V.A.1 Fuel Cell Systems Analysis

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Objectives

- Develop a validated model for automotive fuel cell systems and periodically update it to assess the status of technology.
- Conduct studies to improve performance and packaging, to reduce cost, and to identify key research and development (R&D) issues.
- Compare and assess alternative configurations and systems for transportation and stationary applications.
- Support DOE/FreedomCAR automotive fuel cell development efforts.

Technical Barriers

This project addresses the following technical barriers from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research, Development and Demonstration Plan:

(B   Cost
(C) Performance
(E) System Thermal and Water Management
(F) Air Management
(G) Start-up and Shut-down Time and Energy/Transient Operation

Technical Targets

This project is conducting system level analyses to address the following DOE 2010 technical targets for automotive fuel cell power systems operating on direct hydrogen:

- Energy efficiency: 50%-60% (55%-65% for stack) at 100%-25% of rated power
- Power density: 650 W/L for system, 2,000 W/L for stack
- Specific power: 650 W/kg for system, 2,000 W/kg for stack
- Transient response: 1 s from 10% to 90% of rated power
- Start-up time: 30 s from -20°C and 15 s from $+20^{\circ}$C ambient temperature
- Precious metal loading: 0.3 g/kW

Accomplishments

- Formulated correlations for water uptake in, and ionic conductivity of, 3M modified perfluorinated sulfonic acid (PFSA) membrane.
- Modified and validated the stack model for nanostructured thin film (NSTF) catalyst structures.
- Developed optimum operating maps by integrating the performance of the nanostructured thin film catalyst (NSTFC) stack, compressor expander module (CEM), and humidification devices.
- Analyzed heat rejection at elevated stack temperatures.
- Supplied performance and component data to TIAX and assisted in their manufacturing cost study.
- Developed and validated models for effects of fuel impurities (N_2, CO, CO_2, H_2S, and NH_3) on the performance of PEFC stacks.
- Analyzed effects of anode gas recycle on performance of polymer electrolyte fuel cell (PEFC) stacks.
- Constructed preliminary maps for stack voltage and efficiency degradation due to fuel impurities.

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Introduction

While different developers are addressing improvements in individual components and subsystems in automotive fuel cell propulsion systems (i.e., cells, stacks, fuel processors, balance-of-plant components), we are using modeling and analysis to address issues of thermal and water management, design-point and part-load operation, and component-, system-, and vehicle-level efficiencies and fuel economies. Such analyses are essential for effective system integration.

Approach

Two sets of models are being developed. The GCtool software is a stand-alone code with capabilities for design, off-design, steady-state, transient, and constrained optimization analyses of fuel cell (FC) systems. A companion code, GCtool-ENG, has an alternative set of models with a built-in procedure for translation to the MATLAB/SIMULINK platform commonly used in vehicle simulation codes such as PSAT.

Results

Our analysis of a pressurized PEFC system with finely dispersed Pt on high surface area carbon support showed that meeting the target of 50% system efficiency at rated power requires the stack to operate at 0.7 V/cell or higher, and results in stack specific power and power density being less than the targets of 2,000 W/kg and 2,000 W/L. The Pt loading exceeds 1 g-Pt/kW even if the efficiency target is relaxed to 46% (2005 fuel cell system [FCS]). Durability of the dispersed Pt catalyst and the PFSA membrane under dynamic conditions is a concern, as well. Also, heat rejection from the PEFC stack operating at 80°C is problematic.

In order to overcome these limitations we considered an alternative membrane electrode assembly (MEA) design. We chose 3M's modified PFSA membrane that has shown enhanced durability at low relative humidities. We also selected 3M's nanostructured thin film ternary-Pt catalyst, supported on organic whiskers, for low Pt loading, diminished electrochemically active surface area (ECSA) loss with potential cycling, and stability at high potentials [1]. We developed and validated correlations for the 3M membrane that describe water uptake as a function of temperature and relative humidity, and ionic conductivity as a function of water uptake and temperature.

We modified the stack model in GCtool for the NSTF catalyst structure. We analyzed the experimental data on NSTFC mass activity and specific activity and derived correlations for the ORR (oxygen reduction reaction) exchange current density as a function of the ECSA and Pt loading [1]. We formulated a model for water transport in the modified PFSA membrane and validated it against the measured high-frequency resistance [2]. An empirical model for water flooding of the NSTF catalyst was formulated consistent with the optimum dew point temperature observed at different operating conditions [2]. The NSTFC stack model was validated with the measured polarization curves at different pressures, temperatures and dew points [2].

A method was developed to determine the optimum operating conditions by integrating the NSTFC stack with the compressor expander module and an enthalpy

wheel humidifier for the cathode air feed, and a membrane humidifier for the anode hydrogen feed. Figure 1 indicates that because of the increase in cathode relative humidity (RH) with the decrease in current density (i.e., mass flow rate), the cathode stoichiometry must be raised in order to prevent flooding of the thin catalyst layers at part load conditions. At the optimum operating conditions, the spent gases at the stack outlet are just saturated, although liquid water does form in the catalyst layers. The higher operating temperature (90°C vs. 80°C with Pt/C) and lower inlet RH (50% vs. 60% for Pt/C) imply that the NSTFC stack runs much hotter and drier than the stack with the dispersed catalyst, and the problems of water management in the gas diffusion layers and the flow fields are considerably simplified. Under normal operating conditions, there is no liquid water in the cathode flow fields but the thin NSTF catalyst layers are prone to flooding if the inlet RH and cathode stoichiometry are not properly controlled.

In fuel cell vehicles, heat rejection is generally most challenging when driving on a 6.5% grade at 55 mph. We have looked at the possibility of making the radiator more compact by allowing the stack temperature to rise during such rather infrequent driving conditions where heat rejection is a problem. Figure 2a indicates that the cathode stoichiometry must be reduced from the optimum value shown in Figure 1 if the stack
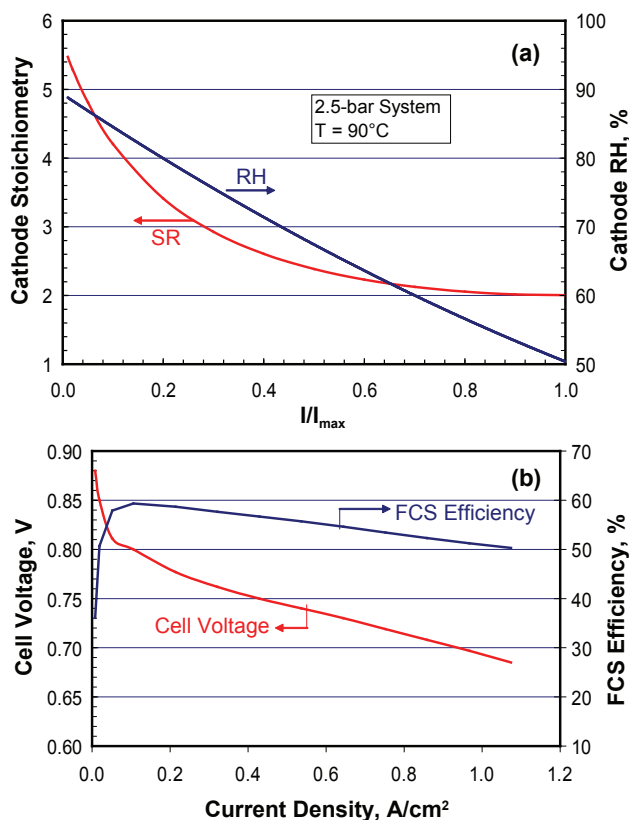

FIGURE 1. Optimized System Operating Conditions and Performance

temperature is allowed to rise, otherwise the membrane dries out, the stack efficiency decreases, and more waste heat has to be rejected. Figure 2b shows the effect of stack temperature on the radiator depth and frontal area needed to reject the waste heat produced in the fuel cell system. The results are for a radiator with 25-louver fins/inch and a 500-W blower that first cools the low-temperature radiator and an air conditioner (A/C) condenser. Figure 2b indicates that, compared to an internal combustion engine (ICE) for the same vehicle platform, a 20-30% larger frontal area is needed if the radiator depth is 25 mm and the stack temperature is allowed to rise to 92–98°C while driving on grade. Although the FCS radiator is larger than its ICE counterpart, it is significantly more compact than the radiator needed for the 2005-FCS [3] for at least three reasons: the higher peak coolant temperature (87–93°C vs. 75°C) means that a 33–50% larger temperature difference driving force is available for rejecting heat to the ambient air at 40°C; improved catalyst durability and use of a thinner membrane (30 μm vs. 50 μm) allow the stack to operate at a higher cell voltage (684 mV vs. 650 mV) with lower Pt loading; and a smaller amount of waste heat is generated because the stack is more efficient at the higher cell voltage (54.7% vs. 51% stack efficiency at rated power). The latent heat load

is negligible, whereas a significant fraction of water is formed as a liquid if the stack is operated at 80°C.

Table 1 summarizes the important results from our analysis of an 80-kW (net) pressurized PEFC with NSTF catalyst and the preliminary projection of the high-volume manufacturing cost by TIAX [4]. The projection is based on a bottom-up, activities-based costing method for the stack components; it does not include the cost of stack conditioning and the OEM's markups. Table 1 indicates that an NSTFC-based PEFC stack has the potential of meeting the 2010 DOE cost target of $25/kW. Also, the stack technology has reached a level of maturity that attention can be turned to the balance-of-plant (BOP) components. The data in Table 1 suggest that the cost of BOP components must be reduced by nearly one-half to meet the system cost target of $45/kW. Given below are some ways of accomplishing this.

- A bottom-up cost study is needed to determine whether the CEM modules can be assembled for less than $400 at high volume manufacturing. The $1,080 estimated cost for the CEM unit is based on consensus and extrapolation of experience with non-automotive applications.
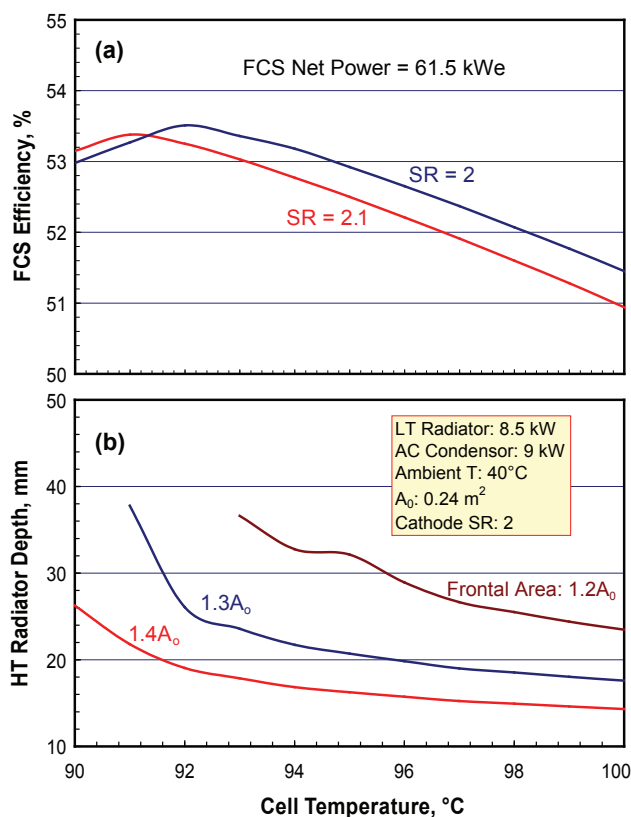

FIGURE 2. Effect of Stack Temperature and Cathode Stoichiometry on Heat Rejection from the Fuel Cell System

TABLE 1. Summary of System Analysis Results

Characteristic	2005 Status	2007 Status	2010 Target
System Cost, $/kWe	108	67	45
System Efficiency @ 25% Rated Power, %	57	60	60
System Efficiency @ Rated Power, %	46	50	50
System Specific Power, W/kg	710	790	650
System Power Density, W/L	590	640	650
Stack Cost, $/kWe	62	30	25
Stack Efficiency @ 25% Rated Power, %	59	62	65
Stack Efficiency @ Rated Power, %	52	55	55
Stack Specific Power, W/kg	1860	1900	2000
Stack Power Density, W/L	1730	2070	2000
MEA Cost, $/kWe	55	21	15
MEA Performance @ Rated Power, mW/cm^2	670	740	1280
MEA Degradation Over Lifetime, %	>90%	TBD	10
PGM Cost, $/kWe	44	16	8
PGM Content (peak), g/kWe	1.1	0.4	0.3
PGM Loading (both electrodes), mg/cm^2	0.75	0.3	0.3
Membrane Cost, $/m^2	24	16	40
Bipolar Plate Cost, $/kWe	3	3	5
CEM System Cost, $	1080	1080	400

- The anode-gas membrane humidifier (MH) is projected to cost about $110. One way of reducing the MH cost is to enhance mass transfer by using tubes with thinner walls. As the stack membranes continue to get thinner (18-μm composite membranes are available), it may be possible to eliminate the anode-gas humidifier altogether and rely only on water transfer from the cathode to the anode within the fuel cell stack.

- A bottom-up cost study may show that the centrifugal fans ($340 estimated cost of the fuel management system) can be less expensive or that the alternatives, such as vane recirculation pumps, are cheaper to build. The anode gas system may have to be simplified (e.g., by doing away with the ejector) to reduce cost, perhaps with a small decrease in efficiency.

- The heat rejection system is expensive ($270 estimated cost) and bulky. Alternative methods and layouts (e.g., side-by-side arrangement of high-temperature and low-temperature radiators) need to be considered.

We have initiated work on analyzing the effects of fuel impurities on the performance of PEFC stacks. The work to date indicates that, at low CO concentrations, the data on poisoning of Pt is consistent with CO adsorption on bridge sites, followed by electrochemical oxidation at high anode overpotentials. We are able to simulate much of the existing data on CO_2 poisoning by postulating reverse water-gas shift reaction between CO_2 and adsorbed H_2 to produce CO. We find that the literature data on H_2S poisoning can be explained by a reaction mechanism that includes at least three steps for reversible associative adsorption of H_2S, irreversible dissociation to form a Pt_2S-like species, and electrochemical oxidation of Pt_2S at high overpotentials. Our work also shows that the literature data on the effect of NH_3 can be modeled by considering its reversible uptake in the membrane and the ionomer in the catalyst. We have obtained results on the effect of anode gas recycle on the buildup of impurities. We have also determined preliminary impurity limits as functions of stack design (membrane thickness, Pt loading), operating conditions (P, T, RH, H_2 and O_2 utilization), and acceptable degradation in stack voltage and efficiency.

Conclusions and Future Directions

- Nanostructured thin film catalyst structures offer advantages of enhanced stability and durability. The NSTF ternary catalyst formulation has the potential to meet the DOE-2010 target of 0.3 g-Pt/kW.

- Our analyses indicate that it is possible to run the NSTFC stack hotter (90°C) and drier (50% inlet gas RH), resulting in a considerable simplification of the heat rejection and water management subsystems.

The oxygen stoichiometry must be carefully controlled, however, to avoid flooding the thin catalyst layer at part-load conditions.

- Further simplification of the BOP components is needed to meet the cost target of $45/kW at high volume manufacturing.

- In FY 2008, we will analyze system configurations suitable for use with high-temperature membranes capable of operating at 120°C without humidification.

- We will expand our work to include other fuel impurities and air impurities.

- We will include long-term degradation effects (durability issues) in systems analyses.

- We will continue to support DOE/FreedomCAR and fuel development efforts.

FY 2007 Publications/Presentations

1. R. K. Ahluwalia and X. Wang, "Rapid Self-Start of Polymer Electrolyte Fuel Cell Stacks from Subfreezing Temperatures," *J. Power Sources*, 162, 502–512, 2006.

2. R. K. Ahluwalia and X. Wang, "Buildup of Nitrogen in Direct Hydrogen Polymer Electrolyte Fuel Cell Stacks," *J. Power Sources*, in press, 2007.

3. R. K. Ahluwalia and X. Wang, "Effects of Impurities on Performance of Fuel Cells - Part 1: N_2," *Scientific Advances in Fuel Cell Systems Conference,* Turin, Italy, September 13–14, 2006.

4. R. K. Ahluwalia and X. Wang, "Effects of Impurities on Performance of Polymer Electrolyte Fuel Cells," *Fuel Cell Seminar,* Honolulu, HI, November 13–17, 2006.

5. D. Myers, X. Wang, D. Papadias, R. Ahluwalia, X. Wang, and R. Kumar "Impurity Effects on Membrane Electrode Assembly Components," *2nd MEA Manufacturing Symposium*, Dayton, OH, August 22–23, 2006.

6. R. K. Ahluwalia, X. Wang, and R. Kumar "Fuel Cell Systems for Transportation – A Status Report," *ASME International Mechanical Engineering Congress and Exposition*, Heat Transfer Division Banquet, Chicago, IL, November 7, 2006.

7. R. Kumar and R. K. Ahluwalia, "The Automotive Fuel Cell System: A Mechanical Engineering Perspective," *ASME Saginaw Valley Section Meeting*, Kettering University, Flint, MI, January 24, 2007.

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1. M. K. Debe, S. J. Hamrock, and R. T. Atanasoski, "Advanced MEAs for Enhanced Operating Conditions," FY 2006 Annual Progress Report, DOE Hydrogen Program, pp. 692–697, November 2006.

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3. R. K. Ahluwalia, X. Wang, and R. Kumar "Fuel Cell Systems for Transportation: Status and Trends," *EET-2007 European Ele-Drive Conference*, Brussels, Belgium, May 30-June 1, 2007.

4. S. Lasher, J. Sinha, Y. Yang and S. Sriramulu, "Direct Hydrogen PEMFC Manufacturing Cost Estimation for Automotive Applications," 2007 DOE Hydrogen Program Review, Arlington, VA, May 15-18, 2007.