly ash removal systems.

## Technical Barriers

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

(A) Durability
(B) Cost
(C) Performance
(E) System Thermal Management

## Technical Targets

This project is directed at the development of a kilowatt scale coal-based SOFC technology. This project will develop a technological basis for the scale up of power generation capability of a kW SOFC to megawatt scale. A current density of 100 mA/cm$^2$ at 0.4 V was the initial target for demonstration of a carbon-based SOFC.

## Accomplishments

Year 2009 was focused on the following milestones:

- The activity of Ni-anode for electrochemical oxidation of solid carbon was investigated at 850°C. CO was the major product of this reaction. Cu and Ce oxide will be added to the Ni-anode to increase the selectivity of $CO_2$ to more than 80% in the products of carbon electrochemical oxidation reaction.
- 500 h of stable operation of Ni-anode, LSM (lanthanum strontium manganese) cathode and a low-cost interconnect was achieved for $H_2$ fuel. Durability test for low ash carbon fuel will be conducted.
- Power density of 0.18 W/cm$^2$ at 0.4 V was achieved at 850°C for low ash carbon fuel. A further improvement of the power density for carbon-based fuel will be the focus.
- Fabrication equipment for developing large-scale fuel cell components has been purchased and tested. Fabrication variables such as slip quality and heating cycles will be evaluated
- Fabrication of coal injection and fly ash removal system has been initiated.

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## Introduction

The direct use of coal in the SOFC to generate electricity is an innovative concept for electric power generation. The coal-based fuel cell could offer significant advantages: (i) minimization of $NO_x$ emission due to its operating temperature range of 700 – 1,000°C, (ii) high overall efficiency because of the direct conversion of coal to $CO_2$, (iii) the production of a nearly pure $CO_2$ exhaust stream for the direct $CO_2$ sequestration, and (iv) low investment and maintenance cost due to simplicity of the process. This technology also promises to provide low cost electricity by expanding utilization of U.S. coal supplies and relieving our dependence on foreign oil.

A small-scale coal fuel cell system including coal injection and fly ash removal parts will be fabricated. The main objectives of this project are (i) improvement of anode catalyst structure and the interface between electrodes and electrolyte, (ii) developing and refining the coal-based fuel cell fabrication techniques, and (iii) fabrication and testing of a small-scale coal fuel cell system. Successful development of this novel coal fuel cell technology will significantly enhance energy security of the U.S. and bridge the gap between a fossil fuel-based economy and the future hydrogen-based economy.

## Approach

Anode supported fuel cells were fabricated by tape casting procedure and tested in a steel reactor with a gas inlet port, a coke feeding mechanism, and a gas exhaust outlet port connected to the mass spectrometer and an off-stream gas chromatograph injection port. Prior to testing of the fuel cell, current collection components were attached to the cathode and Ag paste was coated along the perimeter of the anode to enhance the contact between anode and steel reactor. The performance of the fuel cell is determined by introducing the fuel ($H_2$, coke and coal) and monitoring the current produced at a certain load applied to the fuel cell. A voltage-current polarization plot is also recorded under these conditions for comparison purposes.

Low cost reactors, interconnects, and current collectors based on alloy were tested for durability, stability, and efforts were focused on exploring the processes with high-volume manufacturability for producing fuel cells. A variety of tests were performed to optimize the fuel cell fabrication process, and an optimized process is currently practiced at our fuel cell manufacturing facility. X-ray fluorescence, scanning electron microscopy and energy dispersive X-ray spectroscopy techniques are used to characterize the fuel cell for analyzing the microstructure, before and after the fuel cell testing and monitoring the reproducibility of the fuel cells fabricated in-house. Low- and high-ash content coal and coke were tested for the voltage-current performance and energy conversion efficiency for carbon-based fuel was also calculated. Once sufficient understanding was gained for the (i) reaction mechanism of electrochemical oxidation of carbon-based fuel on the anode, (ii) anode microstructure in terms of porosity, tortuosity and stability, (iii) fuel cell fabrication procedure, (iv) interconnect durability and stability and (v) coal feeding, efforts were expanded to the development of new anode catalysts, fuel cell fabrication methods, and current collection assemblies for kW stack fabrication.

## Results

In the past year, the main focus remained on finding a low-cost interconnect material, enhancing the power density of the fuel cell for $H_2$ and carbon-based fuel, determining the energy conversion efficiency and enhancing the long term stability of the fuel cell performance.

Figure 1 shows the impedance and voltage-current performance plots of a low-cost interconnect at 800°C over a 500 h period during stability testing for $H_2$ fuel. An ohmic resistance of 3.8 $\Omega cm^2$, polarization resistance of 7 $\Omega cm^2$ and power density of 0.11 $W/cm^2$ were recorded initially. The power density of the cell increased to 0.22 $W/cm^2$ after 122 h due to a complete reduction of NiO to Ni under $H_2$, further confirmed by decrease in the ohmic and polarization resistance to 1.1 $\Omega cm^2$ and 1.5 $\Omega cm^2$, respectively at 240 h. The current density continued to decrease after 122 h to 410 h and became constant at 0.17 $W/cm^2$ till 509 h. No decrease in the ohmic resistance was observed on the impedance plot after 240 h indicating a stable performance of the low-cost interconnect material. The

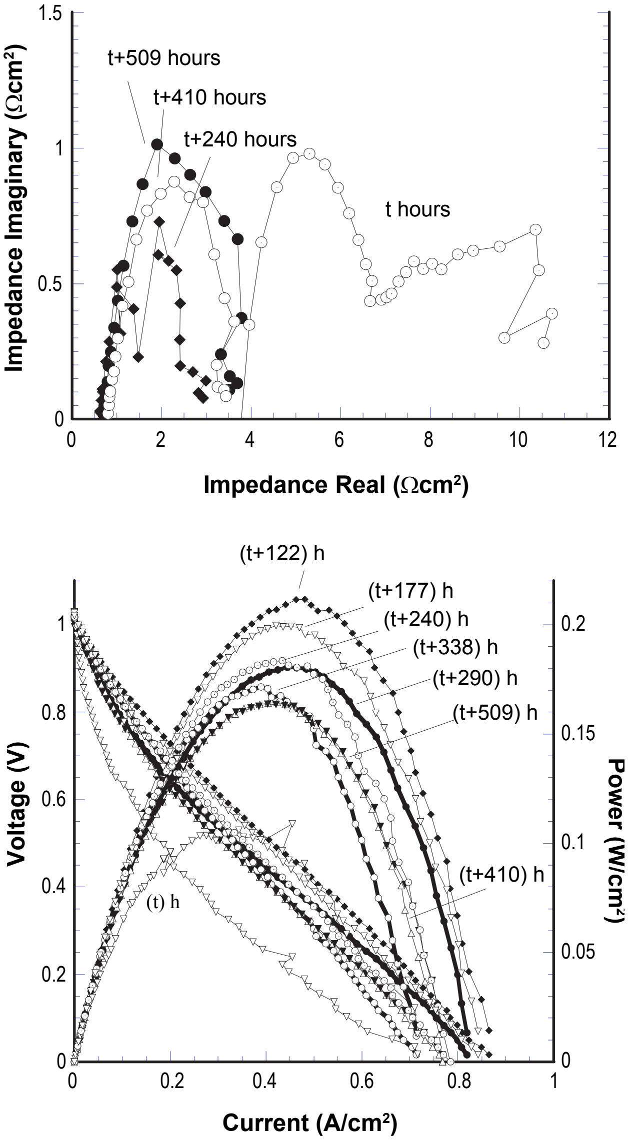

**FIGURE 1.** Impedance and voltage-current polarization plots for the stability testing of a low-cost interconnect at 800°C under $H_2$ fuel.

polarization resistance increased after 240 h showing microstructure of the anode became less porous and resulted in a concentration polarization which led to decrease in the maximum power density. A decrease in porosity could occur from sintering of the anode after long-term exposure at 800°C and led to decrease in the power density till 410 h.

Figure 2 shows the voltage-current polarization plots of (a) He(50 cc/min)/low ash carbon fuel and He/$H_2$(80/80 cc/min)/low ash carbon fuel at 800, 825 and 850°C. A power density of 0.05 W/cm$^2$ was produced at 800°C for low ash carbon fuel which increased to 0.18 W/cm$^2$ at 850°C and did not improve after further heating to 860°C. This result shows that a high activation energy pathway exists for the electrochemical oxidation of carbon at the Ni-anode at 800°C which was lowered at 850°C. Addition of a catalyst is necessary for enhancing the electrochemical oxidation of carbon at 800°C producing power comparable to 850°C. The power produced by low ash carbon has surpassed our 2009 goal of producing 64 mW/cm$^2$ at 0.4 V. The power produced by low ash carbon mixed with $H_2$ was significantly higher than low ash carbon which did not improve significantly on increasing the temperature from 800 to 850°C. This result suggests that $H_2$ is easier to be electrochemically oxidized than carbon at the anode, and reaches a performance level limited by the microstructure of the anode. Low ash carbon seems to be a promising fuel for SOFC technology.

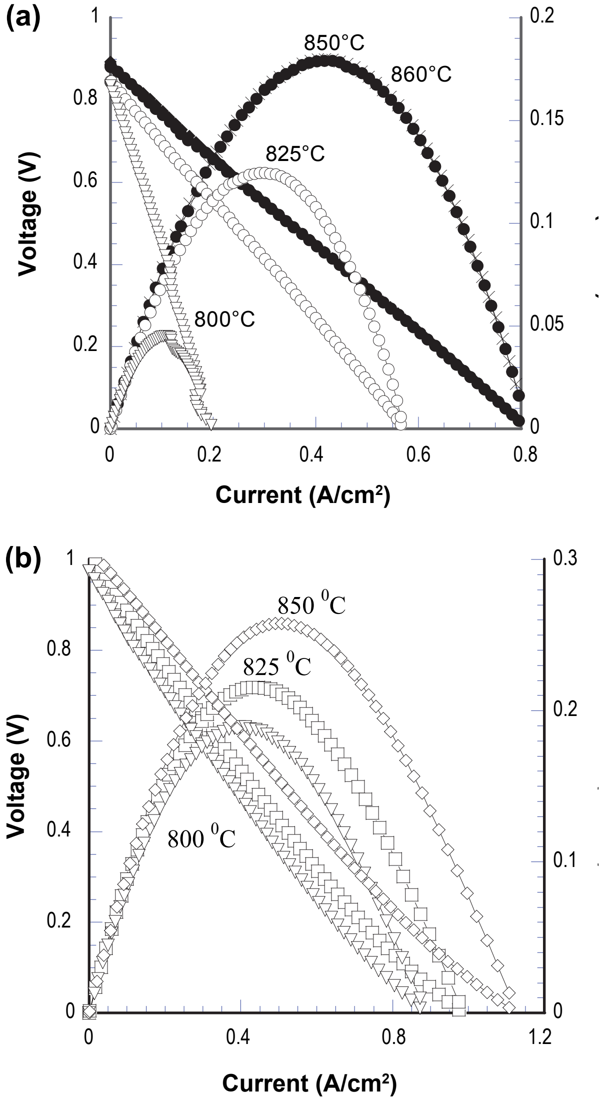

**FIGURE 2.** Voltage-current polarization plots of (a) He(50 cc/min)/low ash carbon fuel and (b) He/$H_2$(80/80 cc/min)/low ash carbon fuel at 800, 825 and 850 and 860°C over Ni/yttria-stabilized zirconia anode.

Figure 3 presents the mass spectrometry profiles at the exhaust of the reactor and current produced at 0.56 V during electrochemical oxidation of Ohio #5 coke at 800°C. The information presented in Figure 2 was used to calculate the energy efficiency, as it is described in the following section. The thermodynamic efficiency $\xi_T$ was obtained by relating $w_e$, the electrical work performed by the fuel cell, and the enthalpy change of the oxidation reaction $\Delta H$. The electrical work $w_e$, was calculated based on the current produced by the cell at the operating voltage resulting in 102.8 J. The enthalpy change of reaction $\Delta H$ was estimated considering the amount of $CO_2$ produced based on the gas chromatograph results, and the coke lower heating value resulting in 204.2 J. The thermodynamic efficiency $\xi_T$ was calculated a:

$$\xi_T = \frac{102.8\ J}{204.2\ J} = 50.3\%$$

A similar energy conversion efficiency was calculated for the low ash carbon which resulted in 58.6% on the lower heating value basis of coke.

## Conclusions and Future Directions

- Stable performance for 500 h of a low-cost interconnect has been demonstrated with $H_2$ fuel and air.
- Maximum power density of 0.18 W/cm$^2$ achieved at 850°C for low ash carbon.
- Thermodynamic efficiency of more than 50% has been demonstrated for a carbon-based SOFC at 800°C.

The proposed future research would be focused in the following directions:

- Identification of the catalyst composition for a long-term electrochemical oxidation of solid carbon fuels at 800°C.

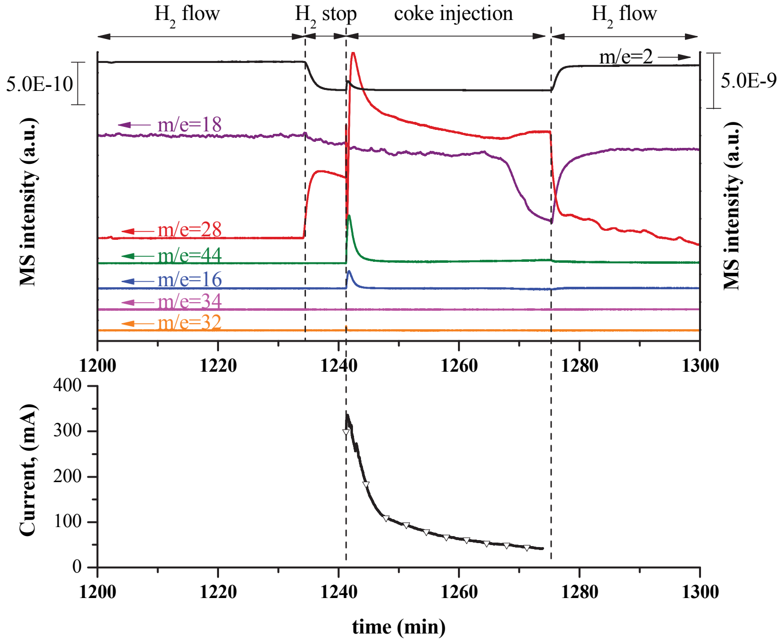

**FIGURE 3.** Mass spectrometer profile and current produced vs time plot for Ohio #5 coke SOFC at 800°C.

- Further investigation of low-cost interconnects materials which cost 50% less than the present available interconnects.
- Completion of design of the fuel cell stack and testing of a small-scale (1-10 kW) coal fuel cell system.
- Fulfill reporting obligations and preparing manuscripts for publications and conference presentations.

## Patents Issued

**1.** "Catalyst Compositions for Use in Fuel Cells," U.S. Patent Application, filed by the University of Akron on Sept.13, 2006.

## FY 2009 Publications/Presentations

**1.** 2009 DOE Hydrogen Program Review – Washington, D.C. – May, 2009.