Reduced-Temperature Solid Oxide Fuel Cells Fabricated by Screen Printing

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Electrolyte films of samaria-doped ceria (SDC, Sm\textsubscript{0.2}Ce\textsubscript{0.8}O\textsubscript{1.9}) are fabricated onto porous NiO-SDC substrates by a screen printing technique. A cathode layer, consisting of Sm\textsubscript{0.5}Sr\textsubscript{0.5}CoO\textsubscript{3} and 10 wt % SDC, is subsequently screen printed on the electrolyte to form a single cell, which is tested at temperatures from 400 to 600°C. When humidified (3% H\textsubscript{2}O) hydrogen or methane is used as fuel and stationary air as oxidant, the maximum power densities are 188 (or 78) and 397 (or 304) mW/cm\textsuperscript{2} at 500 and 600°C, respectively. Impedance analysis indicates that the performances of the solid oxide fuel cells (SOFCs) below 550°C are determined primarily by the interfacial resistance, implying that the development of catalytically active electrode materials is critical to the successful development of high-performance SOFCs to be operated at temperatures below 600°C.

A solid oxide fuel cell (SOFC) is one of the cleanest, most efficient, and versatile technologies for chemical-to-electrical energy conversion. While existing SOFC technology has demonstrated much higher energy efficiency with minimal pollutant emission over conventional energy technologies, the cost of the current SOFC systems is still prohibitive for broad commercialization except for a small niche market. To be economically competitive, both the cost of materials and the cost of fabrication for SOFC systems must be dramatically reduced. One effective approach to cost reduction is to reduce the operating temperature. For example, if the operating temperature of SOFCs can be reduced to 400-600°C, the interconnect, heat exchangers, and structure components may be fabricated from relatively inexpensive metal components.\textsuperscript{1,2} Accordingly, extensive efforts have recently been focused on reduced-temperature SOFCs. Doshi et al.\textsuperscript{3} reported a maximum power density of 140 mW/cm\textsuperscript{2} at 500°C for a H\textsubscript{2}/air fuel cell based on a thin film GCO (Gd\textsubscript{0.2}Ce\textsubscript{0.8}O\textsubscript{1.9}) electrolyte film, a Ni-GCO anode and an ANLC-1 cathode.\textsuperscript{4} The anode-supported cell was prepared by a multiplayer tape-casting technique. The total cell resistance at 500°C was 1.3 Ω cm\textsuperscript{2}, of which 0.7 Ω cm\textsuperscript{2} was attributed to the electrolyte and nearly 0.6 Ω cm\textsuperscript{2} to the cathode, while the polarization of the anode was negligible. Barnett et al.\textsuperscript{5} reported a direct electrochemical oxidation of methane in SOFCs that generated power densities up to 370, 250, and 125 mW/cm\textsuperscript{2} at 650, 600, and 550°C, respectively. The electrolyte film, an 8 μm thick YSZ, was prepared on a La\textsubscript{0.8}Sr\textsubscript{0.2}MnO\textsubscript{3} substrate. The cathode slurry consisting of Sm\textsubscript{0.5}Sr\textsubscript{0.5}CoO\textsubscript{3}, 10 wt % SDC, and a Heraeus binder (V006) was also applied to the electrolyte by screen printing. The cathode was fired at 950°C in air for 4 h with heating and cooling rates of 300°C/h. Shown in Fig. 1 are the microstructures of each cell component as characterized using a scanning electron microscope (SEM, Hitachi S-800). The SDC electrolyte layer (with thickness of 30 μm) is dense while both anode and cathode are porous. The porosities of the cathode and anode are 27 and 35%, respectively, as estimated from quantitative microscopy.

The single cell was sealed on an alumina tube support with silver paste (Heraeus, C8710). The cell was horizontally mounted into an electric furnace. Electrochemical characterizations were performed at ambient pressure at temperatures from 400 to 600°C. Fuel cell performances were measured with an EG&G potentiostat/galvanostat (model 273A) interfaced with model 270/250 Research Electrochemistry software. Humidified (3% H\textsubscript{2}O) hydrogen or methane was used as fuel and stationary air was used as oxidant. The impedances of the cell under open-circuit conditions were measured with an EG&G lock-in amplifier (model 5210) and an EG&G potentiostat/galvanostat (model 273A) interfaced with model 398 electrochemical impedance software.

Shown in Fig. 2 are the I-V curves and the corresponding power densities for an SOFC tested at temperatures from 400 to 600°C with humidified hydrogen as fuel. Each datum point was recorded about 30 min after the cell reached the steady state. As shown in Fig. 2, an open-circuit voltage (OCV) of 1.02 V was observed at 400°C indicating that the electronic condition in SDC is insignificant. The maximum power densities were 128, 188, 240, and 397 mW/cm\textsuperscript{2} at 450, 500, 550, and 600°C, respectively. Although fuel cells based on SDC electrolyte that were 0.25-0.50 mm thick have been tested at temperatures above 600°C,\textsuperscript{6} the performance of the SOFCs below 600°C is not available in the literature. The observed current and power densities at low temperatures (400-600°C) of the fuel cells fabricated by screen printing represent significant progress in low-temperature SOFCs.

Figure 3a shows the impedance spectra of a single cell with humidified hydrogen as fuel, while in Fig. 3b the interfacial polar-
ization resistance and electrolyte resistance as determined from the impedance spectra are shown. At temperatures below 550°C, the interfacial resistance is much greater than the resistance of the electrolyte. The ratio of interfacial resistance to electrolyte resistance increased dramatically as the operating temperature was reduced, implying that the performance of the SOFCs to be operated at low temperatures depends critically on the interfacial resistance. Accordingly, the development of catalytically active electrodes and interfaces is critical to successful development of SOFCs to be operated at low temperatures (<600°C). Mesoporous mixed-conducting electrodes and nanostructured interfaces may significantly improve the performance of low-temperature SOFCs.

It is noted that the interfacial polarization resistance was 1.0 Ω cm² at 500°C, about two-thirds the cell resistance, but reduced to 0.07 Ω cm² at 600°C, only about one-sixth of the total cell resistance. This implies that the cell performance at temperatures above 600°C can be significantly improved by reducing the thickness of the electrolyte, at these temperatures both the cathode (Sm₀.₅Sr₀.₅CoO₃ and 10 wt % SDC) and the anode (NiO and 35 wt % SDC) are sufficiently active. For example, the cell performance at 600°C may be increased by 100% by reducing the electrolyte thickness from 30 to 10 μm because the total cell resistance may be reduced from 0.48 to 0.24 Ω cm².

When humidified methane was used as fuel, the maximum power densities were 78, 195, and 304 mW/cm² at 500, 550, and 600°C, respectively, as is shown in Fig. 4. The power density at 600°C (304 mW/cm²) observed in this work is higher than those

Figure 1. Cross-sectional view (SEM graphs) of (a) a single cell, (b) the porous Ni-SDC anode, (c) the dense SDC electrolyte, and (d) the porous Sm₀.₅Sr₀.₅CoO₃ + 10 wt % SDC cathode.
obtained with SOFCs based on thin film (8 μm thick) YSZ prepared by magnetron sputtering (250 mW/cm²).  

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