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SOLID OXIDE FUEL CELLS FOR MAN-TRANSPORTABLE UNMANNED VEHICLES

Thomas Westrich
Ultra Electronics, AMI
Ann Arbor, MI

ABSTRACT

The military has a need to source propulsion systems that have enhanced efficiencies, lower noise signatures, and improved lifetimes over existing power systems. This is true for energy storage systems on unmanned ground vehicles and for manned vehicles (i.e., Auxiliary Power Units). Fuel cells have the promise to achieve all of these goals. However, to be truly effective, these advanced systems should integrate seamlessly with the current supplies of energy storage (batteries) and energy sources (logistics fuel). The largest fuel cell development hurdle to date has been the ability to handle sulfur concentrations present in logistics fuel. Secondly, the reformer must be capable of several thousands of hours of operation utilizing logistics fuels without loss of performance due to sulfur or carbon deposition. Advancements in several key technologies have the potential to allow development of a logistics fueled solid oxide fuel cell with similar size, weight, and power Advancements in the areas of liquid sulfur-removal, sulfur trap regeneration systems, and reformate purification may all contribute to the development of a logistics-fueled 250W solid oxide fuel cell system. This paper will outline the development of logistics-fueled fuel cell systems for advanced propulsion, specifically focusing on man-transportable robotic systems and the technical barriers and innovations which could allow for the development of deployable solid oxide fuel cell systems.

CURRENT STATE OF THE ART

Unmanned ground or aerial power systems have strict specific power and energy requirements. Traditional power sources (batteries) span a wide range of specific power targets, but have limited specific energy. Therefore, to increase unmanned mission duration, advancements in power systems' specific energy targets or system efficiencies are required. Further, said advancements should not be at the expense of system specific power targets (i.e., higher fuel efficiency with little added mass).

Currently, advanced technology Solid Oxide Fuel Cell (SOFC) power systems with very high system specific power (100 W/kg) and specific energy (1,200 Whr/kg) are produced for the Lockheed Martin Stalker XE Tier 1 unmanned aerial vehicle. This technology combines a high power density solid oxide fuel cell with a high energy density liquid hydrocarbon fuel (propane) to produce a system with the power density required by the Stalker, while also extending mission duration by 4-8 times over a battery-powered Stalker vehicle. Further, this system has been designed to handle environmental and operational stresses, such as vibration, sudden impacts and a wide range of ambient temperatures and altitudes.

A JP-8 fueled SOFC power system with high specific power (50-100W/kg) would have direct application across the military in unmanned vehicle applications that currently rely on various battery technologies. Further, this technology could also have a significant impact in other areas of the military, such as remote battery charging, small manned power systems, or even for low-power FOB charging applications.

ADVANCED POWER SYSTEM DEVELOPMENT

The consumption of liquid fuels, such as JP-8, in an SOFC is much more challenging than gaseous fuels like propane or methane. As a result of the chemical composition of liquid fuels, technical risks during reforming and SOFC operations are greatly increased. Carbon deposition (coking), reformer thermal stability, and reformer control are examples of the elevated risk areas. For example, carbon deposition rates are known to increase in the presence of aromatic hydrocarbons and olefins (so called coke precursors) [1, 2]. Complex reformer control parameters or algorithms are required to minimize the production of these coke precursors.

Sulfur species also increase the complexity of reforming heavy hydrocarbons. Sulfur species are common in gaseous and liquid hydrocarbon feeds, whether as a result of addition, in the case of flammable gas detection, or as a result of the high cost of separation during refining. Sulfur is well known to act as a catalytic poison for both reforming catalysts and solid oxide fuel cell anodes. Point-of-use separation of sulfur from gaseous systems is relatively easy as compared to separation from liquid systems, due to the molecular differences between thiols and propane. However, the high levels of sulfur found in logistics fuels (up to 3,000ppmwS) necessitate some mechanism of sulfur separation or mitigation prior to use as a fuel in an SOFC system.

Advanced Sulfur Filtration System Development

Separation of sulfur species from gaseous hydrocarbons or hydrocarbon reformate gases is relatively simple, and can be conducted with an adsorbent bed. Granulated zinc oxide adsorbent beds are used industrially to remove hydrogen sulfide from flue gases. However, separation of sulfur species from liquid hydrocarbons is more challenging due to the inherent similarity between sulfur-containing hydrocarbons and sulfur-free hydrocarbons. Further, liquid sulfur adsorbents have a significantly lower capacity [3, 4], which leads to increased adsorbent masses for a designed system lifetime. For example, optimized gas-phase sulfur adsorbent materials are capable of retaining as much as 0.35-1.0g sulfur per gram of adsorbent media; in contrast, liquid adsorbents retain 0.003g sulfur per gram adsorbent. As a result, equivalent breakthrough times (or mission duration) require 100x more media for liquid-fuel solutions than gas-fueled solutions. This extreme mass increase decreases the likelihood of reaching the specific power requirements of unmanned vehicles. New liquid-phase sulfur adsorption technologies with higher capacities could bridge this technology gap.

Advancements in oxidative desulfurization (ODS) of liquid fuels have lead to desulfurization materials that can achieve reasonable mission durations with relatively low adsorbent material masses. In the ODS process, once the media has been saturated with sulfur from liquid hydrocarbons, an oxidant (typically air) is used in conjunction with temperature to oxidize the sulfur deposits. Therefore, ODS requires either a single or parallel regeneration loop, depending on the designed mission duration or breakthrough time. With the ODS process, improvement of both specific power and energy can be realized through improvements in either sulfur adsorption capacity or auxiliary process equipment mass reductions.

Further, sequential sulfur adsorption beds can increase the final system specific power and energy by optimizing media beds to achieve the end result of a maximum sulfur concentration delivered to the solid oxide fuel cell system.

Filter-Free Fuel Cell Development

To maintain a high specific power ratio for fuel cell power systems operating on sulfur-laden fuel, the size and mass of sulfur separation systems must be minimized. In the ideal example, an absolute minimum mass would be allocated to sulfur removal. Recently, prototype SOFC technology has been developed that retains as much as 90% of the specific power of traditional SOFC systems. This technology uses hydrogen selective membranes, which are extensive used in the industrial purification of hydrogen and more recently used in steam reformers.

A schematic of the cell and membrane design used in this work is shown in Figure 1. Sulfur-laden fuel species are introduced to the interior of the hydrogen-selective membrane, and can pass through the length of the SOFC tubular cell. When hydrogen is present in the reformate stream, an equilibrium partial pressure of hydrogen is transferred to the anode of the SOFC. As hydrogen is electrochemically consumed, a hydrogen partial pressure gradient is established across the membrane, causing a flow of hydrogen through the membrane to the SOFC anode. As a result, further reforming and shift reactions are favorable within the membrane, leading to higher utilizations of light

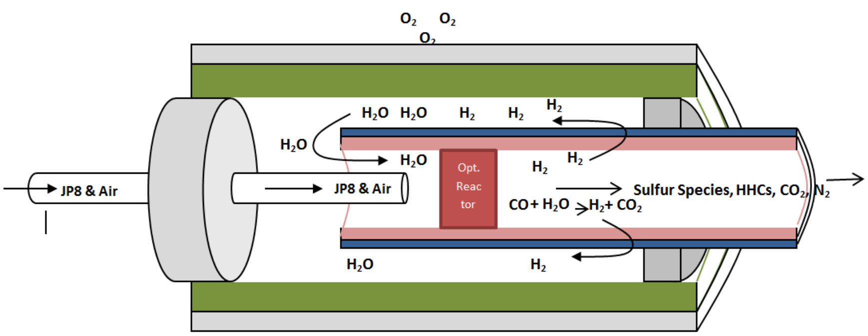

Figure 1: Schematic of the membrane-based JP-8 fueled SOFC cell design.

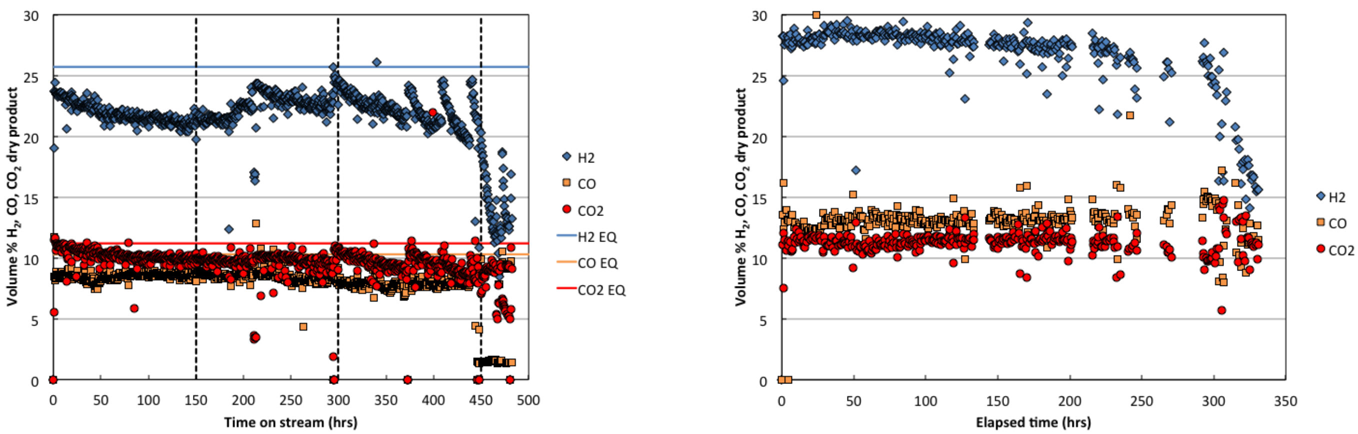

Figure 2: Endurance reforming results for Rh-based Ceria catalyst, tested with 300ppmwS JP-8. Results for endurance operation (right) and cyclic operation (left) are shown.

hydrocarbons. The sulfur present in the fuel stream flows through the interior of the membrane and into the SOFC combustion chamber, bypassing the SOFC entirely and preventing SOFC poisoning or degradation.

As a result of this mixed technology approach, it is possible to produce high power densities at the cell level, while minimizing the required sulfur mitigation mass. In contrast to absorption-based filtration, this technology does not require the membrane to be scaled in size or mass to the required mission duration or system lifetime. Additionally, electrochemically generated steam can be easily recirculated into the fuel stream to aid in steam reforming or water-gas-shift reactions and increase system efficiency. During the course of technology developments, several improvements upon this design were made, however, the illustrated design shown in Figure 1 represents the core technology.

As this system must operate a reformer in the presence of sulfur, catalyst selection, reformer control and reformer lifetime were investigated during early development work.

Catalytic reforming experiments with 300ppmwS JP-8 were conducted with several catalyst formulations. The formulation with highest endurance performance was a Rhodium-loaded ceria-zirconium catalyst [5]. Catalytic partial oxidation reforming endurance results are shown in Figure 2. The rhodium catalyst operated with near-equilibrium product distributions for most of the 450hr test. Catalyst deactivation near the end of the test resulted in the formation of additional light hydrocarbons, and the accumulation of heavy hydrocarbon tars near the reactor effluent. It is also possible to minimize technical risk to the performance and lifetime of the reformer in the presence of sulfur by utilizing specialized reformer technology developments of 3rd party reforming technology companies. in future development work.

To achieve the required SOFC power density and system specific power, the membranes were required to transfer a minimum rate of hydrogen to the SOFC anodes. These flux test results are shown in Figure 3, along with the minimum required flux to meet power density requirements. The palladium-based hydrogen permeable membranes were made with 3 thicknesses: 13.1µm, 17 µm, and 25 µm. The hydrogen flux results followed the accepted membrane governing equations. For example, the measured hydrogen flux is inversely proportional to the membrane thickness, as can be seen in Figure 3.

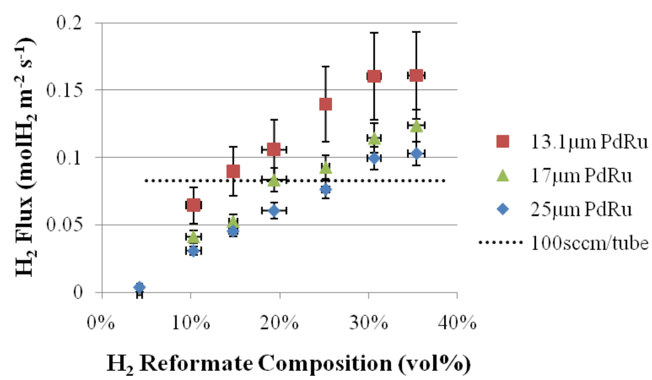

Figure 3: Hydrogen permeable palladium membrane flux results for membranes with varied thicknesses.

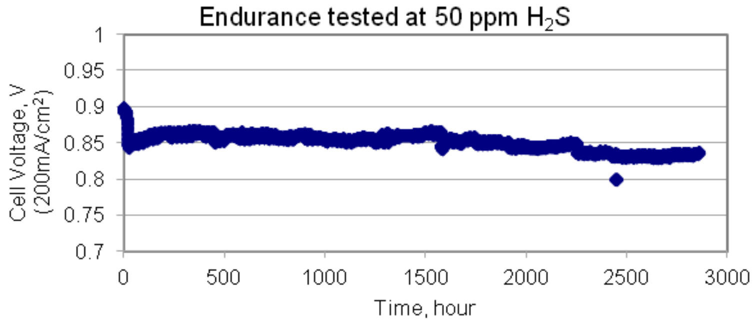

Figure 4: Demonstration of the performance of a SOFC cell and membrane assembly operating with 50ppm H_2S for 3,000hrs at approximately 200mA/cm^2.

Qualitative membrane selectivity measurements were conducted using a single cell/membrane bundle. Upon exposure to H_2S concentrations in excess of 10ppb, the SOFC experiences significant performance degradation. A 2,800 hour experiment using a fuel source with 50ppm H2S in hydrogen successfully proved high membrane selectivity to hydrogen, and the endurance capability of the membrane (Figure 4).

Once the individual components were shown to meet performance requirements, a single cell/membrane bundle was tested with JP-8 reformate. Power curves for several cells operating on reformate are shown in Figure 5. For each cell, the thermal-to-electrical efficiency was calculated to be approximately 15%.

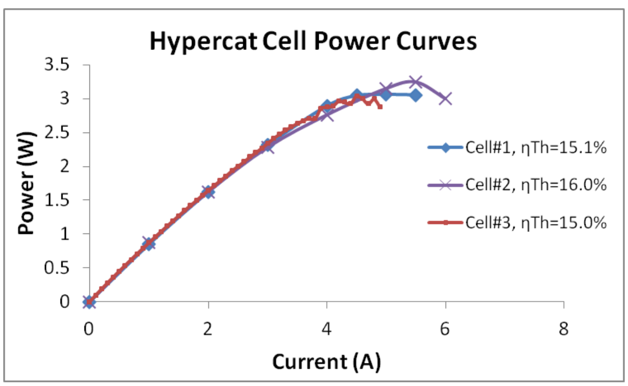

Figure 5: Single cell (SOFC tube and membrane bundle) power curve operating utilizing JP-8 reformate (approximately 40ppmv H_2S).

The demonstrated thermal efficiencies of this technology are limited by two prevailing factors. Firstly, an energy balance reveals that significant energy is not transferred across the current palladium membrane. This energy balance shows that nearly 50% of the thermal energy input to the system (in the form of JP-8) is not transferred across the membrane, either as a result of: sulfur-poisoned reforming reactions, low water recirculation rates, and sulfur-induced changes in hydrogen permeability. Secondly, the use of a primary catalytic partial oxidation reforming reaction significantly decreases the potential system efficiency; effective water recirculation within the cell/membrane bundle is an important integration challenge for future development efforts.

Further development of this technology into a high-TRL product will be required, specifically in the areas of JP-8 reforming (catalyst endurance), membrane manufacturing (low-cost, high-permeance, high-selectivity membranes), and improved system integration. The four 250W prototype units built for this effort experienced either lifetime or performance issues which are related to catalyst selection and endurance as well as membrane manufacturing and system integration issues. These known issues can be addressed with future targeted development efforts. For example, significant developments have been made in the field of palladium-based hydrogen separation membranes over the past 40 years. Sulfur-tolerant [6] and very thin membranes [7] with high hydrogen permeance have been reported in literature and used in commercial hydrogen purification processes. Use of this advanced membrane technology in the presented SOFC design would result in improved system performance, reliability and robustness – key criterion for unmanned vehicles. Further use of advanced reformer or catalyst design would also improve reformer reliability and endurance performance.

This technology has the capability to significantly increase system specific power and energy relative to a comparable reformer/sulfur filtration/SOFC system. The use of a continuous and non-consumable sulfur filtration device, such as a hydrogen-selective membrane, results in the complete absence of large, heavy sulfur filtration beds. Therefore, this design approach has a minimal impact on system specific power and could be an excellent option for JP-8 fueled unmanned aerial or ground vehicles.

Very High Power Density System Development

Another facet of ongoing development is the improvement of fuel cell current and power density. Increased system specific power realized from this effort can be leveraged across SOFC platforms, regardless of the fuel source. As a result of this development, very high power density SOFC systems are possible (140-200W/kg, as opposed to the current 100W/kg systems).

Improvement of system specific power expands the capability of the unmanned system and allows for increased payload power (systems, flight/ground speed, etc.) and longer mission duration.

FUTURE DEVELOPMENT OPPORTUNITIES

As the use of unmanned power systems increase, the system capability must also expand to meet specific user requirements. Specific user requirements that impact the power system are typically: specific or peak power, specific energy or mission duration, lifetime, and cost. As a result, future directed development into high-power density fuel cell systems, high thermal-to-electric efficiency systems, robust system and fuel cell design, and low-cost system design will lead to improved power system specifications and enhanced user capabilities. Given the current state of the art in sub-kilowatt portable power systems, the largest development opportunity is the improvement of system thermal-to-electric efficiency.

High Efficiency Power System Development

The introduction and development of improved fuel reforming strategies have the capability to double or even triple the thermal-to-electric efficiency and mission duration of current SOFC power systems. For example, large stationary solid oxide fuel cell systems are able to reach 40-60% net electrical efficiency utilizing advanced reforming strategies such as steam reforming or anode gas recirculation. In contrast, catalytic partial oxidation reforming has a limited net efficiency of 15-25%. However, adoption of advanced steam reforming technology to the sub-kilowatt mobile SOFC systems is technically challenging and has limited adoption thus far.

For example, it is well known in the SOFC community that steam reforming is able to provide significantly higher electrical efficiencies than catalytic partial oxidation. However, increased electrical efficiency from steam reforming is at the expense of increased system parasitic losses and system complexity. At a minimum, steam reforming requires a water storage tank, a mechanism to fill said storage tank from a condenser, a water delivery or pumping system and a water vaporization system. All of these components add significant mass to the power system, thereby reducing the system specific power.

Alternatively, it is possible to use the unutilized anode gas stream as a source of water vapor for reforming purposes. In this strategy, no liquid water handling equipment is required, which has the potential to reach equivalent system specific power as traditional SOFC power systems, while exceeding the system specific energy by 2-3 fold. However, a large technical investment is required in this design: development of a variable speed, high-temperature tolerant blower to recirculate anode gas, the development of advanced control algorithms to safely operate the fuel cell amongst the complicated system interactions, and the development of high temperature, electrically insulative fuel cell tube manifolding to act as an anode gas plenum.

In addition to the development of systems with increased efficiency, continued development efforts to increase power, lifetime and cost are also important. The lessons learned in such development efforts may be directly applied to fuel cell systems operating on propane or logistics fuel, thereby maximizing the impact of development efforts.

CONCLUSIONS

Developments in hydrocarbon reforming, sulfur capture or sulfur filtration show significant promise to achieve technical goals required of unmanned vehicles' SOFC power systems. Based on the technical progress of propane-fueled SOFC systems used in the field, logistics-fueled SOFC systems can be developed with a wide range of sulfur mitigation and reforming options. As a result of this potential parallel design path opportunity, the associated development risks can be mitigated. Further improvements to system specific power, thermal-to-electric efficiency, system lifetime, and cost will markedly increase system capability and performance.

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