

**PROMISE AND PROBLEMS OF SOLID OXIDE FUEL CELLS
FOR TRANSPORTATION***

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ABSTRACT

Fuel cells are being developed to provide clean, efficient propulsion power in transportation applications. This paper discusses the promise and problems of the solid oxide fuel cell (SOFC) for the automotive application. The SOFC system requires a simple, compact fuel processor, even for complex fuels such as gasoline and diesel fuel. Because of the similar operating temperatures, the fuel processor and the fuel cell stack can be close-coupled, offering a high degree of thermal integration for a high net power density for the system. Although the startup of the SOFC system would not be near-instantaneous, it would not necessarily require more time than for other types of fuel cell systems operating on conventional or alternative transportation fuels. Simulations of system performance show that high efficiencies (~40%) should be achievable. Developments in advanced interconnect and other cell and stack materials are expected to greatly improve the thermal and mechanical ruggedness of the fuel cell stack. Although the balance-of-plant is relatively simple, some components still need development, such as an air preheater offering high heat transfer in a small volume.

1. INTRODUCTION

Most of the world's major automobile companies are exploring and/or developing fuel cells for providing clean, efficient propulsion power onboard light-duty and heavy-duty vehicles. While the phosphoric acid and the alkaline fuel cells have been used in limited demonstration vehicles, the majority of fuel cell cars and buses are powered by the polymer electrolyte fuel cell (PEFC). The major driver for the selection of the PEFC for automotive use is its relatively low operating temperature of 80°C. Even at room temperature, the PEFC can deliver useful power, if hydrogen is the on-board fuel. Thus, the PEFC has been perceived as being capable of near-instant start, similar to today's internal combustion engine.

So far, the solid oxide fuel cell (SOFC), which operates at 700–1000°C, has been dismissed as being unsuitable for automotive applications because it does not offer near-instant start from room temperature. While this may be true, it is not necessarily true that a PEFC system can begin to deliver useful power faster than an automotive SOFC system if the on-board fuel is other than hydrogen. In particular, if the fuel is gasoline or diesel, the startup time for the PEFC-based system may be controlled by the startup time for the fuel processor rather than by that of the fuel cell stack. Furthermore, the fuel processor and the balance-of-plant in an automotive SOFC system are likely to be much smaller and simpler than in an equivalent PEFC system. This paper discusses these and other features that recommend SOFCs for automotive propulsion power. Also covered are the issues that must still be resolved before the solid oxide fuel cell can be successfully developed for this application.

2. PROMISE OF SOFC FOR TRANSPORTATION

The SOFC system requires a relatively simple fuel processor, even for complex fuels such as gasoline and diesel fuel. The reforming reactor only needs to break down the larger hydrocarbon molecules to hydrogen, carbon monoxide, and carbon dioxide, along with lighter hydrocarbon species. Since the carbon monoxide is not a poison for the SOFC (indeed, CO can be electrochemically oxidized in the SOFC and, therefore, can be used as a fuel by itself), there is no need for one or more water-gas shift reactors or for final CO clean-up devices, such as preferential oxidation or methanation reactors. In addition to being simpler than what is needed for PEFC systems, this fuel processor is considerably smaller, as discussed below.

Furthermore, the reforming reactor in an SOFC system does not need to achieve complete conversion of the fuel to H₂ and CO to obtain high system efficiencies. Indeed, designing the reformer for incomplete conversion of the fuel makes it possible to use the water and heat produced by the electrochemical cell reaction to steam-reform (within the stack) the hydrocarbons in the reformat from the fuel processor. Thereby, the cooling requirements in the fuel cell stack can be reduced. Operating in this mode would lead to lower cathode air flow rates with a decreased air-blower power requirement, potentially resulting in increased system operating efficiencies. Thus, the reforming reactor itself can be much smaller than would be needed for complete conversion of the fuel.

The SOFC-based automotive propulsion system offers excellent potential for thermal integration between an autothermal reformer (ATR) for gasoline or diesel and the fuel cell stack. Recent developments in ATR technology for gasoline reforming have driven the reforming temperature to the range of 700–1100°C [1]. The lower end of this temperature

range is preferred for enhanced PEFC system efficiencies [2]. This temperature range is similar to the range of operating temperatures for the solid oxide fuel cells under development. Since the reformer and the fuel cell stack are two of the major components in the SOFC system, this match in operating temperatures permits close coupling of the two, reduces thermodynamic irreversibilities, and eliminates the need for the temperature matching that is required in PEFC systems.

Solid oxide fuel cell systems are also attractive for automobile propulsion because of their high component and system power densities. Our analyses have shown that cross-flow, planar SOFC stacks can achieve power densities above 1 kW/L, particularly for designs based on anode- or interconnect-supported cell and stack structures. Since the entire SOFC system consists of only a few major components, system-level power densities of >250 W/L can be achieved, with efficiencies approaching or exceeding 40% based on the lower heating value of gasoline fuel [3].

Furthermore, the SOFC makes possible an energy conversion system that is essentially all solid state [4]. Because of the elevated temperature of operation, there is no liquid water within the fuel cell stack, and no need for water management for the purpose of achieving the humidification levels needed to maintain electrolyte conductivity. The absence of liquid water and the all-solid-state fuel cell stack lead to a high degree of flexibility in system configuration and component placement and orientation.

3. ISSUES WITH SOFC FOR TRANSPORTATION

Several major issues must be resolved before SOFCs can be used effectively in transportation applications. These include the system startup time and fuel energy consumption, sizes of the fuel cells and stacks, sizes of the balance-of-plant components, thermal and mechanical ruggedness of the ceramic components, and the materials and designs of the thermal enclosure for the elevated temperature components of the SOFC system.

The major component that governs the startup time and fuel consumption for the SOFC system is the fuel cell stack. Although a single SOFC can be heated up very quickly without damage, the mechanical constraints imposed by the stack housing and manifolds could dictate the maximum safe rate at which the fuel cell stack can be heated up. With the appropriate stack designs and materials, however, these constraints can be greatly reduced. For example, with a metallic interconnect that also provides the structural support for the fuel cell stack, stack heat up rates of 2°C/s would yield a startup time of approximately 400 s for a stack operating at 800°C. Based on the electrochemically active fuel cell area, gravimetric stack

power densities of approximately 1.0 kW/kg may be achievable with advanced stack designs and materials. Allowing for the manifolding and other stack hardware, the corresponding stack-level power density would be 0.5 kW/kg. Assuming an average heat capacity of the stack to be 0.54 J/g-K (similar to that of zirconia in the range 25–900 C), and a net efficiency of 36% at rated power, the average fuel feed rate during the startup time of 400 s would be ~75% of the value at the rated power.

This represents a significant amount of fuel consumed during startup from room temperature, and would have to be considered along with the fuel cell system's duty cycle to assess the viability of the SOFC system for a particular application. For vehicles with a minimal duty cycle, say, 1 h of operation with two start-stop cycles over 24 h (i.e., a very light-duty vehicle used only for commuting to and from work), the startup fuel consumption could be comparable to the amount of fuel used during vehicle operation. With longer duty cycles, the startup fuel consumption would be a progressively smaller fraction of the total fuel consumed. The startup fuel consumption can also be reduced by appropriate thermal insulation to reduce heat loss from the system during shutdown, such that the startup is from warm rather than cold conditions.

A second major issue is the size of the cells, stacks, and other components in the automotive SOFC system. Much of the developmental activity in various types of planar SOFC cells and stacks is with ~100-cm² active area cells (e.g., 10×10-cm square, or 11-12 cm diameter circular). With single-cell power densities of 0.4–0.5 W/cm², this translates to 40–50 W/cell. A 1-kW stack would then require 20–25 such cells. In practice, stack-level power densities are often lower, thus requiring a larger active area in the stack. For the light-duty vehicle application, fuel cell systems of 30-50 kW or higher power rating may be needed for a fuel cell/battery hybrid power plant. These power requirements would dictate the use of larger active area cells. Planar single cells as large as 40×40 cm have been developed. Also, "window-pane" cell designs have been developed that have multiple cells on one plate. These larger sizes are comparable to the cell sizes developed for automotive PEFC stacks.

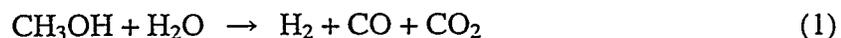
The solid oxide fuel cells are air cooled. Therefore, the larger cells require a significant increase in the size of the air flow channels to limit the parasitic power needed to overcome the air-side pressure drop to 10% (or less) of the gross power generated by the cell. Our analyses have shown, as discussed below, that by increasing the size of a thin-electrolyte, cross-flow planar cell from 6×6 cm to 15×15 cm, the net power increases from 21 W/cell to 105 W/cell, but the net power density decreases from 2 kW/L to 0.6 kW/L of the active volume of the stack. Using a systems approach, however, and absorbing some of the stack

waste heat in the endothermic steam reforming of part of the fuel, the cooling air requirements may be reduced substantially. This would decrease the required air channel height and increase the effective net power density, greatly reducing the active stack volume. The specific magnitude of this reduction would depend on the design of the close-coupled autothermal reformer and the fuel cell stack, but combined ATR/stack net power densities of ~1 kW/L should be achievable.

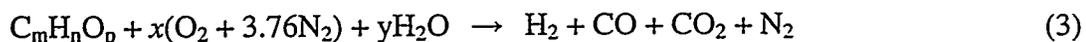
The ceramic structure of the SOFC raises issues of thermal and mechanical ruggedness of the individual cells, stack, and the whole assembly. While these issues have not been fully addressed, some of the SOFC developments underway are expected to alleviate the related problems. For example, the use of ferritic steels or other metallic bipolar plates in the lower temperature SOFCs (those operating at 700–800°C) would increase the mechanical strength and vibration/shock resistance of the stack. Additionally, the use of flexible or compressible gasketing [5] would permit some differential movement of the stack components, thereby increasing the stack's mechanical ruggedness. At the cell level, use of an electrolyte of partially stabilized 3 mol% Y₂O₃-ZrO₂ rather than fully stabilized 8 mol% Y₂O₃-ZrO₂ offers much greater mechanical strength at only a relatively small loss in cell performance. Similarly, other approaches in cell and stack design, such as the “windowpane” approach of making multiple cells on each cell plate, would help to reduce the incidence of failures from thermal and mechanical stresses.

4. FUEL PROCESSORS FOR GASOLINE/DIESEL

For a fuel cell system to operate on a conventional or alternative transportation fuel (i.e., gasoline, diesel, methanol, ethanol, natural gas, or propane), the on-board fuel must first be converted to hydrogen in a fuel processor [6]. The first step in such a process is to break down the carbonaceous fuel into a gaseous mixture containing H₂, CO, CO₂, H₂O, N₂, etc. This may be achieved by steam reforming, e.g., by the unbalanced reactions:

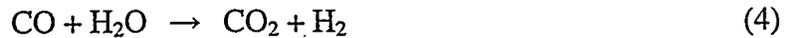


or by a combination of partial oxidation and steam reforming (autothermal reforming),

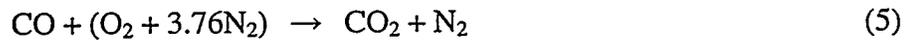


There may also be other byproduct and intermediate species, as well as unconverted input fuel. These reforming reactions are carried out at elevated temperatures, from a relatively low temperature of ~250°C (for the catalytic steam reforming of methanol) to a high temperature of ~1300°C (for the non-catalytic partial oxidation reforming of the liquid hydrocarbon fuels).

Although a fuel cell can operate on the hydrogen in this gas mixture, the low-temperature PEFC requires that this raw reformat be processed further to reduce the CO content to only a few parts per million (ppm) to avoid poisoning of the anode catalyst. This is generally carried out in multiple steps. The bulk of the CO is converted to CO₂ and additional H₂ by reaction with steam according to the water-gas shift reaction,



The water-gas shift reaction is generally carried out over suitable catalysts in two stages, with cooling between the stages to remove the heat of reaction. The CO content in the reformat after the shift reaction is ~0.5% (by volume). The final reduction of CO to <50–100 ppm is achieved by preferential oxidation (with the injection of a controlled small amount of air) or preferential methanation in a catalytic process:



Because of the relative reaction kinetics and the heat exchange required, the major fraction of the total weight and volume of the fuel processor (typically, >80% [7,8]) is due to these CO reduction/removal steps and the associated thermal management components.

For a solid oxide fuel cell, on the other hand, the fuel processor does not need any of the CO removal steps. In addition, as discussed above, even the initial reforming reactions (1), (2), or (3) do not need to be carried to complete conversion. Thus, the fuel processor for an automotive SOFC system is expected to be one-fifth to one-tenth the size of a fuel processor for a PEFC system having comparable fuel processing throughputs.

Fuel processors for automotive PEFC systems operating on methanol and hydrocarbon fuels are being developed by several organizations [1,6,7]. A much simplified version of these can be readily adapted for use in SOFC systems, with considerable savings in weight, volume, and cost. An added significant benefit of the SOFC fuel processor is the much reduced startup time. As discussed in [7], the startup time for the initial reforming reactor is approximately one-tenth of the startup time for the total PEFC fuel processor. Therefore, the startup time for an SOFC fuel processor would be only one-tenth (or less) that of an equivalent PEFC fuel processor.

5. CELL AND STACK DESIGNS AND MATERIALS

Various cell and stack designs are being explored by the different SOFC developers. The largest SOFC power generators to date, 25–100 kW, have been built by Siemens-Westinghouse. These generators are based on their seal-less tubular cell design and operate at

a nominal temperature of 1000°C. Smaller (10–25 kW) systems have been developed by Ceramic Fuel Cells Limited using planar fuel cell and stack designs. Fuel cell stacks of 1–10 kW have been built by Sulzer-Hexis, Honeywell, McDermott, Tokyo Gas, Global Thermoelectric, and others, using square or circular planar cell geometries.

The ~1000°C operating temperature of conventional solid oxide fuel cells limits the materials that can be used for the various cell components. Lowering the operating temperature of the SOFC increases the flexibility of materials selection and reduces cost, while maintaining fuel cell performance. Consequently, a considerable amount of the present R&D activity is focused on developing SOFC materials for use at 800°C, which is low enough that alternative materials become viable. Further reduction in operating temperature to 700°C or even 600°C would increase the materials selection flexibility while yielding shorter startup times and added cost savings. In addition, the lower SOFC operating temperatures result in higher Nernst and cell voltages, leading to increased energy conversion efficiencies. The reduced operating temperature would also improve mechanical robustness and reduce thermal stresses and chemical interactions among the various cell components.

Successfully lowering the SOFC operating temperature does, however, require the development of new materials and/or designs. The conventional yttria-stabilized zirconia electrolyte and the nickel-zirconia anode can be used at temperatures down to 650°C, but the electrolyte layer thickness must be reduced to 5–10 µm. At this thickness, the electrolyte can no longer function as the structural support for the cell, and electrode-supported or interconnect-supported cell and stack structures must be used. The cathode performance decreases at lower SOFC temperatures due to the lack of ionic conductivity in the manganite-based perovskites now used. Alternative materials being investigated include mixed conducting perovskites containing Co, Fe, or Ni on the B-site, and mixed conducting layered structures such as the high-critical temperature superconductors. Additionally, the newer cathode materials may be usable as the supporting structure in the SOFC; the added flexibility would result in certain benefits during cell and stack fabrication.

A significant change that can be realized in SOFCs operating at 800°C or below is the move from ceramic to metallic interconnects. In addition to lower costs, metallic interconnects would improve the mechanical strength of the stack and allow better thermal management during operation. Development of metallic interconnects is still at an early stage. The most significant problem that they face is oxidation of the metal or alloy on the air side of the bipolar plate. New alloys and/or protective coatings are being explored to overcome this problem.

6. STACK AND SYSTEM PERFORMANCE

We have analyzed the performance of a cross-flow planar SOFC cell and stack using an electrochemical performance model developed earlier [9]. The analysis examined the influence of various design and operating parameters on fuel cell efficiency, net power, and net power density. The parameters we examined include the operating cell voltage (0.6–0.85 V), geometric parameters (e.g., electrolyte thickness of 25–200 μm), materials properties (e.g., cathode interfacial resistances of 0.064–1.0 $\Omega\text{ cm}^2$), cell size (6×6 cm, 9×9 cm, and 15×15 cm), and fuel utilization (from 20% to 80%). The cell operation was constrained by using constant average and maximum cell temperatures of 800°C and 850°C, respectively, and limiting the maximum temperature gradient in the plane of the cell to 100°C/cm. In addition, the parasitic power consumption to overcome the pressure drop in the air flow channels was maintained at 10% or less of the gross power generated by the cell.

The base case was a 9×9-cm cell supported by a 200- μm -thick electrolyte and operating on H_2 /air with 80% fuel utilization. The calculated performance is a maximum net power density of 0.68 kW/L (of active stack volume) at a cell voltage of 0.65 V and an efficiency of 36% LHV. In contrast, for an anode-supported cell (9×9 cm) with a relatively thin (25 μm) electrolyte, the maximum net power density increases to 1.4 kW/L at 0.77 V, yielding an efficiency of 43%. Thus, the thin electrolyte design increases both the maximum net power density and the stack efficiency at that power density. Reducing the electrolyte thickness further, to 5 μm , increases the maximum net power density to ~1.5 kW/L at 0.8 V, for an efficiency of 45%. However, these maximum power densities and efficiencies are reduced significantly (to 0.9 kW/L and 39%, respectively), if the cathode interfacial resistance is increased from 0.064 $\Omega\text{ cm}^2$ to 0.5 $\Omega\text{ cm}^2$. Increasing the cell size increases the power generated per cell, but decreases the stack's maximum net power density. For example, by increasing the size of the anode-supported cell from 6×6 cm to 15×15 cm, the maximum net power increases by a factor of 5 (from 21 W to 105 W per cell), but the net power density decreases by a factor of 3 (from 2 kW/L to 0.6 kW/L).

The SOFC has a particular advantage for use in automotive applications where the power demands vary widely and rapidly over time. By decreasing the fuel utilization (i.e., providing excess fuel to the fuel cell), the cell's power output can be increased sharply, even when operating at the same cell voltage. This is due, in part, to a significant increase in the Nernst voltage for the exit fuel and air conditions. For example, for a cell operating at 800°C with H_2 /air, the exit Nernst voltage with 20% fuel utilization is 998 mV versus 874 mV with 80% fuel utilization. Thus, our analyses show that with a fixed cell and stack geometry, the power

output can be increased by 55% or more without decreasing the cell voltage or violating any of the operational constraints discussed above. Of course, operating with low fuel utilization would decrease the overall system efficiency and would not be recommended except for meeting high power demands for short periods.

We have also analyzed the performance of a relatively simple SOFC system operating on diesel fuel for an auxiliary power unit [3]. The analysis would apply as well to an automotive-sized system operating on gasoline. In addition to the fuel cell stack, the major components in this system are a small catalytic autothermal reformer, a combined spent gas burner-air preheater, a fuel pump, and an air blower. Since the autothermal reformer requires some feed water, the system also includes a small condenser to recover the water from the anode exhaust. Calculations were made for a system having 80% fuel utilization and a relatively high air flow rate to the cathode to provide all the stack cooling with the air temperature rise limited to 50°C. In this case, the overall system efficiency was 31%. By increasing the fuel utilization to 90% and reducing the air flow rate to one-third the original value, the calculated system efficiency was increased to 38%. With increased thermal integration to use the fuel cell exhaust to preheat the inlet air, system efficiencies of 40% or higher were obtained. Thus, these analyses show that the projected high efficiency of the automotive SOFC system operating on hydrocarbon fuel would be attainable.

7. OTHER CONSIDERATIONS

Other considerations of SOFCs for transportation applications include the sizes of the various components and the issues of water management, heat rejection, and sulfur in the fuel.

The fuel cell stack is likely to be the most expensive single item in the system. It would, therefore, be designed for the highest practical power densities. Assuming 0.4 W/cm² of active cell area, a 40-kW gross rated power (36 kW net) system would require an aggregate active area of 10 m². The largest component in the system would be the spent gas burner-air preheater, which the above analyses showed could require an active heat transfer area of 30 to 60 m² (i.e., three to six times the active area in the fuel cell stack), depending on the operating temperature difference driving forces. If this heat exchanger is of a conventional design that offers ~0.1 m² heat transfer area per liter of volume, a 300 to 600 L unit would be needed. Novel designs for compact heat exchangers offering much higher active area per unit volume, such as some microchannel concepts, would be needed to reduce the size of this heat exchanger. All of the other components are considerably smaller and would not add significantly to the total volume of the SOFC system.

The catalytic autothermal reformer requires the use of water to avoid excessively high temperatures that would otherwise be needed to prevent carbon formation. For use in automotive systems, this amount of water must be recovered from within the system. Overall, excess water results from the conversion of the gasoline or other fuel. Recovering the needed water, however, involves the use of a suitably placed, air-cooled condenser. If reforming processes can be developed that do not need water as a reactant, or the cell reaction water can be recycled to the reformer without phase change, the resultant SOFC system would be inherently simple and easily controllable.

On the other hand, rejection of the waste heat from the SOFC system is straightforward. All of the waste heat is rejected either in the process exhaust stream or at the air-cooled condenser. The latter operates with high temperature-difference driving forces and is quite compact, approximately one-fiftieth the size of the air preheater. Thus, heat rejection from the SOFC system can be easily handled, even at high ambient temperatures.

One other significant consideration for automotive SOFC systems is the potential sulfur content of the fuel. While some of the alternative transportation fuels are essentially sulfur-free (e.g., methanol, ethanol, and propane), the petroleum-derived gasoline and diesel have sulfur contents ranging from 300 to 1000 ppm by weight. Even the reduced sulfur fuels of the future are likely to contain 30 to 80 ppm of sulfur. The reformat derived from these fuels would have a sulfur content of 3 to 8 ppm of H_2S by volume. This concentration may still be too high for SOFCs using conventional materials, and sulfur-tolerant fuel cell materials and designs would be needed. Without those, the SOFC system would need to incorporate sulfur removal, which would add a significant degree of complexity to the system.

8. CURRENT DEVELOPMENTS

Improved materials, processes, and components for use in low-temperature (700–800°C) SOFCs are being developed at many universities, national laboratories, and commercial organizations around the world. These include research groups at the universities of Birmingham, California (Berkeley), Keele, Hannover, Tokyo, and Pennsylvania, as well as Northwestern University, Imperial College, Forschungszentrum-Jülich, ECN (Netherlands), Argonne National Laboratory, Honeywell, McDermott, Global Thermoelectric, Sulzer Hexis, and Ceramic Fuel Cells. Many commercial organizations are also developing complete prototype systems for a variety of applications, including distributed generation, portable and transportation power, and auxiliary power units.

Recent advances include improved cathode and interconnect materials, enhanced single-cell and stack performances, near-commercial manufacturing of cells and stacks of designs tailored to customer specifications, and demonstrations of prototypic systems of power output ranging from <100 W to >100 kW. Specifically for transportation applications, Delphi Automotive Systems and Global Thermoelectric have recently reported the successful testing of an SOFC-based auxiliary power unit for automotive use [10].

9. CONCLUSIONS

- The SOFC system requires a very simple and compact fuel processor, even for petroleum-derived fuels, such as gasoline and diesel fuel. The fuel processor does not need to achieve complete conversion of the fuel and does not include any process steps to remove carbon monoxide from the reformat. As a result, the fuel processor in an automotive SOFC system is expected to be one-fifth to one-tenth the size of the fuel processor in a PEFC system of equivalent power rating.
- The SOFC system has the potential of delivering high power densities, in excess of 1 kW per liter of active stack volume for 10×10-cm cells. Similar power densities may be achievable with larger (up to 40×40 cm) cells by close coupling of the autothermal reformer and the fuel cell stack.
- The SOFC system offers excellent potential for thermal integration between the fuel processor (a catalytic autothermal reformer) and the fuel cell stack as they both operate in the 700–1000°C temperature range. Current R&D has the goal of reducing the operating temperatures toward the lower end of this range, which will result in higher efficiencies, reduced materials and processing costs, and thermally and mechanically rugged designs.
- The startup time and fuel consumption for the SOFC system is primarily governed by the heat-up requirements of the fuel cell stack. Based on a heat-up rate of 2°C/s, the stack startup time would be ~400 s, with a fuel feed rate during this time of ~75% of the value at the rated power.
- The startup fuel consumption would be comparable to the fuel used during operation in a very light-duty vehicle (one hour operating time with two deep thermal cycles per day). With longer duty cycles and/or with warm starts, the startup fuel consumption would be very much less.
- The thermal and mechanical ruggedness of SOFC stacks must still be addressed. The use of metallic bipolar plates and advanced stack designs is expected to greatly alleviate these problems.

- Simulation of an SOFC-based auxiliary power unit has shown potential efficiencies of 40% or higher, based on the lower heating value of the fuel.
- The spent gas burner/air preheater is likely to be the largest component of the SOFC system. Novel heat exchanger designs are needed that offer active areas much higher than the $\sim 0.1 \text{ m}^2/\text{L}$ available with conventional heat exchangers.

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