Low Cost, High Efficiency Reversible Fuel Cell Systems

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Objectives

- Improve performance of reversible solid oxide stacks (capable of operating in both fuel cell and electrolysis modes) by reducing polarization and rate of degradation.
- Demonstrate an integrated fuel cell hot subassembly with a stack of about 50 reversible type cells operating on natural gas.
- Evaluate the economic impact of reversible solid oxide fuel cell/electrolyzer systems and consider applications where a competitive advantage may be achieved.

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Production section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year R,D&D Plan:

- A. Fuel Processor Capital Costs
- B. Operation and Maintenance (O&M)
- Q. Cost
- R. System Efficiency
- S. Grid Electricity Emissions
- T. Renewable Integration
- U. Electricity Costs

Approach

- Evaluate alternate materials and cell geometry parameters to reduce cell polarization.
- Reduce operating temperature to improve stack life.
- Test a stack of up to 50 reversible cells in a new integrated system assembly.
- Perform cost and environmental evaluations of alternative equipment and system configurations.

Accomplishments

- Optimized cell geometry by modeling the stack and system.
- Designed, fabricated, and evaluated reversible stack performance over a range of temperatures and operating conditions.
- Demonstrated stable operation for >1200 hours in small reversible stacks at acceptable efficiency.
- Met all stack performance targets.
- Completed preliminary economic analysis and identified applications where reversible electolyzer/fuel cell systems may have a competitive advantage.

Future Directions

- Fabricate and test a stack of up to 50 reversible cells using standardized processes.
- Complete evaluation of alternative stack operating conditions.
- Complete cost and environmental assessment of alternative system configurations.
- Complete present project in Fall of 2003.

Introduction

The Technology Management, Inc. (TMI) reversible (fuel cell - electrolyzer) system employs a stack of high temperature solid oxide cells to electrochemically produce either electricity (from fuel and air or fuel and oxygen) or hydrogen (from water and supplied electricity). A fuel cell version of the system operating at atmospheric pressure could operate on natural gas (or other carbon-containing fuel) and air. A high-pressure reversible electrolyzer version would make high-pressure hydrogen and oxygen from water and also operate in reverse (on demand) to generate electricity from these gases. The novel reversible stacks require simpler and more efficient auxiliary equipment than conventional fuel cell systems. Calculations have shown that the proposed systems could achieve higher efficiencies in both fuel cell and electrolysis modes than existing alternatives.

During the current phase of the project, TMI has demonstrated reversible cells and stacks which bettered the quantitative performance targets set for polarization voltage, operating efficiency, rate of performance loss, and seal leakage rate. This success resulted from a cell and stack development program that studied the effects of component dimensions and fabrication details, assembly procedures, startup procedures, and operating conditions. A complete test system for stacks up to 50 cells has been designed, built, and operated (with stacks up to 19 cells thus far). Cost and engineering studies were extended significantly to identify cases where vehicle-grade, high-pressure hydrogen could be produced at the refueling point from natural gas at costs below DOE targets. The use of the proposed reversible electrolyzers for energy storage also appears to offer sizeable advantages over the expected cost, size, and weight of known rechargeable batteries.

Approach

The overall goals for reversible stack performance are (a) high efficiencies at selected power levels and (b) low degradation rate (power loss over time at constant flows). High efficiencies, in turn, require low polarization voltages and low seal leakage rates. Low polarization voltages in solid oxide stacks result from low concentration polarization and low resistive polarization, the latter being quantified by area specific resistance (ASR, expressed in Ohm-cm^2). Seal leakage rates may be computed as a valve coefficient (C_v, a relative measure of flow through an orifice). Simulations and prior experiments have shown that achieving the desired performance of TMI's reversible stacks is significantly more difficult in fuel cell mode than in electrolysis mode. The current phase uses the following goals in fuel cell mode.

- ASR < 1.00 Ohm-cm^2
- Cv < 10^{-5}
- Degradation rate < 5% per 1000 hours

Fabrication improvements, materials substitution, temperature optimization, and assembly improvements were evaluated to reduce ASR. Dimensional optimization and startup details are being used to minimize seal leakage. A lower degradation rate is achieved via improvements in fabrication details and seal leakage. Concentration polarization was reduced by combining increased porosity, improved dimensional stability, and adjusted component dimensions.

The design of a larger system for operation with up to 50 cells is based upon prior work by TMI on other test systems augmented by additional system-specific engineering calculations.

Cost and engineering studies build on work performed during the prior phases of this project as well as cost studies by TMI for other projects. Data on natural gas and electricity pricing are from the Energy Information Administration. Hydrogen cost targets are taken from public DOE hydrogen and fuel cells documents.

Results

Reversible Stack Development. At the start of the present phase, measurements of reversible cell performance were poor and insufficient to achieve targets. There were also considerable variations from cell to cell. Concentration polarization was initially high, thereby limiting maximum possible current densities to only about 100 mA/cm^2. Major improvements have been the result of increased porous layer thicknesses and improved dimensional stability, thereby maintaining higher effective porosities.

Early ASR measurements ranged from about 2.2 to over 3.0 Ohm-cm^2, which were in excess of the goal of 1.00 Ohm-cm^2. ASR was improved by reducing seal leakage, optimizing electrical contact between cell layers, and by slightly increasing temperature. Studies have found that ASR varies with both fuel utilization (fuel oxygen potential) and current density. Recent ASR measurements have ranged from 0.84 to about 1.4 Ohm-cm^2 in single cells and stacks, increasing at high fuel utilization.

Early seal leakage rates varied, sometimes being more than ten times the target C_v of 10^{-5}. Improvements in dimensional tolerances, assembly procedures, and startup procedures have resulted in measured C_v values below the target, with the best values being less than 10^{-6}.

Early degradation rates were highly variable, sometimes exceeding 20% per 1000 hours. One 19-cell stack operated for over 360 continuous hours on natural gas with less than 2% per 1000 hours degradation. However, if a stack has one or more significant leaks, considerably higher rates are still seen. The temperature dependence of degradation rate has not yet been quantified.

A limited number of cells and stacks were tested in electrolysis mode, with the results confirming those from earlier work. Measured ASR in this mode equaled that in fuel cell mode. In projected commercial solid oxide electrolyzers, allowable ASRs are higher than those for highly efficient fuel cell systems due to electrolyzer heat balance considerations.

Cell and Stack Fabrication. Evolutionary improvements and modifications were made to existing TMI materials specifications, fabrication methods, and cell dimensions to accommodate the reversible cell and stack design. Minor modifications of standard TMI fabrication procedures were sufficient to produce thicker fuel and oxygen electrodes. Larger diameter, thicker seals were made using methods from internally funded programs. Over one hundred reversible cells have been fabricated for this project thus far.

Integrated System Demonstration. An important objective of this phase is to demonstrate a complete working system containing a sizeable reversible stack because stack performance is strongly dependent upon the system variables.

Figure 1 shows the test system for stacks up to 50 cells. This system has been designed, built, commissioned, and operated with stacks up to 19 cells thus far. The cylindrical assembly includes the reversible stack, heat exchanger, fuel processing, air blower, startup heater, electrical connections, thermocouples, stack mounting assembly, and insulation. The rectangular enclosure contains test instrumentation, active electrical load circuit, detachable startup systems, and fuel control valve.

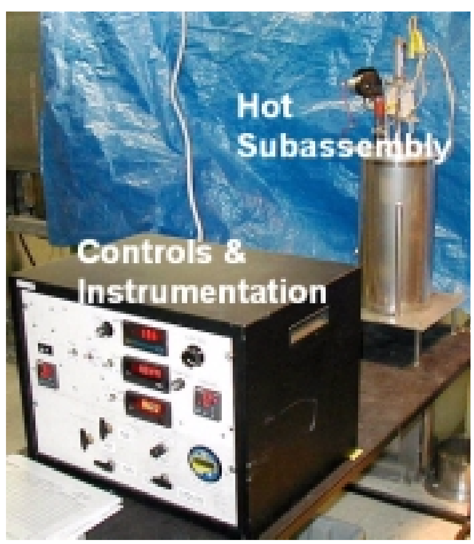

Figure 1. Laboratory System Assembly

The best performance parameters measured to date on this system (with 9- to 19-cell stacks) are shown in Table 1 (seal orifice coefficient was estimated from open-circuit voltage: up to 1.12 V/cell).

Table 1. Best Performance Parameters for Reversible Stack System

Parameter	Units	Goal	Measured
Area specific resistance	Ohm-cm^2	< 1.00	0.96
Power degradation rate	per 1000 hrs	< 5%	< 2%
Seal orifice coefficient		< 10^{-5}	< 10^{-6}

Cost and Engineering Studies. The installed and operating costs of systems producing hydrogen for vehicles are critical. The ultimate widespread use of hydrogen vehicles will only occur if and when affordable hydrogen is readily available. Vehicular hydrogen must also be very pure and available at pressures up to about 80 MPa (11,600 psi) for refilling vehicle tanks rated at about 69 MPa (10,000 psi). Hydrogen production processes that result in the lowest delivered-to-vehicle costs are expected to eventually dominate the market.

TMI believes it may have a candidate technology for achieving very low delivered hydrogen costs. Configured as shown in Figure 2 and sited at vehicle refueling sites, TMI reversible stack and system technology would be employed for both fuel cell system modules (using natural gas or other carbonaceous fuel) and the reversible electrolyzer subsystem, capable of producing 80 MPa pure hydrogen for vehicles as well as performing energy storage (battery replacement). The calculated projected energy conversion efficiencies in fuel cell, electrolyzer, and energy storage modes, as well as the overall natural gas to high-pressure pure hydrogen mode, can be shown to be superior to known competing technologies.

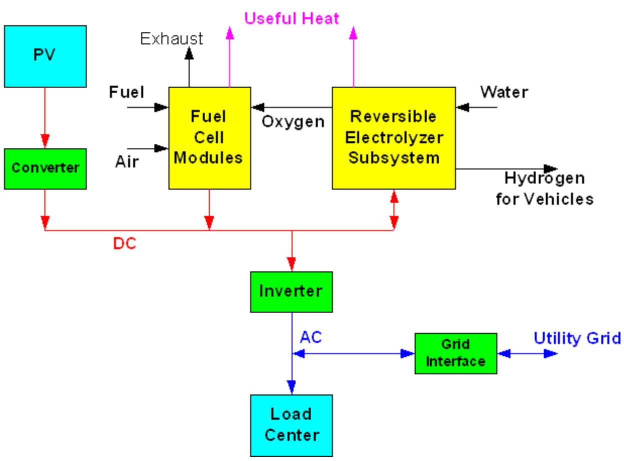

Figure 2. System Concept

TMI cost studies to date show that the lowest vehicle-delivered costs (below $1.50 per kg hydrogen) are projected for residential systems which can co-produce vehicle hydrogen, AC power, and hot water.

Conclusions

- Improved reversible solid oxide stack performance has been achieved.
- Sizeable reversible solid oxide fuel cell/electrolyzer stacks and a complete test system have been demonstrated.
- Projected cost (at the refueling nozzle) of vehicle hydrogen from natural gas has been computed to be as low as $1.50 per kg hydrogen.
- Projected solid oxide electrolyzer efficiencies near 95% have been achieved.
- Highly versatile system concepts using reversible electrolyzers and fuel cells have been proposed.

FY 2003 Publications/Presentations

1. "Low Cost, High Efficiency Reversible Fuel Cell Systems", R. C. Ruhl, DOE Hydrogen and Fuel Cells Annual Merit Review, Berkeley CA, May 20, 2003