ABSTRACT

Pacific Northwest National Laboratory (PNNL) is developing technologies for low-cost modular solid oxide fuel cell (SOFC) systems to accelerate the development of such systems for stationary, mobile, and military applications. This work is being pursued with funding from several United States Department of Energy (DOE) programs, including the Solid State Energy Conversion Alliance (SECA) Core Technology Program, the Advanced Research and Technology Development (AR&TD) Program, and a Co-operative Research and Development Agreement (CRADA) with Delphi Automotive Systems. Experimental and modeling studies are being conducted on a number of important challenges, including low-cost materials and fabrication processes, seal and interconnect stability, cell and stack designs, rapid system startup, thermal cyclability, and long lifetime. This paper summarizes the progress made in these investigations.

INTRODUCTION

High efficiency, superior environmental performance, long-term reliability, and compact size of planar solid oxide fuel cells (SOFCs) make them very attractive for power generation. These cells are capable of achieving very high power densities (1-4). Additionally, sizeable cost reductions are possible through a concept called mass customization that is being pursued in the U.S. Department of Energy's Solid State Energy Conversion Alliance (SECA). This concept involves the development of a core SOFC module, approximately 3 to 10 kW in size, that can be mass produced and used for residential or auxiliary power unit applications. Multiple mass-produced core modules can be combined for applications with larger power needs in distributed stationary power generation, transportation, and military market sectors, thus eliminating the need to produce custom-designed and inherently more expensive fuel cell stacks to meet a specific power rating.

Pacific Northwest National Laboratory (PNNL) is engaged in several activities in support of the design and development of such modular SOFC systems. These activities include:

- Development of optimized materials and cost-effective fabrication techniques for high power density anode-supported planar SOFCs for operation at reduced temperatures (≤800°C).
- Development of cost-effective, reliable, and durable seals and interconnects for the SOFC stacks.
- Application of thermomechanical and electrochemical modeling to aid in the design of SOFC stacks capable of rapid startup and thermal cyclability, two properties which are particularly important for transportation applications.
- Development of optimized, cost-effective reformation of hydrocarbon fuels (natural gas, gasoline, diesel, etc.) integrated with the cell stack.
- Fabrication, testing, and evaluation of prototypical cell stacks to verify performance and performance stability.

PNNL is also assisting in the development of an on-board auxiliary power unit (APU) for application in the transportation industry through a CRADA with Delphi Automotive Systems (5).

This paper reports on the progress that has been made in the development of the anode-supported planar cells and in the development of the auxiliary power unit.

**ANODE-SUPPORTED PLANAR SOFCs**

Among all designs of the solid oxide fuel cells investigated to date, the most progress has been made with the tubular design (6,7). However, the long current path around the circumference of tubular SOFCs makes their electrical resistance high, and specific power output (W/cm²) and volumetric power density (W/cm³) low (7). These low power densities make tubular SOFCs unattractive for transportation applications where space is at a premium; typically the volume target for an automotive APU is about 10 liter/kW. For this reason, only planar SOFCs are considered suitable for use in the transportation sector.

Currently, three basic designs of SOFCs are under development; these are electrolyte-supported, cathode-supported, and anode-supported. In the electrolyte-supported cells, the thickness of the electrolyte, typically yttria-stabilized zirconia (YSZ), is 50 to 150 μm, with thin electrodes fabricated on its two sides. In such cells, the ohmic resistance is high due to high electrolyte resistivity and such cells are suitable for operation at ~1000°C. In electrode-supported designs, the electrolyte thickness can be much lower, typically 5 to 20 μm, and hence the ohmic resistance is much lower than in the electrolyte-supported cells. Thus, the electrode-supported cell design, in principle, is better suited for operation at lower temperatures. Lower operation temperature results in less degradation of cell and stack components, makes feasible use of inexpensive metallic interconnects in place of expensive lanthanum chromite based ceramic interconnects, is less demanding on seals, and aids in faster heat up and cool down. The anode is generally selected as the supporting electrode because, in comparison to the cathode material (typically a doped lanthanum manganite), the anode material (typically a Ni/YSZ cermet) provides superior thermal and electrical conductivity, superior mechanical strength, and minimal chemical interaction with the YSZ electrolyte at high temperatures encountered during cell fabrication.
Using relatively thin Ni+YSZ anode and porous Sr-doped LaMnO₃ (LSM) cathode with thin YSZ electrolyte, several research groups have demonstrated very high performance in anode-supported cells (1-4). Kim et al (1) reported a maximum power density of ~1.8 W/cm² at 800°C in anode-supported single cells with 10 μm thick YSZ electrolyte; Figure 1 shows their data. Such high power densities make anode-supported planar cells very attractive for use in the core SOFC module being developed for stationary, mobile, and military applications.

At PNNL (8,9), similar anode-supported cells are being developed using ~10 μm thick tape cast YSZ electrolyte and ~600 μm thick tape cast Ni/YSZ anode which are laminated together and co-sintered at about 1350°C for 1 hr. Cathode consists of either Sr-doped lanthanum manganite (LSM), LSM+YSZ, or Sr-doped lanthanum ferrite (LSF), which is applied to the electrolyte by screen printing and then sintered. Cells with LSF cathode and a Ce₀.₈Sm₀.₂O₁.₉ interlayer between the LSF and the YSZ electrolyte showed a maximum power density of 0.52 W/cm² at 0.7 V at 800°C with air as oxidant and a mixture of 97% H₂+3%H₂O as fuel. Studies to develop optimized cathode and interlayer materials and optimized microstructures to achieve higher and stable performance are presently in progress.
SOFCs FOR AUXILIARY POWER UNITS

The polymer electrolyte membrane (PEM) fuel cell is generally regarded as the fuel cell of choice for transportation applications. However, PEM fuel cells have several challenges to overcome. Firstly, PEM fuel cells are seeking to replace the internal combustion engine which is well entrenched and highly competitive in cost, specific power, and reliability (though still very inefficient). Secondly, PEM fuel cells require pure hydrogen, with no carbon monoxide (CO), as fuel to operate successfully. However, it may be a very long time before a hydrogen infrastructure is established around the world, and on-board reformer systems to produce hydrogen from existing fuel base (gasoline, diesel, etc.) are technically challenging, complex, and expensive. Furthermore, it is difficult to eliminate all the CO from the reformate stream.

In contrast, SOFCs are highly efficient, capable of achieving fuel to electric efficiencies of over 50%. More importantly, SOFCs can use CO along with H₂ as fuel, and their higher operating temperature and generation of water on the cell anode side makes feasible either on-cell or in-stack reformation of hydrocarbon fuels (gasoline, diesel). Also, in contrast to PEM fuel cells, no noble metal catalysts are used, reducing the cost of the cells. Furthermore, it is expected that the initial application of SOFCs will be in on-board auxiliary power units (APUs) instead of for the drive train to replace the internal combustion engine. Such APUs will operate on existing fuel base (gasoline, diesel) and will supply the ever-increasing electrical power demands of the luxury automobiles, recreational vehicles, and heavy duty trucks; Figure 2 shows the historical trend of peak electrical power requirements in automobiles. The estimates of peak electrical power requirements are listed in Table I.

![Figure 2. Peak Electrical Power Requirements in Automobiles.](image-url)
Table 1. Peak Electrical Power Requirements for Automotive Components.

| Component                | kW  |
|--------------------------|-----|
| Electric suspension      | 12  |
| Air conditioning         | 5   |
| Heated windshield        | 2.5 |
| Electric valve control   | 2.4 |
| Electric power steering  | 1.3 |
| Anti-lock brake system   | 0.67|
| Catalyst heater          | 0.6 |
| Diesel direct injection  | 0.47|
| Electric coolant pump    | 0.3 |
| Compartment fan          | 0.3 |

Use of SOFCs in auxiliary power units for the transportation sector will enable high volume manufacturing, increase their reliability and durability, and reduce cell and system cost, making possible their successful application in other market sectors as well. Once the cost of SOFC-based systems is reduced significantly through mass customization, their use for automotive propulsion might also become feasible some day.

The APU being developed by Delphi Automotive Systems (5) under the CRADA with PNNL is to provide power in the 5 kW range at 42 Vdc. This APU is intended to power an electric air conditioning system with the engine power off, and will operate on gasoline. The requirements for this system include most of the normal automotive items such as physical size, weight, power density, vibration levels and cost. However, this unique application also requires fast heat up and ability to be thermally cycled often. These last two requirements necessitate innovations in cell design, use of metallic interconnects, and use of low mass insulation.

The building blocks of an APU consist of the following sub-systems:

- Solid oxide fuel cell stack incorporating anode-supported planar SOFCs
- Fuel (gasoline) reformation system
- Waste energy recovery system
- Thermal management system
- Process air supply system
- Control system
- Power electronics and energy storage (battery) system

In the initial proof-of-concept unit developed by Delphi Automotive Systems (5), above sub-systems were fabricated separately and assembled together in one package. In the future, innovative integration of the sub-systems will be explored to achieve smaller size, robustness, and lower cost. PNNL will explore the possibility of integrating fuel reformation with the cell stack because of the heat and water being available from the cells, and comparable temperatures of cell operation and hydrocarbon fuel reformation. Application of microchannel technology to hydrocarbon fuel reformation will also be
explored; successful use of this technology will enable significant cost savings by reducing component size without sacrificing performance.

Also, APUs require fast heat up and ability to be thermally cycled. At PNNL, thermomechanical modeling is being conducted to compare different planar stack designs to optimize startup performance (10). Thermal-fluids and stress modeling is also being performed to predict transient temperature distribution and to determine the thermal stresses based on the temperature distribution experienced by interconnects in the cell stacks. Such modeling should aid in achieving stack and APU designs that provide required startup times and thermal cyclability.

SUMMARY

Pacific Northwest National Laboratory is conducting a variety of investigations to develop cost-effective, high power density, anode-supported planar solid oxide fuel cells under the sponsorship of U.S. Department of Energy. Such fuel cells will be used in a core SOFC module of 3-10 kW size capable of being mass produced for use in stationary power generation, mobile, and military applications. Current work at PNNL is aimed at achieving higher stack power density, faster startup, thermal cyclability, hydrocarbon fuel reformation either on the cell or integrated with the cell stack, and lower cost.

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