VII.7 Novel Electrode Materials for Low-Temperature Solid Oxide Fuel Cells

Meilin Liu (Primary Contact), Shaowu Zha, Yin Liu, Luis Aguilar, Yuelan Zhang
School of Materials and Engineering, Georgia Institute of Technology
771 Ferst Drive
Atlanta, GA 30332-0245
Phone: (404) 894-6114; Fax: (404) 894-9140; E-mail: meilin.liu@mse.gatech.edu

DOE Project Manager: Lane Wilson
Phone: (304) 285-1336; E-mail: Lane.Wilson@netl.doe.gov

Objectives

- Characterize the microscopic features of composite anodes and correlate them with the ionic and electronic transport properties as well as the catalytic activities for oxidation of fuels.
- Minimize interfacial polarization resistances of anodes through processing modifications, microstructure improvements, and new materials development.
- Develop new catalysts for pre-reforming of hydrocarbon fuels to smaller molecules or for *in-situ* reforming.
- Gain a profound understanding of the principles of composite anodes for fuel cells to be operated at low temperatures.

Approach

- Synthesize and optimize NiO-GDC (gadolinium-doped ceria) composites as anode materials.
- Develop low-temperature solid oxide fuel cells (SOFCs) with active electro-catalyst for hydrocarbon fuels.
- Test catalyst consisting of 1 wt.% Pt supported by Gd-doped ceria for pre-reforming of propane at low steam/carbon ratio.
- Fabricate highly porous and nano-structured electrodes using combustion chemical vapor deposition (CVD) and characterize their performance as electrodes for low-temperature SOFCs.

Accomplishments

- Four types of anodes with two kinds of NiO and GDC powders were investigated. By carefully adjusting the anode microstructure, the GDC electrolyte/anode interfacial polarization resistances were reduced dramatically. The interfacial resistance at 600°C decreased from 1.61 $\Omega\ cm^2$ for the anodes prepared using commercially available powders to 0.06 $\Omega\ cm^2$ for those prepared using powders derived from a glycine-nitrate process.
- Anode-supported SOFCs with an electrolyte of 20 μm-thick GDC were fabricated by co-pressing, and both Ni- and Cu-based anodes were prepared by a solution impregnation process. At 600°C, SOFCs fuelled with humidified H_2 and methane reached peak power densities of 602 and 519 mW/cm^2, respectively.
- A catalyst (1 wt.% Pt dispersed on porous Gd-doped ceria) for pre-reforming of propane was developed with relatively low steam to carbon (S/C) ratio (~0.5), coupled with direct utilization of the reformate in low-temperature SOFCs. Propane was converted to smaller molecules during pre-reforming, including H_2, CH_4, CO, and CO_2. A peak power density of 247 mW/cm^2 was observed when pre-reformed propane was directly fed to an SOFC operated at 600°C.
- Highly porous, excellently bonded and nano-structured electrodes fabricated by combustion CVD exhibit extremely high surface area and remarkable catalytic activity.

Future Directions

- Fabricate anode-supported SOFCs based on yttria-stabilized zirconia (YSZ) electrolyte and further modify anode surfaces for low-temperature (500-800°C) operation.
- Further minimize interfacial resistances through processing modifications and microstructure improvements using combustion CVD technology.

Introduction

It is the interfacial resistances that limit the performance of SOFCs at temperatures below 550°C. The overall objective of this project is to develop novel electrode materials for SOFCs to be operated at low temperatures in order to significantly reduce the cost of SOFC technology, thus achieving the goals of the Vision 21 coal-based power plants. More specifically, in this work we will (1) characterize the microscopic features of composite mixed-conducting electrodes and correlate with the ionic, electronic, and ambipolar transport properties as well as with the catalytic activities for pertinent electrochemical reactions; (2) minimize interfacial polarization resistances through processing modifications, microstructure improvements, and new materials development; and (3) gain a profound understanding of the principles of composite mixed-conducting electrodes.

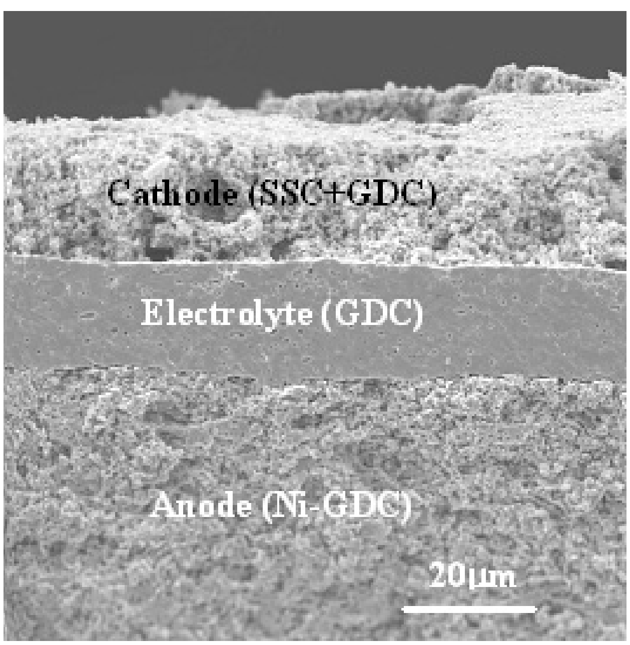

Figure 1. A Cross-Sectional View (SEM photograph) of a Ni-GDC Anode-Supported SOFC with a Thin GDC Electrolyte Layer

Approach

It has been demonstrated that the length of the triple-phase boundary (TPB) correlates well with the reaction rate for electrochemical oxidation of hydrogen; thus, the extension of the TPB becomes a determining factor in improving anode performance. This can be achieved mainly by optimizing the microstructure of the cermet anode through the adjustment of powder morphologies and particle sizes for the precursor NiO and ceria, and/or developing a favorable electroding process.

Carbon deposits can be suppressed by using anode compositions that do not catalyze hydrocarbon cracking, especially when running SOFCs at relatively low temperatures. In this study, we developed new anode materials for direct oxidation of methane and propane in low-temperature SOFCs. Anode-supported SOFCs with thin Gd-doped ceria electrolyte were fabricated by co-pressing, and both Ni- and Cu-based anodes were prepared by a solution impregnation process. We also developed novel catalysts for pre-reforming of propane, coupled with direct utilization of the reformate in low-temperature SOFCs.

We employed the combustion CVD technique to fabricate nano-structured electrodes for low-temperature SOFCs. These combustion CVD-derived electrodes exhibit extremely high surface area and remarkable catalytic activity.

Results

Four types of anodes with two kinds of NiO and GDC powders were investigated. It was found that fuel cell performance depends strongly on the anode microstructure, which is determined by the anode composition and fabrication conditions. By carefully adjusting the anode microstructure, the GDC electrolyte/anode interfacial polarization resistances were reduced dramatically. The interfacial resistance

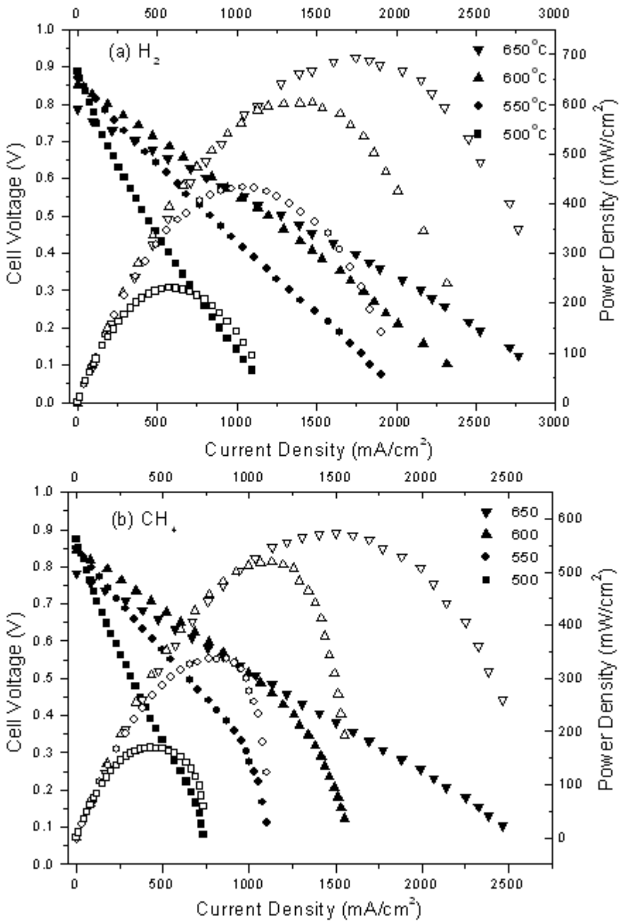

Figure 2. Cell Voltages and Power Densities as a Function of Current Density for a Cell with Ni-GDC Anode Fabricated by Co-Pressing Operated at Different Temperatures (500 - 650°C) with Different Fuels: (a) H_2 and (b) CH_4

at 600°C decreased from 1.61 Ω cm^2 for the anodes prepared using commercially available powders to 0.06 Ω cm^2 for those prepared using powders derived from a glycine-nitrate process.

The critical issues facing the development of economically competitive SOFC systems include lowering the operation temperature and creating novel anode materials and microstructures capable of efficiently utilizing hydrocarbon fuels. Anode-supported SOFCs with an electrolyte of 20 µm-thick GDC were fabricated by co-pressing (Figure 1), and both Ni- and Cu-based anodes were prepared by a solution impregnation process. Figure 2 shows that SOFCs fuelled with humidified H_2 and methane reached peak power densities of 602 and 519

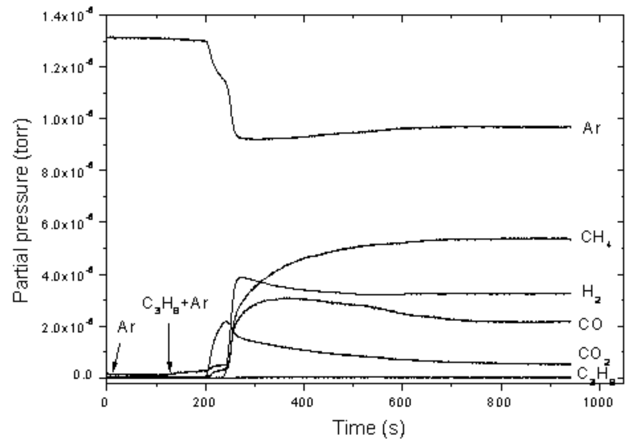

Figure 3. A Typical Mass Spectrum of the Reformed Propane on Catalyst Pt-GDC at 650°C (S/C = 0.5)

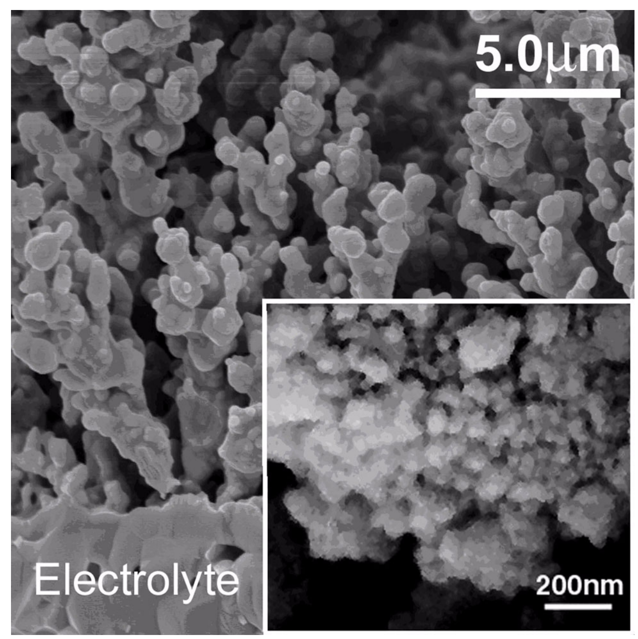

Figure 4. Cross-Sectional Views of an SOFC with Cathode Fabricated Using Combustion CVD

mW/cm^2, respectively, at 600°C. Both microstructure and composition of the anodes, as fabricated using a solution impregnation technique, greatly influence fuel cell performance.

A catalyst (1 wt.% Pt dispersed on porous Gd-doped ceria) was tested for pre-reforming of propane with relatively low steam to carbon (S/C) ratio (~0.5), coupled with direct utilization of the reformate in low-temperature SOFCs. Figure 3 shows that propane is converted to smaller molecules during pre-reforming, including H_2, CH_4, CO, and CO_2. A peak power density of 247 mW/cm^2 was observed when pre-reformed propane was directly fed to an SOFC operated at 600°C. No carbon deposition was observed in the fuel cell for a continuous operation of 10 hours at 600°C.

The development of a nano-structured cathode yielded interesting results. Highly porous, excellently bonded and nano-structured electrodes for low-temperature SOFCs have been successfully fabricated using the combustion CVD approach, as shown in Figure 4. These combustion CVD-derived electrodes consisting of nano-grains less than 50 nm in length exhibit extremely high surface area and remarkable catalytic activity.

Conclusions

The performance of SOFCs at low temperatures depends strongly on the interfacial resistances between the electrodes and the electrolyte. By carefully controlling the microstructures of the electrodes, relatively high performances have been achieved at temperatures below 650°C due to a significant reduction in interfacial polarization resistances. Nano-structured electrodes fabricated by combustion CVD show extremely high surface area and remarkable catalytic activity.

Ni-GDC anode-supported fuel cells are capable of running on methane directly at low temperatures. A catalyst (1% Pt-GDC) was developed for pre-reforming of propane at low steam/carbon ratio to smaller molecules such as H_2, CH_4, CO, and CO_2.

FY 2004 Publications

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