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## Operation of an LSGMC Electrolyte-Supported SOFC with Composite Ceramic Anode and Cathode

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An all-perovskite-based intermediate-temperature fuel cell was fabricated from materials synthesized using glycine-nitrate process (GNP) combustion synthesis and modified Pechini synthesis routes. Yttrium-doped strontium titanate (SYT) was chosen as the conductive ceramic anode component to avoid problems associated with Ni-based anodes.  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.1}\text{Co}_{0.1}\text{O}_{3-\delta}$  electrolyte-supported solid oxide fuel cells (SOFCs) with composite ceramic anode and cathode exhibit a relatively high power density of 0.246 W/cm at 800°C at 0.5 V.

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Operation of a fuel cell in the “intermediate” temperature range of 650–800°C has the potential to reduce fuel cell system costs significantly by allowing use of less exotic materials for balance-of-plant components, by reducing thermal degradation, and by allowing the use of gaskets.<sup>1</sup> Fuel cell stack materials must be chosen to be mechanically compatible with matching coefficient of thermal expansion, chemically compatible such that no deleterious or nonconductive phases are formed at interfaces of dissimilar materials and chemically stable in both the anode’s strong reducing environment and the cathode’s oxidizing environment.<sup>2</sup>

Doped  $\text{LaGaO}_3$  perovskites have been reported to be good electrolyte material candidates. Ishihara et al. reported high pure ionic conductivity of strontium and magnesium-doped lanthanum gallate (LSGM).<sup>3</sup> Subsequent reports have found that additional doping of the b site with cobalt (LSGMC) further enhances ionic conductivity. The total conductivity of LSGMC was found to be optimal at the composition  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.1}\text{Co}_{0.1}\text{O}_{3-\delta}$ . In the present work, all cells were tested using this composition for the electrolyte.

The use of composite electrodes in fuel cells has been reported as a method of improving the cell performance and decreasing polarization losses.<sup>4</sup> In the present work, composites of electronically conductive ceramic, catalytic material, and ionically conducting ceramic are used in both the anode and the cathode.

Anodes used in intermediate-temperature fuel cells must exhibit mechanical compatibility (coefficient of thermal expansion) with the electrolyte as well as chemical compatibility. Chemical incompatibility with conventional nickel-based anodes due to the formation of  $\text{LaNiO}_3$ <sup>5</sup> have led to the investigation of other potential anode materials. Hui et al. reported high electronic conductivities for yttrium-doped strontium titanate (SYT) in strongly reducing atmospheres and tested it as a potential anode material with yttrium-stabilized zirconium (YSZ) electrolytes.<sup>6,7</sup> SYT of the composition  $\text{Sr}_{0.86}\text{Y}_{0.08}\text{TiO}_{3-\delta}$  was used as the electronically conducting component of the anode in the present work. LSGMC of the same formulation as the electrolyte was added as an ionically conducting component to extend the electrochemically active area further into the electrode. Lanthanum-doped ceria (LDC) was added as a component to catalyze the anode reactions and decrease the activation overpotential.

The SYT used in this work was synthesized using a modified Pechini process due to the high cost of titanium nitrate that would otherwise be required for the glycine-nitrate process (GNP). The SYT synthesis was followed by calcining in air at 900°C for 8 h, which was found to be optimal by testing a range of calcining temperatures (800–1400°C) and durations (4–8 h). All other materials used in cell fabrication were synthesized using GNP. The resulting powders were calcined at temperatures and durations determined to be optimal by characterizing the powders with X-ray diffraction

(XRD, Bruker 5000) to analyze phase purity. Scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS) were used to verify the microstructure and composition of the powders.

GSAS software using EXPGUI graphical interface<sup>8,9</sup> was used to determine the lattice parameter of our material for use in calculating theoretical density.

Electrolyte-supported cells were fabricated by first pressing a pellet of LSGMC with binders and plasticizers in a 32 mm die and sintering in air at 1400°C for 6 h. The resulting pellets had nominal thicknesses of 1 mm. The density of the sintered pellets was measured using Archimedes principle to be 6.57 g/cm<sup>3</sup>, a value of 97% of the theoretical density based on the lattice refinements. SEM examination of the cells after testing also confirmed that a high density electrolyte was achieved.

The anode was made from a mixture of 60% SYT, 20% LDC, and 20% LSGMC by weight. An ink of the composite anode was made using a mortar and pestle to mix the anode powder with an organic binder (Heraeus V006).

Strontium-doped samarium cobaltite (SSC) of the composition  $\text{Sr}_{0.5}\text{Sm}_{0.5}\text{CoO}_{3-\delta}$  was chosen as the primary component of the composite cathode for this cell. In previous work, the composite cathode 0.7SSC-0.3LSGMC (wt %) was found to have good performance using symmetric cells of (SSC-LSGMC)/LSGMC/(SSC-LSGMC).<sup>10</sup>

The anode was applied using ink made from the desired composition of anode materials using a screen printer to deposit an anode with an area of 1 cm<sup>2</sup>, after which the anode was fired in air at 1250°C for 2 h. The cathode was applied in a similar manner with an area of 1 cm<sup>2</sup> and was fired at 1100°C for 2 h. Finally, current collectors made from platinum gauze were applied to the anode and cathode using platinum ink (Engelhard A3877A), and reference electrodes were applied at three times the electrolyte thickness away from the electrodes on both sides of the pellet. Platinum ink was used to affix the current collectors, which were then fired in air at 950°C for 1 h. Representative interfaces are presented in Fig. 1 and a representative cell cross section in Fig. 2.

The cell was installed in a button-cell test rig between two 1 in. alumina tubes and sealed with Aremco 503 ceramic cement. The anode gas was 100% dry hydrogen at a flow rate of 200 sccm. The cathode was dry air with a flow rate of 400 sccm. Flow rates were controlled using Brooks 5850s Smart digital mass-flow controllers. The cell was tested in the temperature range of 650–800°C. A Solartron 1260A frequency analyzer coupled with a 1480A multiplexer potentiostat was used to measure electrochemical characteristics of the cell in tests. AC impedance spectroscopy was used to determine anode and cathode interfacial characteristics and potential stair-step was used to collect the current-voltage responses. A three-probe configuration was used to separate out the impedance contributions of the anode and cathode interfaces.

The anode was allowed to reduce in a dry hydrogen environment for 5 h at 800°C to improve the electronic conductivity of the anode

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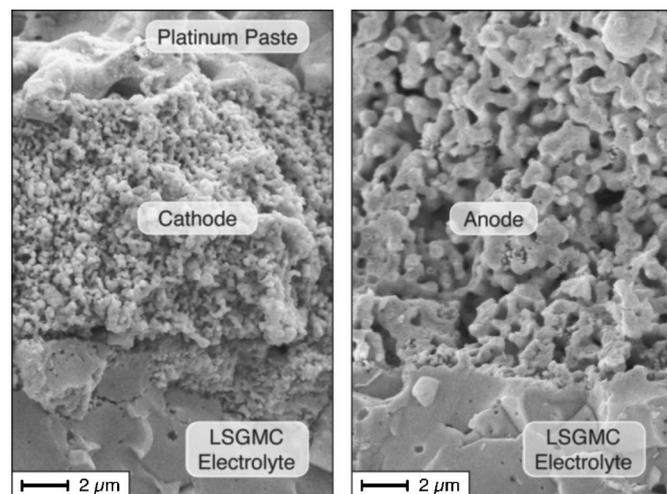


Figure 1. SEM micrographs of representative anode/electrolyte and cathode/electrolyte interfaces.

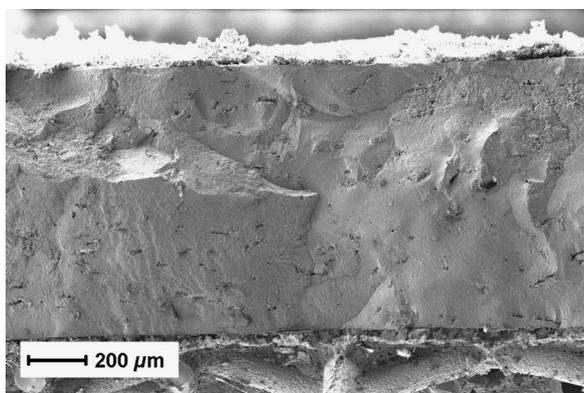


Figure 2. SEM micrograph of a fracture surface of the post-test cell cross section with the cathode at top and anode and Pt current collector at bottom.

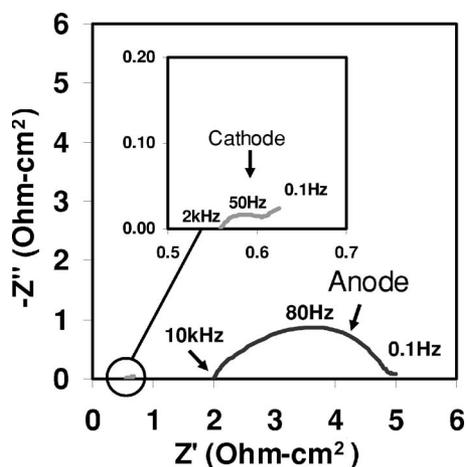


Figure 3. Impedance spectra for cathode/electrolyte and anode/electrolyte interfaces at 800°C with air as oxidant and dry hydrogen as fuel.

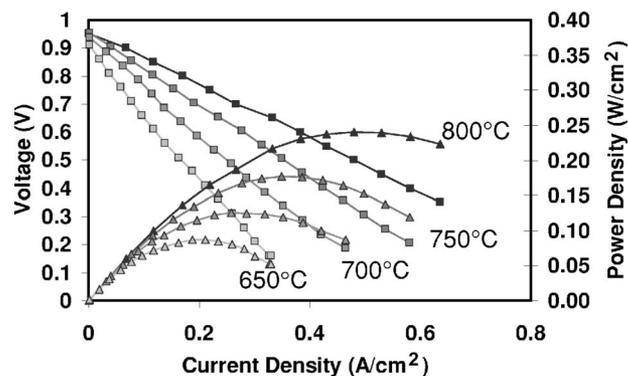


Figure 4. Operational performance curves for the full cell.

before the cell was tested. A decrease in total cell area specific resistance (ASR) of  $0.1 \Omega \text{ cm}^2$  was observed after the reduction treatment.

The ac impedance spectra of the full cell, shown in Fig. 3, reveals that little of the measured ASR of the entire cell is due to the cathode or cathode-electrolyte interfacial impedances. The anode ac impedance spectrum shows that much of the full cell measured ASR is due to the anode's contribution, in bulk resistance and in interfacial resistance. Values measured for full-cell ASR are within expected values for this temperature range using this materials set, though there is room for improvement of anode ASR.

Open-circuit voltages of 0.91–0.95 V were observed as shown in Fig. 4. This number is lower than the expected Nernst voltage for these temperatures. This could be due to reactant cross leakage or minor carrier conductivity introduced by the presence of Co in the electrolyte. Further work is planned to isolate the ionic and electronic contributions to electrolyte conductivity.

The peak power density of  $0.246 \text{ W/cm}^2$  at 800°C is respectable for this material and temperature. These figures compare favorably to the power densities achieved by Hui and Petric<sup>7</sup> using doped SYT anodes on a YSZ-based solid oxide fuel cell at a higher temperature (1000°C). Although Hui and Petric did not report detailed anode performance, their cell performance was likely limited by the SYT anode as illustrated by the anode and cathode interfacial impedance results of Fig. 3. The reason for improved cell performance in this work is likely due to use of a different electrolyte (LSGMC vs YSZ), a composite anode vs a noncomposite anode as well as a different synthesis route for SYT (modified Pechini process vs solid-state reaction).

Future work on dual-mode (fuel cell and electrolyzer) operation of this cell is also planned. Investigation of the durability of this materials set is also planned, as well as operation of anode-supported cells with much thinner LSGMC electrolyte membrane layers. Concerns about the suitability of this material as a conductive anode component remain due to the oxygen partial-pressure dependence of its conductivity. Necessity of reduction pretreatments must be addressed to successfully use this materials set as an intermediate-temperature fuel cell.

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