

MATERIALS AND DESIGN FOR SOFC AUXILIARY POWER UNITS

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Introduction

Traditionally, the development of solid oxide fuel cells (SOFCs) has focused on stationary power applications. However, a recent systems modeling study [i, ii] suggests that an SOFC-based unit operating on diesel fuel warrants consideration for generating auxiliary power for "hotel" loads (e.g., heating, air-conditioning, on-board electronics) and cargo needs (e.g., refrigeration) on heavy-duty vehicles and long-haul trucks. An SOFC-based auxiliary power unit (APU) would address a number of shortcomings with current technologies for generating auxiliary power, such as excessive engine wear with continuous engine idling, and regulatory issues, such as emission restrictions and anti-idling bans. A key technical barrier is the need to reduce the SOFC operating temperature from 1000°C to $\leq 800^\circ\text{C}$ to address materials issues, such as chemical stability in reducing and oxidizing environments and thermomechanical compatibility at 1000°C. If the SOFC operating temperature can be lowered to 800°C, metallic components can replace expensive ceramic ones in the balance-of-plant equipment. In addition, the ceramic bipolar plate can be replaced with a metallic alloy, which would improve mechanical robustness, thermal management, and electrical performance within the SOFC stack. For the SOFC-based APU system discussed below, system modeling suggests that an increase in overall system efficiencies from 35.0% to 40.3%, as well as, an increase in the Nernst voltage from 0.733V to 0.827V may be realized when the operating temperature is decreased from 1000°C to 800°C.

A major hurdle to the development of lower temperature SOFC units is the poor performance of the cathode. Increased resistance of the present cathode as the temperature is reduced significantly lowers the power density attainable. Research into a number of perovskite (ABO_3) and layered structures has identified some candidate cathode materials for use on yttria-stabilized zirconia (YSZ) electrolytes at 800°C.

Auxiliary power generation on heavy-duty vehicles and long-haul trucks would require an SOFC-based APU capable of generating 3- to 10-kWe net power. Catalytic autothermal reforming of diesel fuel would be used to generate H_2 as fuel. The fuel processor requirements for a SOFC-based system would be considerably simpler than those for a polymer electrolyte fuel cell (PEFC) system because CO cleanup would not be required. The following sections will discuss the current research into cathode materials that operate at lower temperatures and the tasks needed to develop a 3- to 10-kW integrated brass-board demonstration system.

Development of Lower Temperature Cathode Materials

The conventional cathode material used on YSZ is a strontium-doped lanthanum manganite (LSM) [iii]. This material operates satisfactorily at 1000°C, but its performance degrades markedly below 900°C because it has poor oxygen-ion conductivity [iv]. A mixed conducting cathode material is desirable at lower temperatures to increase the surface area for the reduction and transfer of oxygen in the air stream to oxygen ions in the electrolyte. A number of perovskite systems have previously been studied for use with YSZ at 1000°C but were discounted due to reactivity or thermal mismatch concerns [v]. At 800°C or lower, these problems are mitigated.

We have studied numerous perovskite-based materials, as well as layered structures. The perovskites consist of lanthanum on the A-site with 20 mol% strontium doping, and either cobalt, iron, or nickel on the B-site with 20 mol% doping of cobalt, iron, or nickel. The best-performing layered structure was the widely studied superconducting material $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO). All cathode materials (except $\text{YBa}_2\text{Cu}_3\text{O}_7$, which was obtained commercially) were prepared by glycine nitrate synthesis, and all were processed in an identical manner to form a cathode ink, which was then slurry coated onto YSZ substrates and sintered. Each cathode was initially evaluated by AC impedance measurements, followed by a long-term thermal stability test for the better-performing materials. Finally, polarization curves were obtained for the best-performing material.

Figure 1 shows the results of the initial electrical evaluation for the better-performing cathodes plotted in terms of area specific resistance (ASR) versus temperature. The initial target was an ASR $<1 \Omega\cdot\text{cm}^2$ at the lowest temperature possible. The temperature range studied was 650-850°C in air. Clearly, the best-performing class of cathode material was the lanthanum ferrate-based compositions, with $\text{La}(\text{Sr})\text{FeO}_3$ (LSF) giving the lowest ASR. The ASR for LSF was below $1 \Omega\cdot\text{cm}^2$ down to 750°C. Other promising cathode materials include lanthanum nickelate (LN) and YBCO, but these showed a greater increase in ASR as the temperature was reduced.

Figure 2 shows the thermal stability of LSF and LN held at 800°C for 500 h. Both show stable performance after an initial increase in resistance, which is thought to be the result of microstructural rearrangement. The LSF displays a very stable performance with an ASR below $1 \Omega\cdot\text{cm}^2$. The LN has a higher ASR and shows a less stable behavior with time, but further improvement may be possible by choosing better dopants. Figure 3 displays the polarization curve for LSF at 800°C after current conditioning. The cathode overpotentials are similar to those of the present cathode material, LSM, at 1000°C. From these results, LSF would be the most likely candidate to replace LSM at lower operating temperatures, but full cell testing will be needed to confirm the performance of this material.

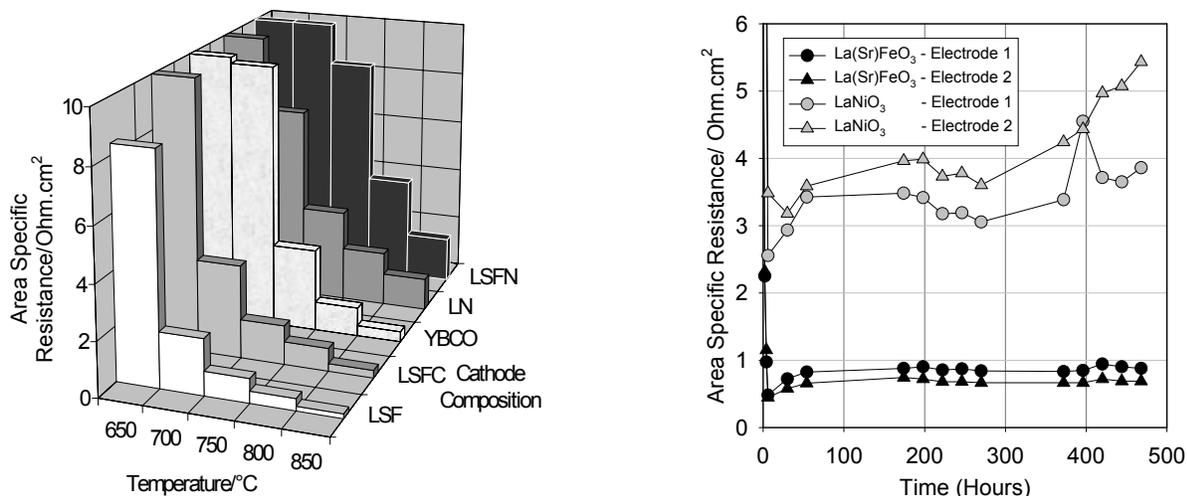


Fig. 1. Area specific resistance of best-performing cathodes. Letters represent first initial of element present, except YBCO, which is defined above.

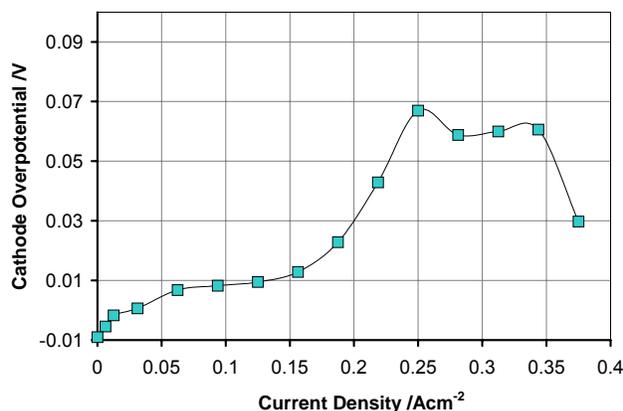


Fig. 3. Polarization curve for LSF at 800°F after current conditioning for 2 weeks at 250 mA·cm².

Fig. 2. Long-term ASR performance of LSF and LN at 800°C.

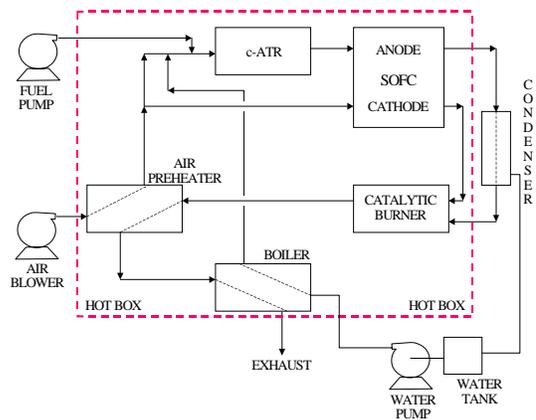


Fig. 4. Conceptual design for SOFC-based APU operating on diesel fuel.

Development of a 3-10 kWe SOFC-based APU

A conceptual design for an SOFC-based APU operating on diesel fuel is shown in Figure 4. The major components of the system are the catalytic autothermal reformer (c-ATR), the fuel cell stack, the air preheater for the cathode and anode air, and the catalytic burner for the spent fuel. All of these components are housed in an insulated thermal enclosure (i.e., "hot box"). Additional system components include the condenser for water recovery, a boiler to generate steam, fuel and water pumps, and an air blower. In this system, fuel, preheated air, and steam are fed to the c-ATR to produce reformat, which flows directly to the SOFC. Water needed for the c-ATR is condensed out of the anode exhaust from the SOFC. The H₂ and CO in the anode exhaust gas are burned in the catalytic burner using the cathode exhaust. The exhaust gas from the catalytic burner is used to preheat the process air and to generate steam. Although the air preheater and catalytic burner are shown schematically as two separate units, the total system weight and volume can be significantly reduced by integrating the air preheater and catalytic burner into a single unit. It is estimated that the total volume of the system would be approximately 100 L.

Fuel is processed by catalytic autothermal reforming to yield a gas mixture consisting of H₂, CO, CO₂, H₂O, N₂, and trace amounts of CH₄, and possibly higher hydrocarbons. Since the SOFC can operate using CO as a fuel, either by direct electrochemical oxidation of CO or by generation of H₂ via the water-gas shift reaction [vi], the fuel processing for a SOFC-based system is much simpler than that for a PEFC-based system. Furthermore, since the SOFC operating temperature of ~800°C is close to the c-ATR operating temperature of 750-850°C, there is excellent potential for thermal integration of these two key chemical and electrochemical components in the APU system for efficient power generation [vii].

On the basis of the lower heating value (LHV) of the fuels, SOFC-based APUs are expected to reach system-level efficiencies of 30%. For example, using GCtool [viii], we have analyzed a 5-kWe APU system, shown schematically in Figure 4, for various operating conditions and efficiencies. As shown in Table 1, an overall energy conversion efficiency of 31% is predicted for this system, despite the fact that the system design is rather conservative with little thermal integration other than the close temperature coupling of the c-ATR and the SOFC. The balance-of-plant, as well as the overall system efficiency, can be increased significantly by operating and design changes. For example, by increasing the fuel utilization from 80% to 85% or 90%, the system-level efficiency can be increased to 34% and 38%, respectively. By using the heat

rejected at the anode condenser to provide some preheated air, efficiencies can be further increased. System efficiencies based on the net power production could exceed 40%; however, some of these efficiency gains would be at the expense of system simplicity and could create additional thermal and mechanical stresses in the ceramic stack components.

Table I. Operating parameters for the base-case 5-kW SOFC system.

Fuel Cell Stack	
Cell voltage, V	0.750
Cell temperature, °C (average)	800
Cathode air feed rate, g/s	63
Anode fuel gas feed rate, g/s	3
Cell efficiency, %	56.2
Autothermal Reformer	
Fuel flow rate, g/s	0.35
Air feed rate, g/s	1.95
Water/steam feed rate, g/s	0.70
Steam-to-carbon, mole ratio	1.50
Reforming temperature, °C	760
Reformer efficiency, % (LHV)	77
Overall System	
Efficiency, % (LHV)	31.1
Balance-of-Plant	
Efficiency, %	72

Clearly, the level of development of planar SOFC stacks, reformer technology, and other system components has reached a stage where it appears practical to develop a 3- to 10-kWe SOFC-based APU demonstration unit. Several issues still need to be addressed before a practical APU can be built: (i) A more detailed engineering design based upon various tradeoffs in system design and operating parameters; (ii) The design and development of planar SOFC stacks capable of generating 3 to 10 kWe at stack-level power densities of $\sim 0.4 \text{ W/cm}^2$; (iii) Design of a fuel processor for specific fuel and determination of its operating envelope, (iv) Design and development of an integrated catalytic burner/air preheater, and

(v) Analysis and resolution of system integration and control issues.

Conclusions

Several perovskite-based and layered structure materials have been identified as candidate cathode materials for SOFCs operating at 800°C. A strontium-doped lanthanum ferrate displayed the best area specific resistance, showing stable long-term performance. Analysis shows a SOFC-based APU would have few components and occupy a volume of $\sim 100 \text{ L}$. Efficiencies with diesel fuel could approach 40% LHV.

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