FABRICATION AND PERFORMANCES OF A SMALL SOFC STACK USING DOPED LANTHANUM GALLATE ELECTROLYTE

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ABSTRACT

A 4-cell tubular solid oxide fuel cell (SOFC) stack using strontium- and magnesium-doped lanthanum gallate electrolyte was built and electrically tested. Doped lanthanum gallate is a potential alternative to the commonly used yttria stabilized zirconia (YSZ) in SOFCs for use at intermediate temperatures (typically 600-750°C). $\text{La}_{0.6}\text{Sr}_{0.4}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{2.8}$ (LSGM) powder was mixed with a polymer-based binder system and extruded into tubes. After sintering, the electrolyte tubes were approximately 6 mm outside diameter, 0.55 mm wall thickness and 100-200 mm long. The tubes were then fabricated into single SOFCs, using optimized $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3}$ (LSCO) cathode, $\text{Ce}_{0.8}\text{Sm}_{0.2}\text{O}_{1.9}$ (SDC) interlayer, and Ni anode. The electrical performances of the single cells, as well as the 4-cell stack, were tested using dry hydrogen as a fuel. A power of 2.5-3 W per cell and 10 W for the 4-cell stack were obtained at 800°C. The maximum power density of the LSGM supported single tubular fuel cell with LSCO cathode, SDC interlayer and Ni anode was 372 mW/cm² at 800°C. This paper will present the fabrication procedure and parameters, single fuel cell, and 4-cell stack performance.

INTRODUCTION

Small-scale tubular solid oxide fuel cells (SOFCs), as described by Kendall and co-workers (1, 2), and later by Sammes and co-workers (3, 4), have many advantages over other designs, including excellent thermal shock properties, rapid start-up ability, and high mechanical integrity. The traditional electrolyte for use in these and other SOFC systems has been doped-ZrO₂, due to its good ionic conductivity at 900 to 1000°C, its stability in both oxidizing and reducing environments encountered in the SOFCs, and its good mechanical integrity (5, 6). The requirement for lower temperature SOFC systems, giving increased efficiencies and materials cost reduction, means that alternative electrolytes are sought which have superior oxygen ion conductivities.

In the search for new electrolyte materials, perovskite based systems (ABO₃) have been considered as alternative options, particularly because ABO₃ can take on a number of different structures and can be doped with aliovalent cations on both the A (for example Sr) and B (for example Mg) sites. They can also accommodate very large concentrations of anion vacancies into their structures.
Doped-LaGaO$_3$ has been reported by a number of workers to have a superior ionic conductivity and a transference number close to unity (7, 8). L$_{0.8}$Sr$_{0.2}$Ga$_{1-x}$Mg$_x$O$_{2+y}$ (x = 0.1 to 0.2) for example, has been reported as having the highest ionic conductivity of any electrolyte system, except for doped-Bi$_2$O$_3$ and doped-CeO$_2$, above 700°C; 7.14 S/cm was reported for L$_{0.8}$Sr$_{0.2}$Ga$_{0.8}$Mg$_{0.2}$O$_{2.85}$, and L$_{0.8}$Sr$_{0.2}$Ga$_{0.8}$Mg$_{0.2}$O$_3$ was found to be stable at partial pressures as low as 10$^{-25}$ atm. (9, 10). Thus, the material has great potential as an electrolyte in a SOFC system. Ishihara et al. (11) have shown that a single SOFC cell made from L$_{0.8}$Sr$_{0.2}$Ga$_{0.8}$Mg$_{0.2}$O$_3$ can produce a power output of 0.44 W/cm$^2$ at 800°C, using a Ni anode and a Sm$_{0.6}$Sr$_{0.4}$CoO$_3$ (SmSrCo) cathode. Later work by Marie et al. (12) observed 0.425 W/cm$^2$ in a cell using L$_{0.8}$Sr$_{0.2}$CoO$_{3.8}$ (LSCo) cathode and a Ni-samaria doped ceria anode on a 500 µm thick L$_{0.9}$Sr$_{0.1}$Ga$_{0.8}$Mg$_{0.2}$O$_3$ electrolyte, at 800°C. Further work by Huang et al. (13) showed that 0.9 W/cm$^2$ was attainable from a solid oxide cell fabricated using a 600 µm thick L$_{0.9}$Sr$_{0.1}$Ga$_{0.8}$Mg$_{0.2}$O$_3$ electrolyte layer, SrCo$_{0.8}$Fe$_{0.2}$O$_{3.8}$ (SCF) cathode, Ce$_{0.6}$La$_{0.4}$O$_{1.8}$ (LDC40) interlayer and LDC40+Ni anode, at 800°C. The use of an interlayer was required as the authors considered the mobility of La leads to second phases in the presence of NiO. Most recently, Fukui et al (14) reported that a thin film (130 µm) L$_{0.9}$Sr$_{0.9}$Ga$_{0.8}$Mg$_{0.2}$O$_3$ cell with LSCo cathode and SDC-Ni anode exhibited a power density of 0.7 W/cm$^2$. This paper will describe the fabrication and electrical performance of single micro-tubular solid oxide fuel cells and a 4-cell stack based on extruded lanthanum gallate electrolytes.

**EXPERIMENTAL**

**Cell Preparation**

Single tubular fuel cells consisted of anode, electrolyte, cathode, and current collectors as illustrated in Figure 1. Electrolyte tubes were fabricated using the L$_{0.8}$Sr$_{0.2}$Ga$_{0.8}$Mg$_{0.2}$O$_{2.8}$ (LSGM) powder and the process developed in our previous work (6). Anodes were formed from NiO, by reducing NiO to Ni in hydrogen atmosphere prior to fuel cell operation. A Ce$_{0.6}$Sm$_{0.2}$O$_{1.9}$ (SDC) interlayer was used to enhance the cell performance. High performance cathode material L$_{0.8}$Sr$_{0.4}$CoO$_{3.8}$ (LSCo) was optimized from our previous study (12).

Anode and cathode were first made into slurries to apply them onto the electrolyte support tubes. The ceramic powders were ball milled in ethanol with PVP/MEKET binders until the mean particle size was less than 1 µm. The anode slurry was dip coated inside the electrolyte tubes by sucking the slurry into the tubes using a syringe-pipe configuration. The coat was 15-20 µm thick after firing at 1200°C for 2 hours. The cathode slurry was brush painted onto the electrolyte tubes after the completing anode. The cathode coat was fired at 1100°C for 2 hours and was 20-30 µm thick after firing.

The cathode current collector, 99.9% silver wire (Silver State Wire), was tightly wound onto the outside of the tubes after the anode and cathode had been fired on. The anode current collector was then prepared and inserted into the tubes to complete the single cells. The effective dimensions of the cells prepared were: OD 6 mm, wall thickness 0.55 mm, active length 50 mm.
Figure 1. Schematic of micro-tubular SOFC single cell, based on LSGM electrolyte.

**Single Fuel Cell and Stack Test Setup**

Single fuel cells and a 4-cell stack were tested using a furnace test station setup shown in Figure 2 to evaluate the electrical performances. Dry hydrogen was fed inside the tubular cell at 50 ml/min at one end of the cell and the exhaust gas was combusted at the other end of the cell. Air was used as the oxidant gas. The stack was run at 800°C and I-V/I-P data were collected using a potentiostat/galvanostat.

Figure 2. 4-cell stack test setup.

**Single Cell Performance**

Typical current density-voltage/power density curves of the single fuel cell are shown in Figure 3. An open circuit voltage (OCV) of close to 1.2 V indicated a good gas tight electrolyte seal. The single cell performance was observed to increase over time. A maximum power density of a single cell reached 372 mW/cm² at 800°C after two days operation, and repeatable power of 2.5-3 W per cell was obtained. Typical single cell power output is shown in Figure 4. These two cells were run separately for about two days at 800°C and 850°C, respectively, at a loading of 0.7 V.
Figure 3. Cell voltage and power density as a function of current density at various temperatures and operation time. The cell consists of air, LSC/LSGM/SDC/Ni, H₂. The LSGM thickness is 0.55 mm.

Figure 4. Cell power as a function of operation time, operated at 0.7 V and 800° and 850°C. Other temperatures are included for comparison. The cells consisted of air, LSC/LSGM/SDC/Ni, and H₂. The LSGM thickness is 0.55 mm.
It is clear that the performance of these two cells showed similar increasing trends, power increased by 17-18% during the first day and 5-6% over the second day. The stable power outputs were 2.5 W for cell (●) at 800°C, and 2.8 W for cell (○) at 850°C, respectively. Figure 5 further illustrates the effect of cell operation time and temperature on the cell performance for cell 2D. After the first day’s running, the peak power/maximum power density tended to be stable, especially at lower temperatures. There was no sign of degradation observed for the cells tested in this work; instead, it took a day for the cell to reach the maximum performance.

Figure 5. Peak power and maximum power density at different temperatures as a function of operation time.

A Small Stack Performance

In order to evaluate the performance of a group of cells, a simple 4-cell stack was built by wiring the four single cells together in series. The stack was tested at 800°C for 50 hours. The results are shown in Figure 6. Similar to the single cell, the stack output increased about 20% during the first day of operation and tended to be stable at 10 W for the rest of the test time. Although longer operation time is needed to better evaluate the performance of the stack, degradation was not an issue during the two day test for the LSC/LSGM/SDC/Ni system.

Microstructural Analysis of the Fuel Cells after Test

Microstructural analysis of the cells after the test (Figure 7) shows that the anode and cathode current collectors were in good contact with the fuel cell. The thickness of the cell components was determined to be LSC cathode (20 μm), LSGM electrolyte (550 μm), SDC interlayer (10-20 μm), and Ni anode (15-20 μm).

The electrolyte was dense and gas tight. On the anode side, however, some of the interlayer had cracked before the anode layer was applied, and the anode coating had filled the gaps caused from the cracks.
SUMMARY

Tubular solid oxide fuel cells with doped lanthanum gallate (LSGM) electrolyte, nickel (Ni) anode, samaria-doped ceria (SDC) interlayer and strontium-doped lanthanum cobaltite (LSCo) cathode were fabricated and electrically tested. Single cell power of 2.5–3 watts at 800°C, and a four-cell stack power of 10 watts were obtained. The single cell or stack performance did not show any degradation over the test period of time; instead, the performance increased, although long-term performance of the single cell and the stack are required.
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